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M. NAKAZAWA and M. TAKEBE
M. Miyahara, S. SASAKI, I. TOUCHI
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on Radiation Detectors and Their Uses

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AND THEIR USES

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Edited by

M. Miyajima and S. Sasaki
National Laboratory for High Energy Physics

T. Iguchi and M. Nakazawa
Tokyo University

M. Takebe
Tohoku University
National Laboratory for High Energy Physics, 1995

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FOREWARD

The third joint workshop, "Radiation detectors and their uses", was held on the January 17, 18 and 19, 1995 at the National Laboratory for High Energy Physics (KEK). This was held as the 9th meeting with the same title of the Radiation Safety Control Center, KEK, the 3rd meeting of "Radiation detectors in the next generation" of the Nuclear Engineering Research Laboratory, Tokyo University and the 1st meeting of "Ion Swarm" group of Japan. This also corresponded to the ten years anniversary of the meeting because we had missed a meeting in 1992. However, we had experienced a miserably large scale of earthquake at Hanshin district on the early morning of the first day of the workshop.

Although we had the earthquake, researchers more than 120 registered to the workshop and fifty one papers were presented from many fields while only two papers were canceled in this meeting. Among those papers, forty two were published in this proceeding. Two invited papers were presented by the visitors from Ukraine and Czechoslovakia.

At this workshop we had a special session entitled "Contact Points between Atomic and Molecular Physics in Gas, Liquid and Solid States and Radiation Detectors", in which three topics were presented from different fields and active discussions were made concerning the topics. Furthermore, we had a poster session for students in the undergraduate and graduate courses to be acquainted with researchers from various fields.

Mitsuhiro Miyajima
Shinichi Sasaki
Radiation Safety Control Center
National Laboratory for High Energy Physics

Tetsuo Iguchi
Masaharu Nakazawa
Department of Quantum Engineering and System Science
Toyko University

Masahiro Takebe
Department of Nuclear Engineering
Tohoku University
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PRESENT STATUS AND PERSPECTIVES OF DOUBLE BETA DECAY RESEARCH

V. I. Tretyak and Yu.G. Zdesenko
Institute for Nuclear Research, 252028 Kiev, Ukraine

1. INTRODUCTION

During last decade due to sharp intensification of double beta decay research the remarkable results have been obtained which level looked like unattainable during all history of these investigations beginning from its theoretical prediction by M. Goeppert-Mayer in 1935 [1] and the first experimental work in 1948 [2]: half-life limit of greater than $10^{24}$ y for $0
\nu$2$\beta$ decay of $^{76}$Ge [3-6]; the large scale experiments on $^{76}$Ge using enriched HP Ge detectors [5, 6]; half-life limits of greater than $10^{23}$ y for $0\nu2\beta$ decay of $^{136}$Xe [7] and $10^{22}$ y for $^{82}$Se [8], $^{100}$Mo [9], $^{116}$Cd [10] and $^{130}$Te [11]; development of the high energy resolution TeO$_2$ cryogenic detectors [11] and $^{116}$CdWO$_4$ crystal scintillators [10] for study of $2\nu$ decay of $^{130}$Te and $^{116}$Cd; observation in direct counting experiments of the $2\nu2\beta$ decay of $^{76}$Ge [4-6], $^{82}$Se [8], $^{100}$Mo [12-14], $^{116}$Cd [15, 16] and $^{150}$Nd [13, 17]; progress in the theoretical interpretation of double beta decay [18-20].

These achievements are related with particular role of $2\beta$ decay investigations among many approaches to study the neutrino properties. At present there are several indications of a physics beyond the Standard Model - the solar and atmospheric neutrinos deficit and mixed dark matter models which require the degenerate neutrino mass scenario with $m_\nu \sim 1 - 2$ eV. The extended SO(10) grand unified model with S4 horizontal symmetry was recently proposed [21] which leads naturally to such a scenario. The decisive test of this model is observation of neutrinoless $2\beta$ decay.

Therefore one can conclude that double beta decay research continue to be interesting and important field of investigations because it can provide an unique and a very sensitive probe of a new physics beyond the Standard Model.

In two next sections the present theoretical situation and experimental status of $2\beta$ decay investigations will be discussed briefly. The goal of present article is not to cover the whole material contained in capital and excellent reviews [18-20, 22-25].
Instead, main attention will be paid only to recent developments in investigations of double β decay of atomic nuclei.

2. THEORETICAL SITUATION

There are two main modes in $2\beta^-$ decay: two-neutrino (2ν) and neutrinoless (0ν)

\[
(A, Z) \rightarrow (A, Z+2) + 2e^- + 2\nu_e \quad \text{(1)}
\]

\[
(A, Z) \rightarrow (A, Z+2) + 2e^- . \quad \text{(2)}
\]

Analogous equations can be written also for $2\beta^+$ decay as well as for electron capture with $\beta^+$ decay (e$^+$) or for double electron capture (2e).

The two-neutrino decay (1) is a conventional (but very rare) decay described in the SM as a second order weak transition between two even-even nuclei. The second process (2) is forbidden in the SM and requires a violation of lepton number conservation and a non-vanishing Majorana neutrino mass or/and an admixture of a right-handed weak interaction to the left-handed one. In gauge theories, which suggest that neutrinos are Majorana particles, there are three ways to produce a Majorana mass: explicit B-L breaking and spontaneous breaking of a local or global B-L symmetry. In last case $2\beta$ decay could occur with emission of light pseudo-scalar Goldstone boson called Majoron:

\[
(A, Z) \rightarrow (A, Z+2) + 2e^- + M. \quad \text{(3)}
\]

Different models for Majoron as extensions of the SM were proposed [26-29]. Recent LEP measurement of $Z^0$ width [30] ruled out possible triplet [26] and doublet [28, 29] Majoron but existence of singlet Majoron [27] or mixture of singlet and doublet /triplet Majorons is still possible.

Trustworthy restrictions for neutrino mass (right-handed admixtures and other parameters) from measured limits on 0ν2β decay rates are possible only in case of reliable calculations of nuclear matrix elements which enter the half-life formulae. Since the 0ν2β decay has not been observed up to now, it is impossible to check the theoretical 0ν2β rates directly and only way is to use the positive 2ν2β decay data to test the assumptions made in nuclear structure calculations.

The 2ν2β decay can be described in second order perturbation of weak interaction as successive Gamov-Teller (mainly) transitions via virtual intermediate $1^+$ states. The shell model and (in last decade) quasiparticle random phase approximation (QRPA) were used most intensively to calculate nuclear matrix elements for 0ν2β and 2ν2β decay. But full scale computations in the shell model were unattainable because the number of basic states in model space increased explosively for nuclei heavier than $^{48}\text{Ca}$
and various simplifications had to be introduced. From this point of view the QRPA is more attractive so far as the new vacuum is determined for quasiparticles and nuclear wave function can be described in terms of small number of quasiparticles' degrees of freedom. Calculations in the framework of QRPA revealed that the $2\nu 2\beta$ decay rates could be strongly suppressed by taking into account the ground state correlations in nuclei which are enhanced by the particle-particle interaction. Predicted $2\nu 2\beta$ decay rates are found to be very sensitive to the value of the strength of the particle-particle interaction $g_{pp}$, and calculated half-lives can be tuned to all experimental data in region of $g_{pp} \approx 1$ (see reviews [18, 19] and references therein). In order to remove or at least decrease these difficulties several attempts were undertaken. For instance, in [31] the QRPA calculations of $2\nu 2\beta$ nuclear matrix elements for $^{76}\text{Ge}$, $^{82}\text{Se}$ and $^{100}\text{Mo}$ were performed with effective Paris and Bonn potentials taking into account the self-consistent self-energy corrections to the single particle spectra. In this way the QRPA instability around $g_{pp} \approx 1$ was avoided and a reasonably good agreement of calculated matrix elements with experimental values was achieved. Recent paper [32] reports the results of the investigation of high order corrections upon the QRPA nuclear matrix elements for several $2\beta$ emitters under approach developed in [33]. It was found that HQRPA corrections display a weak dependence on a particle-particle strength $g_{pp}$ and become important in region $g_{pp} \approx 1$ where QRPA values vanish.

Tedious calculations of matrix elements in shell model and drastic sensitivity of $2\nu 2\beta$ decay rates on the value of $g_{pp}$ in QRPA stimulated the development of alternative approaches to compute the $2\nu 2\beta$ decay amplitude. In operator expansion method (OEM) (see [18, 34] and references therein) the intermediate $1^+$ energy spectrum is not explicitly used. In this way the dependence on $g_{pp}$ is highly reduced and $2\nu$ nuclear matrix elements become comparatively constant in physical region of $g_{pp}$. So the dependence on $g_{pp}$ in the QRPA is denoted as artificial [18]. Another approach eliminating the explicit summation over intermediate states is proposed [35] for exact evaluation of Green's function based on a Lanczos algorithm for inverting linear operators. So this method removes the need of Green's function approximation used in OEM. See also [18-20, 24, 36] for discussion and comparison between calculations of matrix elements in shell model, QRPA and OEM.

The $2\nu 2\beta$ decay to $2^+$ excited state of daughter nuclei is highly suppressed by few orders of magnitude as compared with transition to a ground state due to a few factors (available energy is smaller, transition's amplitude contains higher powers of the final leptons' momenta, leptonic phase space factor is antisymmetrical in two electrons and two neutrinos). However in recent work [37], based on QRPA formalism with effective
Bonn-OBEP two-body interaction, it was found that $2\nu 2\beta (0^+ - 2^+)$ matrix elements do not strongly depend on the value of $g_{\nu\nu}$. So definite predictions for $2\nu 2\beta$ decay to $2^+$ excited state of daughter nuclei are accessible in the QRPA.

As regards the $0\nu 2\beta$ decay, QRPA calculations of $0\nu$ rates are comparably insensitive to various types of ground state correlations (and to particle-particle correlations in particular). This is because in neutrinoless mode the transitions are possible not through intermediate states with $J^\pi = 1^+$ only but mainly through another multipoles $J^\pi$ whose corresponding matrix elements are much less affected by the change of $g_{\nu\nu}$. Therefore, most of the authors agree that comparably certain computation of $0\nu 2\beta$ decay rates are possible (see [18-20, 22-24, 31-36]).

### 3. EXPERIMENTAL STATUS

There are two different classes of the direct double $\beta$ decay experiments: a) with active source (source = detector); b) with passive source. Both of them could be divided into two kinds - with detector for measurement of the energy of the electrons (energy detector) and more complicated with tracking and energy detectors. In the first class of the experiments with energy detectors the required effect can be distinguished on the basis of only one property of the $2\beta$ decay - the distribution of the total energy of the electrons. In the experiments with passive source and energy detectors it is possible to use time coincidence between two electrons and to measure the single and sum energy distributions. Full information about all properties of $2\beta$ decay events can be obtained in the most complete class of the experiments - time coincidence, tracks and vertex of the electrons, energy and angular distributions.

Let us consider briefly some recent results of the direct counting $2\beta$ decay experiments. In order to obtain the restrictions on the neutrino mass in the same scale we will use the calculations [38] with the most extensive list of nuclei.

$^{76}\text{Ge}$. Since 6 years Max-Planck Institute (Heidelberg) in collaboration with Kurchatov Institute (Moscow) have organized and performed the most advanced experiment to study $^{76}\text{Ge}$ by using HP Ge semiconductor detectors enriched in $^{76}\text{Ge}$. They used 16.9 kg of enriched $^{76}\text{Ge}$ to produce HP Ge detectors for ultra low-background measurements. At first three HP Ge detectors (0.927, 2.758 and 2.6 kg) enriched in $^{76}\text{Ge}$ to 86% with good energy resolution (2.5 keV at 1.3 MeV) were running in the Gran Sasso Underground Laboratory (Italy). The background rate around 2 MeV.
was 0.25 counts/y/keV/kg. From 1133 kg-d of operation time the following half-life limit for the $0\nu 2\beta$ decay of the $^{76}$Ge has been obtained: $T_{1/2}^{0\nu} > 1.9 \times 10^{24}$ y (90% CL) [5]. This result corresponds to a neutrino mass less than 1.1 eV. Now the experiment is running with two more HP Ge enriched detectors (1.7 and ~ 2 kg). Therefore during the next 5 years one can expect a limit $T_{1/2}^{0\nu} \geq 10^{25}$ y, which corresponds to neutrino mass less than 0.4 eV. Also $2\nu 2\beta$ decay of $^{76}$Ge [4] has been confirmed with high statistics: $T_{1/2}^{2\nu} = (1.42 \pm 0.03 \pm 0.13) \times 10^{21}$ y [5].

There is another big international project (IGEX) to study $2\beta$ decay of $^{76}$Ge [6]. Three detectors, each composed of one kg of germanium enriched to 87.5% of $^{76}$Ge, were produced and tested so far. The limit $T_{1/2}^{0\nu} > 1 \times 10^{24}$ y (90% CL) was reached and $2\nu 2\beta$ decay of $^{76}$Ge [4] was proved [6]. The goal of IGEX project is to obtain five ~3 kg detectors which could provide a test of $0\nu 2\beta$ decay of $^{76}$Ge with sensitivity of greater than $10^{25}$ y for half-life limit $T_{1/2}^{0\nu}$ [6].

$^{82}$Se. The two neutrino $2\beta$ decay of $^{82}$Se has been measured in University of California by Irvine group with time projection chamber (TPC) placed in magnetic field of ~700 G [8]. The TPC records the electron tracks, and the energies and opening angle are determined from each track. The discovery of $2\nu 2\beta$ decay of $^{82}$Se with $T_{1/2}^{2\nu} = 1.08 \pm 0.26 \times 10^{20}$ y (68% CL) has been claimed after ~21000 h run and limit $T_{1/2}^{0\nu} > 2.7 \times 10^{22}$ y (68% CL) has been set for $0\nu 2\beta$ decay of $^{82}$Se [8].

$^{100}$Mo. Five groups have made the measurements with $^{100}$Mo. In 1982 INR (Kiev) have used the plastic scintillator-wafer stacks with sheets of the $^{100}$Mo [39]. The limit $T_{1/2}^{0\nu} > 2.2 \times 10^{21}$ y (90% CL) has been set [39]. Recently about the same technique (but with semiconductor-wafer stacks) was used by LBL+MHC+UNM+INEL collaboration [9]. The limit $T_{1/2}^{0\nu} > 4.4 \times 10^{22}$ y (68% CL) has been reached in this work. Three another groups used the apparatus with tracking and energy detectors and all groups have claimed the discovery of $2\nu 2\beta$ decay of the $^{100}$Mo: Osaka group result - $T_{1/2}^{2\nu} = 1.15 \times 10^{19}$ y [12]; Irvine group - $T_{1/2}^{2\nu} = 1.16 \times 10^{19}$ y [13]. Recently the Osaka and Irvine groups' results were confirmed by the NEMO collaboration with help of so called NEMO 2 apparatus with good tracking device (source square ~ 1 m²) and 2x64 plastic scintillators: $T_{1/2}^{2\nu} = (1.0 \pm 0.08 \pm 0.2) \times 10^{19}$ y [14]. The NEMO collaboration is going to build the NEMO 3 tracking detector scale up in comparison with previous one by factor of 20 in size (source square ~ 20 m²) and by factor of 1000 in sensitivity. With source's mass ~10 kg of $^{100}$Mo they plan to reach the limit $T_{1/2}^{0\nu} \sim 10^{25}$ y. It will be the biggest device for $2\beta$ decay research worldwide.
To study this nucleus $^{116}$CdWO$_4$ crystal scintillators enriched in $^{116}$Cd to 83% have been developed by Kiev group (Institute for Nuclear Research) [10, 40]. The experiments were performed in the Solotvina Underground Laboratory with three enriched crystal scintillators $^{116}$CdWO$_4$ (19, 14 and 13 cm$^3$) as well as with natural CdWO$_4$ (8, 9 and 57 cm$^3$) detectors. Subtracting the spectra of natural CdWO$_4$ scintillators from the spectra of enriched $^{116}$CdWO$_4$ detectors the positive effect has been seen which is in accordance with $2\nu 2\beta$ decay of $^{116}$Cd with half-life $T_{1/2}^{2\nu} = [2.7^{+0.5}_{-0.4}(\text{stat.})^{+0.9}_{-0.6}(\text{syst.})] \times 10^{19}$ y [16]. The background rate of the enriched crystals was equal to 0.57 counts/y/keV/kg in the energy interval 2.7 - 2.9 MeV where the resolution was about 7.5% at 2.6 MeV. After 5822 h run a lower limit $T_{1/2}^{\nu\nu} = 2.9 \times 10^{22}$ y (90% CL) has been set, which corresponds to neutrino mass less than 4.1 eV [10, 16]. In the near future it is possible to improve this limit to value of $10^{23}$ y ($m_\nu < 2.3$ eV). INR (Kiev) and MPI (Heidelberg) have recently proposed [16] a project of the big scale experiment with $^{116}$CdWO$_4$ crystal scintillators (~20 kg of the $^{116}$Cd) in which for $0\nu 2\beta$ decay of the $^{116}$Cd the limit $T_{1/2}^{0\nu} > 10^{25}$ y can be reached ($m_\nu < 0.3$ eV). In the second phase of this experiment $^{116}$CdWO$_4$ crystals could be used as the cryogenic thermal detectors with high energy resolution (~5 keV at the energy 1 MeV has recently been obtained in the first test of CdWO$_4$ crystal with mass of 58 g [41]).

$^{130}$Te. Milano group used the crystal TeO$_2$ (334 g) as a cryogenic thermal detector to search for $2\beta$ decay of $^{130}$Te [11]. Energy resolution of the crystal is about 10 keV at 1.3 MeV. After 9234 h of operation time in the Gran Sasso Underground Laboratory the background rate inside a 16 keV window at 2.53 MeV was about 3 counts/y/keV/kg. The limit $T_{1/2}^{0\nu} > 1.8 \times 10^{22}$ y (90% CL) or $2.8 \times 10^{22}$ y (68% CL) has been established for $0\nu 2\beta$ decay of the $^{130}$Te ($m_\nu < 5.2$ eV) [11]. In the near future with four TeO$_2$ crystals (full mass = 1.5 kg) the limit $T_{1/2}^{0\nu} > 10^{23}$ y can be set ($m_\nu < 2.2$ eV) and with enriched $^{130}$TeO$_2$ crystal level of $10^{24}$ y can be reached during several years ($m_\nu < 0.7$ eV). It should be mentioned that development of this technique is an important step in $2\beta$ decay research because it can be applied to different crystals which contain the possible $2\beta$ active nuclei.

$^{136}$Xe. Caltech (Pasadena)+Institut de Physique (Neuchâtel)+Paul Scherrer Institute (Villigen) collaboration [7] have constructed the time projection chamber with an active volume of 207 l, which has operated with 5 atm of xenon enriched in $^{136}$Xe to 62.5%. The energy resolution of the TPC is 6.6% at 1.6 MeV. The track reconstruction capability of the TPC provides a powerful means of background rejection. As a result a background rate around 2.48 MeV (within FWHM energy interval) is 0.01
counts/y/keV/kg. From 6218 h of data taking in the Gotthard Underground Laboratory the limit $T_{1/2}^{0\nu} > 3.4 \times 10^{23}$ y (90% CL) has been set for $0\nu 2\beta$ decay of the $^{136}$Xe [7]. Upper limit of the neutrino mass is equal 2.5 eV. At the near future and after some improvements of the apparatus this collaboration plan to reach the limit $T_{1/2}^{0\nu} > 10^{24}$ y ($m_\nu < 1.5$ eV).

One can note that equipment developed in this work joints the advantages of both classes of direct $2\beta$ decay experiments: the reasonable high efficiency and tracking information.

As the natural next step in this direction - large-volume liquid Xe TPC [42] and self-triggered drift chamber filled by liquid enriched $^{136}$Xe [43] - have been proposed allowing to reach the sensitivity's level of $\sim 10^{25}$ y ($m_\nu < 0.5$ eV). In order to overcome even this level a new approach was viewed which used the coincident detection of $^{136}$Ba$^{2+}$ ions (result of $2\beta$ decay of $^{136}$Xe on the atomic level) and $0\nu 2\beta$ signal of $^{136}$Xe with energy 2.5 MeV in a drift chamber or TPC filled by liquid Xe [44, 45]. Such apparatus with background-free 1000 kg of $^{136}$Xe can investigate the Majorana neutrino mass in 0.01 eV region [45]. The first step in this direction has been performed by KEK group (Tsukuba, Japan) [46]. They have developed the collector of positive ions in liquid xenon (PIC) and time-of-flight mass spectrometer (TOFMS) of daughter Ba ions using techniques of Resonant Ionization Spectroscopy. The positive ions collector was operated for 847 h with natural liquid xenon of 3 liters (about $3.6 \times 10^{24}$ nuclei of $^{136}$Xe). No $^{136}$Ba ions were detected by TOFMS. Take in account the full efficiency (less than $10^{-4}$) it gives the sensitivity concerning $T_{1/2}$ of $^{136}$Xe at the level of $10^{19}$ y. Now the KEK group plans to increase efficiency (and consequently sensitivity) by two order of magnitude [46].

$^{150}$Nd. In early experiment of Moscow group the plastic scintillators with sheets of $^{150}$Nd were used and the limit $T_{1/2}^{0\nu} > 1.7 \times 10^{21}$ y (95% CL) was established for $0\nu 2\beta$ decay of $^{150}$Nd ($m_\nu < 4.5$ eV) [47]. Irvine group using 6342 h TPC run with 11.2 g of $^{150}$Nd has set the limit $T_{1/2}^{0\nu} > 2.1 \times 10^{21}$ y (90% CL), which corresponds to neutrino mass $m_\nu < 4.0$ eV [48]. Due to the best sensitivity of $^{150}$Nd to neutrino mass (comparing with another candidate nuclei) the new and reasonable scale up TPC with $\sim 400$ g of $^{150}$Nd can provide the investigation of the Majorana neutrino mass in 0.5 eV domain [48]. Two neutrino $2\beta$ decay of $^{150}$Nd was recently observed with $T_{1/2}^{2\nu} = 1.7 \times 10^{19}$ y [17] (the preliminary result of Irvine group is $T_{1/2}^{2\nu} = 1.0 \times 10^{19}$ y [13]).

The results of the recent and most advanced $2\beta$ decay experiments are summarized in the table, where theoretical estimations [30] are presented also. It is clear that research of $^{76}$Ge and $^{136}$Xe give us the most stringent restrictions on the Majorana neutrino mass (1-2 eV) and in other five experiments the obtained limits are ranged in interval 4-6 eV.
Table. Limits on neutrino mass from the most advanced experiments

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Experiment</th>
<th>Theory [38]</th>
<th>Limit on neutrino mass mν, eV</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>T1/2y, y</td>
<td>68% CL</td>
</tr>
<tr>
<td>76Ge</td>
<td>1.9\cdot10^{24} (90% CL) [5]</td>
<td>2.33\cdot10^{24}</td>
<td>1.1</td>
</tr>
<tr>
<td></td>
<td>3.2\cdot10^{24} (68% CL) [5]</td>
<td>0.9</td>
<td></td>
</tr>
<tr>
<td>82Se</td>
<td>2.7\cdot10^{22} (68% CL) [8]</td>
<td>6.03\cdot10^{23}</td>
<td>4.7</td>
</tr>
<tr>
<td>100Mo</td>
<td>4.4\cdot10^{22} (68% CL) [9]</td>
<td>1.27\cdot10^{24}</td>
<td>5.4</td>
</tr>
<tr>
<td>116Cd</td>
<td>2.9\cdot10^{22} (90% CL) [16]</td>
<td>4.87\cdot10^{23}</td>
<td>4.1</td>
</tr>
<tr>
<td></td>
<td>5.4\cdot10^{22} (68% CL) [16]</td>
<td>3.0</td>
<td></td>
</tr>
<tr>
<td>130Te</td>
<td>1.8\cdot10^{22} (90% CL) [11]</td>
<td>4.89\cdot10^{23}</td>
<td>5.2</td>
</tr>
<tr>
<td></td>
<td>2.8\cdot10^{22} (68% CL) [11]</td>
<td>4.2</td>
<td></td>
</tr>
<tr>
<td>136Xe</td>
<td>3.4\cdot10^{23} (90% CL) [7]</td>
<td>2.21\cdot10^{24}</td>
<td>2.5</td>
</tr>
<tr>
<td></td>
<td>6.4\cdot10^{23} (68% CL) [7]</td>
<td>1.9</td>
<td></td>
</tr>
<tr>
<td>150Nd</td>
<td>2.1\cdot10^{21} (90% CL) [48]</td>
<td>3.37\cdot10^{22}</td>
<td>-</td>
</tr>
</tbody>
</table>

### 4. SUMMARY

Up to now only 18 nuclei from all 35 potential 2β⁻ active candidates were studied in direct counting experiments. The highest limits of half-lives for 0ν2β decay have been set for 76Ge - 2\cdot10^{24} y, 136Xe - 4\cdot10^{23} y, 82Se, 100Mo, 116Cd, 130Te - ~10^{22} y, 48Ca, 150Nd - ~10^{21} y. The 0ν2β results for other nuclei are placed in the region of 10^{16} - 10^{19} y. The two-neutrino 2β decay of 76Ge (T_{1/2} ~10^{21} y), 82Se (~10^{20} y), 100Mo, 116Cd and 150Nd (~10^{19} y) were observed surely.

The most sensitive results for 0ν2β decay (76Ge, 136Xe, 116Cd and 130Te) as well as for 2ν decay mode (76Ge) were obtained by using the technique with active
source. The devices with passive source and tracking detectors are more convenient for
detail investigation of 2ν2P decay (the present day level of sensitivity is 10^{19} - 10^{20} y).

There are several projects to increase the sensitivity of 0ν2β decay experiments to
the level of 10^{24}-10^{25} years. The most promising proposals among them are the
following: the development of large volume liquid Xe TPC [42] and self-triggered drift
chamber filled by liquid enriched ^{136}Xe [43]; the building of the biggest tracking detector
for 2β investigations of ^{100}Mo by NEMO collaboration [14]; using in the experiment of
Milano group the enriched ^{130}TeO_2 crystals with mass about 1 kg [11]; the Kiev-
Heidelberg project with enriched ^{116}CdWO_4 crystal scintillators [16]. However for so
sensitive experiments the serious problem exists connected with background occasioned
by the two neutrino 2β decay (M.K. Moe paid attention to this problem for the first time
[45]). It should be stressed that these background events in principle could not be
distinguished from 0ν2β signal because all features are the similar: the same particles
simultaneously emitted from one point, the same energy region and the right angular
correlation. Only difference in the electrons' sum energy distributions can be used. In this
case the energy resolution of device will play a decisive role, like for calorimeters which
measure only the released energy. Simple calculation shows that reaching the sensitivity
of greater than 10^{24} y with poor energy resolution (worse than 10% at the energy 1 MeV)
looks like very questionable especially if we take into account the problems of low
statistics (for instance, in 10 kg of ^{100}Mo only 42 (4.2) 0ν2β decays will happened
during one year if T^0_{1/2} = 10^{24} (10^{25}) y).

The usual question is "why we need to perform the research with several
nuclides, but not with ^{76}Ge or ^{136}Xe only?" There are many reasons to spread the efforts
to more candidate nuclei. First of all, a neutrinoless 2β decay is unobserved phenomenon
still now. Despite the big progress in understanding and interpretation of this process,
new development in calculations of the nuclear matrix elements and half-lives, the present
theoretical situation does not look completely definite. Thus it is rather difficult to suppose
that without any positive experimental results on 0ν2β decay and on the base of theory
anybody can point out the best nucleus for discovery of neutrinoless 2β decay. On the
other hand, a success in these research could be achieved on the border of modern
technology as an alloy of science, experimental state of art and even luck, hence nobody
can know a priori where the highest sensitivity will be reached. The new and sometimes
unexpected advancements in experimental technique could give the advantages to
particular 2β decay candidates. The good example is CdWO_4 crystal scintillators which
were developed and used for study of 2β decay of ^{116}Cd [10, 16]. Recent discovery that
CdWO₄ crystals can work as the cryogenic thermal detectors with high energy resolution (~5 keV) [41] gives the excellent chance to increase the sensitivity of experiment with $^{116}$Cd. So the present experimental and theoretical situation in $2\beta$ decay problem makes necessary to extend the number of nuclides studied with a sensitivity comparable or better than for $^{76}$Ge. If neutrinoless signal will be discovered in $^{76}$Ge (or $^{136}$Xe), existence of such a peak has to be confirmed on other nuclides and also by another technique.

Therefore we can conclude that in course of about 60 years $2\beta$ decay investigations have been and continue to be popular due to their fundamental importance. Last experiments with enriched $^{76}$Ge and $^{136}$Xe, the development of $^{116}$CdWO₄ crystals and TeO₂ cryogenic detectors, discovery of two-neutrino $2\beta$ decay of $^{76}$Ge, $^{82}$Se, $^{100}$Mo, $^{116}$Cd and $^{150}$Nd give us the new time scale for research in this important field of modern physics. In not so far future we can expect the new distinguished results which will provide a test of current theory as well as an unique and a very sensitive probe of new physics beyond the Standard Model.

Figure from [16] shows the actual status of the most advanced experiments (solid lines), the expected improvements of current results (open bars) and the most realistic projects up to 1999 y.

Figure. Actual status and expected improvements of current results in $2\beta$ investigations
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MEASUREMENT OF THE POSITIVE IONS DRIFTING IN LIQUID OR GASEOUS XENON

Eido Shibamura
Saitama College of Health, Urawa, Saitama, 338 JAPAN

Shinichi Sasaki, Hiroko Tawara and Mitsuhiro Miyajima
KEK, Oho-1, Tsukuba, Ibaraki, 305 JAPAN

1. INTRODUCTION

Several tests on injection and transport of positive ions were made in liquid and gaseous Xe, which are parts of our research program of double beta decay of \(^{136}\)Xe\(^{0}\). In measurements a event of double beta decay is identified by detecting a couple of electrons emitted at the decay and the daughter isotope \(^{136}\)Ba with resonance ionization spectrometry, where we believe that we are almost free from the backgrounds in the measurements. The injection efficiency and drift velocity of positive ions are key parameters in the program. We initiated an experiment of injection and transport of \(^{208}\)Tl ions in liquid and gaseous Xe. At present, we obtained reliable results of the injection and transportation. However the drift velocity of the positive ions was not determined yet because of the statistical error in \(^{208}\)Tl decay counts. We here report the methods of the experiments and the results obtained so far.

2. PRINCIPLE OF MEASUREMENTS

\(^{212}\)Pb is a radio-active isotope decaying in the following decay mode, where the branching ratio \(f_B\) to \(^{208}\)Tl from \(^{212}\)Bi is about 36 %.

\[
\begin{align*}
^{212}\text{Pb} & \rightarrow^{212}\text{Bi} & \rightarrow^{208}\text{Tl} & \rightarrow^{208}\text{Pb} \text{ [stable]} \\
\alpha & \tau = 3.05 \text{ m} & \beta \\
\tau = 10.6 \text{ h} & m = 60.6 \text{ m} & \rightarrow^{212}\text{Po} & \rightarrow\alpha & \beta
\end{align*}
\]

A source of \(^{212}\)Pb was deposited on a cathode surface with thorium active
deposit and set with a distance $D$ from a collector. Ions of $^{208}\text{Tl}$ are partly emitted into liquid or gaseous xenon by the recoil of alpha particle. By applying an electric field $E$ between the cathode and the collector at $t = 0$, the ions escaping recombination will start drift towards the collector. During the drift time $T_b$ of the $^{208}\text{Tl}$ ions between the cathode and the collector, a part of $^{208}\text{Tl}$ will decay into $^{208}\text{Pb}$ and the rest of $^{208}\text{Tl}$ will reach the collector. Hence, $^{208}\text{Tl}$ ions will be accumulated on the collector from time $T_b$. We can know $T_b$ and the amount of Tl ions arriving at the collector by detecting the beta rays from $^{208}\text{Tl}$. The number of $^{208}\text{Tl}$ ions $m(t)$ on the collector at $t < T_b$ is zero, and that at $t > T_b$ is given by the following equation,

$$m(t) = \frac{f_s f_s \lambda_s \lambda_T \nu_0}{(\lambda_s - \lambda_T)(\lambda_T - \lambda_s)} \left( e^{-\lambda_T t} - e^{-\lambda_T T_b} - \lambda_T T_b \right), \quad (1)$$

where $n_0$ is the number of $^{212}\text{Pb}$ at $t = 0$, $\lambda_T$ is the probability for $^{208}\text{Tl}$ ions to be emitted into xenon and escape from recombination at $E$, and $\lambda_T$, $\lambda_s$ and $\lambda_T$ are the decay constants of $^{212}\text{Pb}$, $^{212}\text{Bi}$ and $^{208}\text{Tl}$, respectively.

3. EXPERIMENTAL

![Figure 1](image_url)  
A cross sectional side view of a typical set up of the ionization chamber.
Figure 1 shows a schematic cross section of an ionization chamber used in the experiment in the case of $D = 10$ cm. Field-shaping electrodes $G_1 - G_9$ are placed in the gap between the cathode and the collector. A source of $^{212}$Pb was deposited on the cathode prior to the experiments. The collector is made of a thin aluminum foil, under which Si PIN photo-diode (SSD) is fixed to detect the beta rays emitted from $^{208}$Tl. The chamber was filled with gaseous or liquid xenon. The distance $D$ between the cathode and the collector was set to 2.0, 5.0 or 10.0 cm.

![Graph showing pulse height spectrum of beta rays obtained by the photo diode in the case of gaseous xenon at $E = 0$ V/cm.]

Figure 2 A typical pulse height spectrum of beta rays obtained by the photo diode in the case of gaseous xenon at $E = 0$ V/cm.

Figure 2 shows the pulse height spectrum of the beta rays obtained by the diode at a zero electric field in the case of gaseous xenon, where beta rays from $^{212}$Pb on the cathode are detected since the range of beta rays may be larger than the gap $D$ and the aluminum foil of 25 μm. The beta rays were also detected even in liquid xenon. These were probably emitted from the neutral $^{208}$Tl diffusing near the collector in liquid. Figure 3 shows the counts $n(t)$ of beta rays per 10 seconds whose pulse heights are larger than the threshold $P_T$ shown in Fig. 2. The electric field of $E = 400$ V/cm was applied at
the time \( t \) between \( 0 < t < T_h = 1200 \) sec, where the increase in count rate is clearly shown. For \( t > T_h \), the count rate was decreasing as expected.

\[ c(t) = A_1 \left( e^{-\lambda_1 t} + A_2 n_1(t) / f \right), \]

where \( A_1 = k_1 n_0, A_2 = \lambda_2 k_2, f = k f, k_1 \) and \( k_2 \) are parameters including the efficiency of detecting beta rays from the cathode, and from the \(^{208}\text{Tl}\) on the collector, respectively. The first term expresses background counts and the second term the beta rays from \(^{208}\text{Tl}\) on the collector. In eq. (2) all unknown factors are included in \( T_h, A_1 \) and \( A_2 \). The parameter \( A_2 \) is proportional to the probability for \(^{208}\text{Tl}\) ions to escape from recombination.

---

Figure 3  Count rates as a function of time \( t \)
We determined the parameters $A_3$, $A_2$ and $T_b$ by fitting eq. (2) to the experimental count numbers as shown by the solid curve in Fig. 3. Table 1 shows the parameters obtained for gaseous xenon in the case of $D=5$ cm. The decrease in $A_3$ with increase in $E$ is apparent and is due to the decrease in $^{208}\text{Pb}$ with $r = 10.6$ h. In gaseous xenon $A_2$ increases with $E$ below $E = 500$ V/cm and it seems not to increase appreciably over $E = 600$ V/cm, which indicates that most of $^{208}\text{Tl}$ ions can be collected over this electric field in gaseous xenon. Such a saturation of ion collection is not observed in liquid xenon although the increase in the count rate with increase in $E$ is clear. The drift time $T_b$ of ions is not determined yet with a reasonable error both in liquid and gaseous xenon.

### Table 1 Fitting parameters

<table>
<thead>
<tr>
<th>$E$ (V/cm)</th>
<th>$T_b$ (sec)</th>
<th>$A_2$</th>
<th>$A_3$</th>
</tr>
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<tbody>
<tr>
<td>100</td>
<td>- 4.121</td>
<td>1.646 $10^6$</td>
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<td>200</td>
<td>0.072</td>
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<td>900</td>
<td>12.97</td>
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</tr>
</tbody>
</table>

5. SUMMARY

We detected positive ions of $^{208}\text{Tl}$ emitted into liquid and gaseous xenon after drifting over the distance between the cathode and the collector at an electric field. The drift velocity of the positive ions both in liquid and in gaseous xenon was not determined yet.

Reference

RF DRIFT-VELOCITY FILTER FOR ION-SWARM EXPERIMENTS

Koichi Iinuma, Tsuyoshi Hamano, Tsunehito Ohtomo, and Masahiro Takebe
Department of Nuclear Engineering, Tohoku University

1. Introduction

Mobility, diffusion coefficient, and reaction rate of gaseous ions are important kinetic coefficients for studying complicated transport and reactive processes in radiation chemistry. In general, these coefficients are determined by ion-swarm experiments. Drift-tube method is one of the most conventional and useful techniques in them. Mass spectrometry is essential for correct identification of ionic species in an ambient gas in drift-tube apparatus. Usually, a differential pumping technique is employed to operate a mass spectrometer at high gas pressure. However, this technique is accompanied by some unfavorable problems such as the effect of mass-discrimination and the adiabatic expansion of neutral gas, which occasionally make the measured kinetic data doubtful.

Recently, we started a research for developing an RF drift-velocity filter which operates at high gas pressure in order to avoid these problems. Our present and future objectives are as follows: (i) design of an ion filter using the difference of mobility; (ii) design of an RF mass spectrometer operating at gas pressures ranging between 0.1 Torr and 760 Torr; (iii) combining it with a drift-tube apparatus; and (iv) its application to the experiments in radiation chemistry, gas phase ion chemistry, and atmospheric ion chemistry.

Although the resolution of present filter made on trial is still extremely low, its basic principle of operation is proved. This paper deals with an explanation of its principle and some results obtained by our preliminary experiments for (Li$^+$.Cs$^+$)/CO$_2$ and Li$^+$.CO$_2$/CO$_2$ systems using a drift-tube apparatus.

2. Operational principle of RF drift-velocity filter

A schematic view of the RF drift-velocity filter is shown in Fig.1. The filter consists of eight gold foils, 0.05 mm thick, 2.0 mm wide, placed in parallel 2.0 mm apart on a ceramic holder. The filter operates in the following way: we supply two crossed electric fields, $E_x$ and $E_y$, to
the filter. $E_x$ is the main DC drift field in which ions drift with drift velocity $v_x$. $E_y$ is a periodic AC field perpendicular to $E_x$ and in which the ions oscillate between the foils with drift velocity $v_y$. For a given AC frequency an ionic species with a sufficiently large value of $v_y$ will be collected by the foils and cannot pass through, whereas an ion with a smaller $v_y$ will not be captured and can transmit the filter.

Based upon a simplified kinetic model we can derive two approximate formulas for the cut-off frequency $f_0$ and the transmission efficiency $\xi$. In the present experiment $E_y$ is produced by a square-wave AC potential of frequency $f$. Let us now assume that all the ions traverse the gap length between the foils, $2\delta$, with the same drift velocity $v_y$ during the half period $1/2f$. We also ignore the field distortion (or non-uniformity) of $E_x$ and $E_y$ near the edge of the foils, and assume $E_y >> E_x$. A critical value of drift velocity $v_{dr}$, above which all the ions will be captured, is expressed by $f_0$ as

$$v_{dr} = 4 \delta f_0 \quad (1)$$

Since the reduced mobility of an ionic species $K_0$ is related to $v_{dr}$ (cm/s) by

$$K_0 = 3.7215 \times 10^{-3} \frac{v_{dr}}{E/N} \quad \text{cm}^2/\text{Vs} \quad (2)$$

where $E$ is the electric field strength and $N$ the gas number density ($E/N$ in Td: $1$Td=$10^{-17}$Vcm$^2$). Substituting Eq. (1) into Eq. (2) we obtain

$$f_0 = 67.2 \frac{K_0 \cdot E_y/N}{\delta} \quad \text{Hz} \quad (3)$$

with $\delta$ in cm. We define the transmission efficiency $\xi$ as
\[ \xi = 1 - \frac{f_0}{f} \quad \text{for all } f \geq f_0 \]  

(4)

In the limit \( f \to \infty \) all the ions can pass through the filter and \( \xi = 1 \). Eqs. (3) and (4) tell us that \( \xi \) increases with increasing \( f \), but decreases with increasing \( E_y/N \).

3. Experiments

A schematic view of our drift-tube apparatus with an RF drift-velocity filter is shown in Fig. 2. To produce ideally a homogeneous cylindrical electric field a continuous guard ring \(^{14}\) is employed. The filter is mounted in front of the ion collector. Two thermonic ion sources independently emit both \( \text{Li}^+ \) and \( \text{Cs}^+ \) ions into the drift region. The Tyndall four-gauge method \(^{11}\) is employed in order to measure the times at which the ions arrive at the detector. The profile of ion current with respect to the drift time is called the arrival time spectrum (ATS), which roughly estimates the ions presented in the drift region. Experimental condition is as follows: (\( \text{Li}^+, \text{Cs}^+ \))/CO\(_2\) system, gas pressure=0.2Torr, gas temperature=295K, \( E_x/N=62 \text{Td}, 75 \text{Td} \leq E_y/N \leq 600 \text{Td}, \) and \( 125 \text{kHz} \leq f \leq 3 \text{MHz} \). To obtain the ATS a high speed electrometer (Keithley model 427; shown as CA in Fig. 2) and a laboratory-made digital delay scanner (shown as DDS in Fig. 2) are utilized.

![Fig. 2. A block diagram of drift-tube apparatus. DDS, digital-delay-scanner; PI, photo isolator; DMM, digital multimeter; SWG, square-wave generator; CA, current amplifier; PS, power supply.](image)

4. Results and Discussion

Measured transmission efficiencies for \( \text{Li}^+/\text{CO}_2 \) and \( \text{Cs}^+/\text{CO}_2 \) systems are shown in Figs. 3 and 4, respectively. As is already mentioned, Eqs. (3) and (4) simply explain the overall tendency of those two curves. It is
Fig. 3. Transmission efficiency of Li\textsuperscript{+} ions in CO\textsubscript{2} plotted versus the RF frequency.

Fig. 4. Transmission efficiency of Cs\textsuperscript{+} ions in CO\textsubscript{2} plotted versus the RF frequency. An arrow at the bottom of the figure shows the frequency at which the curve of $V_{RF}=\pm 4V$ starts to rise.

noted that the Cs\textsuperscript{+}-curve starts to rise at lower frequency than the Li\textsuperscript{+}-curve. This difference indicates that the separation of Li\textsuperscript{+} and Cs\textsuperscript{+} ions is possible: when $f$ and $V_{RF}$ are fixed at 1MHz and $\pm 8V$, respectively, more than 90% of Cs\textsuperscript{+} ions can pass through the filter, but most of the Li\textsuperscript{+} ions are captured on the foils. Performance of this filter is tested using our drift-tube apparatus. Fig. 5 shows two ATS of a mixture of Li\textsuperscript{+} and Cs\textsuperscript{+} ions in CO\textsubscript{2}: one ATS drawn by a dushed curve is measured without operating the filter, while the other ATS drawn by a solid curve is measured with it. A ramp between Li\textsuperscript{+} and Cs\textsuperscript{+} peak in the profile indicates the Li\textsuperscript{+}•CO\textsubscript{2} cluster ions produced by the folloowing ion/molecule reaction:

$$\text{Li}^+ + \text{CO}_2 + \text{CO}_2 \rightarrow \text{Li}^+ \cdot \text{CO}_2 + \text{CO}_2 \quad .$$  \hspace{1cm} (5)  

Comparing these two ATS it is obvious when the filter operates almost 90% of the Li\textsuperscript{+} ions is trapped, but the ATS of Cs\textsuperscript{+} ions as well as Li\textsuperscript{+}•CO\textsubscript{2} cluster ions is not seriously affected by its operation. Unfortunately, resolution of the present filter is still extremely low, so that the separation of the mixture of Cs\textsuperscript{+} and Li\textsuperscript{+}•CO\textsubscript{2} ions is not yet sufficient.
Finally we briefly discuss how to increase the resolution of the present filter. Basically, resolution strongly depends on the position of an ion mixture located in the filter when it operates. Diffusion of ions toward ±y-direction is, therefore, undesirable; thus, the operation of RF-filter at high gas pressure is recommended. Also, when an ion is introduced in the filter the ion should be initially located in the middle of the gap between the foils. An entrance aperture in front of the filter or any bunching electric field are useful to make it possible.

5. Conclusions

To separate a mixture of ions into two groups at high gas pressure we have developed and tested an RF drift-velocity filter for \((\text{Li}^+, \text{Cs}^+)/\text{CO}_2\) system at 0.2Torr using a drift-tube apparatus. Measured ATS confirm that it efficiently traps the Li\(^+\) ions, whereas most of the Cs\(^+\) ions as well as the Li\(^+\)·CO\(_2\) cluster ions can safely pass through it. The basic principle of its operation is experimentally proved.

References

1. INTRODUCTION

Secondary electrons produced by ionizing radiation lose their energy by collisions with environmental molecules and are eventually thermalized. The electron thermalization in gaseous media has been a subject of intensive experimental\(^\text{[1]-[7]}\) and theoretical\(^\text{[8]-[14]}\) studies for the past twenty years. Studies of the electron thermalization provide us with knowledge of fundamental nature of electron-atom (or molecule) interactions. The knowledge helps us in understanding the radiation-chemical consequences depending on the environmental conditions and in developing many useful devices utilizing radiation effects including radiation detectors. Most previous investigations for the electron thermalization have been focused on the gaseous media of simple gases such as rare gases or some inert, non-reactive, gases. Except the studies of electron thermalization in air\(^\text{[6]}\) or oxygen\(^\text{[7]}\), which capture electrons moderately, no experimental investigation has been made for systems where some strongly-electron-attaching compounds are involved. In the present study we have observed the thermalization processes in argon in the presence of several electron attaching gases with different characteristics in the electron attachment rates. High-energy electrons have been produced by pulse radiolysis, and the time variation of electron density as well as the degradation of electron energy have been monitored using the microwaves as a probe. We have obtained for the first time the electron-energy loss rate coefficients and the thermalization times for these electron attaching gases.

2. EXPERIMENT

The pulse radiolysis microwave cavity method used in the present study was the
same as employed previously except no microwave heating unit attached. The gas sample in a cylindrical quartz cell placed in a resonant cavity was irradiated by a 3 ns X-ray pulse from Febetron 706. An X-band microwave circuit having a frequency-discriminator unit was used for the signal detection. The outputs were amplified by a differential amplifier and fed to a storage oscilloscope.

The gas pressures were measured with "Baratron" capacitance manometers. Ar (99.999%, Teisan Co.) was used as received. Compounds studied and their purity were as follows; CCl₄ (> 99%, Wako Chemicals), CHCl₃ (> 99%, Takachiho Shoji Co.), and C₆H₅I (> 97%, Wako Chemicals). They were used after degassing at 77 K. All the measurements were carried out at room temperature (298 ± 3 K).

3. Results and Discussion

Following pulsed X-ray irradiation of Ar atoms, high-energy electrons produced by ionization lose their energies by collisions with Ar atoms and are eventually thermalized. The electron thermalization during electron attachment to an electron-attaching gas in Ar buffer gas has been observed. Time profiles of the conductivity signals observed for pure Ar (70 Torr) and Ar (70 Torr) containing CCl₄ are shown in Fig.1(A). For pure Ar the signal increases slowly, shows a maximum (not clearly seen though), and attains a flat level. This feature is attributed to the presence of the Ramsauer minimum in the electron energy dependence of the momentum transfer cross section for Ar. When electrons are thermalized the signal becomes flat. The signals for mixtures with CCl₄, on the other hand, show decreases by the electron attachment reaction. This reaction may be influenced by the electron thermalization process. The amplitude of the microwave conductivity signal reflects not only the electron density but also the time-variation of the electron-collision frequency determined by the momentum-transfer cross sections, as discussed in detail for pure Xe system. For pure Ar the signal involves this effect. Therefore, in order to see only the change in the electron density for mixtures each signal for a mixture has been divided (or normalized) by that observed in pure Ar. The results are shown in Fig.1(B). Generally the rate of disappearance of electrons by attachment to molecules is given by Eq.(1).

\[ \frac{d \ln[e^{-}]}{dt} = - k(e) [CCl_4] \] (1)
where \([\cdot]\) denotes the density, \(k(e)\) is rate constant as a function of the mean electron energy \(e\). If the results of electron densities dependence on time are the first-order decay, differential equation (1) can be solved, to obtain the rate constants. However, it turns out that the decays are not of the first-order. This indicates that the apparent attachment rate constant depends on time. Thus, we have differentiated directly the \(\ln[e']\) vs time curve to obtain the derivative \(-k(e) [CCl₄]\) at respective time \(t\). The rate constants as a function of time so obtained are shown in Fig.2. Since the rate constants of electron attachment to CCl₄ as a function of the mean electron energy have been reported\(^{15}\), the time-variation of the rate constants can be converted to that of the mean electron energy, as shown in Fig.3. The figure 3 shows that at a high concentration of CCl₄ (> 10⁻⁵ Torr) the energy of electrons never relaxed to the thermal value, and most of the electrons are captured by CCl₄ before they are thermalized. In other words, the mean electron energy remains at a certain high energy level through the deformation of the electron-energy distribution function due to a high probability of electron attachment of electrons with very low energies. This is an example of the "attachment heating" as suggested by Shizgal\(^{14}\) in his theoretical studies of electron attachment to CCl₄ and SF₆.

The rate of energy loss can be defined by the following equation\(^{1}\).

\[
de\epsilon/dt = -K(e) (e - e_{th}) N
\]  

(2)

where \(e_{th}\) is the thermal energy, \(K(e)\) is the energy loss rate coefficient, and \(N\) is the density of the total gas molecules in the mixture. Equation (2) is rewritten as

\[
d [\ln(e - e_{th})]/dt = -K(e) N
\]

(3)

Then the derivative of a plot of \(\ln(e - e_{th})\) as a function of time \(t\) gives the value of \(K(e)\) at time \(t\). The value of \(K(e)\) can be regarded as a linear combination of the contribution from buffer gas Ar and that from the solute molecules;

\[
K(e) N = K(e)_{Ar} N_{Ar} + K(e)_{S} N_{S},
\]

(4)

or

\[
K(e) = K(e)_{Ar} N_{Ar} + K(e)_{S} (N_{S}/N_{Ar})
\]

(5)
when \( N = N_{\text{Ar}} \), where the subscripts \( \text{Ar} \) and \( S \) correspond to the buffer gas \( \text{Ar} \) and the solute compound, respectively. Shown in Fig.4 is an example of the plots of \( K(e) \) as a function of \([\text{CCl}_4]/[\text{Ar}]\) at selected values of the mean electron energy obtained for mixtures containing \( \text{CCl}_4 \) at very low pressures, \(< 10^{-5}\) Torr. From the intercepts and the slopes we can obtain the values of \( K(e)_{\text{Ar}} \) and \( K(e)_{\text{CCl}_4} \) as a function of the mean electron energy.

Similar analyses have been carried out for the data for \( \text{CHCl}_3 \) and \( \text{C}_6\text{H}_5\text{I} \) in \( \text{Ar} \) buffer gas. The data of electron attachment rate constants as a function of the mean electron energy\(^{15,16}\) have been utilized for the analyses. The mean electron energies as a function of time for \( \text{CHCl}_3 \) and \( \text{C}_6\text{H}_5\text{I} \) are shown in Figs. 5 and 6, respectively. It should be noted that at a high pressure of \( \text{CHCl}_3 \) the value of \( e \) falls with time down to below thermal energy (-0.04 eV). This is understandable because the electron attachment cross sections for \( \text{CHCl}_3 \) have a maximum at the electron energy of \( \sim 0.3 \) eV\(^{17}\), and the electrons are captured efficiently at this energy during the course of thermalization resulting in less high-energy electrons in the electron-energy distribution. This can be called "attachment cooling" as suggested by Crompton et al\(^{18}\). There can be seen no such an effect in the case of \( \text{C}_6\text{H}_5\text{I} \). This is consistent with the fact that this compound does not capture electrons as effectively as \( \text{CCl}_4 \) does and the cross sections show relatively flat dependence on the electron energy between 0 and 2 eV\(^{16}\).

The values of \( K(e) \) as a function of the mean electron energy are shown in Fig.5 for the compounds studied. Warman and Sauer\(^{11}\) reported the values of \( K \) being \( 1.5 \times 10^{-8} \) cm\(^3\)s\(^{-1}\) or lower for various compounds assuming that they are nearly constant over the mean electron energies between thermal and about 1 eV. The present results show that all the values of \( K(e) \) exhibit marked dependencies on the mean electron energy, and the absolute magnitude of \( K(e) \) for \( \text{CCl}_4 \) are very high, \( 10^{-7}-10^{-8} \) cm\(^3\)s\(^{-1}\), while those for \( \text{Ar} \), which has been obtained from the data for \( \text{CCl}_4-\text{Ar} \) system, are quite low, down to \( \sim 10^{-15} \) cm\(^3\)s\(^{-1}\) at thermal energy, which is much lower than a constant value \( 1.3 \times 10^{-15} \) cm\(^3\)s\(^{-1}\) reported by Warman and Sauer\(^{11}\). The values for \( \text{Ar} \) can also be obtained from the time-profile of the conductivity signal for pure \( \text{Ar} \). This is shown in Fig.5 for comparison, and the agreement between both results seems to be good. It is very interesting that the values of \( K(e) \) for \( \text{CCl}_4 \) are very large. At present we cannot give a reasonable interpretation for this result, though it may be related to a high electron-attaching capability of \( \text{CCl}_4 \).
The thermalization times for electrons, defined as the time taken for the electrons with the mean energy of 1 eV to relax down to an energy of 10% above the thermal energy \(^1\), can be calculated from the equation,

\[
\tau_{th} = \int_1^{1.1} \frac{1}{K(\varepsilon) \left( \varepsilon - \varepsilon_{th} \right)} \, d\varepsilon
\]

The values of \(\tau_{th} N\) in units of sec-Torr are 0.1, 3.0 \times 10^{-9}, 2.9 \times 10^{-8}, and 1.4 \times 10^{-8} for Ar, CCl₄, CHCl₃, and C₆H₅I, respectively. The values for the electron-attaching gases are comparable to the known values for some polyatomic gases\(^1\),\(^5\), but the present value for Ar is much larger than those (on the order of 10⁻³) obtained by previous experimental and theoretical studies\(^1\). The present large value (or long thermalization time) may be primarily due to very low values of \(K(\varepsilon)\) at the electron energies near thermal. The discrepancy has to be explained clearly in a future study. Furthermore, the data analysis must be refined to obtain more accurate values of \(K(\varepsilon)\), because taking a second derivative of the observed signals with respect to the time can introduce large errors into the value of \(K(\varepsilon)\). The study of this subject is still under way.

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**References**

Fig. 1

pressure of CCl₄

a 1.45 x 10⁻⁶ Torr  b 3.59 x 10⁻⁶ Torr

c 1.41 x 10⁻⁵ Torr  d 4.24 x 10⁻⁵ Torr
Fig. 2

Pressure of CCl₄

a $1.45 \times 10^{-6}$ Torr  b $3.59 \times 10^{-6}$ Torr

c $1.41 \times 10^{-5}$ Torr  d $4.24 \times 10^{-5}$ Torr

Fig. 3

Fig. 4
e\textsuperscript{-}-C\textsubscript{2}H\textsubscript{6} Collision Cross Sections by Electron Swarm Study

Y. Shishikura, K. Asano and Y. Nakamura
Faculty of Science and Technology, Keio University
3-14-1 Hiyoshi, Yokohama 223, Japan

1. INTRODUCTION

Physics of electron collisions with saturated hydrocarbons has long been studied theoretically and experimentally. We also have determined a set of electron collision cross sections for methane molecule by an electron swarm method\textsuperscript{(1)}. Molecules usually have a number of competing collision processes with low energy electrons and it was believed to be difficult, in principle, to determine a unique set of electron collision cross sections for molecules by usual electron collision study. In order to avoid this difficulty and to use full advantage of swarm study we use electron swarm parameters both in dilute molecule-rare gas mixtures and in pure molecular gas. Electron swarm parameters in dilute molecule-rare gas mixtures (mixing ratio of molecular gas is usually less than a few percent and argon is used as a buffer gas) are almost insensitive to usually unknown elastic cross section of the molecule because of the elastic cross section of the major rare gas, but they depend mostly on the elastic momentum transfer cross section of argon and also on the vibrational excitation cross sections of the molecule. The drift velocity and longitudinal diffusion coefficient in these mixtures actually very sensitive to changes in the vibrational excitation cross sections for the molecule and, therefore, sensitive determination of them is possible. The vibrational cross sections determined by using swarm data measured in the mixtures are then used to analyze swarm data in pure molecular gas and to derive the momentum transfer cross section for the molecule. The momentum transfer cross section and the vibrational cross sections can also be used to analyze the ionization coefficient and to derive the total inelastic cross section with threshold energy immediately below the ionization threshold. The ionization cross section for most gases are usually known with sufficient accuracy. This procedure can actually be applied to a wide variety of molecules and metal atoms and we have so far determined a number of molecules including SiH\textsubscript{4}\textsuperscript{(2)}, CF\textsubscript{4}\textsuperscript{(3)}, GeH\textsubscript{4}\textsuperscript{(4)} and so on other than CH\textsubscript{4}.

In this study we extend our electron swarm study to ethane molecule whose collision
cross sections with low energy electrons are important to understand phenomenon in particle
detector or in fusion plasmas, and a set of electron collision cross sections for ethane molecule
over the electron energy range from thermal to about 30 eV.

2. EXPERIMENTAL METHOD

The cross sectional diagram of the apparatus, the double-shutter electron drift tube,
used in the present swarm experiments is shown in Fig.1\(^{(5)}\). It consists of a photoelectric electron
source (K), two pairs of electron shutter (S1 and S2), an electron collector (C) and ten guard rings (GRs). The
photoelectric cathode is a back-illuminated gold thin film deposited on a fused quartz window and ultraviolet
light from a deuterium lamp supplies initial electron current. Each electron shutter consists of a pair of parallel wire
grids. The second shutter (S2) and the electron collector (C) are mounted on an insulator disk and the distance between
S1 and S2 (electron drift distance) can be varied from 1 to 10 cm by a linear motion feedthrough. Guard rings are
used to maintain uniform electric field. The two shutters are usually biased so that electrons are blocked there. The bias to S1 is canceled out by applying periodical short pulse voltage in order to inject narrow electron pulses into the drift region periodically. Another pulse voltage with the same frequency is applied also to S2. The phase difference of the S2 pulse relative to the S1 pulse is varied slowly by using a digital delay pulse generator. The collector current is proportional to the number of electrons arriving at S2 at instantaneous delay time after they started at S1. The current is amplified by an electrometer and is recorded as the arrival time spectrum of electrons (ATS). An typical example is shown in Fig.2. At each E/N usually eight ATSs are recorded at eight different drift distances from 2 to 9 cm. By assuming that each ATS is a shifted Gaussian distribution, the electron drift velocity \( W \) is derived from the relationship
between the peak time of each ATS and the drift distance, and the longitudinal diffusion coefficient DL is determined from the relationship between the characteristic width of each ATS and the drift distance. This so-called differential determination of swarm parameters is effective to remove the effects of finite pulse width of the shutter pulses and of non-uniform electric field around electron shutters, and so on, on the resultant electron swarm parameters. Measurements are repeated at three different densities of gas at least at each E/N. Maximum error limits estimated for the present experimental system are 2% for drift velocity and 7% for longitudinal diffusion coefficient. But actual scatters of measured drift velocity and the product of longitudinal diffusion coefficient and the gas number density, NDL, were usually less than 1% and 3%, respectively.

All measurements were carried out at room temperature (22-25 °C). The sample mixtures were composed of pure ethane (purity 99.9%) and pure argon (purity 99.999%). The actual mix ratio was determined by using gas chromatography test. Specially purified ethane (purity 99.99%) was used for the measurement of pure ethane.

3. EXPERIMENTAL RESULTS

Measurements were carried out over the E/N range of 0.03 - 3 Td and the gas pressure range of 40 - 1000 torr in the 0.206% C2H6-Ar mixture, 0.03 - 25 Td and 4 - 1000 torr in the 1.98% C2H6-Ar mixture, and 0.03 - 300 Td and 0.2 - 1050 torr in pure ethane. The results of the...
Fig. 3 The electron drift velocity $W$ and the product of the longitudinal diffusion coefficient and the gas number density $ND_L$ as a function of $E/N$ in the 0.206% C$_2$H$_6$-Ar mixture. The solid symbols show the present measurement. The broken and solid lines show values calculated with the initial cross section set of Hayashi and with the present cross section set, respectively for methane.

Fig. 4 The electron drift velocity $W$ and the product of the longitudinal diffusion coefficient and the gas number density $ND_L$ as a function of $E/N$ in the 1.98% C$_2$H$_6$-Ar mixture. Symbols are the same as in Fig. 3.
measurements are summarized in Figs. 3 - 5. The measured electron drift velocities in both C$_2$H$_6$-Ar mixtures show clear negative differential conductivity (NDC) and this feature indicates the dominant role of vibrational excitation processes of the C$_2$H$_6$ molecule in the mixtures. There are several experimental reports on the electron swarm parameters in C$_2$H$_6$-Ar mixtures$^{[6]}$. However, they all are not in the present range of mix ratios and direct comparison of the results is not possible. In pure ethane gas electron drift velocity enhancement due to the vibrational excitations is apparent but the effect is not so strong as in pure methane$^{[4]}$. There are also a few measurements of electron swarm parameters in pure ethane over rather limited E/N range$^{[7,8]}$ and they agree well with the present results. The present new results in E/N < 0.08 Td and E/N > 80 Td were in fact important to determine the elastic momentum transfer cross section for ethane.

4. BOLTZMANN EQUATION ANALYSIS OF PARAMETERS AND ELECTRON COLLISION CROSS SECTION SET FOR C$_2$H$_6$ MOLECULE

The drift velocity and the longitudinal diffusion coefficient were calculated by the use of
the James Cook University multi-term Boltzmann code of Ness and Robson\textsuperscript{(9)}. The electron collision cross section set for argon atom determined by Kurachi and Nakamura\textsuperscript{(10)} was used throughout the present study. The cross section set for ethane molecule compiled by Hayashi\textsuperscript{(11)} was used as the starting set. It consisted of an elastic momentum transfer ($Q_m$), a pair of vibrational excitation cross sections ($Q_{v24}$ and $Q_{v12}$), an attachment cross section ($Q_a$), two electronic excitation cross sections ($Q_{e1}$ and $Q_{e2}$), a dissociation cross section ($Q_d$) and an ionization cross section ($Q_i$). This initial set for ethane is shown by broken lines in Fig. 8. Electron swarm parameters calculated using this cross section set are shown by broken curves in Figs. 3 - 5. This set is moderately consistent with the experimental drift velocity at least but there are deviations beyond the experimental error limits over the whole $E/N$ range. Worse discrepancy was found for NDL both in the mixtures and in pure ethane in higher $E/N$ range.

As explained before, the initial cross section set for C\textsubscript{2}H\textsubscript{6} molecule was modified according to the following two steps in order that the calculation using resultant cross section set agreed with the experimental data.

(1) STEP 1: Modification on inelastic (mainly vibrational cross section) cross sections in order that calculated swarm parameters in the two mixtures agree with present experimental results.

(2) STEP 2: Modification on elastic momentum transfer cross section for C\textsubscript{2}H\textsubscript{6} molecule so that the calculated swarm parameters in pure ethane agree with present experimental results, when inelastic cross sections determined in STEP 1 are not altered.

4.1 Vibrational excitation cross sections for C\textsubscript{2}H\textsubscript{6} molecule (STEP 1)

The prominent $E/N$ dependencies in drift velocity and longitudinal diffusion coefficient in the present mixtures in low $E/N$ depend mainly on the elastic momentum transfer cross section of argon atom and on vibrational cross sections of ethane molecule, and the elastic momentum transfer cross section of ethane molecule has only very weak contribution to swarm parameters in the mixtures because concentration of ethane in the mixtures is low and electron energy loss through elastic collisions with ethane molecules is small. The elastic momentum transfer cross section of argon atom is known far more precisely than that of ethane and, therefore, it is possible to determine vibrational excitation cross sections for ethane molecule without respect to its elastic momentum transfer cross section from electron swarm parameters measured in these dilute C\textsubscript{2}H\textsubscript{6}-Ar mixtures.

It is well known that ethane molecule has twelve normal vibrational modes. Boesten et al\textsuperscript{(12)} measured absolute differential and integral cross sections for electron impact vibrational excitation of ethane molecule for incident energies from 3 to 20 eV and confirmed strong shape resonance at 7.5 eV. In the present study we arbitrary assume three vibrational composites, $Q_{v1}$
(bending, energy loss 0.11 eV), Qv2 (bending, energy loss 0.16 eV) and Qv3 (stretching, energy loss 0.36 eV) instead of twelve cross sections. Cross section for torsional oscillation (v4, 0.0358 eV) is not included here because a trial calculation indicates that its threshold cross section has little effect on swarm parameters in the present E/N range in the mixtures. Our choice of these three composites depends on the reported energy loss spectrum by Boesten et al (12).

Starting from its threshold each vibrational cross section was modified by trial-and-error manner so that agreement between analyzed and measured swarm parameters was improved. There is necessarily arbitrariness in relative magnitudes of three compounds over a part of energy range, but contributions from different threshold cross sections of the three are separable and we can trust the experimental relative magnitudes around their shape resonance peaks of Boesten et al (12). Fig. 6 shows percentage deviations of calculated swarm parameters from the measurement. Open symbols show those calculated by using the initial cross section set of ethane and closed symbols those by using adjusted vibrational cross sections through STEP 1.

Vibrational cross sections from thresholds to 2 eV contribute mainly to swarm parameters in E/N < 0.6 Td for the 0.206% mixture and in E/N < 2 Td for the 1.98% mixture.
Magnitude of resonance peaks of vibrational cross sections determines drift velocity in these mixtures in higher E/N. The integrated vibrational cross sections of Boesten et al. are multiplied by a factor of 1.3 in order that the calculated electron drift velocity in mixtures agree with the experimental results within error limit. Error limit claimed by Boesten et al is about 30%. One of the electronic excitation cross sections, Qe1 (threshold 1.2 eV), is almost nullified in order that the calculated NDL for the 1.98% mixture in E/N > 10 Td is improved.

Deviations of the calculated swarm parameters after STEP 1 are compared by solid symbols in Fig. 6. The resultant drift velocity and NDL in two mixtures agree with the measurements within estimated maximum error limits. The swarm parameters in pure ethane are also improved by modifying the vibrational cross sections except in E/N < 1 Td where deviation in the drift velocity is increased on the contrary. The elastic momentum transfer cross section of ethane molecule is modified in the following step in order that the experimental swarm parameters measured in pure ethane can be reproduced by the Boltzmann equation analysis.

**4.2 Momentum transfer cross section for C2H6 (STEP 2)**

As was shown in Fig. 6 the deviation of calculated electron drift velocity in pure ethane after STEP 1 exceed the present experimental error limit in E/N < 0.5 Td and E/N > 80 Td.

![Fig. 7 The percentage deviations of calculated electron swarm parameters from the experimental values. Open symbols are those calculated with the set using modified vibrational excitation cross sections (STEP 1) and closed symbols are those with our final cross section set (STEP 2).](image-url)
By using the set of vibrational cross sections determined in STEP 1 the initial elastic momentum transfer cross section was modified so that the calculated swarm parameters in pure ethane agrees with the present measurement over the whole E/N range. Electron drift velocity in the lowest E/N range ( < 0.5 Td ) and in the highest E/N range ( > 80 Td ) is sensitive on the elastic momentum transfer cross section on the left hand side of its Ramsauer-Townsend minimum and on the right hand side of the shape resonance peak, respectively. Deviations of the calculated swarm parameters after STEP 2 are compared in Fig. 7. It is apparent from the figure that the final cross section set can reproduce all swarm parameters measured within maximum error limit except a few points for NDL in the mixtures.

The final result of momentum transfer cross section is shown in Fig. 8 by solid line and it is compared with the beam experiment of Tanaka et al \(^{(13)}\) multiplied by a factor of 0.88 ( 1 < e < 200 eV, solid circles ). Claimed error limit of Tanaka et al is 15-22%.

4.3 Electronic excitation cross sections for C\(_2\)H\(_6\) and Townsend's first ionization coefficient

The initial cross section set includes two electronic excitation cross sections, Qel and Qe2, and the former is almost nullified because of the measured NDL in the mixtures as explained in section 4.1. The magnitude of Qe2 is adjusted in order to reproduce the experimental results of Townsend's first ionization coefficient ( Heylen\(^{(14)}\) and Watts and Heylen\(^{(15)}\) ). The final Qe2 is also shown in Fig. 8 by solid line and with this modification the calculated ionization coefficient agrees with the measurement within 10% over entire range of E/N. It is also confirmed that the modification does not change drift velocity and NDL in mixtures and in pure ethane.

4.4 Discussion on anisotropy of electron scatterings

In the preceding sections all scatterings are assumed isotropic in the analysis and we succeed to determine a set of cross sections for ethane molecule which can reproduce all electron swarm parameters measured in two ethane-argon mixtures and in pure ethane over the most of the E/N range. However, the set fails to reproduce the measured NDL in high E/N. It was expected that the observed discrepancy between experimental and analytical NDL might be reduced or removed by introducing correct anisotropy in scatterings. The effects of anisotropy is investigated by introducing available differential scattering cross sections shown in Table 1. The inclusion of these DCS, however, only changes all electron swarm parameters by less than 1% from those assuming isotropic scatterings, less than a half of the estimated experimental uncertainty of the drift velocity.
Table 1. Sources of differential scattering cross sections for ethane and argon

<table>
<thead>
<tr>
<th>Source</th>
<th>Method</th>
<th>Energy Range (eV)</th>
<th>Authors</th>
</tr>
</thead>
<tbody>
<tr>
<td>ethane elastic</td>
<td>beam (1-100)</td>
<td>Tanaka et al (13)</td>
<td></td>
</tr>
<tr>
<td>vibrational</td>
<td>beam (3-20)</td>
<td>Boesten et al (12)</td>
<td></td>
</tr>
<tr>
<td>vibrational</td>
<td>beam (2.81)</td>
<td>Mapstone et al (16)</td>
<td></td>
</tr>
<tr>
<td>argon elastic</td>
<td>R-matrix (0-3)</td>
<td>Bell et al (17)</td>
<td></td>
</tr>
<tr>
<td>elastic</td>
<td>PO-method (0.13-55)</td>
<td>Dasugpta et al (18)</td>
<td></td>
</tr>
<tr>
<td>elastic</td>
<td>R-matrix (60-150)</td>
<td>Fon et al (19)</td>
<td></td>
</tr>
<tr>
<td>inelastic</td>
<td>beam (16-100)</td>
<td>Chujian et al (20)</td>
<td></td>
</tr>
</tbody>
</table>

Fig. 8 Electron collision cross sections for ethane molecule. ———, present results; ……., Hayashi (11); ○, Tanaka et al (13) multiplied by a factor of 0.88; ★ and ■, Boesten et al (12) multiplied by a factor of 1.3.
5. CONCLUSIONS

The drift velocity and longitudinal diffusion coefficient of electrons in the 0.206% and 1.98% ethane-argon mixtures and in pure ethane are measured in the E/N range 0.03 - 3 Td, 0.03 - 25 Td and 0.03 - 300 Td, respectively. The measured swarm parameters are used to determine vibrational excitation and elastic momentum transfer cross sections for ethane molecule. The determined vibrational and elastic momentum cross sections are moderately consistent with reported results by beam experiments over their energy ranges. The present result of the cross sections is further modified with use of the Townsend's first ionization coefficient. Realistic anisotropies in scatterings have little effect on electron swarm parameters.

6. ACKNOWLEDGMENTS

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1. INTRODUCTION

The swarm analysis by means of evolving transport equations derived from the Boltzmann equation is often available as the moment method. Up to date, the authors have made various investigations in connection with transport equations\(^1\) \(-\) \(^2\), where the collision terms supposed to be physically proper have been assumed. The main purpose of the present study is to analytically derive the collision terms in transport equations of an electron swarm from the ones in the Boltzmann equation. Then, transport equations with new collision terms are arranged and the characteristics of the electron swarm are numerically calculated. A few results are already reported in the case of so weak electric field that inelastic collisions are negligible\(^3\) \(-\) \(^4\). Here, the treatment is extended to the higher electric field in consideration of inelastic collisions. The calculated characteristics of drift velocity and mobility are compared with the existing experimental values.

First, let us treat a temporally steady state, a spatially uniform medium (the drift velocity and the energy of charged particles are uniform) and one-dimensional space, the coordinate being \(z\) with the direction of the electric field. Then, the electron velocity distribution function \(f\left(w, z\right)\) (\(w = \left|w\right|\), \(w\): electron velocity)
assumes the two term approximate form,

$$f(w, z) = f_0(w, z) + f_1(w, z)\cos \theta ,$$

(1)

where $f_0$ and $f_1$ are equilibrium and non-equilibrium velocity distribution parts, respectively and $\theta$ is an angle to the electric field $E$.

2. ANALYSIS OF COLLISION TERMS IN TRANSPORT EQUATIONS

The analysis is carried out by taking moments of collision term in the Boltzmann equation\(^5\), namely,

$$\left(\frac{\partial f}{\partial t}\right)_m = \frac{m}{M_n} \frac{1}{w^2} \frac{\partial}{\partial w} \left( w^3 \nu_m f_0 + \frac{kT_n}{m} w^2 \nu_m \frac{\partial f_0}{\partial w} \right) - \nu_m f_1 \cos \theta ,$$

(2)

where the index $m$ denotes the momentum transfer collision, and $w$ is the electron velocity, $\nu_m$ is the momentum transfer collision frequency by an electron, $T_n$ is the gas temperature, $m$ and $M_n$ are the masses of electron and gas particles. Collision terms of each moments in transport equations are calculated as follows,

for the first moment equation:

$$\int \left( \frac{\partial f}{\partial t} \right)_m d w = \frac{m}{M_n} \int_0^{\infty} \frac{\pi}{w^3} \left( w^3 \nu_m f_0 + \frac{kT_n}{m} w^2 \nu_m \frac{\partial f_0}{\partial w} \right) d w = 0 ,$$

(3)

for the second moment equation:

$$\int m w \left( \frac{\partial f}{\partial t} \right)_m d w = - \frac{1}{3} m \int_0^{\infty} \frac{4}{w^3} \nu_m f_1 d w = - n m \bar{w} \nu_m ,$$

(4)

for the third moment equation:

$$\int \frac{1}{2} m w^2 \left( \frac{\partial f}{\partial t} \right)_m d w = - \frac{2m}{M_n} \left[ \int_0^{\infty} \frac{4}{w^2} \left( \frac{1}{2} m w^2 \right) \nu_m f_0 d w - U_n \left\{ \int_0^{\infty} \frac{4}{w^2} \nu_m f_0 d w + \frac{1}{3} \int_0^{\infty} \frac{4}{w^2} \left( \frac{d \nu_m}{d w} \right) f_0 d w \right\} \right]$$

$$= - \frac{2m}{M_n} \nu_m \left( \epsilon + \frac{1}{3} U_n \frac{d \ln f_0}{d \ln w} \right) ,$$

(5)

where $\epsilon$ is the kinetic energy $(1/2) m w^2$ and $U_n$ is the gas particle random energy $(-3/2 kT_n, k$: Boltzmann's constant, $T_n$: gas particle temperature).
3. EFFECTIVE COLLISION FREQUENCIES

In the above collision terms, the following effective collision frequencies \( \langle \nu_m \rangle \) are defined as follows.

\[
\int m w (\frac{\partial f}{\partial t}) m w \, d w = - n m w \langle \nu_m \rangle = - n m w \langle \nu_m \rangle = - n m w \langle \nu_m \rangle , \quad (6)
\]

\[
\frac{1}{\langle \nu_m \rangle} = - \frac{1}{3} \frac{d \ln f_0}{d \ln w} , \quad (7)
\]

or

\[
\frac{1}{\langle \nu_m \rangle} = \frac{1}{\frac{\bar{e}}{\nu_m}} , \quad (8)
\]

\[
\int \frac{1}{2} m w^2 (\frac{\partial f}{\partial t}) m w \, d w = - n \frac{2 m}{M_n} (\bar{e} - U_n) \langle \nu_e \rangle , \quad (9)
\]

\[
\langle \nu_e \rangle = \nu_m (\bar{e} + \frac{1}{3} U_n \frac{d \ln f_0}{d \ln w}) / (\bar{e} - U_n) , \quad (10)
\]

\[
\bar{e} = U + (1/2) m w^2 , \quad (11)
\]

where \( w \) is the drift velocity \( (v = |w|) \), \( \bar{e} \) is the mean kinetic energy and \( U \) is the random energy \( = (3/2)kT_e \), \( T_e \): electron temperature). Here, \( \langle \nu_m \rangle \) and \( \langle \nu_e \rangle \) are called the effective momentum transfer collision frequency and the effective energy transfer collision frequency, respectively. It is remarkable that the effective collision frequency \( \langle \nu_m \rangle \) is expressed in two different forms as shown in Eq. (7) or (8) from the electric field drift or the diffusion flow term, respectively.

4. TRANSPORT EQUATIONS AND THEIR SOLUTIONS

Transport equations with the above new collision terms are collected as follows.

continuity equation:

\[
w \cdot \frac{1}{n} \nabla n = 0 , \quad (12)
\]

momentum conservation equation:

\[
w = \mu E - D \frac{1}{n} \nabla n , \quad (13)
\]
\( \mu \equiv \frac{e}{m\langle \nu_m \rangle} \); \( D \equiv \frac{(2/3)U}{m\langle \nu_m \rangle} \), \hspace{1cm} (14)

Now, since \( \nabla n = 0 \) from Eq. (12), Eq. (13) becomes

\[
\mathbf{v} = \frac{e}{M_n \langle \nu_m \rangle} \mathbf{E},
\] \hspace{1cm} (15)

Energy conservation equation:

\[
e \mathbf{E} \cdot \mathbf{v} = \frac{2m}{M_n}(\mathbf{\overline{e}} - U_n)\langle \nu_\star \rangle = \frac{2m}{M_n}(U - U_n + \frac{1}{2} m \mathbf{v}^2)\langle \nu_\star \rangle.\] \hspace{1cm} (16)

From these transport equations, the following solutions are obtained,

\[
\mathbf{v}^2 = 2\frac{m}{M_n}(\mathbf{\overline{e}} - U_n)\langle \nu_\star \rangle,\] \hspace{1cm} (17)

\[
(e \mathbf{E})^2 = 2\frac{m^2}{M_n}(\mathbf{\overline{e}} - U_n)\langle \nu_m \rangle\langle \nu_\star \rangle,\] \hspace{1cm} (18)

\[
U = (\mathbf{\overline{e}} - U_n)(1 - \frac{m}{M_n}\langle \nu_\star \rangle) + U_n.\] \hspace{1cm} (19)

5. SOME CALCULATED CHARACTERISTICS

A few examples of results calculated from the above solutions when the equilibrium energy distribution \( f_0 \) is the solution of the Boltzmann equation are illustrated. Gases are He, Ne, Ar, Kr and Xe at the gas temperature \( T_n = 300K \). Figures 1 and 2 show the characteristics of the drift velocity \( \mathbf{v} \) and mobility \( \mu_0 = \mu N \) versus the electric field \( E/N \), respectively (solid lines), \( N \) being the gas particle density. Dotted lines stand for the calculated results from various transport equations with the mean collision frequency \( \overline{\nu_m}/N \) in place of the effective collision frequencies \( \langle \nu_m \rangle/N \) and \( \langle \nu_\star \rangle/N \). It is seen that the differences of the characteristics between the solid and the dotted lines are rather large in Ar, Kr and Xe, the Ramsauer gases. Solid lines calculated with the effective collision frequencies agree well with the existing experimental data. The effect of inelastic collisions does not appear in the weak electric field below about \( E/N = 4Td \). Numerical factors \( F_{\nu_\star} \) and \( F_{\nu_m} \) in these figures indicate the
multipliers to the excitation and the ionization cross sections whose products will be added to the momentum transfer cross section.
Figure 1: The characteristics of the drift velocity $v$ versus the electric field $E/N$, for (a) He, (b) Ne, (c) Ar, (d) Kr and (e) Xe at $T_n=300K$ (solid lines). Dotted lines stand for the calculated results with $\overline{\nu_m}/N$ in place of $<\nu_m>/N$ and $<\nu_e>/N$. 
2(b) Mobility $\mu = \mu N$ [1/cm.s.V] vs Electric Field E/N [Td] for TN=300.0[K] and GAS=NEON

2(c) Mobility $\mu = \mu N$ [1/cm.s.V] vs Electric Field E/N [Td] for TN=300.0[K] and GAS=ARGON

2(d) Mobility $\mu = \mu N$ [1/cm.s.V] vs Electric Field E/N [Td] for TN=300.0[K] and GAS=KRYPTON
Figure 2: The characteristics of the mobility $\mu_N = \mu N$ versus the electric field $E/N$, for (a) He, (b) Ne, (c) Ar, (d) Kr and (e) Xe at $T_n = 300K$ (solid lines). Dotted lines stand for the calculated results with $\nu m/N$ in place of $\langle \nu m \rangle/N$ and $\langle \nu e \rangle/N$.

References

EFFECTS OF ANISOTROPIC SCATTERING ON ELECTRON TRANSPORT PROPERTIES IN GAS

Hisanori Tanaka and Nobuaki Ikuta
Tokushima University

1. INTRODUCTION

Electron transport properties in gases under an electric field have been analysed by solving the Boltzmann equation for the velocity distribution function, usually in an expanded form with density gradients. One aim to use the density-gradient expanded (DGE) velocity distribution is to calculate the time-of-flight (TOF) transport properties in reactive gases and another is to calculate the diffusion coefficient. However, even in steady state in time, the density gradient has no steady values, and the DGE velocity distribution seems to be used in a conceptual form not dependent on position and time. On the other hand, in the flight time integral (FTI) analyses, the Boltzmann equation integrated in position space is solved, where the density gradient can not be considered. However, the transport properties are obtained from the flight behavior of electrons along their trajectories through the time. The flight time is determined by the integral (total) collision frequency but not by the partial collision frequencies such that of momentum-transfer. In this paper, the effects of anisotropic scattering on the electron transport properties in non-reactive gas, particularly on the transverse diffusion coefficient $D_T$ are discussed. The definitions of transverse diffusion coefficient are described first. Anisotropy in scattering adopted here is explained next, and the effects of anisotropic scattering on the electron transport properties are discussed with the results of analytical and FTI calculations.

2. TRANSVERSE DIFFUSION COEFFICIENT $D_T$

The transverse diffusion coefficient $D_T$ is defined as the increasing rate of the transverse dispersion in position space referred to the electric field as

$$D_T = \frac{d<R^2(t)>}{4dt},$$

(1)
where $R(t)$ is the radial distance from the center axis. At the same time, $D_r$ is defined as a constant relating the electron flux $\Gamma_y$ in a transverse direction $y$ with the density gradient in $y$ direction $\nabla_y = \partial n/\partial y$ as

$$\Gamma_y = -D_r \nabla_y n.$$  \hspace{1cm} (2)

These two definitions are equivalent, of course. In the usual Boltzmann equation analyses, the definition in eq.(2) has been used with the density-gradient expanded (DGE) velocity distribution function. However, it is considered that eq.(2) is rather a macroscopic definition. From the microscopic viewpoint, electrons move without suffering any effects from the density gradient. The collective properties of respective electrons in successive microscopic flights gives the macroscopic transport such as the diffusion and drift. Therefore, the electron transport essentially has to be evaluated from the microscopic motion through successive flights. In an steady state in time, the starting velocities in successive flights have a distribution, and a normalized form of starting rate distribution $\Psi_{sn}(v_0)$ is adopted in FTI analyses. In which, the transport properties are calculated with the flight behavior of electrons in one flight instead of those in successive flights by using the transport functions $Gz(v_o)$ or $Gfz(v_o)$. $Gz(v_o)$ gives the mean $z$ value just before collision for electrons started from $v_o$, and $Gfz(v_o)$ implies the mean integrated value of $z$ through a flight for electrons started from $v_o$. The averages for all the electrons in a swarm are obtained by integrating them with the normalized starting rate distribution $\Psi_{sn}(v_0) \, dv_o$ over the velocity space of $v_o$.

In extended FTI method\(^{(3,4)}\) considering anisotropy in scattering, the normalized starting rate distribution $\Psi_{sn}(v_0)$ is given in two terms as

$$\Psi_{sn}(v_o) = \Psi_{sn0}(v_o) + \Psi_{sn1}(v_o) \cos \theta_0,$$ \hspace{1cm} (3)

and $Gz(v_o)$ and $Gfz(v_o)$ are also written in two terms

$$Gz(v_o) = Gz_0(v_o) + Gz_1(v_o),$$ \hspace{1cm} (4)

$$Gfz(v_o) = Gfz_0(v_o) + Gfz_1(v_o).$$ \hspace{1cm} (5)

Here, $Gz_1(v_o)$ and $Gfz_1(v_o)$ have definitions including $\cos \theta_0$ for convenience.\(^{(3,4)}\)

For the calculation of $D_r$, the following expression has been used.\(^{(5)}\) Since $R^2(t) = X^2(t) + Y^2(t)$ for an electron is written as

\[ -52 - \]
Through a number of cascaded flights with a total flight time $t$

$$t = \tau_1 + \tau_2 + \tau_3 + \cdots + \tau_{n-1} + \tau_n,$$  \hspace{1cm} (7)

the cross terms in eq.(6) vanish due to the independence of each flight, and only the square terms remain in a form

$$R^2(t) = \sum_{i=1}^{n} (v_i^2 \sin^2 \theta_i \tau_i^2).$$  \hspace{1cm} (8)

This expression may be regarded for $j$-th electron. Averaging for all the electrons (number of $m$) in a swarm is given as although $n$ values for respective electrons may not be always the same

$$<R^2(t)> = < \sum_{j=1}^{m} \sum_{i=1}^{n} (v_{ji}^2 \sin^2 \theta_{ji} \tau_{ji}^2) >,$$  \hspace{1cm} (9)

where the vanishment of cross terms is further reliable. The average values of $<R^2(\tau)>$ in a flight is obtained in FTI analyses as

$$<R^2(\tau)> = <G(v_0^2 \sin^2 \theta_0 \tau^2)> = 2<Gf(v_0^2 \sin^2 \theta_0 \tau).$$  \hspace{1cm} (10)

The value of $<R^2(\tau)>$ is reduced for an electron by integrating with the normalized
starting velocity distribution $\Psi_{s_n}(v_0)$, and $D_T$ is obtained using the mean flight time $\langle \tau \rangle = \langle Gf1 \rangle$ as

$$D_T = \langle G(v_0^2 \sin^2 \theta_0 \tau^2) \rangle / 4<Gr> = \langle Gf(v_0^2 \sin^2 \theta_0 \tau) \rangle / 2\langle Gf1 \rangle.$$ \hspace{1cm} (11)

In this expression for $D_T$, the flight time $\tau$ depends on the total collision frequency $v_T$ but not on the momentum-transfer collision frequency $v_m$. Even if anisotropic scattering takes place, the distribution of starting velocity distribution in the transverse direction must be isotropic, and $D_T$ is considered predominantly depend on the total collision frequency.

3. ANISOTROPIC SCATTERING

Elastic scattering probability with dependence on deflection angle ($\chi$) is usually described by the differential cross section $\sigma_d(v, \chi)$. Here, collisions are assumed all elastic. The integral (total) cross section $q_d(v)$ is obtained as

$$q_d(v) = \int_0^{\pi} 2\pi \sigma_d(v, \chi) \sin \chi d\chi,$$ 

and the momentum-transfer cross section $q_m(v)$ is defined as

$$q_m(v) = \int_0^{\pi} 2\pi \sigma_d(v, \chi)(1-\cos \chi) \sin \chi d\chi.$$ \hspace{1cm} (13)

The total collision frequency $v_T(v)$ and the momentum-transfer collision frequency $v_m(v)$ for electrons in gas of density $N$ are given as $Nq_d(v)v$ and $Nq_m(v)v$, respectively.

Here, the most simple anisotropy in scattering independent on ion velocity $v$ is assumed.

$$4\pi \sigma_d(v, \chi) = q_d(v) g(\chi) = q_d(v)(1+\alpha \cos \chi),$$ \hspace{1cm} (14)

where

$$\int_0^{\pi} g(\chi)(1/2) \sin \chi d\chi = 1.$$

In eq.(14), "$\alpha$" is the anisotropy index having values between -1 and 1. The positive
value of "d" implies a forward dominated scattering, and "d" of negative value gives a backward dominated one. Anisotropy in this form does not give any harmonics in scattered results in itself. The average of the momentum-transfer (loss) ratio in this case is given as

\[ \int_0^\pi (1 - \cos \chi)(1 + a/cos \chi)(1/2) \sin \chi d\chi = 1 - a/3, \quad (15) \]

and the mean momentum conservation ratio is \( a/3 \). It is noted that the mean velocity conservation ratio is \( a/3 \) for arbitrary components of incident velocity. From eq.(15), the momentum-transfer collision frequency is obtained as

\[ v_m(\nu) = v_\alpha(\nu)(1 - a/3) = Nq_\alpha(\nu)\nu(1 - a/3), \quad (16) \]

and the total collision frequency is given as

\[ v_T(\nu) = v_m(\nu)(1 - a/3)^{-1}. \quad (17) \]

When \( v_m(\nu) \) is maintained constant independent of the anisotropy, the total collision frequency \( v_T(\nu) \) varies in proportion to \((1 - a/3)^{-1}\) as are seen in Fig.1. If \( v_T(\nu) \) is maintained constant, \( v_m(\nu) \) changes in proportion to \((1 - a/3)\). In addition, it has to be noted that the velocity distribution in the transverse direction has to be completely isotropic because there are no source to produce anisotropy.

4. ANALYTICAL DERIVATION OF FORMULAE

In the most simple special condition of constant collision frequency, the transport of electrons are analytically calculated. The drift velocity \( W \) is obtained as the ratio between the forward displacement \( <Gf\nu_\nu> \) and the flight time \( <\tau> = v_T^{-1} \) in a flight as

\[ W = \int_0^\pi \int_0^\pi \int_0^\pi (\nu_\nu \cos \theta_\nu + eEt/m)( \Psi_{5\nu}(\nu_\nu) + \Psi_{5\nu1}(\nu_\nu)\cos \theta_\nu ) \] 
\[ \times \exp[-v_T\nu_\nu]d\nu_\nu d\theta_\nu d\nu /<\tau> \]
\[
\begin{align*}
&= \int_0^\infty [ (eE/mv_T^2)^{n_0} + (1/3)v_0^{n_0}v_T^2 ] d\nu_0/(<\tau> \\
&= (eE/m)/(<v_T> + <v_x>) \quad (<\tau> = 1/v_T)
\end{align*}
\]

where \( <v_x> \) under constant collision frequency is given by \( aW/3 \) from eq.(15). From eq.(17), we obtain

\[
W = (eE/m)(1-a/3)v_m^{-1} + aW/3,
\]

\[
= (eE/m)v_m^{-1} = (eE/m)(1-a/3)v_T^{-1}.
\]

\( e \): electronic charge, \( E \): electric field, \( m \): electron mass.

As is seen above, the anisotropic scattering provides an initial forward velocity \( <v_x> = aW/3 \), and the drift velocity \( W \) is determined only by the momentum-transfer collision frequency \( v_m \), in this case, \( 4.4482 \times 10^8 \) \([s^{-1}]\) as

\[
W = (eE/m)v_m = 3.9540 \times 10^4 \quad [\text{ms}^{-1}]
\]

This value is accurately agree with the data by FTI analysis.

The transverse diffusion coefficient \( D_T \) defined in eq.(11) is obtained as

\[
D_T = \frac{<v_0^2}}{4<\tau>}
\]

\[
= \int_0^\infty \int_0^\pi \int_0^\pi (v_0\sin\theta_o)^3 v_o \exp[-v_T] d\theta_o d\phi d\theta_d/4<\tau>
\]

\[
= \frac{<v_0^2>}{3v_T} = \frac{<v_0^2>}{3v_m}.
\]

The denominator is \( v_T \) but not \( v_m \). The diffusion is considered not dependent on the momentum-transfer collision frequency \( v_m \) but obeys to the total collision frequency \( v_T \). Nevertheless, in the kinetic theory, the diffusion coefficient has hitherto been described as
\[ D'_f = \langle v^2 \rangle / 3 \langle v_m \rangle \]
\[ = \langle v^2 \rangle / 3 \langle v_m \rangle. \quad \text{ (in const. coll. freq.)} \quad (22) \]

From eq. (21), the value of \( 3D_f / \Omega K \) is obtained as
\[ 3D_f / \Omega K = \left( \frac{\langle v_0^2 \rangle \langle v_f \rangle}{\langle v_m \rangle} / 2 \left[ (eE/m) / \langle v_m \rangle \right] \right) \]
\[ = \left( v_m / v_f \right) (m v_0^2 / 12 e) = (1 - a / 3) \langle \varepsilon_0 \rangle. \quad \text{[eV]} \quad (23) \]

This result shows that the \( 3/2 \) times of characteristic energy \( D_f / K \) gives the \( (1 - a / 3) \) times of the starting energy \( \langle \varepsilon_0 \rangle \) but not of the mean energy \( \langle \varepsilon \rangle \) in flight. Meanwhile, the problem due to the difference between \( \langle \varepsilon_0 \rangle \) and \( \langle \varepsilon \rangle \) gives only a small effect because \( \langle \varepsilon_0 \rangle \) and \( \langle \varepsilon \rangle \) have close values as will be seen below. Equation (23) shows, however, that the Einstein relation does not hold in the presence of anisotropic scattering owing to the change of the total collision frequency. It is considered natural that the diffusion coefficient is proportional to the reciprocal of the total collision frequency.

The mean energy in a flight of an electron started with \( \Psi_{sn}(v_0) \) in eq. (2) is expressed as
\[ \langle \varepsilon \rangle = \int_0^\infty \int_0^\pi \int_0^{\pi / 2} (m/2e)[v_0^2 + 2v_0 \cos \theta_0 (eE/m) t + (eE/m)^2] \]
\[ \times \exp[-\nu t] d\theta \left[ \Psi_{sn0}(v_0) + \Psi_{sn1}(v_0) \cos \theta_0 \right] \sin \theta_0 d\theta_0 / \langle \tau \rangle 
\]
\[ = (m/2e) \langle v_0^2 \rangle + \left[ \langle v\rangle \langle \tau \rangle + (eE/m) \langle \tau \rangle^2 \right] E \]
\[ = \langle \varepsilon_0 \rangle + EW \langle \tau \rangle. \quad \text{[eV]} \quad (24) \]

In the case only elastic collision occur, \( \langle \varepsilon_0 \rangle \) is far larger than \( EW \langle \tau \rangle \) due to small energy loss at collision as will be seen in the results (see Tables I and II).
5. RESULTS

In order to understand the most fundamental properties of electron transport in gas under electric field assuming that only elastic collision occurs with constant collision frequency (CCF). Two conditions for the constant collision frequencies are given. One is that the momentum-transfer collision frequency \( v_m \) is constant, and another is that the total collision frequency \( v_T \) is constant, both regardless the change of variable "d" in anisotropy \( g(\chi) = (1 + a/\cos \chi) \). The mass ratio between an electron and a gas atom \( m/M \) is assumed to be \( 10^{-2} \) for the sake of stable analysis, and the average momentum loss ratio at scattering is taken to be \( (m/M)(1-a/3) \). Accordingly, the mean residual ratio of energy through a scattering is given as \( [1-(m/M)(1-a/3)]^2 \).

In Tables I and II, electron transport properties obtained by extended FTI method under constant collision frequencies \( v_m \) and \( v_T \), respectively, are listed with the change of anisotropy index "a" from \(-1\) to \(+1\). Relative changes of main transport quantities are shown in Figs. 1 and 2, in addition to Tables I and II. Assumed conditions and the main changing tendencies of transport properties with the anisotropy index "a" are summarized below.

<table>
<thead>
<tr>
<th>Condition I</th>
<th>Condition II</th>
</tr>
</thead>
<tbody>
<tr>
<td>( v_m ) = constant; ( 4.4482 \times 10^8 ; [s^{-1}] )</td>
<td>( v_T ) = constant; ( 4.4482 \times 10^8 ; [s^{-1}] )</td>
</tr>
<tr>
<td>( v_T = (1-a/3)^{-1} v_m )</td>
<td>( v_m = (1-a/3) v_T )</td>
</tr>
<tr>
<td>&quot;d&quot; = (-1), (-1/2), backward scatt.</td>
<td>&quot;d&quot; = (-1), (-1/2), backward scatt.</td>
</tr>
<tr>
<td>= 0, isotropic scatt.</td>
<td>= 0, isotropic scatt.</td>
</tr>
<tr>
<td>= (1/2), 1, forward scatt.</td>
<td>= (1/2), 1, forward scatt.</td>
</tr>
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</table>

Results I (Table I, Fig.1) Results II (Table II, Fig.2)

- Drift velocity \( W \) = const. \( W \propto (1-a/3)^{-1} \)
- Mean energy \( \langle \varepsilon \rangle \) = const. \( \langle \varepsilon \rangle \propto (1-a/3)^2 \)
- Diffusion coeff. \( D_V, D_T \propto (1-a/3)^{1} \) \( D_V, D_T \propto (1-a/3)^{2} \)
- Mean coll. freq. \( \langle v_T \rangle \propto (1-a/3)^{1} \) \( \langle v_T \rangle \) = const. \( \langle v_T \rangle \) = const.
- Mean flight time \( \langle t \rangle \propto (1-a/3)^{1} \) \( \lambda \propto (1-a/3)^{-1} \)
- Mean free path \( \lambda \propto (1-a/3)^{1} \) \( D_q/K \propto (1-a/3)^{1} \)
- Charact. energy \( D_q/K \propto (1-a/3)^{1} \)
Table I Transport data of CCF electrons for model anisotropy (g(X)=(l+acosX))
with $\gamma_m = 4.4482\times10^8$ [s$^{-1}$], $m/M=0.01$, $N=1\times10^{15}$[m$^{-2}$], $E=100\text{ Vm}^{-1}$.

<table>
<thead>
<tr>
<th>$&quot;a&quot;$</th>
<th>$\gamma_T=\gamma_m/(1-a/3)$</th>
<th>$\gamma_m$</th>
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<tr>
<td></td>
<td>[10$^8$ s$^{-1}$]</td>
<td>[10$^8$ s$^{-1}$]</td>
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<tr>
<th>$\langle v \rangle = \langle v G_{x} \rangle / \langle v_f \rangle$</th>
<th>$[10^8\text{ms}^{-1}]$</th>
<th>$\langle v \rangle = \langle v G_{x} \rangle / \langle v_f \rangle$</th>
<th>$[10^8\text{ms}^{-1}]$</th>
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<td>$3\boldsymbol{D}_{\gamma}/2\kappa$</td>
<td>$[10^{-1}\text{ev}]$</td>
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Table II Transport data of CCF electrons for model anisotropy (g(X)=(l+acosX))
with $\gamma_T = 4.4482\times10^8$ [s$^{-1}$], $m/M=0.01$, $N=1\times10^{15}$[m$^{-2}$], $E=100\text{ Vm}^{-1}$.

<table>
<thead>
<tr>
<th>$&quot;a&quot;$</th>
<th>$\gamma_T = \gamma_m/(1-a/3)$</th>
<th>$\gamma_m$</th>
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<tr>
<td></td>
<td>[10$^8$ s$^{-1}$]</td>
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<th>$\langle v \rangle = \langle v G_{x} \rangle / \langle v_f \rangle$</th>
<th>$[10^8\text{ms}^{-1}]$</th>
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<td>$3\boldsymbol{D}_{\gamma}/2\kappa$</td>
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- 59 -
5. DISCUSSION

The results of trajectory analysis by extended FTI method shown in Tables I and II excellently agree with those analytical calculations. The FTI values and the analytical values using eqs.(20), (21), (23), (24) for \( v_m = 4.4482 \times 10^8 \text{ s}^{-1} \) are compared below,

\[ \nu_m : \text{const.} \]

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<tr>
<th>( a )</th>
<th>-1</th>
<th>-1/2</th>
<th>0</th>
<th>1/2</th>
<th>1</th>
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<tr>
<td>( W ) (FTI) [10^4 ms^{-1}]</td>
<td>3.9541</td>
<td>3.9541</td>
<td>3.9540</td>
<td>3.9538</td>
<td>3.9538</td>
</tr>
<tr>
<td>(ANA) [10^4 ms^{-1}]</td>
<td>3.9540</td>
<td>3.9540</td>
<td>3.9540</td>
<td>3.9540</td>
<td>3.9540</td>
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<tr>
<td>( \langle \tau \rangle ) (FTI) [10^{-9}s]</td>
<td>2.9974</td>
<td>2.6228</td>
<td>2.2481</td>
<td>1.8734</td>
<td>1.4987</td>
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<tr>
<td>(ANA) [10^{-9}s]</td>
<td>1.852</td>
<td>1.0371</td>
<td>0.8889</td>
<td>0.7407</td>
<td>0.5926</td>
</tr>
<tr>
<td>( \langle \varepsilon \rangle ) (FTI) [10^{-1}eV]</td>
<td>0.9735</td>
<td>0.9768</td>
<td>0.9801</td>
<td>0.9834</td>
<td>0.9867</td>
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<tr>
<td>(ANA) [10^{-1}eV]</td>
<td>1.152</td>
<td>1.0371</td>
<td>0.8889</td>
<td>0.7407</td>
<td>0.5926</td>
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<tr>
<td>( D_T ) (FTI) [10^2 ms^{-1}]</td>
<td>4.4725</td>
<td>4.4700</td>
<td>4.4575</td>
<td>4.4555</td>
<td>4.4640</td>
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<td>(ANA) [10^2 ms^{-1}]</td>
<td>4.4741</td>
<td>4.4702</td>
<td>4.4688</td>
<td>4.4628</td>
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<td>( 3D_T/2K ) (FTI) [10^{-1}eV]</td>
<td>5.8050</td>
<td>5.0938</td>
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<td>3.6596</td>
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<tr>
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<td>5.8052</td>
<td>5.0941</td>
<td>4.3778</td>
<td>3.6576</td>
<td>2.9333</td>
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The variations of the data with the change of "a" under constant $v_m$ and constant $v_r$ are quite systematic as are observed in Figs.1 and 2. It is well understood that the electron transport properties such as the drift velocity and the mean energy do not change with the change of anisotropy as far as the momentum-transfer collision frequency $v_m$ is kept constant. However, the constant $v_m$ does not give constant values for the transverse diffusion coefficient $D_T$. When the starting energy $e_0$ is constant independent of anisotropy, $D_T$ changes in proportion to the reciprocal of the total collision frequency $v_T$. If eq.(21) does not hold, the reason for it must be that the vanishment of cross terms in eq.(6) is imperfect. If so, however, the imperfect vanishment of cross terms will not be able just to keep $D_T$ constant regardless of "a". $D_T$ values given by the multi-term analysis (MTA) of the Boltzmann equation have a constant value the same as that in isotropic scattering independent of "a". The reason for it seems to be that $D_T$ must obey $v_m$ as in eq.(22) according to the kinetic theory. Therefore, it is considered that the definitions for $D_T$ in MTA and in FTC are different. Nevertheless, we believe that the microscopic trajectory analysis can provide correct values of $D_T$. It is quite natural that the increase of the total collision frequency $v_T$ will decrease the diffusion coefficient in proportion to the reciprocal of $v_T$. The momentum-transfer cross section can give the memory of anisotropic scattering toward the incident direction in a scattering, but the memory of the incident direction disappears within two or three cascading scatterings as a mean. Therefore, the momentum-transfer cross section seems to be effective only under a continuous force acting on electrons in one direction, such as the electric field force which provides the drift motion being dependent on $v_m$ as was confirmed in this paper. Concerning to the transverse diffusion coefficient $D_T$, there are no applied forces, and $D_T$ will not depend on $v_m$ but may be dependent only on $v_T$.

References

(6) R.E.Robson: private communication.
Response of Imaging Plate to Radiation Energy

Masahiro TAKEBE, Ken ABE, Yoshiyuki SATOH, Manabu SOUDA, Yasuhiro KONDO
Tohoku Univ. Faculty of Engi.

1. INTRODUCTION

Last year, we reported that the stimulation spectra of Imaging plates (IP) show some radiation properties(1). Those results, however, were obtained with relatively rough measurements. We have assured previous data by measuring with a more elaborate instrument than before. The stimulation spectra of imaging plates irradiated with electrons and protons were measured and confirmed the previous observation again. The phenomena may be applied to radiation detection and the discrimination of radiation properties may be valuable in many cases.

2. EXPERIMENTAL

Lights emitted from W-lamp were monochromated with a grating and shorter wave-length lights were cut with a 450nm cut-filter. In order to support multiple read-out measurements, we reduced the light power to about 10% with inserting a ND filter. The illuminated area by stimulation lights on a piece (1 cm x 2 cm) of the IP (original size is 5" x 5" BAS UR3000) was about 5x4mm in oval shape. The photostimulated emission lights were collected with a parabolic mirror and guided to a photomultiplier tube (HAMAMATSU R-955) through a bandpass filter at 400nm having 20nm pass band. The output current of the tube was measured with a TAKEDA RIKEN electrto-ammeter which was connected to an NEC PC-98 personal computer. With scanning wave-length from 440 to 750nm in a 1nm step, we observed the stimulation spectra. PSL intensities of
IP decrease significantly just after radiation irradiation. To minimize experimental error the test pieces of IP irradiated were kept in a dark box and PSL was measured 18hrs after the irradiation. The irradiation amount (particle fluence) was about $2 \times 10^9$ electrons or protons per cm$^2$. The irradiation particle energies of protons and electrons were ranged from 1.5 to 3.2MeV and from 40 to 1250keV, respectively. Because of high intensity of the Dynamitron in our Fast Neutron Laboratory, we reduced and diffused the primary proton beam with using Rutherford scattering of a thin gold foil, 0.2 mm thickness. The electron irradiation was carried out by the three electron microscopes of High Voltage Electron Microscope Laboratory.

3. EXPERIMENTAL RESULTS

It is clearly shown in Fig. 1 that the PSL ratio of 500 to 600nm is decreasing with increasing the proton energy. In other expression, the ratio seems to be proportional to the LET or the initial energy of incident proton beam\(^{(2)}\). A similar feature can be observed in the case of electron irradiation as shown in Fig.2\(^{(3)}\). At high and low energy regions the ratio deviates from a line. We monitored particle fluence with using a current monitor equipped with the electron-microscope. The irradiation condition, however, was drastically different from an ordinary use. It may cause some unexpected error.

4. DISCUSSION

The two reasons for the difference of the stimulation spectra among different irradiation energies are considered. The first is responsible for the readable depth of imaging plate. The phosphor material of BAS3000UR is colored with a blue dye to improve spacial resolution. Therefore red lights are quickly attenuated along with penetrating the phosphor. The attenuation of lights having longer wave length, such as 600nm, results smaller PSL intensities by 600nm than that by 500 nm. In other words, this can be called range effect. This explanation, however, can not be applied to a case of radiations that have high energy enough to go through the IP. We observed the discrimination effect on radiation properties even in the case of high energy electron
irradiation. It suggests another reason. The second is intrinsic LET effect of phosphor that affects the number of electrons in traps of the phosphor. The number of electrons in a trap depends on LET of radiation. This effect may be supported by the following experimental results. According to the experiment by N. Mori\textsuperscript{1}, the electrons having more than 140keV can go through the IP. In the case of electrons having higher energy than 140keV, the range effect hardly interprets the difference between PSL at 500nm and at 600nm.

5. REFERENCES

(3) M. TAKEBE, K. ABE, M. SOUDA, T. SATOH and Y. KONDO. to be published in Nucl. Instr. and Meth.
Fig. 1. PSL ratio of 600nm to 500nm vs. incident proton energy.

Fig. 2 PSL ratio of 600nm to 500nm vs. incident electron energy
LOW ACTIVITY RADIOGRAPHY WITH AN IMAGING PLATE

Ken Abe, Masahiro Takebe, Yoshiyuki Satoh, Yasunori Kokubun and Yasuhiro Kondo
Faculty of Engineering, Tohoku University
Sendai 980-77, Japan

1. INTRODUCTION

The imaging plate (here BAS-UR (1), BaFBrₐ₀.₈₅ₓ₀.₁₅:Eu²⁺ based imaging plate) has many striking performances of radiation detection, such as a simple usage, a large detection area, a high position sensitivity (50-100μm), a high detection sensitivity (100-1000 times as high as an X-ray film), long time dose accumulation, good dose linearity, extremely wide dynamic range of dose (more than 5 decades) and easy erasing for reuse. It can detect even very weak radioactivity of 10⁻⁴ Bq in a spot of 1 mm² or less in area with several hours of exposure (2). The features of the dose accumulation, wide dynamic range and high detection sensitivity will give us a new world of radiography if backed up by a full use of the detectability of extremely low specific radioactivities with further increased S/N (signal-to-noise) ratio, which will surpass the one with the conventional substitution of X-ray films for imaging plates. Recent low level radioactivity imaging of some kind of vegetables (3) and neutron diffraction imaging (4) by use of imaging plates show a step ahead of the radiography.

Enhancement of the S/N ratio as well as reduction of the fading of images due to both temperature and time on the imaging plate is important to low activity radiography because the long time accumulation of exposure dose is indispensable there. Truth to say, improvement of image S/N ratio to the extremely high makes available, for a example, the neutron radiography with very weak neutron sources and/or a rather easy radiography with various radiations of usual radioisotopes. Really, low activity radiography forms the fundamentals common to any type of radiation imaging with the imaging plate.
2. IMAGE BACKGROUND

We do not discuss here the factor S of the S/N ratio at imaging, for example, efficiency of recoil converters for neutron detection, density of spots of PSL (photostimulated luminescence) and others, but do the factor N. The N, mainly usual background fog noises (PSL/cm²/h), could be reduced by good shielding of environmental radiations including the cosmic rays, arrangement of some kind of ROI (region of interest), discrimination of radiation types and energies, and some post procedures. Then first of all, we have studied the shielding of the environmental radiations effective on the imaging plate, and have encountered some difficulties unexpected and unexplainable.

The shielding materials used here are iron box of 92.7 x 93.4 x 111 cm³ in external size equipped with the upper plate of 6 cm in thickness and the other plates of 6.7 cm in thickness that was formed with ancient sunken battleship named Mutsu, completed in 1921, put in CYRIC (Cyclotron and Radioisotopes Center) of our university, iron box of 67 x 57.5 x 63 cm³ in external size with 22 cm thick plates reformed of the ALOKA low level radiation detector system, the observation tunnel 44.5 m underground in Observation Center for Earthquakes and Volcanic Eruptions Prediction of our university, many lead bricks of 5 x 10 x 20 cm³ made about 25 years ago, and a lot of polyethylene bricks, various thickness of lead sheets, metacrylic acid resin plates, oxygen-free copper plates and others.

Fig.1 shows the effectiveness of the fog formation by the environmental radiations on the imaging plates through various thickness of shielding materials composed of iron and lead. The exposure time was 24 hours. The iron was roughly changed to the lead equivalent with conversion factor of mass density ratio of 7.9/11.3. It was found that the reduction of the fog formation is reasonably attained to the extent of nearly 10⁻¹ with a few cm thick lead equivalent, but much more reduction would not be expected with much
thicker lead stacking on the iron box. The effectivenes usually seems to come not only to the bottom for moderate thickness but even to a uphill for extremely thick shield. Much lead shield thicker than 30cm on Mutsu iron box was prohibited due to the endurance limit of the floor. Next, we measured the background fog formation of the imaging plate in the tunnel 44.5m underground with expectation of enough reduction of cosmic rays (mainly muons). Fig.2 shows the result obtained combined with the data given in fig.1. The exposure time was 24 hours. The thickness of lead equivalent was obtained simply with the conversion factor of 2.6/11.3 assuming that the mean mass density of the soil and rocks of the tunnel headgear is 2.6. Radiation background coming from the earth was protected with 5cm thick lead bricks. Total number of the lead bricks was limited to 10 and less in order to avoid troubles given to a lithosphere strain gauge. It is widely known that the muon fluence at the 115mwe deep underground is about $5 \times 10^{-4}$ times as much as the one of the sea level, but in practice the fog noises increased extremely high against such expectation, as is clearly shown in fig.2.

![Fig.2 A combined plot of shield box and tunnel observations](image)

Such a counter result of radiation shielding as the above shown seems fully contradict the common knowledge in the speciality of low activity detection and/or high sensitivity detection always taking with
troublesome and heavy shielding of the cosmic rays, even if it might be somewhat explained by soft showers of the cosmic rays and the extraordinarily low threshold of particle detection of several electron volts after passing through the protection surface layer of the imaging plate, and more others that are, for example, the concentration of radon in air becomes three times as much on the day just after holidays due to the suspension of air conditioning and filtering system during the holidays, the tunnel may have also high density of radon due to bad ventilation and case-dependent beam directions of the cyclotron may often affect the fog formation during the proper exposure and/or transportation of the imaging plate between the shield box and a imaging plate reader. Hereby more supplementary experiments should be done to establish such findings. We are restudying the background fog formation of imaging plate versus thick lead shield so as to get basic data without question, where only lead bricks are used and stacked up in a wide back yard of CYRIC.

3. REGION OF INTEREST OF THE IMAGING PLATE

Fig.3 shows how wide is the dynamic range of exposure dose of the imaging plate in contrast to the X-ray film, which is very often cited to stress the distinguishing merits of the imaging plate (IP). However, is it really a merit even in low activity radiography?

The imaging plate has, roughly speaking, no cutoff threshold of detection in a sense that it can detect even some electron volts radiations and so weak radiations, which makes a distinct contrast with other usual radiation detectors of some keV cutoff and of some spatial intensity cutoff. The nuclear emulsions sensitive to minimum ionization are delicate and limited in use. High gain electronic amplifiers equipped with no discriminator can often hardly work stably. The extremely low threshold
of the imaging plate is a two-edged sword for radiation detection, specially in low activity radiography. Creation of definite ROI windows in energy and/or in dose is fruitful.

4. PARTICLE DISCRIMINATION, POST TREATMENT AND OTHERS

Discrimination of radiation types\(^{(7)}\) has clear utility in low activity radiography with good S/N ratio even without the ROI setting. Energy determination of radiation particles\(^{(8)}\) will also work effectively there. They are, however, concerning the S of S/N which are not discussed here. Further investigations, namely, selective fading of the images, operational reduction of background noises, a new look of muon attenuation versus shielding material for extremely low detection threshold and others with a view to low activity and high sensitivity radiography are in progress.

References

PHOTOSTIMULABLE SrS:Eu,Sm PHOSPHOR AS A STORAGE MATERIAL FOR TWO-DIMENSIONAL X-RAY IMAGE SENSOR


Electron Device System Research Laboratory, Kanazawa Institute of Technology, 7-1 Oogigaoka, Nonoichimachi, Ishikawa 921 Japan

1 Industrial Research Institute of Ishikawa, 1 Tomizu-machi, Kanazawa 920-02 Japan

1.INTRODUCTION

Photostimulated luminescence (PSL) phenomenon is based on the presence of electron and/or hole traps and luminescence centers in storage phosphor materials. During exposure to electromagnetic waves such as X-ray, the traps are occupied with charged carriers such as electrons and holes that are induced by the electromagnetic wave irradiation. Detrapping of these carriers requires energy. The energy is provided by stimulating the phosphor material with visible or infrared light. During a detrapping transition, free carriers created by the absorption of photons from the stimulating radiation field recombine with the luminescence centers, whereby visible photons (PSL) are emitted. Since the PSL intensity is in the most cases proportional to the dose of electromagnetic wave, the phosphor materials which exhibit the PSL phenomenon offer an alternative to conventional two-dimensional imaging technique for ionizing radiation such as X-ray, γ-ray and neutron-ray. In this paper, we report the first observation of intense PSL in Eu and Sm co-doped SrS (SrS:Eu,Sm) phosphor at room temperature (RT) and the PSL characteristics of SrS:Eu,Sm
phosphor as a storage material for two-dimensional imaging sensor for the electromagnetic waves such as X-ray.

2. EXPERIMENTAL

In the preparation processes of SrS:Eu,Sm phosphor ceramics, a mixture of 0.1 [mol%] Eu and Sm co-doped SrS phosphor powders and suitable binders was formed into disk at 0.03 [ton/mm²] and was then sintered at 700-1100 [°C] for 4 [hr] in air. The ceramic disks with 13 [mm] diam. and 1 [mm] thick were used as specimens. X-ray irradiation was carried out at RT in the dark with a copper target operated at 20 [kV] and 2 [mA]. The Xenon-lamp was used as the UV-ray and visible-ray sources. The PSL emission spectra and photoluminescence (PL) spectra were observed using a Hitachi F-3010 spectrofluorometer at RT. The PSL and PL spectra were corrected for the diffraction of the grating and the optical response of the photomultiplier.

3. RESULTS AND DISCUSSION

An efficient PSL peak at about 600 nm was observed when the X-ray-irradiated specimen prepared by sintering at 1100 [°C] was stimulated with 800-1500 [nm] light, although a weak PSL emission was observed for the specimens prepared by sintering below 1000 [°C]. Typical PSL emission spectrum and stimulation spectrum from X-ray irradiated specimen are shown in Fig.1. Figure 2 shows typical PL emission spectrum (solid line) and its excitation spectrum (dotted line) of SrS:Eu,Sm phosphor ceramics. The peak at about 460 [nm] in the excitation spectrum is due to the (4f→5d) transition of Eu²⁺ ions. The peak at about 280 [nm] is due to the band to band transition in SrS since the band gap energy of SrS phosphor is about 4.3 [eV] (280 [nm]). It should be noted that the PSL spectrum as shown in Fig.1 coincides with the PL spectrum. This result strongly suggests that the 600 [nm] PSL peak is attributed to the ionic
transition (5d→4f) of isolated Eu$^{2+}$ ions which occupy cation sites in SrS phosphor. It has been reported that in CaS:Eu,Sm and SrS:Eu,Sm phosphor the Eu$^{2+}$ and Sm$^{3+}$ ions act as hole and electron traps, respectively.\(^{(7)}\)

So, the following excitation and emission mechanisms for the 600 [nm] PSL peak is likely. Part of free electrons and holes produced during X-ray irradiation are trapped at Sm$^{3+}$ and Eu$^{2+}$ ions, respectively, and consequently Sm$^{2+}$ and Eu$^{3+}$ ions are created in SrS phosphor. By stimulation with 800-1500 [nm] light, electrons which are optically released from the Sm$^{2+}$ ions
recombine with Eu$^{2+}$ leading to the excited Eu$^{2+}$ ions from which the 600 [nm] PSL is emitted. It was found that the PSL intensity is increased with increasing X-ray irradiation dose. Figure 3 shows the PSL intensity as a function of X-ray dose. It can be seen that there are good linearity between the PSL intensity and X-ray. X-ray irradiated specimen exhibited a fairly good fading characteristics as shown in Fig.4. These results suggest that the SrS:Eu,Sm phosphor is useful as a storage material for two-dimensional X-ray imaging sensor.
Fig. 3 PSL intensity as a function of X-ray irradiation dose in SrS:Eu,Sm phosphor ceramics.

Fig. 4 PSL fading characteristics of X-ray irradiated SrS:Eu,Sm phosphor ceramics at RT in the dark.
4. CONCLUSION

An intense PSL with a peak at about 600 [nm] was observed by stimulating X-ray irradiated SrS:Eu,Sm phosphors ceramics with 800-1500 [nm] light. Possible excitation and emission mechanisms for the PSL with a peak at about 600 [nm] for the excitation with X-ray are proposed. It is found that the PSL intensity is proportional to X-ray irradiation dose. These results strongly suggest that the PSL phenomenon in SrS:Eu,Sm phosphor is useful as a novel candidate for two-dimensional X-ray imaging sensor utilizing PSL phenomenon.

5. ACKNOWLEDGMENT

We wish to thank Prof. K. Inabe of Kanazawa University for his fruitful discussion and Y. Hirai, S. Ueda, K. Kawabe and S. Ueki for their excellent assistance in the experiments. This work was partly supported by a Grant-in-Aid for Scientific Research (No. 05245214 and No. 06236101) from the Ministry of Education, Science and Culture of Japan.

References


The Development of a Radiation Monitor for Incident Position Detection

Toru Oka, Kazunori Ikegami, Seisaku Imagawa and Teruo Usami
Mitsubishi Electric Corporation
1-1, Tsukaguchi Honmachi 8chome, Amagasaki, Hyogo, 661 Japan

1. Introduction

High reliability is required in the control and measurement system of nuclear power plants in order to meet safety standards. Radiation detectors using a plastic scintillation fiber (PSF) have the potential to be very reliable radiation sensing systems. The main features of the PSF are its flexibility, its possibility of covering a long surface of detection and its immunity to external electromagnetic noise. Recently, the PSF has been developed and used by some universities and corporations\textsuperscript{1,2,3,4}, but mainly for experiments of high energy physics\textsuperscript{5,6}, because they are more sensitive to charged particles than to $\gamma$ rays and X rays.

Several kinds of radiation detectors are installed in a nuclear power plant. The area radiation monitor installed in a plant building mainly measures $\gamma$ rays to prevent overexposure to human body. The monitors are installed only at a few points and their diameter is only about 20cm. So, many radiation monitors are required to observe a large area in a nuclear power plant.

The radiation detector using a PSF has been developed in our study. This detector has the ability of measuring a distribution of dose equivalent in a large area. A more accurate spatial information of the radiation is obtained by using this detector, so safer working places for plant workers can be determined. Consequently, high reliability and safety of the plant are achieved.

In this paper, the experiments of new system using delay fiber which can replace the conventional system will be made.

2. Principle

The time of flight (TOF) method adopted in our experiments measures the time difference between two signals reaching separate detectors. When a radiation crosses the PSF, scintillation occurs in the core of the PSF and produces a light emission. The light trapped in the core travels to both ends of the fiber and finally reaches the extremities connected with photomultiplier tubes (PMTs). The time the light takes to reach the respective PMT is different, depending on the crossing position of the radiation. So if the time difference can be detected, the crossing position of the
radiations can be measured. When the distance from the crossing position to one of the PMTs is \( \ell_i \), the distance to the other PMT is \( \ell_{psf} - \ell_i \), where \( \ell_{psf} \) is the length of the PSF. So the time difference \( T \) is written as follows:

\[
T = \frac{2 \ell_i - \ell_{psf}}{v_i}
\]

where

\( v_i \): the speed of light in the core.

For every light emission produced by a crossing radiation, a signal proportional to the time difference can be observed. By integrating the distribution of counts found at every position, the total number of pulses detected by the system is obtained, this corresponds to the dose equivalent.

The schematic diagram of our radiation detector system using a PSF is shown in Fig. 1. A PMT, a preamplifier and a constant fraction discriminator (CFD) which generates a timing pulse are connected to the respective extremities of the PSF. A time to amplitude converter (TAC) generates an output signal of height proportional to the time difference between the two signals from the CFDs. Last of all, a multi channel analyzer (MCA) counts the pulses from the TAC depending on their height.

![Fig. 1 Principle of our PSF radiation detector](image)

3. Experimental Setup

3-1. Conventional system

In Fig. 2, which shows the setup of our conventional system, \( \ell_i \) indicates the distance from the crossing position of the radiation to PMT1, and \( \ell_{psf} \) indicates the length of the PSF.
The TAC requires two pulse signals, a start pulse and a stop pulse, and generates a pulse proportional to the time difference between start pulse and stop pulse. The TAC has a "dead time" of 5ns, therefore, the time interval between start pulse and stop pulse must be set to be more than 5ns. So, a delay cable or a delay circuit must be connected between the CFDs and the TAC.

The time difference measured by the TAC can be calculated as follows:

The speed of light in vacuum $c_0$ is approximately $3.0 \times 10^8$ m/s, but in the fiber whose refractive index $n$ is approximately 1.6, the speed of light $v_1$ becomes:

$$v_1 = \frac{c_0}{n} = 1.9 \times 10^8 \text{ (m/s)}$$

(2)

The two light pulses are converted into two electric signals at the PMTs and finally both reach the TAC.

Now, the time difference $T_e$ is:

$$T_e = \frac{2 \ell_1 - \ell_{PSF}}{v_1} + \frac{\ell_2 - \ell_b}{v_c}$$

(3)

where

- $\ell_1$: the length of the electric cable between the CFD1 and the TAC,
- $\ell_{PSF}$: the length of the PSF,
- $\ell_2$: the length of the electric cable between the CFD2 and the TAC,
- $\ell_b$: the length of the electric cable between the CFD2 and the PSF,
- $v_c$: the speed of signals in the electric cable.
When a $\gamma$ ray crosses at $\ell_1 = 0$, $T_e$ is minimum. The minimum time difference $T_{\text{min}}$ is:

$$T_{\text{min}} = -\ell_{\text{PSF}}/v_t + (\ell_a - \ell_b)/v_t \quad (4)$$

So, $(\ell_a - \ell_b)$ can be decided as:

$$\ell_a - \ell_b \geq (T_{\text{dead}} + \ell_{\text{PSF}}/v_t) \times v_t \quad (5)$$

where $T_{\text{dead}}$ is the dead time of the TAC.

3-2. New system using a delay fiber

The TAC presented in section 3-1, required that the start pulse appears always on the same connector and the stop pulse had to follow on the other connector with a delay greater than the "dead time". So a delay cable or a delay circuit had to be used on one end. This section proposes a new system with a delay fiber (DF) that is an alternative to conventional systems like the one presented in section 3-1.

The idea is to insert a DF in the center of the PSF and use a TAC that accepts start and stop pulses on anyone of the connectors. Fig. 3 shows the system, where $\ell_1$ indicates the distance from PMT1 to the crossing position of the radiation, $\ell_{\text{PSF}}$ indicates the length of both PSF1 and PSF2, $\ell_D$ indicates the length of the DF.

The time difference $T_e$ measured by the TAC can be calculated as follows:

As the detector of this system is symmetrical, the following equation considers only the case when radiation crosses PSF1.

$$T_e = 2(\ell_1 - \ell_{\text{PSF}})/v_t + \ell_D/v_D \quad (6)$$

where

$v_t$: the speed of the light pulse in the PSF,
$v_D$: the speed of the light pulse in the DF.

When a $\gamma$ ray crosses at $\ell_1 = \ell_{\text{PSF}}$, $T_e$ is minimum and this minimum time difference $T_{\text{min}}$ is:

$$T_{\text{min}} = \ell_D/v_D \quad (7)$$

— 81 —
So the length of the DF can be decided as:

$$\ell_D \approx T_{\text{delay}} \times v_D$$  \hspace{1cm} (8)

One notices that, in this new system with DF, the length of the DF is independent of the length of the PSF. This is not the case in conventional systems, where the length of the delay cable, or, the delay time of the delay circuit, depends on the length of the PSF. When we need a long scintillation fiber, the delay time of a delay cable or circuit must be set long, accordingly. In this case, the timing fluctuation of the input pulse of the TAC gets big. This causes a low position resolution. In addition, the delay function is free from electromagnetic noise. So, the system using delay fiber is effective when the scintillation fiber is long.

![Fig.3 The system using delay fiber](image)

4. Experiments

Several experiments using different types of DFs are made and the fundamental characteristics are evaluated.

When the DF is connected with the PSF there is an insertion loss and as a result the position resolution may go down. In order to evaluate the influence of the insertion loss, three experiments, whose setup are shown in Fig. 4, are made for each type of DF.

In first experiment only a PSF is used, see Fig. 4(a), in second experiment a PSF and a DF are used, see Fig. 4(b), and in third experiment two PSFs and a DF are used, see Fig. 4(c). The diameter and length of the PSF being used are 1 mm and 1.5 m and the distance from PMT1 to the crossing position of the radiation is $\ell_1$. Two types of DF, both of same diameter and length,
respectively 1mm and 1m, are used for each experiment. Table 1 shows the features of the PSF and the DFs.

![Fig. 4 The procedure of the experiment](image)

**Table 1 The features of the PSF and DFs**

<table>
<thead>
<tr>
<th></th>
<th>PSF</th>
<th>DF1</th>
<th>DF2</th>
</tr>
</thead>
<tbody>
<tr>
<td>core material</td>
<td>polystyrene</td>
<td>PMMA</td>
<td>silica</td>
</tr>
<tr>
<td>type of connector</td>
<td>FC</td>
<td>FC</td>
<td>FC</td>
</tr>
<tr>
<td>NA (Numerical Aperture)</td>
<td>0.58</td>
<td>0.50</td>
<td>0.37</td>
</tr>
<tr>
<td>refractive index of core</td>
<td>1.6</td>
<td>1.49</td>
<td>1.46</td>
</tr>
<tr>
<td>transmittance (/m)</td>
<td>0.75</td>
<td>0.98</td>
<td>0.99</td>
</tr>
</tbody>
</table>
5. Results and Discussion

Fig.5 and Fig.6 show the results for DF1 and DF2 respectively. Table 2 and Table 3 sum up the peak counts and FWHM of the curves in Fig 5 and Fig.6 respectively. Below the peak count value is also indicated the percentage compared to the previous peak count in left cell.

![Fig.5 The results for DF1](image)

**Table 2 The peak counts and FWHM for DF1**

<table>
<thead>
<tr>
<th></th>
<th>PSF1</th>
<th>PSF1+DF1</th>
<th>PSF1+DF1+PSF2</th>
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<tbody>
<tr>
<td><strong>peak counts</strong></td>
<td>441</td>
<td>391 (88.7%)</td>
<td>174 (44.5%)</td>
</tr>
<tr>
<td><strong>FWHM (cm)</strong></td>
<td>44</td>
<td>47</td>
<td>49</td>
</tr>
</tbody>
</table>

![Fig.6 The results for DF2](image)
Table 3 The peak counts and FWHM for DF2

<table>
<thead>
<tr>
<th></th>
<th>PSF1</th>
<th>PSF1+DF2</th>
<th>PSF1+DF2+PSF2</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>peak counts</strong></td>
<td>441</td>
<td>322 (73.0%)</td>
<td>160 (49.7%)</td>
</tr>
<tr>
<td><strong>FWHM (cm)</strong></td>
<td>44</td>
<td>45</td>
<td>49</td>
</tr>
</tbody>
</table>

When comparing the peak counts for the experiment of Fig.4(b), one notices that the insertion loss using DF1 is less than with DF2. But for the experiment of Fig.4(c), the percentages of peak counts for DF1 and DF2 are almost the same. The reason is that when light is inserted from a fiber with large Numerical Aperture (NA) to a fiber with small NA, the difference of NAs has a strong influence on the insertion loss. The insertion loss increases when the difference of NA gets bigger. But, on the other hand, when light is inserted from a fiber of small NA to a fiber of larger NA, the insertion loss is independent of any difference of NA.

The reason why the peak goes down anyway for the PSF1+DF+PSF2 setup is mainly due to the attenuation by PSF2. The experiments also indicate that FWHM spreading is very small.

Fig.7 and Fig.8 show comparison between using DF1 and using a delay cable or circuit. In these experiments we use the PSF of 3m long. In all results, almost same FWHM is observed.

It has been demonstrated that the system with DF can effectively replace conventional systems and DF made from plastic (DF1) is more effective than silica (DF2) when the length of the DF is about 1m.
6. Conclusion

The conclusion of this research are summarized as follows.
(1) The system with a delay fiber (DF) can effectively replace conventional systems.
(2) The plastic DF is more effective than silica DF.
(3) Hereafter, we will develop the radiation detector using long PSFs and investigate the effects of the DF.

References

DEVELOPMENT OF A LARGE VOLUME
POSITION SENSITIVE DETECTOR

S.Matsuyama, D.Soda, M.Baba, S.Iwasaki, T.Ohkubo, M.Ibaraki, Y.Nauchi and N.Hirakawa
Department of Nuclear Engineering, Tohoku University, Sendai 980-77, Japan

1. Introduction

We have been conducting measurements of double-differential neutron emission cross sections (DDX) for secondary neutron energy down to 300 keV using a 4.5 MV Dynamitron time-of-flight (TOF) spectrometer for 1-6 MeV, 14 and 18 MeV incident neutrons. A conventional NE213 scintillation detector, 14 cm \( \times \) 10 cm thick or 5" \( \times \) 2" thick, gave sufficient counting rate with suitable energy resolution for 1-6 MeV and 14 MeV neutrons.

Recently we planned to develop a high-efficiency neutron detector to obtain better energy resolution at 18 MeV and to apply the low intensity \( ^{15} \text{N}(d,n) \) neutron source to DDX measurements around 11 MeV. We also planned to make a position sensitive neutron detector to obtain angle-dependent neutron emission spectra in a single measurement. In order to satisfy both requirements, we developed a large volume (6.5 cm \( \times \) 9.5 cm \( \times \) 80 cm) NE213 liquid scintillation detector (LLSD) with two photomultipliers. The LLSD can be used as a single large detector by placing vertically along the scattering plane as well as a wide-angle scattering detector by placing horizontally on a scattering plane. Such a long scintillation detector was applied by other authors. However, those applications were restricted only to neutrons higher than tens MeV. In the present applications, we intend to apply to lower energy neutrons down to \( \sim 1 \text{ MeV} \) and to achieve n-\( \gamma \) discrimination by applying updated data analysis method. This paper presents the details of the design, data acquisition system and the performance of the LLSD.

2. Detector system

2.1 Detector Design

The detector consists of a long scintillator cell and two photomultipliers. The scintillation lights are detected by two photomultipliers. The difference of light traveling time gives position information. On the other hand, this time difference causes ambiguity in determina-
tion of neutron energy. Then, a time compensator is employed to overcome this problem.

In such a long scintillator, the light output depends on event position, since the light receives strong attenuation by absorption in the scintillator and by the tank wall. This light attenuation causes a position dependance of the detection efficiency if detection threshold is set independently of the event position. Furthermore, the manner of position dependance of the detection efficiency changes with neutron energy which causes the ambiguity in the measurements of angular distributions. This problem will be conquered by adopting position-dependent biasing analysis described later.

In designing the LLSD, good timing resolution and mild light attenuation property are required. Timing and light attenuation properties are influenced by reflective materials, shape of the cell and materials of the cell. We selected glass tube for the scintillator cell. Cylindrical and rectangular types were considered for the detector shape. We selected rectangular type, since the rectangular shaped detector was superior properties in light attenuation and transmissibility. There are many types of reflectors. In this study, we considered three types of the reflective materials: 1) specular coating, 2) no coating and 3) black coating. Diffuse reflector used in small scintillators was not considered since it causes the loss of position information.

Timing and light attenuation properties were examined by experiment and calculation\(^5\). Experimental and calculated results showed that the no coating was superior in timing and light attenuation properties. The calculated light attenuation curve for different reflector types is shown in Fig. 1. The result for specular coating shows the highest light correction efficiency, while pulse height variation is largest. The light collection efficiency for the black coating is too low for the present purpose.

The detector size is 80-cm-long, 10-cm wide and 6.5-cm thick in inner dimensions, which was chosen by another physical requirement. The detector thickness was determined considering energy resolution in TOF measurements. The width was chosen to couple directly to a 5" photomultiplier to reduce the loss of light collection.

2.2 Mechanical Design

The schematic view of the LLSD is shown in Fig. 2. The cell is made with 5-mm-thick Pyrex glass and has a long reservoir cell (about 1 l) on the side of the cell. The face of the cell is not coated with a reflector to give best time resolution. Two fast photomultipliers, HAMAMATSU R-1250, are attached directly on both ends of the cell and supported by springs from the frame. The cell, photomultipliers and pre-amplifiers are encased in an aluminum box whose inside is painted in black to avoid light reflection. The size of the box is 198
$X \times 198 \times 1800$ mm. The box has thin windows of 0.5-mm-thick in front and rear sides of the cell.

3. Basic Performance of the LLSD

3.1 Light Attenuation

The light attenuation curve as a function of the distance between neutron incident position and the photomultiplier was measured by observing by the proton edges of 14.1 MeV neutrons. Figure 3 shows the results in comparison with calculated one. The experimental data are normalized to calculated ones at Fig.3. It indicates that the LLSD performs as expected by the simulation. The pulse height variation is large mainly because of large light attenuation in the glass cell. This large pulse height variation can be reduced by separating photomultiplier from scintillator cell as shown by dotted lines for separate contact in Fig 3. In the close contact, photomultiplier is in contact directly with scintillator cell via an optical compound. The difference between close contact and separate contact cases is understood by the difference of critical angle for light reflection ($41.5^\circ$ for glass to glass contact). Figure 4 shows the calculated distributions of incident angle at the end of the scintillator cell as a function of the light emission position. The nearer the emission position, the light component whose incident angle is larger than $41.5^\circ$, thereby, pulse height variation reduces in the separate contact case. As for light attenuation properties, therefore, separate contact case is superior to close contact case. However, the separate contact case proved to deteriorate the pulse shape spectrum which is indispensable for neutron and $\gamma$ separation, Figure 5 shows the pulse shape spectrum for the close contact and separate contact cases. The left and right peaks correspond to neutron and $\gamma$ events, respectively. It shows that the separation between neutron and $\gamma$ peaks is clearer for the contact case than for the separate case, probably light collection efficiency.

The pulse height variation can be reduced by summing both photomultiplier signals, but not eliminated completely. Therefore, we adopt position dependant threshold level described in the next section.

3.2 n-$\gamma$ Discrimination

The performance of n-$\gamma$ pulse shape discrimination is limited by light absorption in the scintillator. Figure 6(a) shows n-$\gamma$ pulse shape spectra for 14.1 MeV neutrons derived from each photomultiplier as a function of the distance of neutron incident positions from the
photomultiplier. Bias level is 2 MeV proton. It shows that the positions of two peaks and the valley move slightly with the distance between photomultiplier and event position. Nevertheless, the separation between n and γ peaks is fairly good in the half region near to the photomultiplier. In early studies, the photomultiplier was equipped with each own n-γ discriminator and the n-γ discrimination signal was taken from the one closer to the event position (OR method). Later, we noticed that n-γ pulse shape spectra using the sum signal of both photomultiplier signals (sum method) provided better result as shown in Fig.7(b). By using the sum signal, position dependance of n-γ properties was almost removed. The sum method is superior not only in performance but also and in simplicity of the circuit.

4. Data acquisition and reduction

4.1 Data acquisition

Figure 7 shows the electronics circuit diagram adopted in this study. Each anode signal from two photomultipliers (PM1 and PM2) is fed into a constant-fraction timing discriminator (CFTD) and converted into a fast logic signal. The fast logic signals are distributed into two circuits for position TOF (P-TOF) and time compensator. P-TOF is derived from the time difference between two photomultiplier signals. Time compensated signals are derived using a mean timer (MT), LeCloy model 624. Those signals are distributed for energy TOF (E-TOF) circuit and for n-γ discriminator. E-TOF is obtained by measuring time differences between the time compensated signals from MT and delayed beam pick-off signals using a time-to-amplitude converter (TAC). The n-γ pulse shape discrimination is made similarly with a zero-crossing method. The dynode signals are summed and fed into timing single-channel analyzer (TSCA), CANBERRA 2035A, and converted into a pulse shape signal. Time difference between TSCA output and delayed MT output is analyzed by TAC and changed to neutron gate signals. Three set of data for E-TOF, P-TOF and pulse height signals are gated by n-γ discrimination signals, and then acquired event by event on a magnetic optical disk by a 3-parameter data acquisition system.

4.2 Data reduction

The three parameter data are analyzed event by event to derive information on neutron energy and incident position after correction for bias, efficiency and backgrounds. The neutron energy was derived from flight time and flight path length for each position.
We set position dependent bias levels by software processing to obtain position independent efficiency using the experimental light attenuation data. Figure 8 shows the bias curve as a function of event position for the proton edges by incident neutrons. The measured relative detection efficiency is shown in Fig. 9 as a function of detector positions for 18 MeV neutrons. The position dependance is eliminated almost completely by the technique. The slant in the PM2 side is due to non uniform dimension of the cell.

Position resolution of the LLSD was measured by using neutron beams collimated to around 4 cm by polyethylene collimator of 40 cm long. Figure 10 shows the position resolution and position linearity for 18 MeV neutrons. The measured resolution is better than 7 cm in FWHM, which correspond to 5 ~ 6 cm inherent detector resolution if the width of neutron beam is considered. The timing resolution is about 650 ps.

5. Application of the LLSD to the neutron scattering experiments

The performance of the LLSD as a position sensitive scattering detector was examined for 14.1 MeV incident neutrons by measuring the secondary neutron TOF spectra from polyethylene. Measurements were carried out using Tohoku University 4.5 MV Dynamitron TOF spectrometer. The experimental setup is shown in Fig. 11. The scattering sample was suspended parallel to the beam axis at 90°. The flight path was around 4 m, which was the shortest one available in the Dynamitron TOF spectrometer. In this arrangement, the scattering angle range covered by the LLSD was around 8° and the scattering cross section data for 7 angles could be obtained.

Figure 12 shows E-TOF vs P-TOF matrix for polyethylene sample. The decrease in neutron energy of the H(n,n)H peak with the increase of scattering angle is observed clearly between elastic and the first-level state inelastic peaks of carbon. Therefore, by using a shorter flight path, the LLSD will provide neutron scattering data over much wider angle range.

The LLSD has been also applied to DDX measurements as a single large detector. The efficiency of the LLSD is three times as large as that of the 14 cm φ × 10 cm thick detector used in previous studies. Therefore, the LLSD is applicable to the measurements employing a low intensity neutron source and a longer flight path without loss of counting rate. We applied the LLSD to the T(d,n) 18 MeV source and the 15N(d,n) 11.5 MeV source with lower strength than 14 MeV source. Previously, we could obtain adequate counting rate with a 14 cm φ × 10 cm thick NE213 detector if the flight path was 4 ~ 5 m. However, the short flight path and detector thickness restricted energy resolution. By using the LLSD, energy resolution could be improved without losing counting rate with an experimental setup almost the same as
that in previous studies\textsuperscript{(1,2)}. The scattering samples were metallic cylinders of elemental silicon. Those were suspended with its vertical axis at 12 cm from the target. The flight path length was 6 m. Typical example of TOF spectra for Si at 18 MeV is shown in Fig.13. Bias level was set at 1 MeV proton. Signal-to-noise is sufficiently good for the DDX measurements. Figure 14 shows the typical results of silicon. The first excited state of Si-28 is sufficiently separated from the ground state.

5. Summary

We have developed a large volume NE213 liquid scintillation detector (LLSD) which is useful as a position sensitive detector and as a high-efficiency neutron detector for the measurements of neutron emission cross sections. The three-parameter date acquisition and position-dependent biasing analysis realized uniform detection efficiency. The performance test showed that the LLSD has good timing resolution and superior signal-to-noise ratio even for bias level around 1 MeV of proton.

We applied the LLSD successfully to the double-differential neutron emission cross section measurements. The high efficiency enabled high-resolution measurements with low intensity neutron source. Furthermore, the position sensitive property realized a spectrometer covering wide range of scattering angle.

The LLSD will be a powerful means not only for DDX measurements but also for other neutron experiments.

References

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(6) S.Iwasaki et al., NETU-49 (Dept. Nucl. Eng., Tohoku Univ.,1987) p.8
Fig. 1 Calculated light attenuation curve for different reflector types.

Fig. 2 Schematic view of the LLSD.

Fig. 3 Measured and calculated light attenuation curve.

Fig. 4 Angular distribution of photons into the cell end as a function of event position.
Fig. 5 Pulse shape spectrum of the LLSD for the close contact and separate contact case.

Fig. 6 Pulse shape discrimination spectra as a function of event position. (a) OR method, (b) sum method.

Fig. 7 Block diagram of the electronic circuit.
Fig. 8 Position dependent bias curve.

Fig. 9 Relative detection efficiency for 18 MeV neutrons.

Fig. 10 Position resolution and linearity.
Fig. 11 Experimental scattering geometry.

Fig. 12 Position vs energy TOF matrix for polyethylene.

Fig. 13 Typical TOF spectra for Si at 18 MeV.

Fig. 14 Double-differential neutron emission spectra of silicon.
STUDY ON THE CALIBRATION OF CR–39 TRACK DETECTOR FOR LOW ENERGY PROTONS AND ALPHA–PARTICLES

T. Yamauchi, K. Oda, H. Matsumoto and H. Miyake

Department of Nuclear System Engineering,
Kobe University of Mercantile Marine

1. INTRODUCTION

The calibration data of CR–39 plastic detector for hydrogen and helium isotopes are essential so as to apply the detector to the personnel neutron dosimetry and the particle identification and/or energy evaluation in the fields of fusion experiments and space dosimetry. Many studies have been performed on the proton response of CR–39 plastic at energy range of a few MeV. Unfortunately, the manner of calibration is not established well. In our opinion, enormous data pile of etch–pit diameter at various incident energy and thickness of layer removed (bulk etch) is not useful in itself. Furthermore, in the case of lower incident energy below 1 MeV for proton, the decrease of particle energy within the thickness of single step etching is unignorable. Namely, we cannot adopt the convenient method of response evaluation in which the track etch rate is assumed to be constant in this case.

One will find in the present paper that the response function, $S(R)$, etch rate ratio against residual range, $R$, can be reconstructed by the suitable calibration experiments in principle. This will give us an unobstruct view of the response and a fundamental information on track registration property of the CR–39 track detector. The purpose of this study is to develop the manner to obtain the complete response function for low energy hydrogen and helium isotopes. We note that $S(E)$ is also obtainable based on the fundamental relation between incident energy, $E$, and particle range from the response function of $S(R)$.

As a promising manner for calibration experiments, the 'growth curve method' has been proposed by several authors. In this method, the response function is reconstructed from the etch–pit growth curve, so that etch–pit radius plotted as a function of the bulk etch. We found, however, there was an application limit for the growth curve...
method owing to the missing track effect \(^{(9, 10)}\). As a result of this effect, we cannot obtain the response near the track end-point from the growth curve in a certain condition. In this paper we described the results of calibration experiments for low energy proton by the growth curve method at the first. Secondly, the theoretical explanation for the missing track effect and the appearance condition on the proton range and incident angle were given. Finally, we proposed a new calibration method of 'profile method' as a countermeasure for the missing track effect. The results of calibration for alpha-particle using this method was also described.

2. PROTON RESPONSE RECONSTRUCTED FROM ETCH–PIT GROWTH CURVE

Experiments for calibrating the response to proton were performed using the following procedure. Samples of 1 mm thick CR-39 plastic (BARYOTRAK, Fukubi-Kagaku, Japan) were bombarded with protons at normal incidence using the tandem Van de Graaff accelerator of Kobe University of Mercantile Marine. Three type of irradiation were made with different energies of 0.4, 0.7 and 1.0 MeV, respectively. After irradiation, the CR-39 samples were chemically etched in a stirred 6N KOH solution at 70 °C. Etchings were interrupted in a few tens minutes for measuring the size of etch-pits. The bulk etch was monitored by the track radius of fission fragments from a \(^{252}\)Cf source. The rate of

\begin{figure}
\centering
\includegraphics[width=\textwidth]{growth_curves.png}
\caption{Growth curves of proton etch–pits}
\end{figure}
bulk etching is found to be about 2.4 μ m/h. The radius of selected etch-pits in each sample were measured as a function of the bulk etch until they fully round-out. This multi-step etching technic provides us a variation of etch-pit radius with bulk etch called a growth curve.

Typical growth curves are shown in Fig. 1. The proton ranges, \( R_0 \), were estimated from the shape of growth curves in spherical phase to be 4.7, 10.5 and 16.8 μ m for incident proton energies of 0.4, 0.7 and 1.0 MeV, respectively. The calibration by the growth curve method (GC method in the below) was based on the following fundamental relation \(^{(6-8)}\),

\[
S(R) = \frac{1 + \beta (G)^2}{1 - \beta (G)^2} \quad \quad \quad (1)
\]

\[
R = R_0 - \left[ G - r \cdot \frac{1 - \beta (G)^2}{2 \beta (G)} \right] \quad \quad \quad (2)
\]

where \( G \) is the bulk etch. \( r \) represents the etch-pit radius at \( G \) and \( \beta \) is the growth rate of \( r \) \((\beta = dr/dG)\). For analyzing the growth curves, we obtained the best-fit polynomial function for each curves firstly. Then, the growth rate was calculated by differentiating it and finally the response function, \( S(R) \), was derived using eqs. (1) and (2).

![Fig.2 Evaluated response function of proton from growth curve](image-url)
Reconstructed response functions by the GC method were shown in Fig. 2, illustrating the etch rate ratio as a function of the residual range. There is a significant difference among the three curves in the shorter residual ranges below 4 \( \mu m \). The response function of the lowest incident energy \( (R_n = 4.7 \mu m) \) has a sharp peak near the track end-point, but we cannot observe the similar peaks in curves of higher energies. In the next section, we will confirm that the response of the lowest energy is most reliable one in the short residual ranges by simulated calculation.

3. MISSING TRACK EFFECT

The disagreement between the reconstructed response functions suggests that there is an application limit of the GC method. The present authors firstly found that the incident range dependence of evaluated response was attributed to the missing track effect (9). In this section, we made simulated calculations of etch-pit growth in order to check the correspondence between a point along the particle trajectory where the 'bulk etch wave' starts and a point on the etch-pit opening where the wave arrived. It is obvious that the use of eqs. (1) and (2) are permitted when the condition of one-to-one correspondence between these two points is satisfied. The missing track effect will occur when this correspondence condition is broken by the geometrical feature of etch-pit different from a pure cone. For details of simulated calculation and used response, the readers should refer to our previous papers (9).

Figure 3 shows a typical results of growth curve calculation (upper) and assumed response (lower) for proton with a range of 35 \( \mu m \). Dotted lines connect the points on the proton trajectory with corresponding points on the growth curve. The direction of the line represents that of the 'bulk etch wave' propagating, of which the angle is equal to \( \cos^{-1}(1/S) \). As shown in this figure, the one-to-one correspondence breaks at several \( \mu m \) of the end-point. At the top left-hand corner of Fig. 3, an enlarged picture of the growth curve in the frame is shown. Passing along the proton trajectory, the points A, B, C, D, E and F appear in this order. The real growth curve is the envelope of these points, so the points among B, C, D and E (lower dotted lines) are imaginary ones. Because these imaginary points are coupled to the response region between b, c, d and e (hatched region), the correspondence between (real) growth curve and response is lost. Then the response in the hatched region cannot be reconstructed from the observed growth curve. We named the hatched invisible region as a missing track segment and this effect as a missing track effect (9).

Because the missing track effect is fundamentally related to the geometry of etch-pit, the appearance of this effect depends sensitively on the particle, particle range and...
incident angle. The calculational results are summarized in Fig. 4 on the distribution of missing track segment for normal incident proton and other hydrogen isotopes with various ranges. In this calculation, for proton, the missing track effect occurred at ranges above 9 \( \mu m \). And the width of the missing track segment were found to increase with range. The effects for deuteron and triton are indicated to appear at ranges more than 17 and 26 \( \mu m \), respectively. The difference in the distributions of missing track segment among hydrogen isotopes were caused by the disparity of their response. It is clear from this figure that the suffering of missing track effect is avoidable if the range is short enough.

![Simulated calculation of growth curve](image)

Fig. 3 Simulated calculation of growth curve
Returning to the calibration results by the GC method in Fig. 2, the disagreement of the obtained functions should be brought out by the missing track effect. The problem may arise in the fitting and its differentiation processes of the growth curve, in which the derivative of the growth curve is regarded as a continuous function at all bulk etch. If the missing track effect occurs, the discontinuous change of slope of the growth curve appears, indicating as a point of \( B(E) \) in Fig. 3. For precise analysis of calibration with longer particles ranges, we must develop a modified GC method. For example, it may usefully divide the growth curve into two or more parts in the step of fitting. In conclusion, the response function for the shortest range of 4.5 \( \mu \)m is the most reliable one in the three response at short residual ranges.

![Fig.4 Appearance condition of missing track effect on particle range](image)

![Fig.5 Appearance condition of missing track effect on incident angle (proton)](image)
Dependence on the incident angle for the distribution of missing track segment was also calculated. The results on proton are summarized in Fig. 5. The width of the segments hardly change with angle up to 25 degree, and they turn to shrink and vanish at about 39 degree for every examined ranges. The obtained calculation results shows the advantage of selection of an oblique incident irradiation for calibration experiment, but it should be note that the critical angle condition let us to sacrifice the most of response at surface side for only several μ m region near track end-point. Then it seems not to be a good selection.

4. RESPONSE EVALUATION FROM ETCH-PIT PROFILE

The growth curve has the most comprehensive information obtained from measurements of etch-pit opening, but there is a great difficulty to reconstruct the complete response from the growth curve due to the missing track effect as described in the previous section. Then we made a new trial evaluating the response from the etch-pit profile. Evolution of etch-pit profile by chemical etching has been intensively studied \(^{13-15}\). However, it should be emphasized that these studies have been made from the viewpoint of drawing profile and little attention has been paid for applying the theory to response calibration experiments. The simulated calculation of etch-pit profile evolution for proton indicated the effectiveness of this profile method (PR method in the below) as a countermeasure for the missing track effect \(^{14}\).

Schematic diagram of etch-pit profile and some important track parameters are shown in Fig. 6. The original point of x—y coordinates is fixed at the track end-point. Position of the track end-point is concordant with the center of circle which is inscribed to the rounded end-point region of etch-pit wall. The relation between the response at residual range of R and the shape of etch-pit wall \(f(x)\) is expressed by the following equations.

\[
S(R) = \sqrt{1 + \frac{1}{f'(x)^2}} \tag{3}
\]

\[
R = f(x) \cdot f'(x) + x \tag{4}
\]

where \(f'(x) = \frac{df(x)}{dx}\) gives a slope of the etch-pit wall at \(x\).

The observation of etch-pit profile was performed non-destructively by optical transmission microscopy \(^{15}\). The CR-39 plastics (BARYOTRAK) with a thickness of about 1 mm were edge-wise polished with 0.2 μ m alumina powder. The polished edges
Fig. 6 Schematic diagram of etch-pit profile

Fig. 7 Photographs of alpha etch-pit (E = 6.1 MeV)

G: bulk etch
(a) G = 19.2 μm
(b) G = 20.2 μm
(c) G = 21.2 μm
were irradiated by alpha-particles from $^{252}$Cf source ($E = 6.1$ MeV), then sequentially etched in 6N KOH solution at 70 °C. In Fig. 7, typical photographs of etch-pit profiles at indicating bulk etches are shown. The shape of etch-pits were found to be clearly observed except for those near the etched-surface.

As the first step of the PR method, the best fit function for the shape of etch-pit profile was determined from the photograph which the etch-pit end-point slightly passed through the track end-point. Then the obtained function, $f(x)$, was analyzed using eqs. (3) and (4). Calibration result for alpha-particle by the PR method is shown in Fig. 8 with a solid line. The response curves evaluated by the GC method are also plotted with broken lines at various incident angles. It is obvious from this figure that the rapid increase and decrease of etch rate ratio near the track end-point is vividly reconstructed by the PR method. But we must note that there is application limit of the PR method that the response at residual ranges greater than $R_{\text{max}}$ is unobtainable in principle (see Fig. 6). Therefore, the PR method and GC method should be applied complementary for the total calibration of response.

Furthermore, at near the etched-surface region, it is difficult to obtain the focused image of etch-pit profile by an optical microscope. This will became larger problem in the experiments for hydrogen isotopes because of their shallower etch-pit. With the use of STM and AFM it will be possible to overcome this difficulty $^{3, 61}$.

![Fig.8 Response curves for alpha-particles](image)
5. SUMMARY

The summary of this study is follows. Results of calibration experiments for low energy proton using the growth curve method showed the significant dependence on incident energy at short residual ranges. This disagreement among the obtained response with different range was confirmed to be caused by the missing track effect from simulated calculation on etch-pit evolution. The theoretical explanation and the appearance condition on the particle range and incident angle for the missing track effect were also given. Moreover, we proposed the profile method as a new calibration method of a countermeasure for the missing track effect. Through the calibration of alpha–particle using this method, it found to be effective to evaluated the response rapidly changed near the track end–point.

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1. INTRODUCTION

We have reported various results of the optical observation of self-quenching streamers (SQSs) in gas counters induced by α-, β- and X-rays for the purpose of understanding the operation mechanism of the SQS mode. As described in our previous papers (1, 2), the dense ionization tracks significantly contribute to the streamer formation when α-rays are irradiated; those streamers develop only in the specific directions limited by the spatial charge distribution along the primary ionization tracks formed by α-rays. This is a direct evidence of another SQS-formation mechanism that is different from the conventional one based on the electron feeding by the photo-ionization process (Atac's mechanism (3)). From this viewpoint, we have proposed an SQS formation mechanism for α-rays.

In order to check the validity of the newly-proposed mechanism, a computer simulation of streamer propagation may be very helpful. Until now, some computer simulations of streamers have been attempted under the condition of nonuniform electric field to investigate the properties of electrical breakdown in gases. On the other hand, to the authors' knowledge, no computational study of the SQS propagation in gas counters has been reported. In the present paper, we will describe a simple computation model of SQS propagation induced by α-rays inside a gas counter. Simulated results will be compared with observed ones.
2. STREAMER FORMATION PROCESS

In the conventional mechanism of the streamer formation, including the SQS mechanism by Atac et al. (3), streamers are promoted by the free electrons which are produced through the photo-ionization process around a streamer head. In contrast with this, we have proposed a streamer-formation mechanism which is based on the electron feeding from the high density ionization-tracks formed by incident α-rays (1, 2). In this mechanism, the electrons that reach the anode wire first produce an initial avalanche near the anode surface; other delayed electrons are fed into the initial avalanche making secondary avalanches, one after another, along the electron-drifting path. Through the process mentioned above, a streamer-discharge channel spreads about a few mm away from the anode surface towards the cathode. The streamer stops the growing when all initial electrons have been consumed for the streamer development. A schematic representation of the mechanism of such a streamer formation promoted by the ionization tracks is shown in Fig. 1.

![Fig. 1](image)

A schematic representation of the mechanism of the ionization-track induced SQS.

3. COMPUTATION MODEL

In the calculation, an electron drifting process, an electron bombarded ionization process and a space charge effect on the electric field are taken into account as fundamental processes. An ion drifting process is not considered because the drift velocity is so slow that those ions almost do not move from their formation points during the electron drifting and the streamer growing.
A. Continuity equations and the characteristic-curve method

To simulate the electron-avalanche growth, a set of continuity equations for electrons and single-charged positive ions (Eqs. 1 and 2) is numerically solved by the characteristic-curve method in a two-dimensional space (5, 6, 7),

\[
\frac{\partial n_e}{\partial t} + \text{div} n_e \mathbf{W} = \alpha n_e \mathbf{W}, \quad (1)
\]

\[
\frac{\partial n_p}{\partial t} = \alpha n_e \mathbf{W}, \quad (2)
\]

where \( n_e \) and \( n_p \) (cm\(^{-2}\)) are the densities of electrons and single-charged positive ions, respectively, \( \mathbf{W} (=1\text{|}\mathbf{W}|) \) (cm/s) the drift velocity of electrons, \( \alpha \) (cm\(^{-1}\)) the ionization coefficient by collisions. The following equations are derived from Eqs. 1 and 2;

\[
n_e(r, t+\Delta t) = n_e(r-W\Delta t, t) \exp(A\Delta t),
\]

\[A = \alpha W - \text{div} W, \quad (3)\]

\[
n_p(r, t+\Delta t) = n_p(r, t) + \alpha W n_e(r, t) \Delta t. \quad (4)
\]

Equation (3) represents the change of the electron density in a coordinate system moving along the drifting path (i.e. the characteristic curve).

Fig. 2 The electron drift velocity measured in a CH\(_4\) gas (4). The solid is expected in the present calculation.

Fig. 3 The ionization coefficient measured in a CH\(_4\) gas (8). The solid is expected in the present calculation.
The counting gas is assumed to be CH₄(100%) at atmospheric pressure. Since electron swarm parameters of CH₄, \( W \) and \( \alpha \), have been measured experimentally, it is expected to obtain reliable data. Analytical expressions of the electron drift velocity, \( W \), and the ionization coefficient, \( \alpha \), in CH₄ are prepared by reference to experimental data (4, 8) as shown in Figs. 2 and 3.

**B. Space lattice and electric-field calculation**

For the simulation, a cylindrical gas counter is supposed, whose inner diameter is 20 mm; the anode-wire diameter is 50 μm. The calculation is carried out in a semicircle region which is divided into 100 meshes at regular intervals in the radial direction (from 25 μm to 10 mm) and 90 meshes at regular intervals in the angular direction (from 0° to 180°) as sketched in Fig. 4.

The strength of electric field produced by the space charges is simply evaluated using the Coulomb's law between all combinations of mesh points inside the calculation space at each time step. Then, their radial components are added to the static electric field produced by the applied electric potential to the anode wire.

![Fig.4 A space lattice used for the simulation and a typical arrangement of initial electrons (not in scale).](image)

**C. Two-region model**

According to the "two-region model" (5, 9), a streamer channel is classified as two different regions: an "active region" around the streamer head and a "passive region" which represents the path between the streamer head and the high-voltage electrode, as graphically...
shown in Fig.5. In the "active region", the charge density is relatively low and the ionization phenomenon is active because of the high electric field. On the other hand, in the "passive region", the charge density is rather high and the ionization phenomenon is inactive. Therefore, the contribution of the "passive region" to the streamer development may be very small. As the streamer develops longer, the "active region" changes to the "passive region" step by step. In the present calculation, the "passive region" is excluded from the calculation space in order to lighten the computer's calculation load. Consequently, the information inside the streamer is lost. Nevertheless, this approximation based on the two-region model may be adequate for the present purpose because only propagation characteristics of streamers are interesting and desirable to estimate.

D. Flow of the calculation

Initial electrons are distributed in a line with simulating ionization tracks of α-rays for each incident condition. The line density is 6.25 × 10⁴ (electrons/cm); therefore, for example, a 1.6 cm long ionization track is formed as the result of about 2 MeV energy loss of an α particle in the counting gas. A typical arrangement of initial electrons is indicated in Fig.4 (not in scale), which will be used for the calculation in Figs.6, 7 and 8(a). In the actual calculation, those initial electrons are put on the corresponding mesh points with proper density so as to reproduce the uniform distribution along the ionization track.

The Meek's streamer condition is adopted to judge a streamer generation (i.e. start of streamer development); when the static electric field on the anode surface is completely cancelled by the influence of space charges, it is assumed that a streamer departs from the anode surface. The streamer growing is treated under the concept of the two region model explained already. The region whose net-charge density reaches 1 × 10⁻⁷ (C/cm²) is considered as "passive region" and the region becomes a new streamer head. The value of 1 × 10⁻⁷ (C/cm²) corresponds to a typical net-charge density inside a streamer region (5). (In the present case, net-charge density is obtained by subtracting the electron-charge density from the ion-charge density.) When the total number of electrons inside the calculation space falls less than 100, it is supposed that the streamer has stopped its growing.

The time step is ordinarily chosen to be 5 ns during the initial electron drifting, 0.1 ns during the initial avalanche growing and 0.05 ns during the streamer growing. If much smaller time step is required, the computer program automatically reduces the time step by the half and executes the calculation over again in that stage; this process is repeated until a proper result is obtained.
4. RESULTS AND DISCUSSION

Figure 6 shows a calculated time variation of a streamer represented by the streamer-head positions. The three-dimensional displays of net-charge distribution at 70.135 ns, 70.163 ns and 70.205 ns are given in Fig. 7 (a), (b) and (c) respectively. For this calculation, initial electrons are arranged as sketched in Fig. 4 with simulating a 1.6 cm ionization track. As indicated in Fig. 6, the streamer starts the growing at 70.070 ns and ends at 70.205 ns to form an about 3500 μm streamer channel. The calculated streamer length shows fairly good agreement with observed one (about 4 mm) in a CH₄ gas (1). The average speed of the streamer propagation is estimated at about 2.6 cm/ns, which seems to be rather faster than the typical ones of photo-mediated streamer propagation (for example about 0.1 cm/ns or less (5)). This may be due to the difference of electron-feeding process between both streamer types. Since a direct measurement of propagation speed of α-ray induced streamers has not been achieved, there are no appropriate data to be compared with this calculated result.

As clearly indicated in Fig. 7, the streamer develops only in the specific directions; the property of bi-directional streamer propagation which has been optically observed is adequately reproduced.

The variations of streamer shape are studied for some initial-electron arrangements. The calculated results in Fig. 8 show the net-charge distributions at the moment when the streamers stop their elongation. The optical images obtained with similar initial-electron arrangements are indicated in the same figure (1, 2). Those calculations are performed with assuming a different sort of counting gas from the one used for the
observations*. And the geometrical configurations for the computations (the shape of the cathode electrode and the initial-electron arrangements) are slightly changed from those used in the observations for simplicity. In spite of these facts, simulated results show a good agreement with observed ones in their shapes and directions for each arrangement of initial electrons. This means the present simple computation model based on the newly-proposed SQS mechanism successfully interprets the essence of the SQS phenomenon induced by α-rays.

*The observations have been performed mainly in a Ne(70%)+ CH₄(30%) mixture because the range of α-rays (²¹⁰Po, 5.3 MeV) in this mixture is longer than that in a CH₄(100%) gas and the arrangements of initial electrons can be selected in full of variety. In Fig.8, the apparent disagreement in streamer length between the calculated results and the observed ones may be due to the fact that different gas mixtures are supposed.

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Fig.7 Three dimensional display of net-charge distribution.
Fig. 8 Streamer shapes calculated for different positions of initial electrons. The optical images taken with similar initial-electron arrangements in a Ne(70%)+CH₄(30%) mixture are shown in the right hand. In this figure, \( h \) is the distance from the anode wire to the initial ionization track and \( d \) the distance from the \( \alpha \)-ray source to the mylar window of the counter. The broken lines represent the ionization tracks of \( \alpha \)-rays.
5. SUMMARY

In order to check the validity of a newly-proposed SQS mechanism for α-rays, a modeling of the phenomenon is made and a computer simulation has been carried out. A set of continuity equations for electrons and single-charged positive ions is numerically solved to evaluate the avalanche growth in a two-dimensional space. The streamer development is treated under the concept of the two region model with an aid of the Meek's streamer condition. A comparison between computed and experimental results shows a good agreement in nature and the validity of the newly-proposed SQS-formation mechanism has been confirmed successfully.

References

GAMMA-RAY RESPONSES
OF PRESSURIZED IONIZATION CHAMBERS
BY MONTE CARLO SIMULATION

Tatsuo Torii *
Oarai Engineering Center, PNC
4002 Narita, Oarai, Ibaraki, 311-13, JAPAN

Yoshio Hino
Electrotechnical Laboratory
1-1-4 Umezono, Tsukuba, Ibaraki, 305, JAPAN

1. INTRODUCTION

To measure the radioactivity of gamma-ray emitting nuclides, calibrated ionization chambers with reentrant source receptacles, as shown in Fig. 1, are ordinarily used. These are filled with nitrogen or argon gas pressures up to 20 atmospheres. The activity of the radioactive source can be obtained by measuring the ionization current of the ionization chambers. Main characteristics of this ionization-chamber system are simplicity of operation and long-term stability. The stability of two pressurized ionization chambers made by different manufactures, which are used in Electrotechnical Laboratory (ETL), are within ± 0.5 percent during these 15 years. Therefore, if the ionization current of such an ionization chamber is calibrated precisely with each gamma-ray emitting nuclides, it can be a detector for secondary radiation standard instrument for radioactivity. The Bureau International des Poids et Mesures (BIPM) uses the ionization chamber pressurized nitrogen gas with 20 atmospheres as the international reference system (SIR) for gamma-ray emitting nuclides, and many data have been accumulated using this chamber (1). ETL also registered data for some typical radionuclides, and the consistency of Japanese radioactivity standardization has been confirmed.

The ionization current of an ionization chamber can be described as a smooth function for gamma-ray energy. Therefore, if the response function is previously obtained by experiment with high accuracy, it can be evaluate the radioactivity of radionuclides that have not yet been calibrated. Furthermore, this method contributes to evaluate the radioactivity of short half-lived nuclides, which it is difficult to measure the radioactivity, and that of other type sources such as gaseous

*1 Present address:
Monju Construction Office, PNC
2-1 Shiraki, Tsuruga, Fukui-ken, 919-12, Japan
nuclides filled in ampoules. To obtain the response function of the pressurized ionization chambers precisely, we measured the ionization current of ionization chambers with standard gamma-ray sources by experiment, and calculated to estimate the continuous response function by Monte Carlo code.

2. SIMULATION

To obtain the gamma-ray response of ionization chambers, we used the Monte Carlo electron-photon transport code system, EGS4 \(^{(2)}\), and calculated the energy deposition in atmospheric gases in ionization chambers. In the present calculation, the transport of low energy electrons, less than 1 MeV, is significant in estimating the energy deposition in the gas. The PRESTA \(^{(3)}\) algorithm has been used, which was developed to improve electron transport in EGS4 code system. From the calculated energy deposition in the gas, \(E_{dep}\) [eV], the ionization current \(I\) [A] \(=e\cdot E_{dep}/W\) and the efficiency \(\varepsilon=I/(A\cdot P)\) were calculated. The charge of an electron is denoted by \(e\) [C], and \(W\) is W-values of an electron in gases (Ar: 26.4 eV, N\(_2\): 34.8 eV) \(^{(4)}\).

In this study, the ionization efficiencies of three types of pressurized ionization chambers were calculated. These are named types IG11/A20, IG11/A10 and IG12/N20, which are manufactured by CENTRONIC Co. Ltd., UK. Former two chambers are filled argon gas at pressures of 20 and 10 atmospheres, respectively. Last one is filled nitrogen gas at a pressure of 20 atmospheres. In the chambers, aluminum electrodes are arranged in coaxial cylindrical structure (see Fig. 1), and inner diameters of the reentrant tubes, which are made of stainless steel, are 1 and 2 inches for the types IG11 and IG12, respectively. The can and electrodes of these ionization chambers were modeled as the cylinders that have equivalent volume and diameter of ionization areas. The chambers simulated under the same geometrical setting with the experimental condition, such as size and position of the acrylic guide tube inserted in reentrant tube and sources. In this calculation, sources were supposed to be a solution with a volume of 3.6 cm\(^3\) filled in a glass ampoule, and monoenergetic photons were emitted from the source in an arbitrary direction using random numbers. To reduce the statistical uncertainty within ± 3 %, the number of histories was set one million for each energy.

3. RESULTS AND DISCUSSION

Figs. 2 and 3 show the results of the calculation and the experiment. These figures show that the calculated response of the argon ionization chamber is a little smaller than experimental result. On the other hand, the nitrogen chamber is a little larger. The reason of this can be understood as the tolerance of the ionization volume and the gas pressure. Therefore, it seems that the calculation agrees well with the experiment practically. All the chambers calculated show
proportional response functions with gamma-ray energy more than about 200 keV, and the response functions of argon chambers make peak at the energy of about 80 keV. To make clear the reason, we calculated the reactions of the photoelectric effect and the Compton scattering in argon and nitrogen gases. From this result, it is confirmed that the peak of the argon chamber’s response at about 80 keV is caused by the photoelectric effect of argon, as illustrated in Fig. 4.

4. CONCLUSIONS

The gamma-ray responses of the pressurized ionization chambers filled with argon or nitrogen are calculated using the EGS4/PRESTA code system. The results are in good agreement with the experiment practically, and describe the energy responses in terms of the detection efficiency. The difference between the experimental data and the calculated result is caused by the errors in simulated model, such as the thickness of the reentrant tube and electrodes, geometrical shapes of electrodes, and gas pressure. If it is improved the model by applying more exact conditions, more reliable response function can be obtained.

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Fig. 1 Sectional View of Pressurized Ionization Chamber

Fig. 2 Response of Pressurized Ionization Chambers filled Argon or Nitrogen
Fig. 3 Response of Pressurized Ionization Chambers filled Argon Gas

Fig. 4 Number of Reactions of Photoelectric Effect (PE) or Compton Scattering (COMP) in Chambers
Diethorn Parameter at Low Temperature of Gases for a Proportional Counter

Kazuko Fukumura, Akio Nakanishi and Takayuki Kobayashi
Department of Physics, Shiga University of Medical Science,
Seta, Otsu, Shiga 520-21, Japan

1 Introduction

A proportional counter is used widely, in particular, for conversion electron Mössbauer spectroscopy (CEMS), because its counting efficiency and resolution for low energy radiations are comparatively high and because it is easy to fabricate a counter for experimental purposes. In order to investigate the temperature dependence of CEMS spectra, the counter, in which a sample material is mounted, has to be operated at low temperature.

The first application of a proportional counter to CEMS was performed by Swanson and Spijkerman at room temperature\(^1\). Later, the counter was operated at low temperature between 77 K and room temperature\(^2,3\). Isozumi \textit{et al.} also had a success in observing CEMS spectra at 5 K\(^4\). In studies with CEMS, the operation of the counter at 5-77 K is also expected and has been done by us\(^5-9\). Consequently, CEMS measurements at temperature above 5 K up to room temperature come to be possible as illustrated in the Fig. 1. However, the mechanism of operation of a proportional counter at low temperature is still unknown. It is, therefore, very
interesting and important to understand the operational properties of a proportional counter at low temperature for stable and reliable CEMS measurements. In the present work, the mean energy required to produce an ion pair in a proportional counter is estimated and compared with the \( W \) value. The effect of photo-electrons to operation of the counter is also discussed.

2 Gas Multiplication Factor in a Proportional Counter

An electron is efficiently multiplied in a proportional counter through electron collisions with gas molecules to form an electron avalanche. The gas multiplication is enhanced with electrons emitted at the cathode by the photo-effect and the neutralization of positive ions and is reduced with electron attachment to gas molecules. When the anode voltage is comparatively low and the above-mentioned effects are negligibly small, the gas multiplication factor \( A \) due to electron collisions in the proportional counter is given by the equation\(^{10}\)

\[
\frac{\ln A}{V} = \frac{\ln 2}{\Delta V \cdot \ln(b/a)} \left\{ \ln V - \ln \left[ K p \ln(b/a) \right] \right\},
\]

where \( V \) is the anode voltage, \( \Delta V \) is the mean energy required to produce an ion pair, \( a \) and \( b \) are the radii of the anode and cathode, respectively, \( p \) is the gas pressure in the counter at room temperature and \( K \) is the minimum value of \( E/p \) above which the gas multiplication takes place. Here, \( E \) is the electric-field intensity in the counter. In the case that the effect of photo-electrons is not small, the gas multiplication factor \( A \) is enhanced according to the equation\(^{11}\)

\[
M = A + \gamma A^2 + \gamma^2 A^3 + \cdots = \frac{A}{1 - \gamma A} \quad (\gamma A < 1),
\]

where \( \gamma \) is the probability of electron emission by the photo-effect per ion pair formed in the counter and \( M \) is the total gas amplification factor.

3 Experimental

The cryostat and counter assembly are described elsewhere\(^5\)). The proportional counter in the system was of the gas-filled type, because the temperature is not well controlled in a gas-flow type counter. The operation of the counter was examined with a \( ^{57} \)Co source of \( \sim 1 \) kBq electroplated on an aluminum foil and mounted on the cathode plate of the counter. Signals from the counter were recorded with a multichannel analyzer through a low-noise charge-sensitive preamplifier and an amplifier. The counter gas examined in the present work are helium, neon, He+1.3%Ar, He+10%CO, He+5%N\(_2\), He+2%CH\(_4\) and hydrogen. The
counter gas was filled to 1 bar at room temperature. Before filling, in the counter, the rare gases were purified with a trap of molecular sieves cooled at 77 K.

The gas multiplication factor $A$ was estimated with Mathieson's method\textsuperscript{12}). When all the ions produced by a radiation with energy $E$ in the counter are collected, the signal voltage $V_\infty$ of the amplifier with the amplifier factor $G$ is expressed as

$$V_\infty = \frac{G A E}{C W} e,$$

where $A$ is the gas multiplication factor, $C$ is the electric capacity of the charge-sensitive preamplifier, $W$ is the $W$ value of the counter gas and $e$ is the elementary electric charge. In the practical situation of measurements, however, a signal is usually clipped before all of the ions are collected. The clipped signal voltage $V_T$ was derived by Mathieson as

$$\frac{V_T}{V_\infty} = \frac{0.87 + 0.797 \log(T/t_0)}{2 \ln(b/a)},$$

where $T$ is the shaping time of the amplifier and $t_0$ is given by

$$t_0 = \frac{a^2 \ln(a/b)}{2 \mu V}.$$

Here $\mu$ is the mobility of the counter gas. The gas multiplication factor $A$ was derived from Eqs. (3)-(5) using the peak voltage $V_T$ corresponding to the conversion and Auger electrons in the spectrum of $^{57}$Co.

4 Results and Discussions

![Gas multiplication factor as a function of anode voltage.](image-url)
The obtained total gas multiplication factor $M$ is shown in Fig. 2 as a function of anode voltage $V$. In most cases the multiplication factor is larger than 1000, but it is much smaller, ranging between 200-300, for pure He and Ne at low temperature. The value $(\ln M)/V$ was plotted as a function of $\ln V$ which we call the Diethorn plot and is shown in Fig. 3. If no photo-electron contributes to the gas multiplication, the plotted value may changes linearly with $\ln V$ according to Eq. (1). A secondary electron emitted as an ion is neutralized at the cathode has no effect on the estimated gas multiplication factor because a signal is clipped before all the ions are collected. As seen in Fig. 3, the value $(\ln M)/V$ increases linearly with $\ln V$ in cases of He and Ne at low temperature, while it goes up more, in other cases, at higher anode voltages. The estimated gas multiplication factors at lower anode voltages were analyzed by using Eq. (1) and the parameters $\Delta V$ and $K$ at temperature were estimated as listed in Table 1. The value of $\Delta V$ is larger than the $W$ value of the corresponding gas except for the cases of the He-Ar mixture and hydrogen. The reductions in the estimated values of $\Delta V$ of hydrogen and the He-Ar mixture is mainly due to the phot-effect and the Penning ionization effect, respectively. Since the effect of photo-electrons is found as well in the other gas mixtures, the parameters $\Delta V$, $K$ and $\gamma$ were estimated by fitting the data to Eqs. (1) and (2) simultaneously with the least-square method. The obtained results are also listed in Table 1. The Diethorn plots of hydrogen with the original data and with the data modified by eliminating the effect of photo-electrons are given in Fig. 4. It is seen that the original curving plot is transformed into the plot being linear.

![Fig. 3 Diethorn plot with the total gas multiplication factor $M$.](image_url)

From Table 1, it is found that $\frac{\Delta V}{W} = 1.6 \approx \frac{W}{I}$ for He and Ne, $\frac{\Delta V}{W} = 2.1 \approx \frac{W}{I}$ for $H_2$, $\frac{\Delta V}{W} = 0.9$ for the He-Ar gas mixture and $\frac{\Delta V}{W} = 1.3 - 1.5$ for the other gas mixtures, where $I$ is the ionization energy of a gas molecule. It is known the parameter $\Delta V$ of the gas mixtures of Ar and CH₄ is larger than $\frac{W}{I}$, while
\[ \frac{\Delta V}{W} = 0.7-0.8 \] for the Penning gas mixtures of Ar, Xe and CO\(_2\)\(^{15}\). These results imply that the mean energy of ionization, i.e. \(\Delta V\), is larger than \(W\) when the gas multiplication is controlled by the quenching process while the Penning effect makes the mean ionization energy to reduce greatly. The results obtained in the present work might be explained in a similar way by introducing a model for the operating mechanism of a proportional counter filled with a rare gas.

Table 1 Diethorn parameters \(\Delta V\) and \(K\), and probability \(\gamma\) of electron emission by the photo-effect per ion pair. The values of \(\Delta V\) and \(K\) listed on the upper line of each gas or gas mixture were estimated with the gas multiplication factors at lower anode voltages in Fig. 2.

<table>
<thead>
<tr>
<th>Counter Gas</th>
<th>Temperature (K)</th>
<th>Max. Gas Multiplication Factor</th>
<th>(\Delta V) (V) (K\times10^{-4}) (V·cm(^{-1})·bar(^{-1})) (\gamma\times10^{4})</th>
<th>W Value (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>He</td>
<td>14.6</td>
<td>3.0\times10^2</td>
<td>68</td>
<td>0.6</td>
</tr>
<tr>
<td>Ne</td>
<td>25</td>
<td>2.0\times10^2</td>
<td>55</td>
<td>0.8</td>
</tr>
<tr>
<td>H(_2)</td>
<td>17</td>
<td>2.0\times10^3</td>
<td>~35</td>
<td>~6.0</td>
</tr>
<tr>
<td>95%He +5%N(_2)</td>
<td>60</td>
<td>9\times10^3</td>
<td>40</td>
<td>1.2</td>
</tr>
<tr>
<td>90%He +10%CO</td>
<td>65</td>
<td>6\times10^3</td>
<td>46</td>
<td>1.9</td>
</tr>
<tr>
<td>98%He +2%CH(_4)</td>
<td>77</td>
<td>1.0\times10^4</td>
<td>29</td>
<td>0.9</td>
</tr>
<tr>
<td>98.7%He +1.3%Ar</td>
<td>77</td>
<td>1.0\times10^3</td>
<td>~16</td>
<td>~0.9</td>
</tr>
</tbody>
</table>

\(*\) assumed value.
The operation of the counter filled with the gas mixtures except for the He-Ar mixture is obviously explained with the ordinary quenching model. Stable operation of a counter filled with a rare gas may owe to the presence of ion clusters at low temperature. A He$^+$ ion, for example, formed by a radiation is converted to an ion cluster He$_n^{+}$ ($n\approx10$) and goes to the cathode. The cluster is neutralized at the cathode and dissociated, with emitting no electron, at the expense of the energy released at the neutralization. Therefore, the cluster plays a similar role to a quenching molecule in a mixed gas and prevents a discharge of the counter. If the counter gas is pure enough, a photon emitted from an excited atom is mostly absorbed by another atom. On the other hand, at higher temperature, the size and density of the cluster ion decreases and the above-mentioned process does not function any more. At much higher temperature around room temperature, impurity molecules are evaporated much enough for the counter to work again with the help of the Penning effect. The ion cluster H$_3^+$($\text{H}_2$)$_n$ may also play an determinant role in operating a counter filled with hydrogen. As can be seen in Table 1, however, the probability $\gamma$ of hydrogen is not small in contrast to the cases of He and Ne.

References
10) W. Diethorn, NYO-6628 (1956).
Space Charge Effect depending on Direction of Charged Particles in $^{3}$He Proportional Counters

N. Takeda and K. Kudo
Electrotechnical Laboratory

Abstract

The variations of pulse height and energy resolution at a full energy peak depending on the direction of charged particles produced by thermal neutrons in cylindrical $^{3}$He proportional counters have been measured using a pulse shape analysis. The difference of peak positions affected by charged particles having mainly "parallel direction" and "any other direction" to the anode wire was found to be 1%, and this difference became larger with increasing bias voltage.

It is concluded that, as a result of space charge effect, the direction of charged particles to the anode wire affects the variations of pulse height, energy resolution and the optimum bias voltage in the proportional counter. The experimental results also suggest that by selection of signal pulses produced by the parallel ion track, the full-width-at-half-maximum (FWHM) of the 764 keV full energy peak can be reduced from 4.9% to 3.9%.

I. INTRODUCTION

$^{3}$He proportional counters are often used for neutron spectroscopy and measurements of neutron fluence, since a $^{3}$He(n,p)T reaction, which is a dominant reaction occurring in the counters, has a large cross section at low energy region. Charged particles of a proton and a triton created by the $^{3}$He(n,p)T reaction have finite straight tracks, and have an isotropic angular distribution for thermal neutrons in the counter. Initial ion pairs, which are produced by an interaction with an ionization gas, are distributed along their paths. The orientation of the ion track relative to collection electrode made variation of the risetime of the output pulse from preamplifier[1]. Recently, Dietz et. al. observed a time profile of output pulses in the $^{3}$He proportional counter[2]. His result was that the long risetime was weakly correlated to an enhanced asymmetry in the peak of pulse height distribution, and it was suggested that a space charge effect account for the asymmetric distribution.

It is the purpose of this study, (i) to report on the use of a pulse shape parameter as a means of determining ion track orientation relative to the anode wire (in chapter II and III),
and (ii) to report the effect of the track orientation on the pulse height and energy resolution of the 764 keV full energy peak in a commercially available cylindrical $^3$He proportional counter (in chapter IV).

II. PULSE SHAPE DEPENDING ON ION TRACK ORIENTATION

Waveforms of pulses from the $^3$He proportional counter were changed depending on the ion track orientation relative to the anode wire and the ion density distribution$^{[9]}$. In figure 1, some examples of simulated pulse shapes of the signals from the $^3$He proportional counter are shown$^{[4]}$. The input parameters were assumed to be 1.27 cm of cathode radius, 15 $\mu$m of anode radius, and 600 kPa (400 kPa of $^3$He gas and 200 kPa of Ar gas) for the counter. The total kinetic energy of a proton and a triton is 764 keV in the $^3$He(n,p)T reaction induced by a thermal neutron. The arrows pointing towards the letters p and T in the above box indicate the directions of proton and triton relative to the axis of the anode wire. The dashed curves show the differentiated signals. The curve of (a) shows the pulse shape due to the tracks of charged particles produced parallel to the anode wire. The pulse rises quickly because all of the ion pairs are equidistant from the anode wire. The curves of (b) and (c) show the pulse shape due to the tracks of charged particles produced perpendicularly to the anode wire. These pulses rise slower than those of (a).

Figure 2 shows the relation between the orientation of the ion track and the maximum gradient of pulses. The cases of (a), (b) and (c) in figure 1 correspond 0 degree, 90 degree and -90 degree, respectively. The orientation of ion tracks had isotropic distribution, and the $^3$He(n,p)T reaction occurred all region in the counter. Then, the maximum gradient was varied depending on the radial position of reaction, even if it had same

Figure 1. Simulated time profile of the pulses from $^3$He proportional counter.
orientation of ion track.

III. EXPERIMENTAL PROCEDURE

The output signals of a cylindrical $^3$He proportional counter (Reuter-Stokes RS-P40806-275) were sent to a preamplifier (ORTEC 142PC, risetime: $\sim$100 nsec., decay time constant: $\sim$50 $\mu$sec.), then directly to a digital waveform analyzer (Analytek 2000, 167 MHz, 12 bits, and 2k-words sampling) and then stored on a hard disk of a computer. A high stability power supply (Matsusada HEPM-3R10-LGS12, stability: 10ppm through 8 hours) was used to apply the bias voltage to the counter. In order to determine the systematic dependency of the direction of charged particles on the pulse height and the energy resolution in the counter, neutron experiments were performed in a thermal neutron field, since a proton and a triton from the $^3$He($n,p$)T reaction induced by a thermal neutron are emitted in the opposite direction from the reaction point and produce the straight ion tracks, as shown in the upper box of figure 1. Counting rate in the experiment was less than 40 cps.

Figure 3 shows a two-parameter representation in which both the pulse height and the maximum gradient of each pulse are the parameters. Here, the pulse height of each pulse was directly obtained from the saturated output voltage from the
The measured pulse height distributions were divided into three groups as described above. An example of the pulse height distributions near the thermal neutron peak is shown in figure 5. The symbol ■ shows the pulse height distribution in group A, and the symbol □ shows the distribution in group B. The counter was operated at a normal bias of 1300V, and the thermal neutrons were collimated to the central part of the counter to avoid an edge effect. The pulse height distribution in group A was symmetrical, while the distribution in group B had an asymmetrical distribution in the lower part of the peak. This asymmetry in group B became
notable with increasing the bias voltage. This result agreed to those presented by Dietz et al. To obtain peak channels and FWHMs of the full energy peaks, it was assumed that the full energy peaks had Gaussian distributions. The Gaussian curves were fitted to the experimental results by using the least square method (solid curves in figure). The peak channels of groups A and B were 536.4 channel and 531.7 channel, respectively. The FWHMs of the full energy peaks were 3.9% and 4.9%, respectively. The uncertainty to determine the pulse height and the resolution was estimated as ±0.3%.

The peak positions of groups A (■) and B (□) as a function of the operating voltage are shown in figure 6. These positions are normalized to the totality detected pulses. Peak resolutions as a function of the operating voltage are also shown. The symbols •, ○ and × refer to the energy resolutions of the groups A, B and total, respectively.

The difference of the peak positions between the groups A and B became greater as increasing the bias voltage. The reason of the loss of pulse height is an influence of space charge effect, which distorts an electric field appreciably in group B. It would be explained that the anode length which collects the electrons in the case of group B is shorter than that of group A, and hence the condensed space charge reduces a gas multiplication. The asymmetry of the peak in the group B was caused by adding together the contribution of charged ion tracks.
having a different amount of space charge effect corresponding to the emission angle to the anode wire.

The energy resolutions in groups A and B show the different tendencies with increasing bias voltage. The optimum voltages for best energy resolution in groups A and B were 1300V and 1200V, respectively. The energy resolution in group B became noticeably worse with increasing voltage compared to that in group A. The energy resolution of the proportional counter was adversely affected by the space charge effect and the parallel component of the charged particles in the group A furnished the better energy resolution.

V. CONCLUSION

In this study, it is concluded that, as the result of space charge effect, the orientation of ion track with respect to the anode wire affects the variations of pulse height, energy resolution and the optimum bias voltage in the proportional counter. The experimental results also suggest the improvement of the energy resolution by simply selecting the "parallel direction" component of ion track to the anode wire.

VI. REFERENCES

DEFECTS IN OPTICAL FIBERS DURING 
14MeV NEUTRON IRRADIATION

Tsunemi Kakuta, Yujiro Ikeda and Yukio Oyama(1) 
Japan Atomic Energy Research Institute(1) 

Tatsuo Shikama and Minoru Narui(2) 
Tohoku University(2) 

Kazuo Sanada and Naoki Shimoto(3) 
The Fujikura Ltd.(3)

1. INTRODUCTION

Looking at previous investigations for the radiation effects on optical fibers, gamma-ray irradiation tests were mainly conducted more than that of other radiation sources\(^{(1-4)}\). From these investigations, some significant optical effects were pointed out for practical application in high dose and high dose rate of radiation environment. The most serious problem of radiation effects on optical fibers is the increase of transmission loss due to the radiation-induced defects. It is known that the high energies of radiation causes ionization and atomic displacements within the molecular bonding network of the optical fibers\(^{(1-4)}\).

When applying to more severe radiation environments in nuclear plants, i.e., diagnostics for fusion reactors, sensors for in-core measuring systems of fission reactors, radiation related subjects should be considered by different radiation sources. Owing to this, heavy neutron irradiation effects on optical fibers were examined with 14 MeV fast neutrons as comparing the results on \(^{60}\)Co gamma-ray irradiation.

This paper mainly outlines the behavior of optical transparency of the pure silica core fibers, particularly the irradiation effects of gamma-ray sources.

2. EXPERIMENTAL PROCEDURE

A pure silica(SiO\(_2\)) core step-index(SI) type optical fiber, which shown in Table 1, was used for this experiment. The fiber having low OH content SiO\(_2\) core with fluorine doped SiO\(_2\) clad. Outer diameter of the core and clad is 200\(\mu\)m and 250\(\mu\)m, respectively.
Irradiated length of optical fiber was 50m.

Table 1 Details of the Pure Silica Core Step Index Type Optical Fiber.

<table>
<thead>
<tr>
<th>Fiber Type</th>
<th>Core</th>
<th>Clad</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Material</td>
<td>Diameter</td>
</tr>
<tr>
<td>Pure Silica Core Step-Index</td>
<td>SiO₂</td>
<td>200μm</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Item</th>
<th>14 MeV Fast Neutron</th>
<th>Gamma-ray</th>
</tr>
</thead>
<tbody>
<tr>
<td>Facility</td>
<td>FNS, JAERI, Tokai</td>
<td>⁶⁰Co, JAERI, Tokai</td>
</tr>
<tr>
<td>Irradiation Length</td>
<td>50 meter</td>
<td>50 meter</td>
</tr>
<tr>
<td>Irrad. Temperature</td>
<td>Room Temperature</td>
<td></td>
</tr>
<tr>
<td>Flux / Dose rate</td>
<td>$8.7 \times 10^8$ n/cm²·s</td>
<td>$7 \text{ Gy} / \text{s}$</td>
</tr>
<tr>
<td>Fluence / Total Dose</td>
<td>$1.0 \times 10^{14}$ n/cm²</td>
<td>$1.3 \times 10^5$ Gy</td>
</tr>
<tr>
<td>Measurement</td>
<td>In-situ Measurement for Optical Transparency</td>
<td></td>
</tr>
</tbody>
</table>

A fast neutron irradiation facility (FNS) in JAERI and a ⁶⁰Co irradiation facility in JAERI were used for this experiment as a 14MeV fast neutron source and gamma-ray source. Irradiation conditions of the optical fibers are listed in Table 2. Figure 1 shows an outline of the measuring procedure. The fiber was examined on its dynamic optical phenomena during...
the irradiation with a 14 MeV fast neutron and a $^{60}$Co gamma-ray. In case of 14 MeV fast neutron irradiation, the optical fiber was irradiated under flux of about $8.7 \times 10^8$ n/cm$^2$/s with 14 MeV fast neutron beam. Total fluence of 14 MeV fast neutron to the optical fiber was up to $1.5 \times 10^{14}$ n/cm$^2$. Gamma-ray irradiation with a $^{60}$Co source was examined under the dose rate of 7.0 Gy/s, and total dose was up to $1.3 \times 10^5$ Gy.

One end of optical fiber was connected to the xenon-lamp of white light source. The other end was connected to the optical spectrum analyzer, and the spectral transmission characteristics during irradiation under different conditions was measured. Measuring wavelength range is 350 nm to 1750 nm.

### 3. RESULTS AND DISCUSSION

An example of the increase in optical absorption of low OH content pure silica core SI type optical fiber during irradiation with a 14 MeV fast neutron and a $^{60}$Co gamma-ray, is shown in Fig. 2. The observed values were little different in comparison with two kinds of radiation source.

**Figure 2** Spectral Induced Absorption of Pure Silica Core Fiber During 14 MeV Fast Neutron Irradiation as Compared with Co-60 Gamma-ray
When the optical fiber was irradiated by 14MeV fast neutron, large absorption loss at wavelength shorter than 700 nm, was appeared. This induced absorption could be separated into an absorption band in the region from UV to visible wavelength and that in the 600nm band. In the UV-visible band, the value of absorption increases inversely as the wavelength \((1/\lambda)\) because shorter than 700 nm. In the region of 600nm absorption band, absorption peak of 630 nm in which caused by non-bridging oxygen hole center(NBOHC), was created(5).

In case of gamma-ray irradiation, the absorption spectrum was appeared widely than that of 14 MeV fast neutron irradiation. The absorption is spread its wavelength region from UV to 1000 nm. This UV-visible absorption band has been observed in many previous experiments, and was attributed to the optical absorption by E'centers(6).

During irradiation of solid state substances by ionizing radiation such as X-ray and/or gamma-rays, charged particle radiation collides with electrons. In the case of glass, depending on the degree of imperfection in their molecular structure even before irradiation, these collisions which involves charged particles and electrons create new defects of color centers. In optical fibers, these defects cause the increase in transmission loss and is detrimental to optical fibers used for signal propagation. The imperfection in glass consists of intrinsic defects such as oxygen deficient centers, non-bridging oxygen hole centers(NBOHC), oxygen

### Table 3  Defects and Color Centers in Silica Glass

<table>
<thead>
<tr>
<th>Absorption Band</th>
<th>Defects in Silica Glass</th>
</tr>
</thead>
<tbody>
<tr>
<td>163 nm</td>
<td>(=) Si - Si = (Si(III) defects)</td>
</tr>
<tr>
<td>215 nm</td>
<td>(=) Si - O - O - (Peroxy radical)</td>
</tr>
<tr>
<td>225 nm</td>
<td>(=) SiH \cdots! O_{3} Si = (E2' center)</td>
</tr>
<tr>
<td>245 nm</td>
<td>(B_{2} \alpha \rightarrow = Si - Si = (Si(III) defects))</td>
</tr>
<tr>
<td></td>
<td>(B_{2} \beta \rightarrow Unknown)</td>
</tr>
<tr>
<td>260 nm</td>
<td>(=) Si - O - (NBOHC)</td>
</tr>
<tr>
<td>325 nm</td>
<td>(=) Si - O - O - Si = (Peroxy)</td>
</tr>
<tr>
<td>360 - 380 nm</td>
<td>Created by Gamma-ray</td>
</tr>
<tr>
<td>600 - 630 nm</td>
<td>(=) Si - O - (NBOHC) \rightarrow 630 nm</td>
</tr>
<tr>
<td></td>
<td>(=) Si - O - \cdots! H - O - Si = (NBOHC + H)</td>
</tr>
<tr>
<td>660 nm</td>
<td>Created by Gamma-ray</td>
</tr>
<tr>
<td>760 nm</td>
<td>Unknown</td>
</tr>
<tr>
<td></td>
<td>Radiation hardning, Photo-bleaching</td>
</tr>
<tr>
<td>945, 1240, 1390 nm</td>
<td>(=) Si - O - H = (OH radical)</td>
</tr>
<tr>
<td>1240 nm</td>
<td>H - H</td>
</tr>
<tr>
<td>1440 nm</td>
<td>(=) Si - O - \cdots! H(^{+}) = (Peroxy radical + H)</td>
</tr>
<tr>
<td>1450 nm</td>
<td>(=) Si - O - O - H = (Hydro-peroxy)</td>
</tr>
<tr>
<td>1520 nm</td>
<td>(=) Si - O - \cdots! H - H = (Peroxy radical + H)</td>
</tr>
<tr>
<td>1000 nm - 1600 nm</td>
<td>Centered at 1240 nm</td>
</tr>
<tr>
<td>(&gt;1600) nm</td>
<td>Created by Gamma-ray</td>
</tr>
</tbody>
</table>
dangling bonds and so on. Table 3 shows the imperfection of SiO$_2$ glass. The better radiation resistivity is dependent on the structure of glass which is determined by the changing of the refractive index of core and clad and the structure of SiO$_2$ with the dopants.

4. CONCLUSION

14 MeV fast neutron irradiation effects on pure silica core optical fiber was observed as comparing the results on $^{60}$Co gamma-ray irradiation. As a result, the following characteristics have become clear.

In case of 14 MeV fast neutron irradiation, induced absorption was concentrated in the wavelength from UV to 700 nm. Induced absorption could be separated into two absorption band. One is the UV-visible band, effected by E'-center, which absorption is inversely proportinal to the wavelength($1/\lambda$) shorter than 700 nm. Other is the absorption peak, centered at 630 nm, which was attributed to the optical absorption by NBOHC.

In case of gamma-ray irradiation, the absorption spectrum is appeared widely than that of 14 MeV fast neutron irradiation. The absorption is spread its wavelength region from UV to 1000 nm.

The experiment of neutron irradiation effects on optical fibers, i.e. knock-on collisions and hence cause the formation of E'centers, Si defects, peroxy-radical, dangling oxygen radicals and other defects, are less than that of gamma ray irradiation. Therefore, future basic researches and developments have to be studied and progressed parallel with the nuclear technology.

References

STUDY ON BUTENES AS SUPERHEATED LIQUID FOR NEUTRON DETECTOR

Teruko Sawamura, Tatsuya Joji and Hastuo Yamazaki*
Department of Nuclear Engineering, Faculty of Engineering,
Hokkaido University

1. INTRODUCTION

Neutron detectors basing on the visible bubble formation induced by radiation in superheated liquid was developed in these ten years. The principle is the same as that of the bubble chamber, except the superheated liquid is in drop form suspended in a matrix material such as viscous liquid or elastic polymer. In the bubble chamber a liquid is kept as superheated state. After the passage of ionizing radiation through this superheated liquid, along its tracks vapor bubbles are formed in the liquid.

The detectors recently developed, which are known as superheated drop detectors or bubble detectors, contain a small tube of clear material in which many sensing liquid drops are dispersed. The liquid has a boiling point lower than the detector working temperature. When the detector is being stored, the liquid is externally pressurized to be insensitive for radiation and, by removing the pressure, the liquid becomes superheated state, which is called sensitization. In other words each liquid drop is an individual bubble chamber. When one of the drops vaporizes and expands its volume, the other drops are not affected and keep the superheated state by deforming of the drop—dispersed matrix. Thus the detector retains the sensitivity until all the drops have been vaporized.

In this paper, Freons and other some liquids having a low—boiling temperature have been studied as sensitive liquids for neutron detection. The response and threshold energy for the neutron detection have been investigated experimentally and compared with the theoretical calculation. The bubble formation process in the sensitive liquids, however, has not been fully understood.

To investigate the basic process of the bubble formation induced by neutrons, the use of simpler liquids is considered to be more advantageous. Four
butenes, indicated in Table 1, are discussed as the sensitive liquids.

2. RESPONSE OF NEUTRON DETECTOR

By neutron interactions with nuclei constituting the sensitive liquid, charged particles are emitted and deposit a part of those energy in a localized area along those tracks. The energy eventually becomes heat and vaporizes the liquid locally. This locally high temperature area is called a nucleus of vaporization and this phenomena is called, nucleation. The nucleus having a larger radius than a critical radius, determined by mechanical equilibrium between pressure inside and outside of the nucleus, grows up to be a visible bubble until whole liquid of the drop where the nucleus produced vaporizes. The nucleus having a smaller radius than that is collapsed by the pressure of the liquid outside of the nucleus.

Thus, the detection sensitivity for neutrons having energy $E_n$, $K(E_n) = \frac{\text{number of visible bubbles}}{\text{neutron fluence}}$, is given as

$$K(E_n) = V \sum_i N_i \sum_j \sigma_{ij}(E_n) S_{ij}(E_n).$$

(1)

$V$: detector volume

$N_i$: number density of $i$-atom constituting the sensitive liquid.

$\sigma_{ij}$: cross section of an interaction of $i$-atom emitting $j$-charged particle (called $ij$-particle in the following).

$S_{ij}$: visible bubble formation probability by $ij$-particle.

The probability, $S_{ij}(E_n)$ is given as

$$S_{ij}(E_n) = \int_{E_{ij}\text{min}}^{E_{ij}\text{max}} \frac{\epsilon_{ij}(E_n, E_{ij})dE_{ij}}{\epsilon_{ij}(E_n, E_{ij})dE_{ij}}$$

(2)

$\epsilon_{ij}(E_j, E_n)$: energy spectrum of $ij$-particle produced by neutrons with $E_n$. Maximum and minimum energy of the particle are $E_{ij}\text{max}$ and $E_{ij}\text{min}$ respectively.

The integral of the numerator is performed for $E_j$ satisfied a criterion of the visible bubble formation described in the next section.

Many experiments for Freon-12 as a sensitive liquid have been reported because of its low boiling point. In the case of Freon, neutron interactions
with the atoms of C, Cl and F contribute to the detection sensitivity. However only atoms of C in butenes have possibility to form the visible bubbles. And there exist four kinds of butene, with nearly the same physical parameters associated with the neutron interactions and different thermal parameters. This is convenient to investigate the basic mechanism of the bubble formation and the neutron detection response.

3. CRITICAL ENERGY OF VISIBLE BUBBLE FORMATION

The bubble chamber was originally developed by Glaser in 1952 and then the bubble formation process was considered by Seitz. He thought the energy imparted by the charged particle propagated near the particle path and resulted in region of extremely hot fluid, 'thermal spike'. According to this theory, the radiation—induced cavitation occurs when the deposited energy in the liquid exceeds the energy forming the smallest bubble, or nucleus, which can grow spontaneously as a result of evaporation of the fluid. The radius of this bubble is given by the equilibrium condition:

\[ P_v = P_i + \gamma / R_{cr}. \] (3)

- \( P_v \) : vapor pressure in the bubble.
- \( P_i \) : liquid pressure outside the bubble.
- \( \gamma \) : surface tension of the liquid.
- \( R_{cr} \) : the bubble radius.

The radius, \( R_{cr} \), is called a critical radius. The energy needed to form the bubble with radius=\( R_{cr} \) is called as the critical energy.

According to the reversible thermodynamic work to form a vapor bubble of critical size

\[ E_c = - (4\pi/3)R_{cr}^3 \Delta P + 4\pi R_{cr}^2 \gamma (T) \] (4)

By substituting \( R_{cr} = 2\gamma / \Delta P (\Delta P = P_v - P_i) \) into (4),

\[ E_c = 4\pi \left\{ (2\gamma / \Delta P)^2 \gamma - (2\gamma / \Delta P)^3 \Delta P / 3 \right\} \]
\[ = (16\pi/3) \gamma (T)^3 / (\Delta P)^2. \] (4')
Seitz gave the different expression for $E_c$ by considering energy of vaporization, assuming that initial bubble is formed so quickly that the energy cannot be furnished from the ambient energy. Thus

$$E_c = (4\pi/3)R_c^3\rho_1 + 4\pi R_c^2 \gamma(T) + (4\pi/3)R_c^3 n_v H_s$$

(5)

$n_v$: the number of moles per unit volume in the vapour at pressure $P_v$.

$H_s$: the heat of the sublimation per mole.

Here the first term is added to the expression in Seitz's original paper, which is work done in expanding the bubble against the pressure $P_1$.

The second term of (4) or (5) was exchanged by the energy of the surface,

$$4\pi R_c^2 \left( \gamma(T) - T(d\gamma/dT) \right),$$

in the expression derived by Bugg. He also added the effect of the temperature difference between liquid and vapor inside the bubble, liquid viscosity and kinetic energy of liquid given by vapour expansion:

$$E_c = -(4\pi/3)R_c^3\partial P + 4\pi R_c^2 \left\{ \gamma(T) - T(d\gamma/dT) \right\}$$

$$+ (4\pi/3)R_c^3 \left\{ n_v H_s + \frac{T_v}{T_1} \right\} + 2\pi \rho_1 R_c^3 r^2 + \alpha(T)$$

(6)

$T_v, T_1$: vapor and liquid temperature.

$\rho_v, \rho_1$: vapor density at pressure $P_v$ and liquid density at pressure $P_1$.

$(C_v)_v$: specific heat of the gas at constant volume and pressure $P_v$.

$2\pi \rho_1 R_c^3 r^2$: kinetic energy owing to radial motion of the surface of the bubble.

$\alpha(T)$: imparted energy to the liquid during the growth of the bubble by the action of viscous force.

Neglecting the last two terms, he estimated the lower limit of $E_c$ under the conditions:

$$T_v \sim T_1$$

$$P_v \sim P_m, P_m: equilibrium vapour pressure at T$$

$$R_c = 2\gamma/\partial P, (\partial P = P_m - P_1)$$

$$E_c = -(4\pi/3)R_c^3\partial P + 4\pi R_c^2 \left\{ \gamma(T_1) - T_1(d\gamma/dT) \right\} + (4\pi/3)R_c^3 n_v H_s$$

$$= (16/3)\pi (\gamma^2/\partial P^2) \left\{ 2n_v H_s/\partial P + 1 - 3(T_1/\gamma)(d\gamma/dT) \right\}$$

(6')
Semenova expressed $\delta P$ in eq.(6) as $(P_i-P_1)(1-V/V'')$, where $V/V''$ was volume ratio of liquid and vapor in the bubble \cite{4}.

If $\delta P$ in the first term, $-(4\pi/3)R_{c}^{3}\delta P$, of the equation (6) is changed by $-P_1$, which means that $P_v\sim0$ during nucleation process, then (6') becomes

$$E_c = (4\pi/3)R_{c}^{3}P_1 + 4\pi R_{c}^{2}(\gamma(T_i)-T_i(d\gamma/dT)) + (4\pi/3)R_{c}^{3}n_vH_s$$

$$= (16/3)\pi(\gamma^3/\delta P^2)(2n_vH_s/\delta P + (P_i/\delta P) - 3(T_i/\gamma)(d\gamma/dT))$$

which is given by Harper \cite{5}. Comparing with each eq.(4), (5) and (7), the largest $E_c$ is resulted by (7) and the smallest $E_c$ by (4) because $(P_v/\delta P)\geq1$ and $-(3/2)(T_i/\gamma)(d\gamma/dT) > 0$.

4. ESTIMATION OF $E_c$

Few experimental values of $E_c$ have been reported by Apfel for Freon—114 and isobutane \cite{5}. Those are shown in Fig.1 together with the estimated values of $E_c$ derived with (4) and (7). Physical parameters necessary for the calculation are derived by referring to the book written by Reid et al \cite{7}. From the figure the experimental values are found to fit on Harpers expression, eq.(7).

We calculated the critical radius and the critical energy due to eq.(7) for butenes and Freons in table 1. and shows in Fig.2 and Fig.3 respectively. The boiling temperature is one of the dominant factors to determine the value of $E_c$. Fig.4 is the relation between $E_c$ at 20 °C and the boiling temperature. Butene has the higher critical energy and then may be less sensitive for neutron detection than Freon—12. From Fig.3 $E_c$ of Freon—12 at 20 °C is nearly equal to that of isobutene at about 40 °C. We could study about a butene detector under an elevated temperature condition.

5. BUTENE DETECTORS AND BUBBLE COUNTING

We made two types of detectors; the matrix material of A—type is glycerine and that of B—type is glycerine+polyacrylamide \cite{8}. Though the A—type detectors are easily made, but the ascending of butene—liquid—drops and the more rapid ascending of produced visible bubbles are serious problem during neutron irradiation. The emitted sound at visible bubble formation was used for the bubble counting of A—type. The B—type detectors get rid of that problem. Polyacrylamide, an elastic
polymer, traps the liquid drops and also the visible bubbles to stay at the position where those are formed. After the polymerization and after the neutron irradiation small quantity of Freon-12 is poured over the detector medium. Saturated vapor pressure at storing temperature of the detector makes the detectors insensitive. Removing the Freon sensitize the detectors before irradiation. Bubble counting by naked eye after the irradiation is possible, however, we also applied the acoustic counting for the B-type as this method is convenient for large number of the bubbles.

Two types of detectors were irradiated with Am-Be neutrons. The detectors were confirmed to be sensitive for the neutrons. The B-type detectors are tested and confirmed, if they have reusability gained by repressurization. The temperature control during the making process and the irradiation of the detectors, however, is insufficient to discuss the detector quality.

6. CONCLUSION

Butenes as sensitive liquids of superheated drop neutron detectors are discussed. The values of the critical energy to form visible bubble are calculated for butenes and Freons. Butenes are expected to be sensitive at an elevated temperature up to about 40 °C. We made two types of detectors with different matrix materials and irradiated those with Am-Be neutrons. Though the detectors were confirmed to be sensitive for neutrons and for B-type to be possible to reuse, quantitative results have not been gained by insufficient testing condition. Improving the conditions and the detector-making procedure are going on.

ACKNOWLEDGEMENTS

We received the instruction from Mr. K. Koizumi in making the detectors. We wish to thank him for his instruction and valuable discussions.
References


<table>
<thead>
<tr>
<th>liquid</th>
<th>density (g/cm³)</th>
<th>boiling point at 760mmHg(°C)</th>
<th>vapor pressure at 20 °C (atm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1-Butene</td>
<td>0.5951*</td>
<td>-6.0</td>
<td>2.6</td>
</tr>
<tr>
<td>cis-2-Butene</td>
<td>0.6213*</td>
<td>+3.7</td>
<td>1.8</td>
</tr>
<tr>
<td>trans-2-Butene</td>
<td>0.6042*</td>
<td>+0.8</td>
<td>2.0</td>
</tr>
<tr>
<td>iso-Butene</td>
<td>0.5942*</td>
<td>-7.0</td>
<td>2.6</td>
</tr>
<tr>
<td>Fleon-12</td>
<td>1.311 **</td>
<td>-29.8</td>
<td>5.8</td>
</tr>
<tr>
<td>Fleon-114</td>
<td>1.455 **</td>
<td>3.5</td>
<td>1.9</td>
</tr>
</tbody>
</table>

Table 1. Liquids and those physical parameters
Fig. 1 Comparison of the experimental values (6) with the calculation by eq. (4) and (7) in the text.

Fig. 2 Relation between the critical radius and temperature at liquid pressure = 1 atm.
Fig.3 Critical energy vs temperature, calculated with eq.(7)
Fig. 4 Relation between the critical energy and the boiling temperatures at liquid pressure=1 atm.
Development of Superconducting Tunnel Junction Type X-ray Detector

M. Ukibe, M. Nakazawa, T. Iguchi, H. Tkahasi
Department of Quantum Engineering and Systems Science Faculty of Engineering,
The University of Tokyo, Hongo, Bunkyo-ku, Tokyo 113, Japan

M. Kisimoto, M. Katagiri
Japan Atomic Energy Research Institute, Tokai-mura, Naka-gun, Ibaraki 319-11, Japan

M. Kurakado
Advanced Materials & Technology Research Laboratories,
Nippon Steel Corporation, 1618 Ida, Nakahara-ku, Kawasaki 211, Japan

1. Introduction

In the field of X-ray Spectroscopy, it is necessary to develop a radiation detector with good energy resolution. Now, a semiconductor detector has the best energy resolution in all kind of detectors. Since present performance of semiconductor detector is almost theoretical limit due to inherent characteristic of the semiconductor, it is very difficult to improve that further. Therefore, a radiation detector using superconducting tunnel junction (STJ) have been investigated very actively in many groups of the world. At present, energy resolution of the STJ X-ray detector is better than that of the other detector. So we began to develop a Superconducting Tunnel Junction Type X-ray Detector that can be used for various experiments practically. A He cryostat having a X-ray window was made for the STJ, so that the STJ X-ray detector can be irradiated by a radiation source outside cryostat and a detection efficiency of the STJ can be measured. By the He cryostat, we investigated a X-ray detection characteristic of the STJ and analyzed radiation signals from the STJ by digital processing.

2. Experimental system

The developed He cryostat is shown in Fig. 1. A STJ was set on a 0.4K stage parallel to this cryostat side. The cryostat had three thin Be windows for passing of X-rays. The outer Be
Fig. 1 External source $^3$He cryostat

3. Characteristic of The STJ

Two STJs, which were fabricated by Advanced Materials & Technology Research Laboratories, Nippon Steel Corporation, were used in this study. In the following, a $178 \times 178 \mu m^2$ STJ on wafer No55 was called A-STJ. A $100 \times 100 \mu m^2$ STJ on wafer No55 was called B-STJ. A- and B-STJ had Nb/Al-AlOx/Nb layers. Two STJs had about 0.7 $\mu m$ in thickness respectively. A $^{55}$Fe X-ray source which was $3.7 \times 10^6$ Bq was set in front of the outer Be window.

Figure 3 shows pulse height spectra of X-rays obtained with A- and B-STJ. Figure 3a shows a X-ray spectrum measured by A-STJ. The energy resolution determined by the full-width-at-half-maximum (FWHM) of the peak was 166 eV for 5.9 keV X-rays. The energy resolution of the pulser peak is 81 eV. Figure 3b shows a X-ray spectrum measured by B-STJ. The energy resolution for 5.9 keV was 273 eV. The energy resolution of the pulser peak is 42 eV. All X-ray peaks were made by X-rays absorbed in the Nb base electrode.

Since the capacitance of STJ increases as the STJ area increases, electronic noise increases with the increase in STJ area. So B-STJ had the better energy resolution of the pulser than A-STJ had. A-STJ had, however, the better energy resolution for 5.9 keV than that of B-STJ. In other words, the energy resolution of X-ray spectra is improved as the STJ area increases. The reason of this result was considered as explained below. Since diffusion length of excited electrons in Nb electrode is considered about 30 $\mu m$ on the average and the ratio of the diffusion length to the scale of STJ area can not be neglected, the quantity of excited electrons which is affected by the window was 250 $\mu m$ thick and the other two were 100 $\mu m$. Distance between X-ray source outside the cryostat and a STJ was 2.5 cm. In consequence X-ray measurements with various radiation sources can be carried out during one cooling cycle.

It takes about five hours to cool the STJ down from room temperature to 0.4 K using the cryostat. The cryostat can keep temperature of the STJ at 0.4 K for about thirty hours.

The block diagram of the detector using STJ is shown in fig.2.
Effect of energy diffusion in B-STJ is much bigger than in A-STJ.

Experimental intrinsic total detection efficiency, absolute peak detection efficiency for 5.9 keV X-ray and intrinsic peak detection efficiency for 5.9 keV X-ray was obtained by Peak analysis for two X-ray spectra. Besides, theoretical intrinsic total detection efficiency was calculated by considering a ratio of X-rays absorbed in Nb electrodes to X-rays hitting STJ. The experimental and theoretical detection efficiencies are shown in Table. 1. In Table. 1, it was considered that the effect of energy diffusion made experimental intrinsic peak detection efficiency for 5.9 keV X-rays in A-STJ better than in B-STJ. And the calculation of theoretical intrinsic total efficiencies were based on only signals generated by X-rays absorbed in Nb base electrode. But in fact phonons generated by X-rays absorbed in the sapphire wafer and SiO₂ around the STJ diffuse to Nb base electrode and make phonon signals. So experimental intrinsic total efficiencies were bigger than theoretical ones for two STJs.

In this study we evaluated only intrinsic total detection efficiency theoretically. So in the future we will take the effect of energy diffusion into account on evaluating theoretical intrinsic peak detection efficiency.

Table. 1 Detection efficiency of two STJs for X-ray

<table>
<thead>
<tr>
<th>STJ</th>
<th>Energy</th>
<th>experimental absolute peak efficiency</th>
<th>experimental intrinsic peak efficiency</th>
<th>experimental intrinsic total efficiency</th>
<th>theoretical intrinsic total efficiency</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>6.5 keV</td>
<td>4.32 x 10⁻⁹</td>
<td>0.0755 %</td>
<td>0.155 %</td>
<td>0.105 %</td>
</tr>
<tr>
<td>A</td>
<td>5.9 keV</td>
<td>3.59 x 10⁻⁸</td>
<td>0.628 %</td>
<td>1.29 %</td>
<td>1.10 %</td>
</tr>
<tr>
<td>B</td>
<td>5.9 keV</td>
<td>8.42 x 10⁻⁹</td>
<td>0.467 %</td>
<td>1.32 %</td>
<td>1.10 %</td>
</tr>
</tbody>
</table>

4. X-ray Signal Analysis using Digital Processing

X-ray signals from the preamplifier and main amplifier measured by a digital oscilloscope, Lecroy Model 9450A. Figure 4 shows a typical pulse shape of output signal from preamplifier for the X-ray peak of 5.9 keV X-rays measured by A-STJ. The Risetime determined by a period between
10% and 100% of the full pulse height was about 0.5μsec. This risetime is called risetime(10-100%). Since the risetime of output signal from preamplifier for a pulser, whose original risetime was 0.05μsec on pulse generator, was 0.4μsec, the risetime(10-100%) of X-ray signal was considered as time constant of STJ. Since the intrinsic tunneling time constant of A-STJ was about 5μsec, it estimated that generating signal charge depended on diffusion time of excited electrons in the STJ. It is confirmed from the result of pulse shape that shaping time constant of the main amplifier above 1.0μsec is necessary to collect signal charge completely. Figure.5 shows the correlation between risetime(10-100%) and signal voltage from the preamplifier for X-ray signals with A-STJ. Roughly the correlation is divided into two regions. It is considered that A-region contains phonon signals and B-region contains X-ray signals.

6. CONCLUSION

1. The absolute peak detection efficiency for X-rays was determined using the External source 3He cryostat.

2. For 5.9keV Xrays, theoretical intrinsic total detection efficiency and experimental were equal within 20%.

3. It is confirmed that X-ray signals can be distinguished from phonon signals in correlation between the risetime and peak voltage of output signals using digital processing analysis.
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A DIGITAL SIGNAL WAVEFORM PROCESSING FOR SEMICONDUCTOR RADIATION DETECTORS

Tomohiko Kurahashi, Hiroyuki Takahashi, Tetsuo Iguchi, Masaharu Nakazawa
Department of Quantum Engineering and Systems Science, Faculty of Engineering, the University of Tokyo, Japan

1. INTRODUCTION

A leading edge of an output pulse signal from a radiation detector contains information about an electric charge collecting process in it \(^{(1)}\) \(^{(2)}\). The goal of signal waveform processing is to improve precision and efficiency of radiation measurements by analyzing signal waveforms. It will be difficult to perform complicated analysis of signal waveforms with a conventional analog signal processing method due to its hardware complexity. But nowadays the development of high speed and extremely precise ADC makes it possible to apply a computerized waveform processing method to digitized output signals \(^{(1)}\).

In this paper, as an example of digital signal waveform processing methods for semiconductor radiation detectors \(^{(3)}\) \(^{(4)}\), an analysis result about a cadmium telluride (CdTe) detector is discussed.

2. SIGNAL WAVEFORM FROM PLANAR SEMICONDUCTOR RADIATION DETECTOR

When electron-hole pairs are produced inside a semiconductor radiation detector by a radiation incidence, the amount of induced charge generated with the travel of the two charge carriers to the \(n^+\) or \(p^+\) layer is usually observed as an output pulse signal from the detector. In the case of planar detectors, the amount of induced charge, \(Q(t)\), is shown as eq. (1).

\[
Q(t) = \frac{q_0}{d} \left( \text{traveling distance of electron} + \text{that of hole} \right) \\
= q_0 \left[ \frac{v_e}{d} \text{min}(t, t_e) + \frac{v_h}{d} \text{min}(t, t_h) \right] \quad (1)
\]
$d$ is thickness of detector, and $q_0$ is the maximum amount of induced charge defined as $q_0 = n_0 e$ ($n_0$ is the number of produced electron-hole pairs and $e$ is electron charge). $v_e$ and $v_h$, $t_e$ and $t_h$ show velocity and maximum traveling time of electron and hole respectively.

When the electric field in the active volume of a detector is supposed to be sufficiently high and uniform, velocity of charge carriers is constant and therefore maximum traveling time varies with the position where the electron-hole pair is produced in the active volume. Fig.1 shows typical examples of signal waveforms from a planar semiconductor radiation detector. When an electron-hole pair is produced at the position which is located at $x$, $t_e$ is defined as $t_e = x/v_e$, and $t_h$ as $t_h = (d - x)/v_h$.

![Diagram](image)

**Fig. 1:** Signal waveforms from a planar semiconductor radiation detector.

![Diagram](image)

**Fig. 2:** Effect of charge trapping and charge detrapping.

(time scale in (b) is much larger than in (a).)

In addition to the produced position of electron-hole pairs, charge trapping and charge detrapping are also important for signal waveforms from a semiconductor.
detector \(^{(6)}\). As shown in Fig. 2(a), due to charge trapping, the leading edge of output signal becomes dull and consequently maximum pulse height decreases. If charge detrapping occurs, the signal waveform will have two typical components as shown in Fig. 2(b).

3. ANALYSIS RESULT

Here is discussed an analysis result of measured data using a \(^{137}\)Cs \(\gamma\)-ray source with a CdTe semiconductor radiation detector \(^{(7)}\) at a room temperature. Fig. 3 shows an example of measured signal waveforms, and Fig. 4 shows a measured energy spectrum (hereafter pulse height is reduced to half in energy spectrums). In this energy spectrum, the number of data on the lower maximum pulse height side of both photopeak and Compton continuum is increasing and consequently photopeak becomes obscure. This is because many numbers of measured signal data are affected by charge trapping during charge carriers (especially hole) collection.

![Fig. 3: An example of measured waveforms.](image)

![Fig. 4: Measured energy spectrum.](image)

The purpose of this analysis is to make a photopeak clear by rejecting signal data that have been affected by charge trapping. We divided each signal waveform into sections at intervals of 5% of its maximum pulse height, and obtained time of sections between 5% and 90% of its maximum pulse height. We thought that this time has the following characteristics:

- in the case that signal waveform in some consecutive sections consists of one straight line, time of those sections is constant.
- in the case that the slant of signal waveform becomes dull by the effect of charge trapping, time in the sections increases in sequence.
• For example, when $T_X$ is time of section $X$ ($X = A, B, C, D, E$) in Fig. 5, the following relations must be held: $T_B = T_A$, $T_E = T_D$ in (a) and $T_A = T_B < T_C < T_D < T_E$ in (b).

• Note that charge detrapping is able to be disregarded in this analysis, because measurement time per pulse is too short for charge detrapping to occur.

![Waveforms with and without the effect of trapping.](image)

**Fig. 5:** Waveforms with and without the effect of trapping.

and actually selected measured data according to our thought.

The dotted line in Fig. 6 shows a original energy spectrum, and the solid line shows an energy spectrum obtained after rejection of events affected by charge trapping. Table 1 summarizes differences between two energy spectrums in Fig. 6. By comparing these two spectrums, it is obvious that both the removal of the effect of charge trapping and the clarification of photopeaks are done by this data selection.

![Energy spectrums.](image)

**Fig. 6:** Energy spectrums.
Table 1: The result of data selection.

<table>
<thead>
<tr>
<th></th>
<th>number of data</th>
<th>peak (pulse height/num of data)</th>
<th>FWHM (pulse height/keV)</th>
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<tr>
<td>before selecting</td>
<td>31169</td>
<td>158 / 132.2</td>
<td>15.0 / 62.65</td>
</tr>
<tr>
<td>after selecting</td>
<td>3302</td>
<td>159 / 79.0</td>
<td>3.1 / 13.08</td>
</tr>
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</table>

4. CONCLUSION

In this paper an example of digital signal waveform processing methods for a semiconductor radiation detector is discussed, and the analysis result indicates that this method is effectively usable for CdTe detectors.

However, as is obvious in Table 1, data selection to make photopeak clear is also a cause of reducing counting efficiency. In order to keep the efficiency, development of a method to revise data affected by charge trapping is necessary.

We plan to make further efforts on this point, which will be a fully intelligent new signal processing in future.

References

Experimental study and numerical simulation in He/Ne/Ar nuclear pumped laser

N.Yamanaka, H.Takahashi, T.Iguchi and M.Nakazawa,
Department of Quantum Engineering & Systems Science
University of Tokyo, Hongo 7-3-1, Bunkyo-ku, Tokyo, 113, Japan

T.Kakuta, H.Yamagishi and M.Katagiri
Advanced Science Research Center
Japan Atomic Energy Research Institute
Tokai-mura, Ibaragi-ken, 319-11 Japan

Abstract

A Nuclear Pumped Laser (NPL) using $^{3}$He/Ne/Ar gas mixture is investigated for a purpose of applying to an optical self-powered neutron detector. Reactor experiments and simulations on lasing mechanism have been made to estimate the best gas pressure and mixture ratios on the threshold input power density (or thermal neutron flux) in $^{3}$He/Ne/Ar mixture. Numerical results show that the best mixture pressure is $^{3}$He/Ne/Ar=2280/60/100 Torr and thermal neutron flux threshold $5 \times 10^{12}$ n/cm$^{2}$/sec, while the reactor experiments made in the research reactor “YAYOI” of the University of Tokyo and “JRR-4” of JAERI also demonstrate that excitational efficiency is maximized in a similar gas mixture predicted by the calculation.

1. INTRODUCTION

Nuclear pumped lasers have been investigated as a promising method to transfer nuclear energy into optical energy. In 1980, B.D.Carter, et al. reported CW lasing on Ne 5s$'[1/2]$-3p$'[3/2]$ transition with $\lambda = 632.8$ nm in $^{3}$He/Ne[1]. The threshold for lasing was achieved at a neutron flux level of $2 \times 10^{11}$ n/cm$^{2}$/sec. However, this result was refuted theoretically by M.A.Prelas and G.A.Schlapper in 1981[2], on the basis of gain calculation data, and experimentally by E.G.Batyrbekov, et al. in 1991[3].
Recently, Ne 3p'[1/2] - 3s'[3/2] of λ = 585.2 nm laser in 3He/Ne/Ar(H2) mixture system has been studied where third material Ar(H2) is added as a quenching material and the threshold neutron flux in 3He/Ne/Ar(H2) nuclear pumped laser was reported 10^{13} ~ 10^{14} n/cm^2/sec[3, 4].

We have previously showed the possibility of an optical self-powered neutron detector based on 3He/Ne NPL through an experimental study[5]. Here are discussed the applicability of 3He/Ne/Ar NPL to the neutron detector from simulation and experiment.

2. SIMULATION for He/Ne/Ar NPL

Based on a plasma process model[3](Fig.1) for nuclear pumped lasing in a He/Ne/Ar mixture, the simulation was made to find the relations among total gas pressure, gas mixture ratio, threshold power density. With this kinetic model there was considered the concentration balance of atomic and molecular ions: He^+, HeO^+, Ne^+, NeO^+, HeNe^+, Ar^+, ArO^+, ArO^2 and excited atoms and molecules: He^*, HeO^*, HeO^*, Ne^*, NeO^*, NeO^*, NeO^*, HeNe^*, HeNe^*, Ar^*, ArO^*, ArO^2, ArO^2. At the same time the level kinetics of Ne atoms 3p and 3s states were studied. Ne atom 3p state population is written as follow

\[
\frac{d}{dt}[\text{Ne}(3pi)] = \delta_i S - [\text{Ne}(3pi)] \left( \frac{1}{\tau_i} + [\text{Ne}] \sum_{k=i+1}^{10} k_{ik} \right) + [\text{Ne}] \sum_{k=1}^{i-1} k_{ki}[\text{Ne}(3pi)] , \quad (i = 1, 2, \ldots, 10) \] (1)

where, \( \delta_i \) is population efficiency[6], [Ne] - Ne atoms concentration, [Ne(3pi)], \( \tau_i \), \( k_{ik} \), and \( k_i \) - respectively concentration, radiative lifetime[7], rate constants intra and intermultiplet relaxation of 3pi levels of the 3p states system. The system of particle balance differential equations supplemented by the equations of charge conservation and electron energy balance was solved by a computersaided numerical method.

Fig.2 showed an example of numerical result on the dependence of the Ne 3p and 3s level populations on the input power density through 3He(n,p)T nuclear reaction in 3He/Ne/Ar=5000/60 /100 Torr mixture. In this case, the threshold energy density which can make inverse population between Ne 3p and 3s levels is 0.1 W/cm^3 corresponding to thermal neutron flux of 5 × 10^{12} n/cm^2/sec.

Fig.3 shows a map of the threshold input power density versus the partial gas pressures of 3He and Ne under the constant Ar gas pressure of 100 Torr. (No lasing occurs if the Ar gas pressure is under 100 Torr.) It is found that the condition of around 60 Torr of Ne and more than 5000 Torr of 3He gas pressures might give the lower threshold of less than 0.05 W/cm^3 corresponding to thermal neutron flux of 2 × 10^{12} n/cm^2/sec, which is comparatively easy to realize in research fission reactors with thermal neutron flux around 10^{14} n/cm^2/sec.
Figure 1: The scheme of basic kinetic processes occurring in He/Ne/Ar plasma

Thermal Neutron Flux [n/cm³ sec]

Figure 2: Ne atoms 3p and 3s level populations versus input power density
3. REACTOR EXPERIMENT

The preliminary neutron irradiation experiments on laser gas cells have been made in the maximum thermal neutron field of \(3.2 \times 10^9\) n/cm²sec of the “YAYOI” reactor in the Nuclear Engineering Research Laboratory of University of Tokyo the experimental setup is shown in Fig.4. Three types of gas cell listed in Table.1 were used, which normal type, optimized type and Ne reinforced type, respectively.

<table>
<thead>
<tr>
<th>No</th>
<th>He [Torr]</th>
<th>Ne [Torr]</th>
<th>Ar [Torr]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1140(1)</td>
<td>540(1)</td>
<td>206(1)</td>
</tr>
<tr>
<td>2</td>
<td>2280(2)</td>
<td>60(0.11)</td>
<td>100(0.49)</td>
</tr>
<tr>
<td>3</td>
<td>2280(2)</td>
<td>760(1.4)</td>
<td>100(0.49)</td>
</tr>
</tbody>
</table>

The value in blanket is ratio to cell NO.1

Table 1: mixture ratios

Light emissions from these cells under neutron irradiation have been observed by using a photo multi spectrum analyzer. As an example of the observed light emission spectra, Fig.5 shows the result from NO.3 gas cell of \(^3\)He/Ne/Ar=2280/760/100 Torr mixture in reactor power
Figure 4: "YAYOI" experiment system

Figure 5: Observed spectrum of NO.3 cell
He/Ne/Ar=2280/760/100 Torr mixture
of 2 kW corresponding to thermal neutron flux of $3.2 \times 10^{19}$ n/cm$^2$sec, where four lines of 585.2, 640.3, 702.3 and 724.5 nm on the Ne 3p-3s transitions are clearly appeared.

Fig.6 illustrates another experimental setup the thermal neutron field of about $10^{12}$ n/cm$^2$sec/MW of the "JRR-4" pool type reactor in Japan Atomic Energy Research Institute (JAERI). The observed spectra in NO.1 cell of Hc/Ne/Ar=1140/540/206 Torr mixture are shown in Fig.7, depending on the reactor power, where thermal neutron flux per unit reactor power [MW] is $1.1 \times 10^{11}$ n/cm$^2$sec. The clear peaks of NeI 585.2 nm are found with an intense and broad peaks in the region between 400 nm and 550 nm from Cherenkov radiation.

4. DISCUSSION

The dependences of NeI 585.2 nm line intensity for three type gas cells on thermal neutron flux are shown in Fig.8. In the thermal neutron flux region less than $10^{12}$ n/cm$^2$sec/sec, the line intensity of Ne 585.2 nm from NO.1 cell of $^3$He/Ne/Ar=1140/540/206 Torr in proportion to the neutron flux, but no lasing occurred.

Table.2 summarizes the "YAYOI" reactor experimental and numerical results of NeI 585.2 nm line intensity and this line intensity per unit Ne pressure, corresponding to exciting efficiency, of the three gas cells at the reactor power of 2 kW. The normalized line intensity in the gas mixture ratio of NO.2 cell is larger than those of other cells, which demonstrates experimentally the validity of numerical results on the gas mixture ratio. But, in numerical results this line intensity of NO.1 cell is smaller than those of NO.2 and NO.3 cell and these numerical results do not agree with experimental results. Thus, it is required to improve simulative model, especially charge transfer process between Ne$^+$ and Ar and quenching of Ne 3p levels in collision with electrons.
Figure 7: Observed spectrum of NO.1 cell
He/Ne/Ar=1140/540/206 Torr mixture

<table>
<thead>
<tr>
<th>cell NO</th>
<th>585.2 nm line intensity</th>
<th>line intensity / unit Ne pressure</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>counts/sec</td>
<td>counts/sec atm</td>
</tr>
<tr>
<td></td>
<td>experiment</td>
<td>calculation</td>
</tr>
<tr>
<td></td>
<td>calculation</td>
<td>experiment</td>
</tr>
<tr>
<td>1</td>
<td>119.95(1)</td>
<td>85.952(1)</td>
</tr>
<tr>
<td>2</td>
<td>80.837(0.67)</td>
<td>359.04(4.18)</td>
</tr>
<tr>
<td>3</td>
<td>575.72(4.80)</td>
<td>890.36(10.3)</td>
</tr>
</tbody>
</table>

The value in bracket is ratio to cell NO.1

Table 2: Ne 585.2 nm line intensities and intensities per unit Ne pressure
Figure 8: NeI 585.2 nm line intensity dependence on thermal neutron flux
NO.1 gas cell He/Ne/Ar=1140/540/206 Torr
NO.2 gas cell He/Ne/Ar=2280/60/100 Torr
NO.3 gas cell He/Ne/Ar=2280/760/100 Torr

5. CONCLUSION

The applicability of $^3$He/Ne/Ar NPL to an optical self-powered neutron detector has been considered through the simulation and the reactor experiments using the research reactors "YAYOI" and "JRR-4". The results show that He/Ne/Ar=2280/60/100 Torr mixture gives the best radiational efficiency for NeI 585.2 nm line, and the line intensity is proportional to the neutron flux less than $10^{12}$ n/cm$^2$sec. Although no lasing occurs in the present mixture ratios, these lasing gas cells are expected to be available as the a kind of in-core neutron flux monitor.

We are planning the next NPL experiments at larger thermal neutron flux $\sim 10^{14}$ n/cm$^2$sec.
References


TEST OF A STACKED NaI(Tl) SPECTROMETER WITH INTERMEDIATE ENERGY PROTONS

H. Murohka, H. Yoshida, O. Iwamoto, Y. Uozumi, A. Nohtomi,
T. Sakae, M. Matoba
Dep. of Nuclear Engineering, Kyushu University

T. Maki
University of Occupational and Environmental Health

N. Koori
Faculty of Integrated Arts and Sciences, University of Tokushima

Abstract
A stacked NaI(Tl) spectrometer which consists of two silicon detectors, 4 rectangular NaI(Tl) scintillators (2"x2"x2") and a cylindrical NaI(Tl) scintillators (φ 8cm x 18cm), has been developed to detect charged particles up to about 400MeV. Tests were performed using 300MeV and 350MeV proton beams from the ring cyclotron at RCNP. The ΔE-E technique has been used to identify particles and also events which are originated to nuclear reactions in the detector material. The tail of peak spectrum is suppressed clearly and the peak efficiency is reasonably estimated.

1. INTRODUCTION
In recent year, wide continuum spectra of charged particle reactions for intermediate energy are investigated to understand the feature of the reaction mechanism from low to high energy transition region. Systematic studies of (p,p'x) reaction were carried out below 200MeV, and there are a few data and some calculations of (p,nx) reaction at GeV energy region. It is desired to measure continuum spectra of charged particle reactions in the intermediate energy region. A wide energy acceptance spectrometer is required to realize this measurement.

A full-energy detector for charged particles in intermediate energy region needs thick material. Therefore, a part of energy is lost with nuclear reactions in the detector material when the detection energy increases. The energy deposit in the detector is smaller comparing with the full energy one. This process results in a "tail" in the energy spectrum. So, it is important to identify the event of the process of full energy deposit or of nuclear reactions in the detector material to use the detector as an energy spectrometer. For this aim, the detector has been frequently divided into several detector components (stacked detectors) and ΔE-E technique has been applied to suppress "tail" events.

In recent results, obtained spectra with stacked spectrometers were not good, and then the performances were not accurately checked, because these tests were not performed with monochro-
matic energy particles\textsuperscript{[8]}. In the present work, recoil protons in pp scattering were detected in coincidence to obtain a quasi-monochromatic energy particle. Detector tests were performed using 300MeV and 350MeV proton beams from the ring cyclotron at Research Center for Nuclear Physics (RCNP) in Osaka university.

\section*{2. EXPERIMENTAL DETAILS}

A schematic diagram of the stacked \textsuperscript{[8]}NaI(TI) spectrometer is shown in Fig.1. It consists of two thin silicon detectors, 4 rectangular NaI(Tl) scintillators (2" × 2" × 2"), and a cylindrical NaI(Tl) scintillator (φ 8cm × 18cm). All detectors were set in line. The overall material thickness of 38cm is enough to stop protons up to about 400MeV.

Two thin silicon detectors of 150μm and 300μm thick, were placed in front of the NaI(Tl) crystals; each of silicon detectors has an effective area of 300mm\textsuperscript{2}. Each rectangular NaI(Tl) crystal is framed in 1mm thick aluminum vessel. It has two optical windows (top and bottom) which are packaged with pyrex film of 4mm thick. Two photomultipliers (PMT: Hamamatsu R329-02) are attached to these optical windows. All optical contacts are kept with optical grease. Between adjacent NaI(Tl) crystals, there exists a dead layer which consists of 100μm thick aluminum isolation foil and 6mm air gap. The last cylindrical NaI(Tl) crystal has an optical contact with photo-multipliers (PMT: Hamamatsu R1307) at the end of the crystal.

Detector tests were performed using 300MeV and 350MeV\textsuperscript{[6]} proton beams from the ring cyclotron at RCNP. A 1.2mm thick polyethylene (CH\textsubscript{2}) was used as a target.

The stacked NaI(Tl) spectrometer was set in the way that its axis was directed along lines from 30° to 70° with angular step 10° and its front face was set at distance with about 80cm from the target as shown in Fig.2.

For \textit{H}(p,p)\textit{H} scattering measurement, a coincidence spectrometer which consists of two thin silicon detectors of 150μm and 300μm thick and a rectangular NaI(Tl) scintillator with a PMT was arranged. Thus, quasi-monochromatic proton events could be selected.
Fig. 2: A typical arrangement to measure 215MeV protons is displayed.

Coincidence signals between two silicon detectors and first NaI(Tl) scintillator in the stacked spectrometer and the coincidence spectrometer were used as gate signals for data acquisition system. The shaping time of each amplifier was set to 1 μsec.

Energies of measured protons from pp elastic scattering were 215MeV, 115MeV, 64MeV, 22MeV at 300MeV incident energy and 250MeV, 160MeV at 350MeV incident energy.

3. EXPERIMENTAL RESULTS

Protons which scattered in the CH₂ target pass through a thin mylar window of the target chamber, and then flies about 80cm in air. Then scattered protons pass through two silicon detectors before entering the stacked NaI(Tl) scintillator. A typical energy spectrum of protons penetrating through the first scintillator at 215MeV is shown in Fig.3.

The energy calibration was performed using maximum energy loss in each rectangular NaI(Tl) crystal from \(^{12}\)C(p,p'x) reaction and also the full energy peak of each angle.

An edge which is observed at about 550 channel corresponds to the maximum energy which is deposited in a single rectangular NaI(Tl) scintillator with protons, that is about 122MeV. Each amplifier gain was adjusted to these edges to be appeared in similar channels. Fine adjustments were done in off-line analysis.

A peak at 760 channel corresponds to protons scattered from H target at 30° which has energy of 215MeV penetrating though two thin silicon detectors and the first NaI(Tl) crystal. That deposited energy in the first NaI(Tl) crystal is about 57MeV. The deposited energy was estimated using a relation between the light output of the NaI(Tl) crystal and the real energy loss in the detector material\(^7\).

Figure 4 shows a typical ΔE-E plot. The horizontal axis shows the energy sum of two silicon detector outputs and NaI(Tl) scintillation detector outputs. The vertical axis shows the energy sum of \(Δ_{SSD1} + Δ_{SSD2} + Δ_i\) where \(Δ_{SSD1}\) and \(Δ_i\) represent the energies which deposit in the i-th silicon detector and the i-th NaI(Tl) scintillator, respectively. Events are not selected in this condition.

Dense dots at 215MeV is due to protons elastically scattered from the H target and energy deposit is almost constant at low energy in the first scintillator in vertical axis.
Fig. 3: A energy spectrum of protons penetrating in the first scintillator at 215 MeV proton.

Fig. 4: $\Delta E$-$E$ plot of elastic scattered protons at energy of 215 MeV. A peak exists at 215 MeV in horizontal axis. Energy deposit is almost constant at low energy in the first scintillator in vertical axis.
4. DISCUSSION

Every event can be identified as events of full energy loss or of nuclear reactions using the \( \Delta E-E \) method. Parameter \( E \) is the full energy of the particle and \( \Delta E \) the energy loss in the first scintillator. The particle identification parameter \( P \) is defined as,

\[
P = \frac{[E^b - (E - \Delta E)^b]}{n}
\]

Here the parameter \( b \) is almost constant and is set to a value of 1.73 for proton. The parameter \( n \) is the number of the \( n \)-th scintillator as \( \Delta E \) detectors.

Figure 5 shows a typical particle identification spectrum which was obtained from \( \Delta E = \Delta_{SSD1} + \Delta_{SSD2} + \Delta_1 \) in the stacked spectrometer at 215MeV proton. Since the peak corresponds to protons of full energy, the Gaussian shape fitting can be applied.

The division region was set by a Gaussian shape fitting to the particle identification spectrum in Fig.5. For example, 5 window settings of \( \pm \sigma_1, \pm \sigma_2, \pm \sigma_3, \pm \sigma_4, \pm \sigma_5 \) were applied, where \( \sigma_i \) means the \( i \)-times of the standard deviation. Using this method, protons which corresponds to elastically scattered from \( H \) were clearly distinguished.

A typical spectrum is shown in Fig.6 together with two spectra of "peak" and "tail". With the window settings from \( \pm \sigma_1 \) to \( \pm \sigma_3 \), the Gaussian shape is reasonably kept. But the window setting over \( \pm \sigma_4 \), its shape was degraded with tail events. The "best" spectrum is obtained in near \( \pm \sigma_3 \) window setting. In this case, the peak efficiency is estimated reasonably using these results.

The peak efficiency was 61\% at 215MeV proton in \( \pm \sigma_3 \) window setting, and energy resolution was about 6\%. Figure 7 shows the \( \Delta E-E \) plot of elastically scattered protons at 215MeV in the same window setting. It can be known that tail events in Fig.4 are completely suppressed.

Figure 8 represents peak efficiencies obtained in last two experiments. Points shows experimental results, and a line shows a result of a MonteCarlo simulation. The results of MonteCarlo simulation are almost same as the experimental results. However in lower energies, the value of peak efficiency in last experiments was rather small by about 10\% compared with a MonteCarlo simulation result. One of origins of this reduction may be the counting loss in the coincidence spectrometer.

5. CONCLUSION

A stacked NaI(Tl) spectrometer for intermediate energy is developed to detect charged particles up to about 400MeV.

Using the particle identification technique, it is easy to suppress tail events which come from nuclear reactions in the detector material. In this results, a beautiful peak spectrum was obtained and the efficiency was reasonably estimated. Therefore, tail events have no effect on final results.

In two detector tests with pp elastic scattering protons at intermediate energies, it was confirmed that the stacked spectrometer was quite useful to register intermediate energy charged particle.
Fig. 5: Particle identification spectrum of protons obtained at energy of 215 MeV.

Fig. 6: Top: Full energy spectrum of protons obtained at energy of 215 MeV.
Middle: "Tail" events were identified using the particle identification technique with ±3σ window setting for the proton peak in particle identification spectrum.
Bottom: "Peak" events with ±3σ window setting. Energy resolution was about 6%.
Fig. 7: $\Delta E$-$E$ plot of elastic scattered protons at energy of 215 MeV using the $\pm\sigma_3$ window.

Fig. 8: The peak efficiencies at 22, 64, 115, 160, 215, 250 MeV. The $\pm\sigma_3$ window setting for the particle identification spectrum is applied. Each error is within 0.7%.
6. ACKNOWLEDGEMENT

We are grateful to the staff of Research Center for Nuclear Physics (RCNP), Osaka University, for support of the experiments at the cyclotron facility. This work was performed at the RCNP under programs E44 and E75.

References

A VERSATILE TWO-DIMENSIONAL BACKGAMMON-DRIFT CHAMBER FOR TRANSMISSION TRAJECTORY COUNTER

Department of Nuclear Engineering, Kyushu University

1. INTRODUCTION

A number of two-dimensional position sensitive gas counters with a backgammon-shaped electrode read-out technique have been constructed in several laboratories\(^\text{1-3}\). However, among these counters no counter to use as transmission type was developed. A new type of two-dimensional position sensitive gas counter with a backgammon-shaped electrode as one dimension read-out has been developed in our laboratory. The counter can achieve two dimensional position information by using a backgammon-shaped electrode read-out for one coordinate and the drift time measurement read-out for another coordinate. This gas counter is designed for using as a transmission trajectory counter which is simple in structure, fabrication and also can be used to investigate the response function of spectrometer for incidence of charged particle in different parts of detector area\(^\text{4}\).

In order to apply this counter to determine the incident angle of charged particles which are emitted from accelerator beam or particle induced reactions, a test experiment is performed with cosmic-rays\(^\text{5}\). For this purpose, a new backgammon-shaped electrode is developed (see fig.2).

In the present paper, the design, the structure and the performance of this counter are described. This counter system is now tested to develop as a neutron detector with a new backgammon-shaped electrode pattern.
2. DESIGN AND CONSTRUCTION OF COUNTER

The structure of the design two-dimensional position sensitive gas counter with a backgammon-shaped electrode pattern is shown in Fig. 1. The gas counter consists of three main parts; a detector body, a drift space body and two covering plates. The detector body holds the drift space body and then both sides of the detector body are covered by the covering plates.

The drift space body consists of 1 anode wire, 6 shaping wires and 18 drift plates including a backgammon-shaped and a ground electrode. The volume of the drift space is $140\times123\times40$ mm$^3$. The anode and shaping wires are of nichrohm (Ni-Cr) of 50 μm in diameter. The anode wire is fixed in the parallel at 15 mm distance from the backgammon-shaped electrode. The six shaping wires are used to form the best shaping of electric field predicted by the calculation of the electric field. The drift plates are made using aluminized mylar sheets with various widths, except for the backgammon-shaped and ground electrodes. The distance between every drift plate is 6 mm.
The signal of the first dimension is obtained from the backgammon-shaped electrode. The backgammon-shaped electrode is a thin conductive layer divided into two regions like a saw-teeth narrow insulating gap. This backgammon-shaped electrode pattern was manufactured by photo-etching on a thin gold layer laminated onto an epoxy glass plate. The final backgammon-shaped electrode pattern is shown in fig. 2. The sensitive area of the electrode is 120x40 mm² and the width of insulating gap is about 0.5 mm.

The signal of the second dimension is obtained from the drift time measurement. To obtain the condition of constant electron drift velocity condition in the drift space, uniform electric field is needed. A uniform electric field is formed by a distributed high voltage with a high voltage divider circuit set in the drift space. The high voltage divider circuit is soldered and placed between the detector and the drift space body. Schematic view of the high voltage divider circuit is shown in fig. 3. To realize the uniformity of the electric
field, the parameters of the electric field shaping are determined from a numerical analysis with a code "The Equipotential Lines Calculation"(6).

![Diagram](image)

**Fig. 3 The high voltage divider circuit**

### 3. EXPERIMENT AND RESULT

Simple tests of the design gas counter as two dimensional positions and transmission trajectory sensing were performed by using β-rays from a $^{90}\text{Sr}$ source. The arrangement of the measurement is shown in fig.4. The gas counter and a plastic scintillation counter are used in this test. The plastic scintillation counter is placed behind the gas counter. The plastic scintillation counter is used to detect the coincidence and start signal in the drift time measurement. The sum of the signals from both sides of the backgammon-shaped electrode is used as the stop signal. To obtain information of two dimensional positions, backgammon-shaped electrode read-out as horizontal position and drift time measurement read-out as vertical positions are performed simultaneously. Coincidence signals between the horizontal and
The vertical positions are recorded by SEM (Status Encode Module) and then these data are taken together using a micro-computer.

The measured linearity curve for a backgammon-shaped electrode and the drift time read-out are shown in fig.5. The linearity of the signal from of the backgammon-shaped electrode (horizontal position) is good over the 70 mm sensitive area of the 120 mm backgammon-shaped electrode. The linearity of the drift time measurement (vertical position) is also good over the 80 mm sensitive area of the 120 mm drift space.

Figure 6 shows that the position resolutions of the counter in the backgammon-shaped electrode read-out measured by x-rays is less than 0.9 mm in full width at half maximum (FWHM).

![Figure 5](image.png)

Fig. 5 The plot of linearity curves of horizontal position (backgammon-shaped electrode read-out) and vertical position (the drift time read-out).

For the test of the response measurement for cosmic-rays of the gas counter two detectors are used the gas counter and a NaI(Tl) scintillation counter. The gas counter is placed upon a NaI(Tl) scintillation counter. The arrangement and the result of the measurement is shown in fig.7.

The argon:methane (70 % Ar + 30 % CH₄) mixture gas is used as the counting gas which flows continuously at atmospheric pressure.
Fig. 6 The position resolution of the gas counter for a backgammon-shaped electrode read-out measured by using x-rays.

Fig. 7 a). The arrangement of cosmic-ray measurement by using the gas counter. b). The response of the gas counter to cosmic-rays where A: Signals from the backgammon left side (L) and B: Signals from the backgammon right side (R).
4. CONCLUSION

A two-dimensional position sensitive gas counter with backgammon-shaped electrode and drift time read-out techniques has been constructed. Tests of the counter by using β-rays and x-rays indicate that this type of the gas counter is useful for two dimensional positions sensing for transmission operation. From the linearity measurement, it is shown that this counter has a sensitive area at least $80\times 80\ \text{mm}^2$ in $120\times 120\ \text{mm}^2$ aperture region for two-dimension.

We are now planning to apply this counter to determine the incident angles of charged particles which are emitted from neutron induced reactions in order to measure the double differential cross section of neutron induced reactions.

REFERENCES

PRESENT STATUS OF A SELF-TRIGGERED LIQUID XENON DRIFT CHAMBER FOR DOUBLE-BETA DECAY EXPERIMENTS

A.G. Prokopets
Department of Accelerator Science,
The Graduate University for Advanced Studies,
1-1 Oho, Tsukuba, Ibaraki, 305 Japan

S.Sasaki, H. Tawara and M. Miyajima
Radiation Safety Control Center,
National Laboratory for High Energy Physics,
1-1 Oho, Tsukuba, Ibaraki, 305 Japan

Yu. G. Zdesenko
Institute for Nuclear Research,
Kiev, Ukraine, 252039

1. Introduction

Nuclear double beta-decay ($2\beta$) is a crucial and sensitive prove for understanding fundamental properties of the weak interaction. Many experimental studies as well as theoretical ones have been made to search for $2\beta$ decays in the last decade [1]. There are two principal modes in the $2\beta$, two-neutrino double beta-decay ($2\nu2\beta$) mode and neutrinoless double beta-decay ($0\nu2\beta$). Recently, there has been found the $2\nu2\beta$ decay for $^{76}$Ge [2], $^{72}$Se [3], $^{100}$Mo [4], $^{116}$Cd [5] and $^{150}$Nd [6], and the experimental lower limits of half-life were set for some other candidates [1]. However, the neutrinoless $2\beta$ decay ($0\nu2\beta$) has not been observed so far and the experimental limits on their half-lives were obtained for them and some other candidates[1].

The experiments on the $2\beta$ decay search require not only to reduce background counts to ultra low level, but also to achieve a good energy resolution in measurements of the summed energy of two emitted $\beta$-rays in order to find a peak corresponding to the $0\nu2\beta$ decays around the end point of $2\nu2\beta$ decays.

The $^{136}$Xe is an unique candidate to search for $2\beta$ decays from the reasons mentioned in the previous papers in detail [7,8], where we also described about designing considerations of a self-
triggered liquid xenon ionization drift chamber for the $0\nu2\beta$ decay experiments.

The apparatus is a single gridded ionization chamber equipped with four photomultipliers (PMT). The collector electrode of the chamber is segmented to determine the two dimensional position of events. The PMT is used to observe scintillation photons from liquid xenon excited by the two emitted $\beta$-rays and trigger the event to measure a drift time of a swarm of electrons ionized by $\beta$-rays. In designing consideration, the two dimensional position was to be determined by a position of a segment, where a swarm of ionized electrons was collected, and a point localized by the charge division method on the resistive segment. Also, the segmented collectors are intended to incorporate an active anti-coincidence method between the segments in order to reduce background events. However, the charge division method seems to have several withdraws from view points that it cannot separate two different points on a resistive segment during a time constant of circuit if two different events appear on the segment at a same time and also largely deteriorate the energy resolution. In order to achieve a good performance on the energy resolution, we have made up a collector with 43 hexagonal segments for the anti-coincidence between the segments.

In this paper we describe the present status of the chamber with the test results of a painted resistive electrode and also those of low temperature performance of the PMTs. We also describe the results of calibration of the chamber obtained by operating it as a gaseous ionization chamber in which alpha-particle sources are incorporated.

2. Liquid Xenon Ionization Drift Chamber

The drift chamber of an effective volume of 1 liter is a single gridded ionization chamber in which the collector electrode is segmented as described below. The system of the drift chamber is described in detail with a gas filling system and a refrigerator [8]. Again, the schematical drawing is shown in Fig. 1. The three electrodes, a cathode, a grid and a collector, are configured in the ionization drift chamber as schematically shown in Fig.2. The distance between the cathode and the grid is 40 mm and one between the grid and the cathode is 5 mm. In the present system, the collector electrode is already replaced to one with 37 hexagonal and 6 irregular shape pads from the reasons mentioned above. In the system, as shown in Fig.3, x- and y-coordinates of an event are localized by a pad with a precision of its size and z-coordinate is determined from a drift time of an electron swarm from a time when scintillation due to two $\beta$-rays is observed. The collector segments, made of 0.1 mm thick copper, were formed on a G10 board of 250 mm in diameter and of 6 mm thick by etching. The arrangement of pads on the collector is shown in Fig.4. A pad is
hexagonal with a side of 14.5 mm and an area of 546.2 mm$^2$, and an irregular shaped pad with an area of 362.8 mm$^2$ located at the outermost places so as to cover the effective area of 179 mm in diameter. The pads are separated by 0.5 mm each other. The grid is also replaced with a new one to avoid distortion of grid wires due to shrinkage of a metal frame at low temperature. The new grid is a ring shaped frame of invar of 6 mm thick and with an outer diameter of 230 mm and an inner diameter of 179 mm. Gold coated tungsten wires of 100 μm in diameter are strung with a spacing of 1 mm on the invar flame. Any distortion of the wires and also the flame was not found at a temperature of liquid nitrogen (-200°C).

The electronic read-out system is schematically shown in Fig.5. Signals from each PMT is fed into a constant fraction discriminator (CFD) throughout each PA (an emitter follower). The output signals from 4 CFDs are fed into a fast four fold logic (AND/OR). Number of photons incident to one PMT was estimated to be 90 to 3200 in a case of 0ν2β events in which an energy deposition due to two emitted β-rays is 2.48 MeV[8]. In the case of lower energy deposition, signals might be possibly missed in one or two PMTs. In order to study various cases of events including background events, a trigger logic such as shown in Fig.5 is used to generate gate pulses and also to record the output pattern of AND/OR circuit. Furthermore, the signal heights from the all PMTs are recorded to increase redundancy of data. Signals from all pads are fed into low noise charge sensitive amplifiers and further amplified with shaping amplifiers (MA). Only outputs from the all MAs during a gate pulse are analyzed with ADC and TDC in a CAMAC crate. The outputs from all the ADCs and the TDCs are recorded on a magneto-optical disk through a micro-computer. Time sequence of signals in this system is schematically shown in Fig. 6. The time constant of amplifiers and the time resolution of the trigger logic circuit should be optimized from measurements, so details are not described here.

3. Test Results of Resistive Electrodes

There was proposed a collector electrode with resistive strips in order to collect a swarm of electrons ionized by two emitted β-rays from a 2β decay and to determine the summed energy of two β-rays and the two dimensional position of the event. The 17 resistive strips were realized to be 10 mm wide and 180 mm long by painting carbon paste on a G10 board with 1 mm spacing. The resistivity of each strip was about 30 kΩ and roughly uniform within 10% of the total resistivity. However, the capacitance of a strip turned out to be very large (220 pF). Noise widths were measured with two charge sensitive amplifiers connected to both ends of a strip by injecting charges ($1.9 \times 10^5$ electrons) with a mercury test pulser through a charge terminator (capacitor)
into a various place on the strip. Those were typically about $1.5 \times 10^3$ electrons in FWHM. From this test results and consideration mentioned above, we replaced to a segmented collector described above. The capacitance of a hexagon pad was about 4.5 pF and was negligible in comparison with a capacitance of a coaxial cable to connect a pad to a PA which is fixed outside the chamber vessel.

4. Low Temperature Performance of Photomultipliers

In this experiments, the PMTs will be operated under the condition they are immersed into liquid xenon to observe scintillation photons. A photomultiplier, R-208-01 (Hamamatsu), has been successfully used to measure the $W_s$-value of liquid xenon previously [9]. However, the characteristics of PMTs are normally different from a PMT to a PMT. We have selected four from ten R-208-01 PMTs for this system by measuring their behaviors during changing temperature in a cryostat. We observed light fed through a glass optical fiber of 0.3 mm in diameter and 1 m long from a blue LED (480 nm with a width of 70 nm in FWHM) which is put outside the cryostat. Furthermore, we also observed scintillation light from a CdWO₄ scintillator (15 mm $\phi$ x 18 mm) attached without optical glue to the PMT entrance window in order to get relative intensities of the LED to scintillation from the CdWO₄ scintillator due to gamma-rays of $^{22}$Na at various temperature, and also to compare those to scintillation from NaI(Tl) scintillator due to gamma-rays at a room temperature. Temperatures of the entrance window and the bottom of a PMT were monitored by two thermocouples. The PMT was cooled down step by step with liquid nitrogen vapor to liquid nitrogen temperature. A typical variation of pulse heights obtained during cooling down with the LED light source is shown with a variation during heating up as a function of temperature in Fig.7. The pulse heights gradually increase as the temperature decreases and abruptly diminish at below -170°C. The temperature rise at -150°C seems due to a delay of the temperature rise of the photocathode. Furthermore, there exists a large discrepancy in the pulse heights between processes at cooling down and at heating up. The almost same discrepancies are observed with the all PMTs. The exact reason is not presently known. However, it might be due to any distortion of electrode structure in the glass envelope by a rapid heat cycle of a few hours. This phenomenon suggests incorporation of a calibration system into liquid xenon.

5. Calibration of Electronic System of the Drift Chamber
In order to calibrate the whole system without the PMT system, the drift chamber was operated as a gaseous ionization chamber with a stainless steel cathode plate and a collector plate (not segmented). Eight spots of $^{241}$Am alpha source of 8.5 mm in diameter were electrochemically deposited on the cathode plate.

Firstly, the performance of grid was studied by filling the chamber with a gas mixture of Ar + CH$_4$(10%) at 1 atm. The pulse height distributions were measured as a function of a ratio of the field strength between the collector and the grid to one between the grid and the cathode, keeping the field strength between the collector and the grid 200 V/cm as shown in (a) of Fig.10. The pulse height clearly saturates above the calculated critical field ratio of 1.92 [8]. Also measurements of pulse height distribution were made as a function of the field strength between the grid and the cathode, keeping the field ratio 3.0. The pulse height also clearly saturates above 80 V/cm. The results show good performance of the grid.

Nextly, the collector was replaced to a segmented one. The chamber was filled with a gas mixture of Ar+CH$_4$(10%) at a pressure of 1 atm and 2 atm in order to attempt not only determining energy resolution of alpha particles, but also searching the spots of alpha-particle sources. The range of 5.5 MeV alpha-particles from $^{241}$Am is about 4 cm in the gas mixture of 1 atm and about 2 cm at 2 atm, respectively. Then, ionized electrons along the tracks of alpha-particles are sensed by one to five pads. The emission of alpha-particles from the sources is isotropic on the cathode and those pulse height distribution observed at the cathode is uniform depending on $\cos \theta$, where $\theta$ is an emission angle of alpha-particles measured from the normal to the cathode plain. The charge sensitive amplifiers were connected to all pads to measure signals due to collected charges and a charge sensitive amplifier system was connected with the cathode to observe signals to generate gate pulses through a single channel discriminator as shown in Fig.8. The amplifier gains were independently adjusted with a well calibrated mercury pulser through a charge terminator (capacitor). The noise widths of the amplifiers were distributed from 950 to 1100 electrons in FWHM. Every pedestal of all channels was also obtained for the conversion of pulse heights to charges (in the number of electrons). The pulse height distribution of the cathode pulses is shown in Fig.11. The upper edge corresponds to alpha-particles emitted parallel to the cathode plain and the lower edge to right angle to the cathode plain. The slight non-uniformity depends on the shaping time constants of the amplifier (both 5µsec in differential and integral time constants) and a small peak at the low pulse height side arises from edge effects which occur with the alpha-particles stopped at a guard ring of cathode and escaped from the effective volume of the ionization chamber. In order to generate trigger pulses for all ADCs which analyze signals from all pads, the cathode pulses due to alpha particles emitted with $\theta = 80^\circ - 85^\circ$ and $\theta = 0^\circ - 10^\circ$ are selected with the SCA to mainly analyze “multi-pads” signals and also “one-pad” signals,
respectively. Alpha-particle spectra as "multi-pads" events are constructed from summing up a pulse height of every pad by off-line data analyze and are shown in (a), (b) and (c) of Fig.12 in the case of pressure of 1 and 2 atm and "one-pad" events at 2 atm in (d). The energy resolution becomes better at smaller emission angles in the case of "multi-pads" events. However, it is still worse than the case of "one-pad" events. The most probable pulse heights seems to well agree at the spectra of "multi-pads" events, but those seem to be slightly different from one of "one-pad" events, even if the statistics of "one-pad" events is poor.

Lastly, we tried to determine the position of the alpha-particle source by simply looking for larger signals from one pad. The numbers of counts per pad are plotted against the pad number as shown in (a) and (b) in Fig.13. The (a) was obtained from the events of which the energy deposition per pad is larger than 640 keV and the (b) from the events with the energy deposition larger than 3.2 MeV. The positions of 7 spots are well identified from 8 spots which we made on the cathode. The last spot was missed because it lies just outside the effective volume of the chamber.

6. Discussion

The self-triggered liquid xenon ionization drift chamber is developing to search for 0v2β decay of 136Xe. We are intending to get the resolution of about 3% or better in the energy measurements of energetic electrons in liquid xenon and further to localize the position of energy deposition due to radiation within several cubic mm in order to study the background suppression efficiency. The drift chamber will be operated as 43 independent detectors in order to incorporate coincidence and anti-coincidence technique for background suppression. The electron swarms drift with a velocity of 3 mm/μsec under an electric field above 3 kV/cm in liquid xenon. The rise time of signals from the pads is 1.7 to about 2.7 μsec and the shaping time constants of the amplifiers should be long enough to obtain good energy resolution. Presently those are selected to be 10 μsec. On the other hand, the signals, which are amplified with those time constants, has about 30 μsec rise time and request about 50 μsec gate signal to be analyzed at an ADC. However, the drift time measurements serve to eliminate signals, which have a drift time of more than 16 μsec, as backgrounds, since the maximum drift time of the swarms is about 13.3 μsec in the region between the cathode and the grid. The anti-coincidence measurements from a pad to a pad serve to eliminate background signals which have a drift time shorter than that time.

The drift chamber was calibrated with a gas mixture as described above. However, we should again calibrate the whole system with liquid xenon without any radiation sources insides in
order to obtain a charge collection efficiency. This may be possible from reconstruction of events irradiated outside with several gamma-ray sources with known energies, because the fine adjustment of the amplifier gains can be made with a high precision test pulser.

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Fig. 1. Cross-sectional view of the LXe drift chamber.

Fig. 2. Configuration of electrodes and PMT in LXe Drift chamber.
Fig. 3. The events localization in the LXe drift chamber.

Fig. 4. The segmented collector with 43 conductive pads.
Fig. 5. Block-diagram of the electronics for LXe drift chamber. (PA) - preamplifiers; (MA) - main amplifiers; (CFD) - constant fraction discriminators; (D) - delay.

Fig. 6. Time sequence of the signals in LXe measurements.
Fig. 7. Temperature dependence of output signal for PMT R208-01 (LED as the light source).

Fig. 8. Block-diagram of the electronic circuits for measurements with Ar-CH₄ gas mixture.
Fig. 9. Time sequence of the signals in measurements with Ar-CH₄ (10%) gas mixture.

Fig. 10 (a) saturation curve for α-particles from $^{241}$Am as function of $Z$; (b) saturation characteristics of the pulse height distribution as a function of the field strength between the grid and the cathode at the fixed $Z$. 
Fig. 11. Pulse height distribution of the cathode pulses.
Fig. 12. Energy spectra of 5.5 MeV α-particles from $^{241}$Am obtained by summation of signals from pads.
Fig. 13. Reconstruction of the location of the sources.
RESISTIVITY DEPENDENCE OF PLASMA DELAY IN A SILICON SURFACE BARRIER DETECTOR

Ikuo Kanno, Satoshi Kanazawa and Itsuro Kimura
Division of Nuclear Engineering, Kyoto University

1. INTRODUCTION

Silicon surface barrier detector (SSBD) has been employed as a charged particle detector. The SSBD has a lot of excellent properties: compactness, low operating voltage, excellent timing and energy resolutions.

Heavy ion measurements, however, revealed unfavorable characteristics of the SSBD. The output pulse heights deviated from linear relationship to the incident kinetic energies of charged particles.\(^{(1),(2)}\) The timing of pulse output retarded some nano seconds from the charged particle incidence.\(^{(3)-(6)}\)

The timing retardation is called the plasma delay. The plasma delay depends on the energy, proton number and mass number of the incident charged particle, as well as the bias voltage applied to the SSBD. Another important parameter of the plasma delay is the resistivity of the silicon substrate in the SSBD.

The physics of the plasma delay was reported by the one of the present authors as a simple model.\(^{(6)}\) The calculated results with the model of the plasma delay clearly reproduced the experimental data of Bohne et al.\(^{(6)}\) In that model, the resistivity dependence of the plasma delay, however, was not included. No theoretical works on the resistivity dependence of the plasma delay were reported.

In the large accelerator facility such as large hadron collider (LHC), the SSBDs are planned to be employed as track detectors for high energy charged particles. In the experimental area of the LHC, there will be high energy parasitic neutrons accompanying the high energy charged particles. The SSBDs will be exposed by the high energy parasitic neutrons. The SSBDs are expected to be intact for more than 10 years, with the neutron fluence of \(10^{13}-10^{14}\) n/cm\(^2\). The neutron irradiation introduces impurities in the silicon substrate and results in the change of the resistivity. Wunstorf et al. reported an n-type silicon increased its resistivity and finally changed to be a p-type silicon with the neutron irradiation.\(^{(7)}\)

The resistivity dependence of the plasma delay is one of the required theme in both the detector physics and the neutron irradiation effect of silicon.
2. MODELS

2.1 Plasma Delay

The plasma delay is the time retardation between the charged particle incidence and the pulse output.

The plasma delay was found by Quaranta et al., for the first time. In those days, the plasma delay was an unknown phenomenon and was discussed whether it had physical meanings or it was caused by electronic circuits. Neidel et al. reported that the plasma delay was proportional to the inverse electric field strength when an applied bias voltage was relatively high and was constant when the bias voltage was lower. The resistivity dependence of the plasma delay was not observed by Neidel et al. Theoretical works were reported on the linear response of the plasma delay to the inverse electric field strength, based on various models. Neidel et al. reported that the plasma delay was proportional to the inverse electric field strength when an applied bias voltage was relatively high and was constant when the bias voltage was lower. The resistivity dependence of the plasma delay was not observed by Neidel et al. Theoretical works were reported on the linear response of the plasma delay to the inverse electric field strength, based on various models. Neidel et al. reported that the plasma delay was proportional to the inverse electric field strength when an applied bias voltage was relatively high and was constant when the bias voltage was lower. The resistivity dependence of the plasma delay was not observed by Neidel et al. Theoretical works were reported on the linear response of the plasma delay to the inverse electric field strength, based on various models.

In 1985, Bohne et al. carried out the experiments of the plasma delay with high energy charged particles, such as $^{40}\text{Ar}$ with 286 MeV and 476 MeV in energy. They reported that the plasma delay had a maximum value as a function of inverse electric field strength. The theoretical works on the linear dependence of the plasma delay on the inverse electric field strength lost their standing points.

The resistivity dependence of the plasma delay was also reported by Bohne et al.: the plasma delay had greater value in higher resistivity SSBD with the same electric field strength. This experimental results predict that the timing of a SSBD output will be delayed when the resistivity of the SSBD increases due to the neutron irradiation.

2.2 Plasma Column and Electric Field Strength

The plasma delay is the outcome of two competing electrical forces which exert on a plasma column, which is created by an incident charged particle.

The plasma column, a region of dense electron-hole pairs, is inserted in the electric field in the depletion layer of the SSBD. An example of the plasma column is shown in Fig. 1. The plasma column behaves as a conductor at the time of the plasma column creation.

The electric field strength inside the depletion layer is shown in Fig. 2(a).
The maximum electric field strength is given\(^{(17)}\),

\[ F_{\text{max}} = \frac{2V}{d} = \sqrt{\frac{2V}{\mu_e \tau}}, \]

where, \( V \) is the applied bias voltage, \( d \) the depletion layer thickness, \( \mu_e \) the electron mobility, \( \tau \) the charge collection time and is proportional to the silicon resistivity. The electric field in the depletion layer can not penetrate the plasma column, which behaves as a conductor.

Due to the density difference of electrons and holes inside and outside the plasma column, the electrons and holes diffuse radially and expand the radius of the plasma column. The radius of the plasma column, however, can not expand freely, under the influence of the electric field strength.

In a high electric field strength, the plasma column radius hardly expands because of the strong compressing force. The electron-hole density in the plasma column remains high and the electric field hardly penetrates the plasma column. On the other hand, the electrons and holes are strongly attracted towards the positive and negative electrodes. With these competing forces, the plasma delay shows complicated behavior to have a peak value as a function of inverse electric field strength.

When the electric field strength is low, the attracting force is weak. The radius compressing force is also weak, then the radius expands easily and the electron-hole density in the plasma column decreases in a shorter time. Although the electric field strength is weak, the time the plasma column begins to erode becomes shorter.

In the former model described above, the effective electric field strength \( F_{\text{eff}} \) (the electric field strength at the 2/3 of the plasma column length) was employed as a representative value.

2.3 Resistivity Dependence of Electric Field Strength\(^{(11)}\)

With the conductor plasma column, the electric field strength outside the plasma column increases and the one inside the plasma column vanishes.

The plasma column has dielectric property when it begins to erode. The electric field strength inside the plasma column \( F_{\text{in}} \) is depressed by the ratio of dielectric constants of silicon \((\varepsilon)\) and the plasma column \((\varepsilon')\).

\[ \gamma(t) = \frac{\varepsilon}{\varepsilon'(t)}, \quad (1) \]

\[ F_{\text{in}}(x) = \gamma(t)F_{\text{out}}(x). \quad (2) \]
Here, $F_{\text{out}}(x)$ is the electric field strength outside the plasma column and $\gamma(t)$ is a function of time and has value 0 at $t = 0$ and finally becomes 1. On the other hand, $F_{\text{out}}(x)$ is enhanced so that the electric potential in the depletion layer is kept constant.

$$F_{\text{out}}(x) = \frac{2V(d - x)}{(d - l)^2 + \gamma(t)l(2d - l)},$$

(3)

Here, $l$ is the plasma column length.

The electric field strength with the conductor plasma column in the depletion layer is obtained with the ratio $\gamma(t)$ as zero,

$$F_{\text{out}}(x) = \frac{2V(d - x)}{(d - l)^2}.$$  

(4)

The electric field strengths in the depletion layer with a conductor and a dielectric plasma column are shown in Fig. 2(b) and (c).

In Fig. 3, the electric field strengths are plotted for $362\,\Omega\text{cm}$ and $2100\,\Omega\text{cm}$ SSBDs with the same $F_{\text{eff}}$ for the plasma column length of $30\,\mu\text{m}$ and the ratio of dielectric constants, $1/10$. The electric field strength facing to the plasma column top in the lower resistivity SSBD is greater than the one in the higher resistivity SSBD. The electric field strength in the side of the plasma column has no influence with the presence of the plasma column and remains unchanged. In short, the carrier attracting force is greater in the lower resistivity SSBD, whereas the radius expansion restricting force is the same value for both SSBDs.

We conclude here that we should employ the enhanced electric field strength $F_{\text{out}}$ for the estimation of the attractive force, and $F_{\text{eff}}$ for the compressing force.

3. EXPERIMENTALS

The plasma delay of $^{16}\text{O}$ ion with $61\text{MeV}$ in energy was measured using the Tandem Accelerator Facility of Japan Atomic Energy Research Institute. Oxygen ions were accelerated and scattered by a gold target foil of $100\mu\text{g/cm}^2$ in thickness. The scattered oxygen ions were extracted into the flight path of $105\text{cm}$ in length with the angle of $45$ degrees. In the flight path, a micro-channel-plate detector (MCPD) was placed. The SSBDs with resistivities of $362\Omega\text{cm}$ and $2100\Omega\text{cm}$ were set at the end of the flight path. The time-of-flight measurement was carried out between the MCPD and the SSBD, with changing the applied bias voltages of the SSBDs. The schematic drawing of the experimental set-up and the electronic circuits are shown in Fig. 4.
The plasma delay was obtained following the method of Bohne et al.\(^6\) The measured and calculated plasma delays are shown in Fig. 5. The calculated values are normalized to the experimental result of 0.5cm/kV with 362Ωcm SSBD.

4. CONCLUSIONS

The electric field strength in the depletion layer changes with the electric property of a plasma column. Two competing electric forces result in the plasma delay. Employing the enhanced electric field strength due to the presence of the dielectric plasma column, the resistivity dependence of the plasma delay was clearly explained. The model calculations reproduced the experimental results of the plasma delay of \(^{16}\)O ions fairly well.

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Fig. 1. Examples of calculated plasma columns created by (a) α particle with 8.78MeV, and (b) 40Ar with 476MeV.
Fig. 2. Electric field strengths in the depletion layer of a SSBD with (a) no, (b) conductor and (c) dielectric plasma column.
Fig. 3. Electric field strengths are drawn by dashed lines for 362 and 2100Ωcm SS-BDs with the same $F_{eff}$. Solid lines show the electric field strength with a dielectric plasma column 30μm in length and $\epsilon/\epsilon' = 1/10$. 
Fig. 4. Schematic drawing of experimental set-up and electronic circuits.

Fig. 5. Measured and calculated plasma delay for $^{16}$O ion with 61MeV in energy. The calculated results were normalized to the experimental data of 362Ωcm SSBD at 0.5cm/kV.
Development of Multichannel CdTe Radiation Detectors

Kenji SATO, Toshiyuki SATO, Masaaki UKITA, Satoshi TOKUDA, Takaaki INOUE, Susumu ADACHI, Takuro IZUO, Naoyuki HORIZ, Kouichi TANABE, Yuriko KONISHI, Shinichiro ISHIDA, and Motosada KIRI

Central Research Laboratory, Shimadzu Corporation, Kyoto, Japan

1. INTRODUCTION

One of the applications using radiation detectors is medical diagnostic imaging. In particular, X-ray radiography is the most general and aged. Nevertheless, detectors used there have been conventional X-ray films up to the present, in contrast to solid state detectors used for Computed Tomography (CT) or Positron Emission Tomography (PET).

Multichannel CdTe radiation detectors have been developed for a new X-ray radiography system operating in the photon counting method, which was named "Quantum Radiography". The Quantum Radiography system has superior performance, and its diagnostic efficiency has been recognized.

This paper is intended as an introduction of the multichannel CdTe radiation detector and the Quantum Radiography system. In addition to this, actual examples of clinical experiments are lastly described.

2. CADMIUM TELLURIDE

First of all, we describe the reason why cadmium telluride (CdTe) was marked out for our multichannel radiation detectors' material.

The semi-insulating CdTe compound semiconductor is a favorable material for use as radiation detectors. The bandgap of 1.5 eV is wide enough to operate at room temperature as compared to that of other semiconductors such as Ge (Eg=0.7eV) or Si (Eg=1.1eV). The high resistivity ($\rho=10^8 \Omega\text{cm}$) gives low dark current. The large atomic numbers of Cd and Te (48 and 52, respectively) offer high detecting efficiency for $\gamma$-rays and X-rays. The charge carrier mobilities of about 1000 cm$^2$/Vs for electrons and about 90cm$^2$/Vs for holes are high enough to operate the detectors at a high counting rate above 1 Mcps.

In addition to these physical properties, many small electrodes can be easily fabricated on surfaces of the CdTe substrate with photolithography. This is an important characteristic for multichannel radiation detectors having a high spatial resolution.
3. THE MULTICHANNEL CdTe RADIATION DETECTOR

3.1. Construction

The multichannel CdTe radiation detector consists of a CdTe substrate having a common-bias electrode on its one surface and a plurality of signal readout electrodes on its other surface, as shown in Fig.1. These each of signal readout electrodes are connected with their respective signal processing circuits composed of four basic circuits, that are a preamplifier, a shaping amplifier, a comparator and a binary counter, as shown in Fig.2.

We now consider the case where an X-ray interacts in a certain channel. To begin with, an X-ray induced charge arises at a signal readout electrode on the channel. It is fed to the preamplifier as a charge pulse. Next, the charge pulse is converted into a voltage pulse with the preamplifier. The voltage pulse is amplified and shaped with the shaping amplifier. And then, the amplified and shaped voltage pulse is discriminated from noise pulses with the comparator if the threshold voltage level (Vth) is set higher than the maximum noise level. Then, a digital pulse is fed to the binary counter. The digital pulse is counted with the counter. Finally, the counted value is fed to an external memory at a certain time interval.

In this way, each channel of the multichannel radiation detector simultaneously operates and individually counts up the number of incident X-ray photons in each channel.

3.2. Detector module

In accordance with above construction, we made 256-channel detector modules whose CdTe detector elements and signal processing circuits were mounted on a ceramic board. A schematic view and a photograph of the module, of which an actual size is 32 mm in width and 55 mm in length, are shown in Fig.3.

In the front row, four CdTe detector elements are arranged. The detector element has 64 square pixels, which are 16 pixels in the direction of the width and 4 pixels in the direction of the length at intervals of 0.5 mm (see closeup below right). In the second and third row, eight 32-channel amplifier ICs and eight 32-channel comparator ICs are respectively arranged at short intervals between each row. In the back side, four 64-channel binary counter ICs are arranged. The counted values of these counters are fed to external memories through the printed circuits on the back end.

These parts except counters are provided with solder bumps on the respective electrodes. By means of surface mount technology using these solder bumps, the compact arrangement was achieved.

3.3. Performance

A wide dynamic range and linear sensitivity are required for the fine quality imaging such as the medical diagnostic imaging. The CdTe radiation detectors operating in the photon counting method have performance to satisfy these requirements.

The typical example is shown in Fig.4. This figure indicates relation between a counting rate of the CdTe radiation detector and an X-ray dose rate. It clearly appears that linearity between both is kept in a wide range above 1 Mcps. Here, this experiment
was carried out as follows: the CdTe detector and an X-ray tube were placed 200 cm apart; the X-ray tube was supplied with a tube voltage of 100 kV and a tube current of 91 mA; the sampling data was measured at intervals of 1.178 msec, then it was normalized to counts per second (cps); the X-ray dose rate was indicated with the thickness of absorber that consists of acrylate, which was placed between the detector and the X-ray tube, so that the scale of the horizontal axis is centimeter (cm).

Next, a comparison of the CdTe detector with an X-ray film was made. The data of the CdTe detector was converted into the linear scale in the range of film density to compare in the same scale. As shown in Fig.5, image density of the X-ray film is slowly increasing at the regions of low dose rate, while, it of the CdTe detector is linearly increasing. This is due to the noiseless operation in the photon counting method.

From this result, the CdTe detector is superior to the X-ray film in regard to the density resolution at the regions of low dose such as the overlapped areas with internal organs, e.g., the parts of the lung overlapped with the heart or diaphragm.

4. THE QUANTUM RADIOGRAPHY SYSTEM

A new X-ray radiography (Quantum Radiography: QR) system was experimentally produced. For this system, the multichannel CdTe radiation detector composed of twelve detector modules was used. A schematic view of the detector is shown in Fig.6. The size of the detective area is 384 mm in length and 2 mm in width, and the number of the channels is 3072.

A schematic diagram of the QR system is shown in Fig.7. Two-dimensional images are obtained by means of vertically scanning operation(7). During this operation, an X-ray focus, slits and the detector have been kept in a linear position.

Furthermore, the CdTe detector is mounted at an inclined angle of about 14° from the horizontal position, consequently, the spatial resolution is effectively improved about four times as shown in Fig.8.

Next, the most important feature inhering in the QR system is shown in Fig.9. In the conventional film-screen system, many scattered X-rays come in the X-ray film as shown in Fig.9 (b). These scattered X-rays make radiographs foggy. On the other hand, in the QR system, since the scattered X-rays are shut by the slits placed before and behind the subject as shown in Fig.9 (a), they hardly come in the CdTe detector, so that radiographs using the QR system are clear.

5. CLINICAL EXPERIMENTS

Clinical experiments using the QR system were tried. The two typical examples are shown in Fig.10 and Fig.11.

A chest radiograph shown in Fig.10 is a case of diffuse panbronchiolitis. Many granular tubercles lying scattered in the lung are clearly represented as compared to conventional film-screen systems. And also, bronchial tramlines with the bronchiectasis are clearly represented especially at the overlapped areas with the heart and right diaphragm. A chest radiograph shown in Fig.11 is a case of renal cell cancer
metastasized to the lung. Several nodules overlapped with the heart and right diaphragm are clearly represented too.

As the above results, the QR system can find out some of diseases that are invisible in the conventional systems. Thus the QR system has been recognized as effective in the clinical diagnosis(8)(9).

6. SUMMARY

Multichannel radiation detectors using CdTe compound semiconductors were developed. The detectors have the following features: room temperature operation, high detecting efficiency, the high counting rate above 1 Mcps and the high spatial resolution below 0.5 mm.

Furthermore, a new X-ray radiography (Quantum Radiography) system was experimentally produced. For this system, the multichannel CdTe radiation detector composed of twelve detector modules was used. The size of the detective area is 384 mm in length and 2 mm in width, and the number of the channels is 3072.

From the clinical experiments, it was found that the high density resolution and excluding effect of scattered X-rays make out the fine quality images, especially at the overlapped areas with internal organs.

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Fig. 1 Construction of the multichannel CdTe radiation detector

Fig. 2 Brock diagram of the signal processing circuit
Fig. 3 Schematic view and photograph of the 256-channel detector module
Fig. 4 Counting rate of the CdTe radiation detector

Fig. 5 Comparison of the CdTe detector with an X-ray film
number of the modules: 12
number of the channels: $768 \times 4 = 3072$

Fig. 6 Schematic view of the multichannel CdTe detector used for the QR system

Fig. 7 Schematic diagram of the Quantum Radiography system
Fig. 8 Improvement of the spatial resolution

(a) QR system

(b) conventional film-screen system

Fig. 9 Difference in excluding effect of scattered X-rays
Fig. 10 Chest radiograph using the QR system

Focal length = 200cm, Tube voltage = 100kV, Tube current = 64mA,
Sampling time = 1.18ms/pixel
Fig. 11 Chest radiograph using the QR system

Focal length = 200cm, Tube voltage = 100kV, Tube current = 64mA,
Sampling time = 1.18ms/pixel
A germanium gamma-ray detector is indispensable to gamma-ray spectroscopy applications. However, it takes much time for keeping liquid nitrogen in a cryostat of the detector. Recently, a maintenance-free germanium detector system has been required from various fields. Therefore, high purity germanium spectrometers cooled by a closed-cycle cryogenic refrigerator were developed by several germanium detector makers. However, the refrigerator system is usually large and heavy. A noise reduction system in the cryostat is required to reduce mechanical vibration produced by the refrigerator. Consequently, it is very difficult to miniaturize the detector system using this refrigerator.

For resolving above-mentioned problems, we developed a small electric-cooled germanium gamma-ray detector using a Stirling refrigerator for cooling a detector element. However, a large germanium detector element having the relative detection efficiency more than 10% cannot be used because the cooling faculty of the Stirling refrigerator is low. Therefore, we have developed a general-purpose electric-cooled germanium detector having the relative detection efficiency more than 10%.

Schematic construction of the developed electric-cooled germanium detector is illustrated in Fig. 1. Two Stirling refrigerators are used to cool down a germanium detector element to liquid nitrogen temperature (77K) instead of a dewar containing liquid nitrogen. A Model SRS-2110 made by Sumitomo Heavy Industry Co. was used as the Stirling refrigerator. The cooling faculty of this refrigerator is 1.5 W at 80K when it works in the operation condition of AC 16V and 3.5A. The refrigerator consists of a compressor, a connection pipe, a cooling system with a disperser and an AC power supply. He gas is compressed in the compressor as synchronizing with 50Hz of
the AC power supply. Two cold heads of the cooling system cooled by the displacer are connected by an inverse-T shape fitting. A germanium detector element is attached at the center of the fitting.

A $85\text{cm}^3 (46.7\text{mm}\times 47\text{mm})$ closed-end high purity germanium detector is used as the germanium detector element. This detector has a relative detection efficiency of 14%. Moreover, the first stage circuit of a preamplifier (ORTEC-120) is installed near the germanium detector element to reduce the microphonic noise which is produced by the vibration of the Stirling refrigerators.

The developed two-refrigerator-type germanium gamma-ray detector is shown in Photo 1. A size of the detector is $60\text{cm} \times 15\text{cm} \times 15\text{cm}$. 

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Photo. 1 General-purpose electric-cooled germanium detector using two Stirling refrigerators
3. PERFORMANCE OF THE DETECTOR

The measurement of cooling characteristics was carried out by operating two Stirling refrigerators with AC 16V of the rated voltage. The result is shown in Fig. 2. The lowest cooling temperature is 77K. The operation time of 6 hrs is necessary to cool down the temperature.

After cooling down, AC voltage was reduce to 12V and the temperature was kept at 89K. The performance of the detector was measured by a $^{60}$Co gamma-ray source. The typical gamma-ray spectrum for the $^{60}$Co source is shown in Fig. 3. The dependency of FWHM energy resolution on shaping time of a spectroscopy amplifier (CANBERRA-2021) was measured. The shaping time is changed from 0.5μsec to 4 μsec. The results are shown in Fig. 4. The best FWHM energy resolution is obtained when the shaping time is 2μsec. The energy resolution for 1.33MeV gamma-rays is 2.06keV and that for the pulser is 1.34keV. On the other hand, the energy resolution for 1.33MeV gamma-rays is 2.08keV and that for the pulser is 1.20keV if the AC power supply of the refrigerators is stopped for a short time. From these results, it is revealed that the developed germanium detector has the FWHM energy resolution as same as that of a conventional germanium detector using liquid nitrogen.
4. ANALYSIS OF ENERGY RESOLUTION

In case of using two-refrigerators-type germanium detector, the microphonic noise produced by two refrigerators was much lower than that produced by one refrigerator. Therefore, the FWHM energy resolution is very good.

The variation of weight(size) of the germanium detector element can be considered as one of the reasons. The dependency of FWHM energy resolution on shaping time of a spectroscopy amplifier was measured by using a 14cm³ germanium detector instead of the 85cm³ germanium detector. The results of comparison with 14cm³ and 85cm³ germanium detector are shown in Fig. 5. Both dependencies of the energy resolution are almost same. The energy resolution for 1.33MeV gamma-rays is 2.17keV and that for the pulser is 1.44keV. On the other hand, the energy resolution for 1.33MeV gamma-rays is 2.58 keV and that for the pulser is 1.88keV in case that this 14cm³ germanium detector was attached at one Stirling refrigerator. It is revealed that the weight of the germanium detector element is not related to the improvement of the energy resolution.

Next, the difference of phase of operational AC voltages between two Stirling refrigerators can be considered as one of the reasons. Measurements of the energy resolution carried out in condition of the same phase and the inverse phase between two Stirling refrigerators. However, both results are same. It is revealed that the phase difference of operational AC voltages is not related to the improvement of energy resolution.

From above experimental results, it is estimated that the reason is the use of two Stirling refrigerators, itself. Therefore, the analysis of the mechanical vibrations produced by the Stirling refrigerators was carried out to inquire the improvement reason of the FWHM energy resolution. Signals of mechanical vibrations were measured by using an accelerator sensor. Fitting positions of the accelerator sensor is illustrated in Fig. 6. The sensor was attached at three positions, e.g., front, side and top of a germanium detector element and the frequency spectra of output signals...
were measured by a spectrum analyzer. The difference of the frequency spectra between a one-refrigerator-type germanium detector and a two-refrigerators-type germanium detector was obtained. The 14cm³ germanium detector was used for these experiments. Typical comparison results at the top position are shown in Fig.7. Two spectra are distributed in the range from 1kHz to 15kHz. The spectrum for the two-refrigerators-type germanium detector is lower of 10dB than that for the one-refrigerator-type germanium detector. Moreover, comparison results for other two position were almost same.

Therefore, it is confirmed that the use of two refrigerators contributes to the improvement of energy resolution for the two-refrigerators-type germanium detector.

5. CONCLUSION

A general-purpose electric-cooled germanium detector having the relative detection efficiency of 14% was developed. The energy resolution of this detector is just as same as that of a commercial germanium detector cooled by liquid nitrogen. Since two Stirling refrigerators were used for cooling down the detector element, the detector is small, light and portable. It is concluded that this germanium detector can be applied to various gamma-ray spectroscopy.

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FABRICATION OF RADIATION DETECTOR USING COMPOUND-SEMICONDUCTOR (PbI$_2$, TlBr)

T.Shoji, K.Sakamoto, K.Ohba, T.Suehiro and Y.Hiratate

1. INTRODUCTION

Lead iodide (PbI$_2$) crystal is a promising material for use as a nuclear radiation detector which can be used over a temperature range between room temperature and about 100°C. It is also possible to fabricate X-ray and low energy γ-ray radiation detectors of small size, because the material has high atomic number (Pb: 82, I: 53) and wide band gap (2.55 eV). The PbI$_2$ crystal is an anisotropic semiconductor which is crystallized in the CdI$_2$ type of layer structure. The development concerning PbI$_2$ radiation detectors has been discussed so far by Roth(1), Manfredotti(2), and Lund(3). Most recently, Lund(4) and Shoji(5) discussed the PbI$_2$ crystal growth method and its characteristics. We have reported(6) performances of PbI$_2$ radiation detectors fabricated from crystals grown by multi-Bridgman method. In this case, the average energy for the production of electron–hole pair of the PbI$_2$ detector could be estimated to be about 30 eV, which is larger than the value expected from the formula proposed by Klein(7). It is considered that some portions of the carriers produced by γ-ray irradiation in the detector are captured by various trap levels or lattice defects in the PbI$_2$ crystal. Consequently, we could not observe the photopeak of γ-rays. After considering these results, we studied the growth method for PbI$_2$ single crystals with higher quality.

On the other hand, thallium bromide (TlBr) crystal is also emerging as a promising material for the development of high resolution X-ray and γ-ray detectors at room temperature. The TlBr crystal has a CsCl type cubic crystal structure, it melts congruently at 480°C and it does not exhibit any solid–solid phase transition. The interest in TlBr crystal is due to its high average atomic number (Tl: 81, Br: 35), high density (7.5 g/cm$^3$) and wide bandgap (2.7 eV). The photon stopping power of TlBr crystal is greater than any of the semiconductors discussed. This material was first investigated as a radiation detector material by Rahman(8,9). However, it is thought that fabrication of the TlBr radiation detector is located in its rudimentary stage, and we have to cope with many problems; growth technique of TlBr single crystals and electric contact with the crystal, etc.

In this paper, we will discuss the fabrication of radiation detectors using the PbI$_2$ and TlBr crystals.
2. EXPERIMENTAL PROCEDURES

2-1. Crystal Growth

The PbI₂ powder with nominal purity of 4 nine is used for the single crystal growth. In this experiment, the crystals are grown by four different methods as follow; 1) the ordinary Bridgman method (B01), 2) only the 60 pass zone refining (zone melting method:Z60), 3) ordinary Bridgman methods after the 3 pass zone refining (B03) and 4) ordinary Bridgman method after the 20 pass zone refining (B20). The growth rate is 20 mm/hr and growth temperature is 450 °C in the Bridgman method. In the zone refining, the system has two narrow heating elements to obtain melting zone of about 3 cm in length. The growth rate and temperature are 10 mm/hr and 450 °C, respectively. The resistivities of all PbI₂ crystals are larger than $10^{10}$ Ωcm.

On the other hand, in the TlBr crystal growth, the TlBr powder with nominal purity of 5 nine is used. The starting material is sealed in a long quartz tube (10 cm) under vacuum of about $10^{-6}$ Torr. The zone refining is carried out only twice because the quartz tube is broken in furnace by reaction between quartz and TlBr. After the 2 pass zone refining, the TlBr crystals are grown by ordinary Bridgman method. The growth rate is 20 mm/hr and growth temperature is 480 °C and then, after cutting off the inferior part of the crystal, the remainder of the crystal is fed into the Bridgman apparatus again. The growth rate and temperature are 10 mm/hr and 480 °C, respectively. The resistivity of the crystal becomes to increase up to about $10^{11}$ Ωcm.

2-2. Fabrication of Detectors

The PbI₂ crystals cleaved into small pieces of 2x2x0.5 mm³ are used for the fabrication of radiation detectors. The surfaces of these pieces are not polished mechanically nor etched chemically. The electrodes are formed on both sides of the pieces by painting them with colloidal graphite (aquadag). The leakage currents of the detector are ranging from $10^{-11}$ to $10^{-12}$ A for the bias voltage 30 V at room temperature.

The fabrication method of the TlBr detector is the same as that for the PbI₂ detector.

2-3. Characterization of Crystals

Characteristics of the crystals are obtained by using the photoluminescence (PL) measurement and response property by the α- and γ-rays.

For PL measurement, a 50 mW He-Cd laser (4416 Å) is used as an excitation source. The PL from the samples is detected by a photomultiplier tube in connection with a 50 cm single grating monochrometer. Output signals from this tube are amplified by a conventional lock-in
amplifier and then fed to an X-Y recorder.

The α-rays with an energy of 5.48 MeV emitted from an 241Am source impinge on the detector and create electron–hole pairs along the trajectory in the crystal. Electric charges collected on the electrodes are supplied to a charge sensitive preamplifier. The output signal is shaped in the main amplifier with a time constant of 2μs. Then signals are analyzed and stored in a 1024 channel pulse height analyzer.

3. RESULTS AND DISCUSSION

In the case of the PbI2 radiation detector, pulse–height spectra for the 5.48 MeV α-particles impinging on the positive electrode are shown for the detector fabricated from the sample grown by the ordinary Bridgman method (1–(a)), from sample grown by ordinary Bridgman method after the 3 pass zone refining (1–(b)) and by ordinary Bridgman method after the 20 pass zone refining (1–(c)). As shown in Fig.1–(a), the α-peak is not observed by the detector fabricated from sample B01 because of insufficient quality of the crystal. On the other hand, in Figs.1–(b) and –(c) the α-peaks with an energy resolution of about 2.8 MeV and 759 keV (FWHM) are clearly observed with an applied detector bias of 35 V and 20 V, respectively. These result show that the detector fabricated from B20 is of higher quality than that from samples B01 and B03. However, the detector fabricated from B20 exhibits no response for γ-rays. When the α-particles impinge on the negative electrode, the peak in the energy spectrum can hardly be observed. This is an experimental evidence that the main contribution to the output signals comes from electron carriers, while hole carriers contribute much less.

The PL characteristics of the PbI2 crystal grown by the ordinary Bridgman method (B01) is shown in fig.2–(a). The result for the crystal grown by the Bridgman method after 3 pass zone refining (B03) is shown in fig.2–(b), while the result obtained for the crystal by the Bridgman method after the 20 pass zone refining (B20) is given in fig.2–(c). These figures depict the intensity of 2.50 eV emission line which is attributed to bound excitons and the intensity of the 2.4 eV emission band which is attributed to donor-acceptor pairs. As shown in fig.2–(a), the intensity of 2.50 eV emission line is much weaker than that of 2.4 eV emission band. On the other hand, for the figs.2–(b) and –(c), the intensity of 2.50 eV line bears comparison with that of the 2.4 eV emission band. From these results, it is suggested that the samples B03 and B20 are of higher quality than B01 sample. From the comparison between fig.2–(b) and fig.2–(c), one can see the emission intensity ratio (2.5eV/2.4eV) of the sample B20 is larger than that of the B03, the sample B20 being of higher quality than B03. The experimental results of the PL measurement agree well with the response property obtained by the α-ray experiment shown in fig.1.

Fig.3 shows the variation of the peak position corresponding to the 5.48 MeV α-rays as a
function of the applied bias voltage on the PbI$_2$ detector fabricated from B20. As shown in this figure, the peak position shifts to higher channels with increasing detector bias. However, the energy resolution becomes worse gradually. The reason for this can be traced back to a speculation that at lower bias electron carriers give main contribution to the output pulses, while at higher bias hole carriers contribute also to the output signals. Hole carriers undergo various capture levels originated from impurities and native defects while traversing the depleted region of the crystal. Although the contribution of hole carriers increases the output pulse heights, this possibly deteriorates the energy resolution.

Fig. 4 shows the variation of the energy resolution (FWHM) and the peak position of the 5.48 MeV peak as a function of the detector bias at room temperature. The FWHM energy resolution of the α-peak becomes markedly degraded at above 30 V. It is suggested that the detector fabricated from the sample B20 suffers from incomplete carrier collection because the property of the crystal grown by this growth method is not sufficient yet for use as a radiation detector.

Therefore, in order to produce better detectors, the number of pass in the zone refining method is increased to 60 (Z60). The detector fabricated from the sample Z60 clearly exhibits response for lower energy γ-rays. Fig. 5 shows the pulse height spectra for the 59.5 keV γ-rays from the $^{241}$Am source obtained at various values of the detector bias. The photopeak originated from the 59.5 keV γ-rays is clearly observed, although it is not well resolved from a large tail at lower channels which is caused probably by electric noises and Compton scattering. Observation of the γ-ray photopeak seems to be a good evidence that the sample Z60 is superior in quality than B20.

Fig. 6 shows the PL characteristics of Z60. The emission intensity ratio (2.48 eV/2.4 eV band) of the sample Z60 is about 1. However, as shown in this figure, the bound exciton is split into two peaks (2.492 and 2.495 eV). It is well known that the construction of PbI$_2$ crystal gives rise to a phase transition from the 2H to the 4H form. The peak splitting can be attributed to this change of crystal construction. As a result, clear evaluation of the crystal quality from the PL measurement meets with difficulty.

Fig. 7 shows the spectrum of $^{241}$Am α-rays obtained for the radiation detectors using the TlBr crystal. Figs. 7-(a) and - (b) show the pulse-height spectra with and without the α-source. Although the response is observed slightly with an applied detector bias of 50 V, no peak is observed probably because the quality of the TlBr crystal used is not good enough. It seems highly necessary to optimize growth conditions such as the temperature and the growth rate to obtain crystals of better quality.

4. SUMMARY

The radiation detectors were fabricated from the PbI$_2$ and TlBr crystals. The PbI$_2$ crystals
were grown by four different methods. For the Pbl₂ detectors fabricated from the samples grown by the ordinary Bridgman method, the α-peak was not observed. In the detector fabricated from the samples grown by Bridgman method after the 20 pass zone refining, the α-peak with an energy resolution of about 759 keV (FWHM) was clearly observed at an applied detector bias of 20 V. However, the photopeak for γ-rays was not observed. The detector fabricated from samples grown by 60 pass zone melting method successfully yielded a photopeak for ³⁴⁸Am γ-rays (59.5 keV).

In the case of the TlBr crystal, some trace of response for α-rays was observed, but the peak in the pulse-height spectrum was not yet observed.

5. REFERENCES

Fig. 1. Pulse-height spectra for the 5.48 MeV $\alpha$-particles in PbI$_2$ detectors
a) ordinary Bridgman method
b) ordinary Bridgman method after the 3-pass zone refining
c) ordinary Bridgman method after the 20-pass zone refining

Fig. 2. PL characteristics of the PbI$_2$ crystal grown by:
  a) ordinary Bridgman method (B01)
  b) ordinary Bridgman method after the 3-pass zone refining (B03)
  c) ordinary Bridgman method after the 20-pass zone refining (B20)
Fig. 3. Variation of the peak position corresponding to the 5.48 MeV \( ^{241}\text{Am} \) \( \alpha \)-rays as a function of the applied detector bias on the \( \text{PbI}_2 \) detector.

Fig. 4. Variation of the energy resolution and the peak position for the 5.48 MeV \( \alpha \)-particles as a function of the detector bias.
Fig. 5. Pulse height spectra for the 59.5 KeV γ-rays from $^{241}$Am source as a function of the applied detector bias

Fig. 6. PL characteristics of the sample Z60

Fig. 7. Spectra of $^{241}$Am α-rays obtained for the radiation detector using the TIBr crystals.  
   a) with the α-source  b) without the α-source
LONG TERM VARIATION ON CHARACTERISTICS OF GERMANIUM DETECTORS

Kunio TERAI
Shimane Prefectural Institute for Public Health and Environmental Science.
582-1 Nishihamasada, Matsue, Shimane 690-01, Japan

Kazuhisa KOMURA
Low Level Radioactivity Laboratory, Kanazawa University.
Wâke, Tatsunokuchi, Ishikawa 923-12, Japan

1. INTRODUCTION

Germanium semiconductor detectors (Ge detectors) have become indispensable in non-destructive measurement of environmental radioactivities and \textit{in-situ} measurement of environmental radioactivities and radiations. Recent development of fabrication technique of Ge detectors make available large volume Ge detectors with the 25 cm relative efficiency defined by ISO (International Standard Organization) higher than 100 %.

It is well known that characteristics of Ge detectors (detection efficiency, energy resolution, liquid nitrogen (LN2) consumption, etc.) deteriorate gradually with time, though deterioration rate depends on the operating conditions and/or fabrication performance of each Ge detector. There seems no paper describing the deterioration of Ge detectors over 10 years\textsuperscript{1}). In the present paper, we report the long term variation on characteristics three Ge detectors having the relative efficiency of 10.2 %, 13.6 % and 40.5 % used in Shimane Prefectural Institute for Public Health and Environmental Science.

2. LN2 CONSUMPTION AND CALIBRATION METHOD OF Ge DETECTORS

2.1 LN2 CONSUMPTION

Characteristics of the Ge detectors examined in the present paper are summarized in Table 1. Weight of LN2 used to fulfill the dewar has been recorded at every time, because the increase of LN2 consumption is considered to be one of the good indicators to know the deterioration of Ge detector. The values given in the text are obtained by least squares method.
Table 1 Comparison of data shown in the attached specification for 10.2%(3/9/1973), 13.6%(July 1973), 40.5%(2/24/1982) Ge detectors and those measured by us.

<table>
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<tr>
<th>Composition</th>
<th>Unit</th>
<th>Ortec</th>
<th>Measured</th>
<th>Canberra</th>
<th>Measured</th>
<th>Ortec</th>
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<td>10.2%Ge(Li)(8001-1023S)</td>
<td>Measured</td>
<td>13.6%Ge (Li)(7219-7500/S)</td>
<td>Measured</td>
<td>40.5%h.p. Ge (GEM-35195-S)</td>
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2.2 CALIBRATION METHOD OF Ge DETECTORS

Calibration sources used in the present study are a set of standard gamma ray sources of $^{241}$Am, $^{109}$Cd, $^{133}$Ba, $^{57}$Co, $^{203}$Hg, $^{54}$Mn, $^{60}$Co and $^{88}$Y with activities less than 370 kBq. Each source was placed at 25 cm from the top surface of Ge crystal and the relative efficiency, the peak to Compton ratio, the FWHM (full widths at half maximum), the FWTM (full widths at tenth maximum) and the FWFM (full widths at fiftieth maximum) were measured by accumulating sufficient peak counts.

3. Ge DETECTORS USED IN THIS STUDY

3.1 10.2% Ge(Li) DETECTOR

Performance of the 10.2% true coaxial type lithium-drifted Ge detector (Ortec 8001-1023S, S/N 13-0176) with a preamplifier equipped with an uncooled FET (operated at room temperature) was observed for a long term. After 6 years of operation, consumption of liquid nitrogen became extremely large due to vacuum deterioration. Therefore, re-drifting of lithium was performed to repair the Ge detector. The size and alignment of Ge crystal and construction materials of the detector were compared with the specification data given by EG & G Ortec at the time of repair. The size of p-core and n$^+$ layer of the Ge crystal were measured after re-drifting of lithium to compare with the values given in the specification card and to know the reason of decrease of the detection efficiency.

3.2 13.6% Ge(Li) DETECTOR

This is a closed-end coaxial type lithium-drifted Ge detector (Canberra 7219-7500/S) with a preamplifier equipped with an uncooled FET. The detector has been operated in an air-conditioned room of 19 - 23 °C and 58 - 62 % of relative humidity. Its characteristics were regularly measured over 6 years. At the time of repair, the size and weight of the Ge crystal were measured and compared with the specification data of Canberra.

3.3 40.5% HIGH PURITY Ge DETECTOR

This is a closed-end coaxial type high purity Ge detector (Ortec GEM-35195-S) with cooled FET. The detector is of a specially designed portable type for field use. Continuous in-situ measurements over several months with this detector were performed to monitor the radioactive fallouts due to Chernobyl Reactor Accident.

It experienced more than 50 times of temperature cycling (warm-up and cool-down cycle) during 10 years of operation. The detector was sent back to EG & G Ortec for...
repair several times.

4. RESULTS AND DISCUSSION

4.1 10.2 % Ge(Li) DETECTOR

This detector was first used in May 1973. After 4 years of use, the detector was shipped to EG & G Ortec for repair and sent back to our laboratory. After 18 months of use the detector conked out again, and supply of liquid nitrogen was stopped.

The size of the Ge crystal measured just before shipping was given in Table 1 and the cross-sectional view of the detector is shown in Fig. 1. The size of the Ge crystal and active volume measured are little bit different from the values given in the specification.

The consumption rate of liquid nitrogen was rather constant during first 4 years (1.32 - 1.42 l per day, Ave. = 1.37 l per day). Then deterioration of vacuum was found from rapid increase of LN2 consumption. Six months after the finding of increase of LN2 consumption as seen in Fig. 2, output signal could not be observed at 4000 V of normal applied voltage. Just before shipping for repairing the detector (October 1977), LN2 consumption was increasing by 0.8 % per day. The relative efficiency and the FWHM at 1332 keV were measured to be 3 % and 2.4 keV, respectively, at 3000 V.

![Fig. 1 Cross sectional views of the 10.2 % and 13.6 % Ge(Li) detectors.](image-url)
During the period of 2 months after repair (July-August 1978), LN2 consumption was fairly constant and 1.35 l per day, which is almost the same as that of the original value under normal condition. About 2 months after the finding of the slight increase of LN2 consumption, LN2 consumption began to increase by 0.3 % per day (November 1978-September 1979, Fig. 2), which is about 1/3 of previous case in 1977, and then after 2 months LN2 consumption had increased by 0.9 % per day. Therefore, liquid nitrogen supply was stopped in February 1980.

![Graph showing LN2 consumption (L/day) vs. Elapsed Time (days) with points and annotations for Vacuum deterioration, Rapid vacuum deterioration, and Gradual vacuum deterioration after repair]

**Fig. 2 Liquid nitrogen consumption of the 10.2 % Ge(Li) detector.**

### 4.2 13.6 % Ge(Li) DETECTOR

This detector was used for the first time in October 1974. The relative efficiency, the peak to Compton ratio, the FWHM, and the FWTM/FWHM during 7 years since February 1983 are given in Table 2 (a).

The rate of decrease of the relative efficiency and the peak to Compton ratio was 1.00 % per year (Fig. 3) and 0.55 % per year (Fig. 4), respectively. This Ge(Li) detector was used for 18 years (until November 1992). Recently, the size and weight of crystal of the 13.6 % Ge were measured. The Ge crystal was supported by 3 sticks made of 4.9 mm φ aluminum and 3.1 mm φ teflon (A, B and C). The lengths of the sticks were found to be different: A = 11.8 mm (Aluminum part : 9.8 mm), B = 12.3 mm (10.1 mm) and C = 18.2 mm (1.5 mm), respectively. A spring was inserted in the aluminum tube C. Long term variation of the relative efficiency, the peak to Compton ratio, the FWHM, the FWTM/FWHM and the FWFM/FWHM of the detector are shown in Figs. 3 to 7, respectively.
Table 2  Change of the characteristics of 13.6 % Ge (Li) and 40.5 % high purity Ge detectors during about 10 years, measured with a $^{60}$Co point source at 25 cm distance from the top surface of Ge crystal.

(a) 13.6% Ge (Li) Detector

<table>
<thead>
<tr>
<th>Date</th>
<th>Relative Efficiency %</th>
<th>Peak Compton</th>
<th>FWHM</th>
<th>FWTM FWHM</th>
<th>FWFM FWHM</th>
</tr>
</thead>
<tbody>
<tr>
<td>2/18/1983</td>
<td>13.8</td>
<td>41.8:1</td>
<td>1.90</td>
<td>1.87</td>
<td>2.72</td>
</tr>
<tr>
<td>3/14/1984</td>
<td>13.1</td>
<td>41.0:1</td>
<td>1.85</td>
<td>1.79</td>
<td>*</td>
</tr>
<tr>
<td>3/ 4/1985</td>
<td>13.5</td>
<td>40.2:1</td>
<td>1.82</td>
<td>1.99</td>
<td>*</td>
</tr>
<tr>
<td>3/ 3/1986</td>
<td>13.2</td>
<td>44.0:1</td>
<td>1.77</td>
<td>1.92</td>
<td>*</td>
</tr>
<tr>
<td>3/ 2/1987</td>
<td>13.4</td>
<td>42.2:1</td>
<td>1.80</td>
<td>1.91</td>
<td>*</td>
</tr>
<tr>
<td>2/22/1988</td>
<td>12.9</td>
<td>41.6:1</td>
<td>1.82</td>
<td>1.93</td>
<td>*</td>
</tr>
<tr>
<td>3/ 6/1989</td>
<td>12.7</td>
<td>38.6:1</td>
<td>1.96</td>
<td>1.89</td>
<td>*</td>
</tr>
</tbody>
</table>

* not measured

(b) 40.5% high purity Ge Detector

<table>
<thead>
<tr>
<th>Date</th>
<th>Relative Efficiency %</th>
<th>Peak Compton</th>
<th>FWHM</th>
<th>FWTM FWHM</th>
<th>FWFM FWHM</th>
</tr>
</thead>
<tbody>
<tr>
<td>4/15/1982</td>
<td>41.6</td>
<td>71.4:1</td>
<td>1.77</td>
<td>1.84</td>
<td>2.41</td>
</tr>
<tr>
<td>11/4/1982</td>
<td>40.4</td>
<td>70.4:1</td>
<td>1.78</td>
<td>1.84</td>
<td>2.42</td>
</tr>
<tr>
<td>4/18/1983</td>
<td>40.5</td>
<td>72.2:1</td>
<td>1.77</td>
<td>1.82</td>
<td>2.40</td>
</tr>
<tr>
<td>10/15/1984</td>
<td>40.5</td>
<td>73.8:1</td>
<td>1.71</td>
<td>1.83</td>
<td>2.45</td>
</tr>
<tr>
<td>8/12/1986</td>
<td>39.9</td>
<td>67.7:1</td>
<td>1.81</td>
<td>1.85</td>
<td>2.46</td>
</tr>
<tr>
<td>5/19/1987</td>
<td>38.5</td>
<td>69.6:1</td>
<td>1.77</td>
<td>1.84</td>
<td>2.43</td>
</tr>
<tr>
<td>8/13/1987</td>
<td>37.2</td>
<td>65.8:1</td>
<td>1.84</td>
<td>1.83</td>
<td>2.49</td>
</tr>
<tr>
<td>10/30/1990</td>
<td>36.9</td>
<td>69.8:1</td>
<td>1.75</td>
<td>1.84</td>
<td>2.45</td>
</tr>
</tbody>
</table>

* 4/25/1991 | 38.0                  | 71.3:1       | 1.70 | 1.87      | 2.46      |
* 2/8/1993  | 38.0                  | 68.6:1       | 1.74 | 1.83      | 2.50      |

* after repair

Fig. 3  Variation of the relative efficiency of the 13.6 % and 40.5 % Ge detectors.
Fig. 4 Variation of peak to Compton ratio of the 13.6 % and 40.5 % Ge detectors.

[P/C] = -0.00132×[days] + 71.7

13.6% Ge (Li)

[PC] = -0.000631×[days] + 42.0

40.5% h.p. Ge

Fig. 5 Variation of FWHM of the 13.6 % and 40.5 % Ge detectors.

[FWHM] = 9.75×10^{-6}×[days] + 1.83

13.6% Ge (Li)

[FWHM] = 5.42×10^{-6}×[days] + 1.76

40.5% h.p. Ge

Fig. 6 Variation of FWTM/FWHM ratio of the 13.6 % and 40.5 % Ge detectors.

[T/H] = 2.51×10^{-6}×[days] + 1.87

13.6% Ge (Li)

[T/H] = 2.26×10^{-6}×[days] + 1.83

40.5% h.p. Ge
4.3 40.5 % HIGH PURITY Ge DETECTOR

This detector has been used since April 1982. Except for the use in the field, the detector was used in an air-conditioned room. Measurement conditions in the field (in situ) were much worse compared with those of room measurements. For example, in the in situ work measurements were made under torrential rain, under extremely low temperature below 0 °C, high temperature and high humidity as in a sauna or hotspring, or in very high gamma-ray background in the presence of fast neutrons from a nuclear reactor, with maximum flux of $10^4$ n cm$^{-2}$s$^{-1}$.

As known from Table 2 (b), the energy resolution of the detector equipped with a cooled FET is better than the 13.6 % Ge(Li) detector with an uncooled FET. After 9 years of use (January 1991), this detector was sent back to EG & G Ortec for repair. The detection characteristics measured just after the repair, are given in the bottom of Table 2 (b). The size of Ge crystal was the same as the values given in the specification. However, thickness of end-cap changed from 1.0 mm to 1.27 mm and the distance from end-cap to the Ge crystal was shortened from 6 mm to 3 mm to get detection efficiency as high as possible.

Long term variation of the relative efficiency, the peak to Compton ratio, the FWHM, the FWTM/FWHM and the FWFM/FWHM of the 40.5 % high purity Ge detector are shown together with those of the 13.6 % Ge(Li) detector in Figs. 3 to 7.

The absolute peak efficiency and the FWHM before and after repair are compared in Figs. 8 and 9. As shown in Table 2 (b) and Fig. 8, detector performance has not completely be restored to that in 1982.

Annual decrease of the relative efficiency and the peak to Compton ratio were 1.36 % per year (Fig. 3) and 0.67 % per year (Fig. 4), and those of the FWTM/FWHM and the FWFM/FWHM were 0.045% per year (Fig. 6) and 0.296 % per year (Fig. 7), respectively.
During 10 years of operation, the relative efficiency decreased by 50 % and 4.4 % at 59.5 keV and 1332 keV (Fig. 8), respectively. The FWHM increased up to 10 % and 0.85 % at 80.9 keV and 1332 keV (Fig. 9), respectively.

This detector has been subjected to more than 50 times of temperature cycling during 10 years. After the repair in 1991, this detector can be used without trouble until
5. CONCLUSION

Long term variation of characteristics of Ge detectors depends, of course, highly on the fabrication performance of each detector and on measurement conditions. Characteristics of these three Ge detectors have been followed for more than 10 years for the liquid nitrogen consumption, the relative efficiency and the peak to Compton ratio. Main causes of deterioration of the Ge detector studied are found to be as follows.

1. Deterioration of vacuum of the 10.2% Ge(Li) detector cryostat which had been repaired twice during 6 years, was caused by a defect in metal-welding between the flange and cryostat.

2. Deterioration of the 13.6% Ge(Li) detector, which had been used for 18 years, was much slower than the 40.5% high purity Ge detector (3). This was mainly due to excellence of fabrication and to careful use in an air-conditioned room.

3. Deterioration of the 40.5% high purity Ge detector was mainly due to frequent temperature cycling of over 50 times during 10 years of field works.

Observations described in present paper are believed to be very helpful to the researchers using Ge detectors for scientific studies and/or the monitoring of environmental radioactivities.

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The authors express their deep thanks to Dr. M. Noguchi, former science staff of the Japan Atomic Energy Research Institute (JAERI) for his continuous encouragement and suggestions to round off this study.

REFERENCES

SYSTEM CALIBRATION FOR PHOTON SPECTROMETERS

Toshiaki Kishikawa *
Faculty of Engineering, Kumamoto University, Kumamoto, 860 Japan

Chushiro Yonezawa and Hideaki Matsue
Department of Chemistry and Fuel Research
Japan Atomic Energy Research Institute (JAERI), Tokai, Ibaragi, 319-11 Japan

Hiroyuki Sawahata
Research Center for Nuclear Science and Technology
The University of Tokyo, c/o JAERI, Tokai, Ibaragi, 319-11 Japan

ABSTRACT
System calibration for precise measurement of photon energy with Ge / Ge-BGO photon spectrometers has been performed by using a precision pulser. Smoothed spline function was applied for the non-proportionality correction function as well as the electronic noise width function.

1. INTRODUCTION

Energy of photon, i.e. gamma-ray or mesonic X-ray, is one of essential quantities representing excited state of nuclide or mesonic atom, respectively. For the energy measurement, photon spectrometers equipped with germanium semiconductor diode detectors of various types are extensively used, because of high resolution of energy. Any of conventional energy calibration methods is based on the linear relation between the gamma-ray energy standard source(s) and the maximum position (the peak position) of the relevant photopeak in the spectrum. However, the method derives deviation of energy values as compared with those measured by crystal diffraction spectrometry.

A new method of energy calibration is based on the instrument function model summarized in Ref. (2). Figure 1 illustrates the concept of new energy calibration method. The method uses "reference standard" source(s), the gamma-ray energies of which are measured by the crystal diffraction method and adjusted by the standard wavelength of 606-nm $^{85}$Kr ($\lambda = 605.780210$ nm). As the energy – wavelength conversion constant, $E\lambda =$

* Corresponding author. Fax.: + 81 (0) 96-342-3783.
Upon incidence of photons the detector generates charge carriers (electrons and positive holes). While the charge carriers are drifting toward relevant electrodes, some of them are trapped in trapping centers. If there is no trapping event, then the resultant photopeak in a memory of a pulse height analyzer (PHA) shows the characteristic shape of normal distribution (ND) due to statistical fluctuation. The centroid of ND is the position reflecting the photon energy. The trapping event causes escape from estimated arriving of the charge carriers onto the electrode. Fluctuation due to the event is expressed in terms of the random escape probability density distribution (REPDED). The resultant, compound distribution function is defined to be the instrument function. Deconvolution of the instrument function, therefore, provides the ND folded in it.

The spectrometer system should be calibrated prior to measurement of standard or sample source(s). The system calibration can be performed by using a precision pulsar. The performance is based on the convolution nature of the ND (source) and the ND (electronic noise) giving the ND (compound). This paper reports the method of the system calibration and an interpolation method of the electronic noise.

2. METHOD OF THE SYSTEM CALIBRATION

2.1. System Calibration

The NDs folded in the instrument function are those of a) statistical fluctuations at time of charge carriers creation $P_C(E, x)$ with incident photons of energy $E$ and b) electronic noise of the spectrometer system $P_N(E, x)$. The centroids of NDs, i.e. $x_{C,C}$ and $x_{C,N}$, of the charge carriers creation and the electronic noise, respectively, are identical because of the folding nature, thus

$$x_C = x_{C,C} = x_{C,N}$$  

1.23984244 x 10^{-6} \text{ eV m ± 0.3 ppm}$$  

is applicable from the 1986 Adjustment of the Fundamental Physical Constants (3).

Fig. 1. Concept of energy calibration.
Therefore, the system calibration can be performed with pulsar signals which generate the noise shapes at relevant positions in the memory channels of PHA(s) corresponding to amplitude voltage values of the pulsar.

We perform the system calibration with a precision pulsar. Measure the peak height distribution of \( i \)th pulsar signal amplitude voltage \( A_i \) in order to find out the apparent centroid \( x_{\text{iap}} \). Then, obtain a pulse amplitude equation with the two points method or with the linear least square method:

\[
A(x) = a_w (x - x_0),
\]

where \( a_w \) is the amplitude voltage-channel conversion coefficient or the slope in V/channel, and \( x_0 \) is the offset channel to give zero amplitude voltage.

The system calibration equation can be given from a deviation \( \Delta x_i \) of \( i \)th pulsar centroid

\[
\Delta x_i = x_{\text{iap}} - x_i,
\]

as:

\[
x_i = x_{\text{iap}} + \Delta x_i - x_0.
\]

A plot of \( \Delta x_i \) versus \( x_{\text{iap}} \) gives the system non-proportionality correction curve.

### 2.2. Interpolation of the system calibration curve

The smoothed spline function is applied in order to interpolate the system calibration curve \(^{(4,5)}\). With the set of system non-proportionality data

\[
\Delta x_i = y_i = y(x_i) \quad (i = 1, 2, \ldots, n),
\]

the smoothed functional value \( f(x) \) for \( x = x_i \) of the system non-proportionality function \( f(x) \) is either close to the measured data \( f(x_i) \approx y_i \) or much more smoothed from the original data. As a measure of smoothing we introduce the smoothing evaluation function:

\[
\sigma(g) = \sum_{i=1}^{n} w_i [(f(x_i) - y_i)^2 + g \int_1^{\infty} [f^{(m)}(x)]^2 \, dx],
\]

with \( g \) \((0 \leq g < \infty)\) being the smoothing parameter, \( w_i \) \((0 \leq w_i \leq 1)\) being the weighting factor and \( f^{(m)}(x) \) being the \( m \)th order differential of spline function. The first term on the right hand side of Eq. (6), i.e. the square of the second power norm, is the quantity term how the smoothed function is to fit the original data points. The second term is a measure of smoothness of the obtained function. If \( g \) is zero, then \( \sigma(g) \) is also zero, thus the functional value in Eq. (6) turns to be \( f(x_i) = y_i \) \((i = 1, 2, \ldots, n)\). Therefore \( f(x) \) gives
folded lines. If \( g \) is infinity, on the other hand, then the functional value is far apart from the original point and \( f(x) \) turns to be zero, i.e. \( \Delta x \) is zero.

The spline function is made up of divided polynomial functions. The system non-proportionality function \( f(x) \) is given by the natural spline of \((2m - 1)\)th order:

\[
f(x) = p_{2m-1}(x) + \sum_{i=1}^{2m-1} c_i (x - x_i)^{2m-1},
\]

with

\[
p_{2m-1}(x) = \sum_{i=0}^{m-1} a_i x_i,
\]

where \( p_{2m-1}(x) \) is the polynomial of \((m - 1)\)th order with \( a_i \) being the \( i \)th parameters, and \( c_i \) is the constant to satisfy \( m \) conditions of

\[
\sum_{i=0}^{2m-1} c_i x_i^r = 0 \quad (r = 0, 1, \ldots, m-1),
\]

and \((x - y)^{2m-1}\) is the truncated power function of \( m \)th order.

The optimum value of smoothing parameter \( g \) can be obtained based on criterion for fitting named trend \((6)\). Let us define residue (as a function of \( g \); see Eq. \((13)\)) be

\[
r_k(g) = \Delta x_i - \Delta x_{\text{spline}} = y_k - f(x_k, g),
\]

since the system non-proportionality function \( f(x) \) is a function of \( g \), then the trend \( R(g) \) can be given by

\[
R(g) = \sum_{k=0}^{g} r_k(g)^2.
\]

If \( g \) has much higher value, then the neighboring residues are, both, either positive or negative. That is, the product of the neighboring residues, i.e. the trend, become positive. Therefore, the optimum smoothing parameter \( g_{\text{opt}} \) can be obtained at the minimum point of the trend-versus-smoothing parameter curve.

Finally, we obtain general equation of the system calibration curve for Eq. \((4)\) by replacing \( x_C = x_p, x_{\text{C,ap}} = x_{\text{ap}} \) and \( f(x_{\text{C,ap}}) = \Delta x(x_{\text{C,ap}}) \):

\[
x_C = x_{\text{C,ap}} + f(x_{\text{C,ap}}) - x_0.
\]

The energy calibration curve is, as a result, given by introducing the estimated centroid data from measured photopeak shapes based on the instrument function:

\[
E(x_C) = c_W x_C,
\]

where \( c_W \) in units of keV/channel is the channel width or energy-channel conversion coefficient.

2.3. Electronic noise width
The noise width is a part of observed width:

\[ s_{\text{obs}}^2(E, x_C) = s^2(E, x_C) + s_n^2(x_C), \]

where \( s(E, x_C) \) is the standard deviation of charge carrier creation and \( s_n(x_C) \) is that of electronic noise. Because of the presence of fluctuation in the noise width value, the width is expressed in the function of \( x_C \). The interpolated value of noise width can be obtained from the smooth spline function \( f(x) \) and measured data \( y_{N,j} = s_{N}(x_{C,j}) \) by replacing, respectively, \( f(x) \) and \( y_{N} \) in Eq. (6).

3. EXPERIMENTAL

Two spectrometers were used in the experiments. One was a typical single mode spectrometer (Ge equipped EG&E Ortec 7450 multichannel analyzer of KRI - Kurokami Radioisotope Lab. Kumamoto Univ.). The other was Seiko EG&G Ge-BGO multi-mode operation spectrometer system. The features of the latter have already been discussed (7); however it is important to note that the system has four modules of PHAs operated at the same time:

- the Compton-suppression mode at 0 – 4 MeV range (PHA-1);
- the Compton-suppression mode at 3 – 11 MeV range (PHA-2);
- the pair mode at 3 – 11 MeV range (PHA-3);
- the single mode at 0 – 11 MeV range (PHA-4).

The spectrometer system is installed at the cold or thermal neutron beam guides at the neutron beam hall of JRR-3M, Tokai Establishment, Japan Atomic Energy Research Institute (JAERI).

A pulser, BNC model PB-4 was used for generating precision signals.

4. RESULTS AND DISCUSSIONS

4.1. Noise centroids

Typical spectrum of noise peaks measurement is shown in Fig. 2 with an appropriate single mode PHA (at KRI). Precision pulsar signals at varied amplitude voltage were introduced with each live time of 100 s at the test input of a preamplifier of the spectrometer system. During an overall acquisition time background photon signals were unavoidably incident on the detector and stored in the memory channels as base components under pulsar peaks.
The pulsar centroid was estimated after subtracting the base component.

The plots of Amplitude versus channel number are shown in Figs. 3a and 3b, respectively, for the PHA of KRI and for the PHA-1, 2 and 4 of the Seiko EG&G Ge-BGO multi-mode operation spectrometer system. The linear relation of the amplitude voltage and channel were obtained according to Eq. (2) with the linear least square method (though, the fitted lines are not inserted in the
Top figures of Figs. 4a-4d give plots of the deviation $\Delta x_i$ against $x_i$, i.e. the figures.

Fig. 4. Deviation versus channel number for different PHAs of Photon spectrometers.
non-proportionality correction curves, for the PHA-KRI and the PHA-1 to 4 systems. Because of the patterns of the figures to show fluctuations of the deviation values, an optimization treatment is necessary for the interpolation of the non-proportionality correction.

4.2. The smoothing parameter

In order to obtain optimum non-proportionality correction curves the trend versus smoothing parameter curves were plotted for varied value of logarithm of the smoothing parameter \( g \). As shown in Figs. 5a and 5b, the curves tend to show minimum positions. From the minimum position we obtain the optimum smoothing parameter \( g_{opt} \).

![Fig. 5. Trend as a function of smoothing parameter for various PHAs.](image)

4.3. The interpolation function for the non-proportionality correction curve

If one applies the optimum smoothing parameter \( g_{opt} \) to the optimum non-proportionality correction function, i.e. the natural spline in Eq. (6), the function \( f(x) \) is the function to minimize the smoothing evaluation function \( \sigma(g) \). Such the condition can be
established when one satisfy the following conditional equations:

\[ f(x_j) - y_j = (-1)^n g_{opt}(2m - 1)! c_j w_j^{-1} \quad (j = 1, 2, ..., n). \]  

Finally, the optimum non-proportionality correction function can be obtained. Minus value of the residual, \(-r_k(g)\)\_gopt = \(\Delta x\)\_spline - \(\Delta x\)\_j, of Eq. (8) are plotted in the bottom figures of Figs. 4a - 4d for the PHA-KRI and the PHA-1 to 4 systems. The non-proportionality correction curves are not inserted in the top figures of Figs. 4a - 4d.

5. CONCLUSION

The system calibration for pulse height analyzers can be established with the use of precision pulser. It is concluded that the system non-proportionality depends essentially on maintenance conditions of the analyzer system, so that occasional or regular checking of the system condition is necessary for the precise measurement of photon energy.

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Signal Processing of Preamplifiers for Superconducting Tunnel Junction X-ray Detector

Keisuke Maehata and Kenji Ishibashi
Department of Nuclear Engineering, Kyushu University
Yuzuru Matsumoto
Kyushu Teikyo Junior College

1. INTRODUCTION

In usual X-ray measurement with Si semiconductor detectors, charge sensitive preamplifiers are used in order to produce a voltage pulse whose height is proportional to incident X-ray energy independent of the fluctuation of the bias voltage of Si detector. Since electron-hole pairs produced by incident X-ray are collected by the strong electric field in the depletion region of the Si semiconductor, the output voltage pulse of the preamplifier with a fast rise time is considered as an impulse response to the integrated circuits[1].

Recently, several superconducting tunnel junction (STJ) detectors have been developed for further high resolution X-ray measurement[2]. The STJ detector consists of three layers i.e. superconductor, insulator and superconductor. In STJ detectors, the deposited energy by incident X-ray breaks Cooper-pairs and creates quasiparticles. The preamplifier of the STJ detector collects the signal charge caused by tunneling quasiparticles remaining in the superconductor during a relaxation process. The signal charge extracted from STJ generates exponentially decaying current. The decaying time constant is defined by the time of relaxation and tunneling of quasiparticles in the STJ and depends on physical properties of material[2]. Actual STJ detectors have the decaying time constant of the signal current in the range from 0.5 to 20 µs[2]. We fabricated a STJ detector with the decaying time constant of 0.6µs and obtained energy spectra of synchrotron radiation in energy range from 4 to 15keV by the use of a commercial charge sensitive preamplifier (ORTEC 142C) for the Si detector[3]. On the other hand, a current sensitive preamplifier was employed in precise measurements of the low energy response of a STJ detector with the decaying time constant of 5µs[4]. Since the decaying current generated by the STJ has quite long time constant compared with the impulse response of the Si detector,
the signal processing of the preamplifier is optimized to obtain an appropriate pulse height of the signal concerned with the decaying current. In this work, we analyze characteristics of signal processing of preamplifier for STJ detector by taking into account of the decaying time constant. Furthermore, several noise contributions to the output voltage of the main amplifiers are estimated.

2. OUTPUT VOLTAGE OF PREAMPLIFIER

A circuit diagram of the STJ detector connected with the preamplifier provided with feedback resistance and capacitance is shown in Fig.1(a). Symbols appear in this paper are listed in the nomenclature. By converting the feedback impedance to the input impedance of the operational amplifier, an equivalent circuit diagram of the preamplifier is drawn as given by Fig.1(b). The current source in Fig.1 indicates the generation of the decaying current by X-rays incident on STJ. The decaying current is given by

\[ i(t) = \frac{qN}{\tau_f} \exp(-t/\tau_0), \]  

where \( N \) is the total number of initial quasiparticles, \( \tau_f \) is the tunneling time constant and \( \tau_0 \) is the time constant of the decay of quasiparticles. The output voltage of the preamplifier for \( i(t) \) in Fig.1(b) is obtained as

\[ V_{\text{out}}(t) = -A \frac{Q}{C_e} \frac{1}{1 - \frac{\tau_0}{\tau_{\text{in}}}} \left( e^{-t/\tau_0} - e^{-t/\tau_{\text{in}}} \right). \]  

In eq.(2), \( Q \) is the total charge collected by the preamplifier and expressed by \( Q = qN\tau_0/\tau_f \). Alternatively, by means of the energy required for single electron collection \( \varepsilon_{\text{eff}} \), the energy \( E \) of the incident X-ray gives the total charge as \( Q = qE\varepsilon_{\text{eff}} \). A decaying time constant \( \tau_{\text{in}} \) in eq.(2) is given by \( \tau_{\text{in}} = C_e R_L \), where the input capacitance \( C_e \) and resistance \( R_L \) of preamplifier is derived from Fig.1(b) as \( C_e = C_d + C_u + (A + 1)C_f \) and \( 1 / R_L = 1 / R_j + (A + 1) / R_j \), respectively. The output voltage of the preamplifier is calculated for the detection of incident X-rays of 6keV on our previous STJ detector (STJ-K) that was employed in the synchrotron radiation measurement. Parameters of the preamplifier ORTEC 142C used in this work are as follows: \( A = 80000, R_f = 500\,\text{M}\Omega \) and \( C_f = 2\,\text{pF} \). The value of \( \varepsilon_{\text{eff}} \) of the STJ-K was found to be 20meV by the measurement. In Fig.2, the solid curve indicates calculated output voltage of the preamplifier for STJ-K. For a comparison, the output voltage for the STJ developed by Lawrence Livermore National Laboratory (STJ-L) is estimated in the case of the operation with the same preamplifier.
and the result is indicated by the dotted line in Fig.2. Electric properties of the STJ-K and STJ-L used in the calculation are listed in Tab.1. The rise time constant of STJ-K and STJ-L is 0.7 and 7 μs, respectively. In measurements with STJ-L, a current sensitive preamplifier of $\tau_{in} = 0.45\mu s$ was employed [4]. The output voltage of the current sensitive preamplifier for STJ-L is obtained by assuming the capacitance component of the feedback resistance of 0.1pF, and shown by the broken line in Fig.2. The output voltage of the current sensitive preamplifier recovers to the zero level in 20μs.

3. SHAPING AND NOISE CONTRIBUTION

The signal from the preamplifier is transmitted to the main amplifier to be amplified and shaped to a convenient form for the further processing such as a multichannel analyzer. Since the signal to noise (S/N) ratio is the most important parameter for the measurement with an excellent energy resolution, shaping characteristics of the main amplifier are analyzed concerned with the signal from the preamplifier in consideration of the noise contribution. Since the Gaussian shaping is popular in commercial main amplifiers, in this work, the CR-(RC)$^4$ shaping is considered in order to simplify the analysis. The CR-(RC)$^4$ shaping circuit consists of CR differentiation followed by 4 stages of RC integration, and the time constant of each stage takes the same value defined by $\tau = RC$. By using the Laplace transform, the transfer function of the CR-(RC)$^4$ shaping is given by

$$G(s) = \left(\frac{1}{\tau}\right)^4 \frac{s}{(s + 1/\tau)^5}. \tag{3}$$

The output voltage after shaping is calculated by the use of eq.(3) for the signal from the charge sensitive preamplifier of the STJ that is shown in Fig.2. Fig.3 shows the output of the main amplifier after shaping in the case of incidence of X-rays of 6keV on the STJ-K for several shaping time constants $\tau$. The peak voltage of the main amplifier becomes constant for the shaping time constant $\tau > 2\mu s$. The recovery time of the output voltage is about 20μs. The signal from the main amplifier of $\tau = 2\mu s$ in Fig.3 can be processed by commercial equipment such as the analog to digital converter and the multichannel pulse height analyzer. On the other hand, the output voltage after shaping is calculated with the assumption of the STJ-L operated with the charge sensitive preamplifier. As shown in Fig.4, the shaping time constant should be taken to be $\tau > 10\mu s$ to obtain sufficient pulse height, and the recovery time of the output is about 0.1ms. It is difficult for commercial equipment
to handle such long voltage pulse. However, the signal from the current sensitive preamplifier for STJ-L has a fast decaying tail with the recovery time of 20μs as shown in Fig.2. By amplifying this pulse without shaping, commercial equipment can be utilized for further signal processing. In this processing, an appropriate filtering should be employed to minimize several noise contributions[4].

Since the signal from the preamplifier contains components of the voltage pulse induced by the incident X-rays and several noises, the noise contribution to the signal should be analyzed. In usual analysis, the noise source is regarded as a current or a voltage source. Fig.5 shows the equivalent circuit for analysis of the noise contribution. The voltage source \( n_a \) and \( n_b \) indicates Johnson noise of the resistance of the STJ and the feedback resistance, respectively. In the operation of the STJ detector, the constant bias current \( I \) is supplied through the junction and \( I \) acts like the gate leakage current in the field effective transistor (FET) of the preamplifier. The current source \( n_c \) represents the noise arises from \( I \). The last voltage source \( n_d \) is generated by the shot noise of the FET, where \( R_{eq} \) and \( T_{eq} \) is the equivalent noise resistance and temperature of the FET, respectively. The root mean square (RMS) value of noise voltage observed at the output of the preamplifier is derived from the equivalent circuit of the noise source in Fig.5 as follows,

\[
V_{n_a}^2 = A^2 \frac{4kT_f}{\sqrt{\left(\frac{1}{R_d} + \frac{A+1}{R_f}\right)^2 + \omega^2 \left(\frac{\omega^2 C_d + C_a + (A + 1)C_f}{R_d}\right)}} d\omega, \tag{4}
\]

\[
V_{n_b}^2 = A^2 \frac{4kR \cdot T_f}{1 + \omega^2 C_f^2 R_f} d\omega, \tag{5}
\]

\[
V_{n_c}^2 = A^2 \left(\frac{1}{R_d} + \frac{A+1}{R_f}\right)^2 + \omega^2 \left(\frac{C_d + C_a + (A + 1)C_f}{R_d}\right)^2 d\omega, \tag{6}
\]

\[
V_{n_d}^2 = A^2 \left(\frac{1}{R_d} + \frac{A+1}{R_f}\right)^2 + \omega^2 \left(\frac{C_d + C_a + (A + 1)C_f}{R_d}\right)^2 d\omega, \tag{7}
\]

where \( V_{n_a}, V_{n_b}, V_{n_c} \) and \( V_{n_d} \) are the RMS values of noise voltage generated by the source \( n_a, n_b, n_c \) and \( n_d \), respectively. The RMS value of total noise voltage \( V_{n_{tot}} \) at the output of the main amplifier is calculated by eqs.(4) - (7) and the transfer function of the shaping network in the form of Fourier transform \( G(j\omega) \). The expression of \( V_{n_{tot}} \) is given by
\[ V_{\text{RMS}}^2 = \int_0^\infty \left[ V_{n_a}^2 + V_{n_b}^2 + V_{n_c}^2 + V_{n_d}^2 \right] G'(j\omega)G'(j\omega) \, df \]

where \( G'(j\omega) \) is the complex conjugate of \( G(j\omega) \). The RMS value of \( V_{\text{RMS}} \) is evaluated for the STJ-K by taking parameters as \( T_d = 0.4K, T_f = 300K, I = 0.13\mu A, T_{eq} = 300K \) and \( R_{eq} = 14\Omega \). The values of \( V_{\text{RMS}} \) evaluated for STJ-K is shown by the solid line in Fig.6 as a function of the shaping time constant \( \tau \). Recently, a GaAs FET was developed for a cryogenic preamplifier operated at 1K[5]. For an example of application, the FET of preamplifier is assumed to be cooled down to 4.2K i.e. \( T_{eq} = 4.2K \) with keeping \( R_{eq} = 14\Omega \). The dashed line in Fig.6 indicates \( V_{\text{RMS}} \) of the cooled GaAs FET preamplifier.

By supposing that the incident X-ray induce a constant pulse height \( V_{\text{peak}} \) at the output of the main amplifier in the absence of noise, \( V_{\text{peak}} \) is superimposed on the noise voltage \( V_n \) at the output to obtain a resultant pulse height \( V_{\text{ph}} \) expressed as \( V_{\text{ph}} = V_{\text{peak}} + V_n \). Since \( V_{\text{RMS}} \) is the RMS of \( V_n \) and \( V_{\text{ph}} \) is the Gaussian distribution, the full width at the half maximum (FWHM) \( V_{\text{FWHM}} \) of \( V_{\text{ph}} \) is given by 2.36\( V_{\text{RMS}} \). When the energy resolution \( \Delta E \) arises from the electric noise alone, the FWHM of the energy spectrum can be written by the relationship as

\[ \frac{\Delta E}{E} = \frac{V_{\text{FWHM}}}{V_{\text{ph}}} = 2.36 \frac{V_{\text{RMS}}}{V_{\text{ph}}}. \]

The resultant pulse height \( V_{\text{ph}} \) of the STJ-K is equivalent to the peak voltage shown in Fig.3. The solid curve in Fig.7 indicates the calculated \( \Delta E \) of the STJ-K for the detection of X-ray of 6keV. In the measurement of the synchrotron radiation, the shaping time consonant \( \tau \) of 2 \( \mu s \) was chosen for the STJ-K, and the value of \( \Delta E \) was obtained to be 160eV by the use of a pulsar[3]. From Fig.7 the calculated value of \( \Delta E \) at \( \tau = 2\mu s \) is indicated to be 170eV. The calculated result agrees well with the measurement. The dashed curve in Fig.7 shows calculated \( \Delta E \) in the case of the cooled GaAs FET preamplifier. The value of \( \Delta E \) is expected to decrease below 100eV in \( \tau < 1\mu s \).

4. CONCLUSION

Characteristics of signal processing of the preamplifier for STJ detector are analyzed by means of equivalent circuit diagrams. In the analysis, the decaying current signal generated by the STJ is taken into account. The signal from the STJ detector of the decaying time constant \( \tau_0 = 0.6\mu s \) is demonstrated to be processed by the charge sensitive preamplifier for further signal processing by commercial equipment with the shaping time.
constant of 2μs. Since the STJ of τ₀ > 5μs has difficulty of the signal processing by the charge sensitive preamplifier because of the long decay voltage pulse at the output, utility of the current sensitive preamplifier with an appropriate filtering is suggested for further signal processing by commercial equipment. The RMS of the noise voltage at the output of the preamplifier is estimated by the use of the noise equivalent circuit. The estimated electronics noise contribution ΔE for STJ-K agrees well with the experimental result. In the case of the cooled GaAS FET preamplifier, ΔE is expected to decrease below 100eV in τ < 1μs.

REFERENCE


NOMENCLATURE

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
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<tbody>
<tr>
<td>A</td>
<td>Open loop gain of the preamplifier</td>
</tr>
<tr>
<td>C</td>
<td>Capacitance of the shaping network</td>
</tr>
<tr>
<td>C_d</td>
<td>Capacitance of the STJ</td>
</tr>
<tr>
<td>C_e</td>
<td>Input capacitance of the preamplifier</td>
</tr>
<tr>
<td>C_f</td>
<td>Feedback capacitance of the preamplifier</td>
</tr>
<tr>
<td>C_st</td>
<td>Stray capacitance of the cable between the STJ</td>
</tr>
<tr>
<td></td>
<td>and the preamplifier</td>
</tr>
<tr>
<td>E</td>
<td>X-ray energy</td>
</tr>
<tr>
<td>f</td>
<td>Frequency of noise voltage</td>
</tr>
<tr>
<td>I</td>
<td>Leakage current of the STJ</td>
</tr>
<tr>
<td>i_s</td>
<td>Signal current of the STJ</td>
</tr>
<tr>
<td>N</td>
<td>Total number of quasiparticles</td>
</tr>
<tr>
<td>q</td>
<td>Electronic charge</td>
</tr>
<tr>
<td>Q</td>
<td>Total charge collected by the preamplifier</td>
</tr>
<tr>
<td>R</td>
<td>Resistance of the shaping network</td>
</tr>
<tr>
<td>R_eq</td>
<td>Equivalent noise resistance</td>
</tr>
<tr>
<td>R_d</td>
<td>Resistance of the STJ</td>
</tr>
<tr>
<td>t</td>
<td>Time</td>
</tr>
<tr>
<td>T_eq</td>
<td>Equivalent noise temperature</td>
</tr>
<tr>
<td>T_f</td>
<td>Temperature of feedback resistance</td>
</tr>
<tr>
<td>T_d</td>
<td>Temperature of the STJ</td>
</tr>
<tr>
<td>v_n</td>
<td>Noise voltage</td>
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</tbody>
</table>
\( V_{\text{ntot}} \) RMS of total noise voltage
\( V_{\text{peak}} \) Constant pulse height without noise
\( V_{\text{ph}} \) Resultant pulse height with noise
\( \Delta E \) Energy resolution
\( \varepsilon_{\text{eff}} \) Energy required for single electron collection
\( \tau \) Shaping time constant
\( \tau_0 \) Decaying time constant of signal
current of the STJ
\( \tau_1 \) Decaying time constant of the output voltage of the preamplifier
\( \tau_d \) Tunneling time constant

<table>
<thead>
<tr>
<th>Tab. 1 Electric properties of the STJ</th>
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<tbody>
<tr>
<td>STJ-K</td>
</tr>
<tr>
<td>( Q )</td>
</tr>
<tr>
<td>( R_d )</td>
</tr>
<tr>
<td>( C_d )</td>
</tr>
<tr>
<td>( C_w )</td>
</tr>
<tr>
<td>( \tau_0 )</td>
</tr>
</tbody>
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Fig. 1 (a) Circuit diagram of the STJ detector connected with the preamplifier

Fig. 1 (b) Equivalent circuit diagram of the STJ detector connected with the preamplifier
Fig. 2 Output voltage of the preamplifier

Fig. 3 Output voltage of the main amplifier of STJ-K

Fig. 4 Output voltage of the main amplifier of STJ-L
Fig. 5 Equivalent circuit diagram of noise sources

![Equivalent circuit diagram of noise sources](image)

Fig. 6 RMS of the noise voltage at the output of the main amplifier

![RMS of the noise voltage at the output of the main amplifier](image)

Fig. 7 The energy resolution $\Delta E$ arises from electronics noise

![The energy resolution $\Delta E$ arises from electronics noise](image)
PULSE SHAPE DISCRIMINATION BETWEEN PROTONS, PIONS AND ELECTRONS WITH NE213 LIQUID SCINTILLATOR

Tatsushi NAKAMOTO, Kenji ISHIBASHI, Naruhiro MATSUFUJI, Nobuhiro SHIGYO, Keisuke MAEHATA
Department of Nuclear Engineering, Kyushu University
Hakozaki, Fukuoka 812-12, Japan

1. INTRODUCTION

The pulse shape discrimination (PSD) method with an organic scintillator is often employed for eliminating gamma-rays from neutrons. The principle of the PSD is based on the difference in decay components of light outputs. The amplitude of the slow decay component depends on the density of ionization. The density of ionization induced by protons or ions is quite higher than that by electrons. The neutron and gamma-ray discrimination, therefore, can be made by the use of the difference in the decay components. Various methods of PSD have been studied, and they are classified into two kinds. One of them is the "zero cross timing method", and the other is the "two-gate integration method". We tested the zero cross timing method for measuring high energy neutrons up to a few hundred MeV. We found that saturation effects occurred on the zero cross time in a pulse-shaping preamplifier, and gamma-rays could not be eliminated from neutrons in such a high energy region. The two-gate integration method was then used and saturation effects did not appear. The two-gate integration method, therefore, was adopted into our spallation neutron measurements induced by protons of GeV energies.

Neutron production double differential cross sections were successfully obtained by using the PSD eliminating gamma-rays. At the next step, we plan to get the emission cross sections of charged particles such as pions and protons from the same experimental data. Pions have never been reported to be identified by the PSD so far. In this work, we attempt to demonstrate that pions are separated from protons and electrons by the two-gate integration method with an NE213 organic liquid scintillator. The $\Delta E$-$E$ method is usually used for the charged particles identification. We consider that it may be possible to make the particles identification more accurate by combining the PSD with the $\Delta E$-$E$ method.

2. EXPERIMENTAL METHOD
The experiment of the PSD was performed with 12 GeV Proton Synchrotron at The National Laboratory for High Energy Physics (KEK). Protons in the range of 0.8 to 3.0 GeV were bombarded on targets of C to Pb. and such particles were measured as protons, pions, neutrons and gamma-rays. Fig. 1 shows the timing scheme of the PSD for the particles identification with the two-gate integration method. The anode signal of a photomultiplier tube (PMT) was integrated by charge sensitive analogue-to-digital converters (ADCs) with "prompt-" and "delayed-" gates. The "prompt gate" was 40 ns in width and covered the fast component of the anode signal. The "delayed gate" with a width of 350 ns was delayed by 150 ns from the prompt gate to collect charges of the decay component. Events induced by various particles in an NE213 organic scintillator were classified into a few types on the basis of the relationship between the fast and decay components of the PMT light output.

Figure 2 shows the block diagram of electronic circuits for the PSD. The NE102A plastic scintillator performed as a veto counter for neutron measurements was located in front of the NE213 liquid scintillator, whereas for the charged particle detection it was regarded as a ΔE counter coincident with the liquid scintillator. The NE213 was 12.7cm in diameter and 12.7cm thick and coupled with a Hamamatsu H1250 PMT. The NE102A was 18cm long and 1cm thick and connected with a Hamamatsu H1949 PMT. The anode signal of the PMT for the NE213 was branched into four by a signal divider (Div.). One of them was converted into a logic pulse by a constant fraction discriminator (CFD 2) and then sent to the coincidence module (Coin.), and the others were transmitted to the charge sensitive ADCs (ADC 1-3) through delay lines (Delay). The signals of incident proton beam monitor were converted to the time-of-flight (TOF) gate signals with a time duration of 150 ns at the gate generator (G.G. 1). When logic pulses from the CFD 2 for the E detector coincided with the TOF gate signal, the trigger signals were sent to G.G.2-4 in order to make gate signals for ADCs. ADC 1 and 2 worked for the PSD with the prompt and delayed gates and ADC 3 was used for determining a threshold level of neutron measurements with the total gate which was 250 ns in width. The anode signal of PMT for the NE102A was sent.
3. RESULTS AND DISCUSSIONS

Before treating the PSD data, we preliminarily carried out the particle identification by two other methods. First, the identification was made by the $\Delta E$-$E$ method and the results are plotted in Fig. 3. It is shown that proton and pion distributions indicate folds around 400 ch in the ordinate. In the $\Delta E$ range above 400 ch, protons and pions are clearly separated since they stopped in the NE213. In the region below 400 ch, high energy particles are supposed to escape from the E detector of the NE213 without depositing their full energies. In addition to the $\Delta E$-$E$ method, we looked into the distribution of the total charge versus the TOF of the NE213. The results are shown in Fig. 4, where vertical and horizontal axes correspond to ADC 3 channels for the total gate and TDC ones for the TOF, respectively. Since the coincidental events of $\Delta E$ and E detectors were chosen, charged particles dominated Fig. 4. However, gamma-rays had a chance to make Compton scattering or pair creation in the $\Delta E$ detector to emit electrons toward the E detector. For
this reason, flash gamma-rays exist around 3300 ch of the TDC in Fig. 4. There is a time-walk effect of the CFD as a curve of the flash gamma-ray distribution. In Fig. 4, proton and pion distributions show folds, again because of the charged particles escape as mentioned above. Proton and pion energies at the folds are obtained by the TOF with corrections, and they were evaluated as 120 and 60 MeV, respectively. In both Figs. 3 and 4, folding and overlapping in distributions disabled us in identifying the particles of partial energies deposition.

Following the particles identification in Figs. 3 and 4, we analyzed the PSD data by the two-gate integration method. Fig. 5-a shows the PSD results for events in which particles completely stopped in the NE213 and deposited their full energies as indicated in the TDC region.
of (A) in Fig. 4. One can see that the distributions were non-linear. In the region from 100 to 400 ch of the prompt gate ADC, the decay component of the light output in the delayed gate ADC decreases with the fast component. This is ascribed to the effect of a reflected pulse generated in cable circuits. The reflected pulse had a very small amplitude and reversed polarity. Its delay time was about 150 to 200 ns and it appeared on the decay component of the primary pulse. The reflected pulse was received without saturation effect in the amplifier unlike the primary pulse, and the reduction effects of the decay component occurred. The decay component rapidly increased with the fast one above the fast gate ADC channels of 500. This is because the saturation effects occurred in the PMT itself for high energy particles and were more influential on the fast components than on the slow ones of the signals. The discrimination results, however, are sufficiently good including pions. It is found that the two-gate integration method is applicable to identifying charged particles which completely stopped in the detector. When only the ΔE-E method is employed for the charged particles identification, ambiguities still remain due to the poor resolution in the ΔE detector. We consider that the PSD in addition to the ΔE-E method make the charged particles identification more accurate.

On the other hand, Fig. 5-b shows the PSD results for events in the region of (B) in Fig. 4 where particles escaped from the detector and deposited energies were relatively small. Proton and pion distributions were shifted to the electron one, and the particles identification was unable to be made. This is because escaped particles took out most of their energies and induced ionization of a lower density. The decay component of light output was reduced and the discrimination was unable to be made.

4. SUMMARY

We identified the charged particles by PSD with the two-gate integration method with the NE213 liquid scintillator. When charged particles deposited all their energies and completely stopped in the detector, the discrimination was sufficiently good and pions were demonstrated to be identified. On the contrary, in the case that incident particles escaped from the detector and most of their energies were taken out, they were unable to be distinguished and the discrimination was unable to be carried out.

References

(1) F. D. Brooks, Nucl. Instrum. Methods 4, 151 (1959)
Fig. 5-a Results of the PSD with the two-gate integration method. Incident particles completely stopped in the NE213. Distributions of proton, pion and electron are clearly separated from each other.

Fig. 5-b Results of the PSD with the two-gate integration method for escaped particles from the NE213. It is shown that proton and pion distributions are shifted to electron one, and the particle identifications are not clear.
BACKGROUND SIGNALS FROM BGO SCINTILLATORS—ORIGIN OF RADIOISOTOPIC IMPURITY OF $^{207}\text{Bi}$—

Research Institute for Advanced Science and Technology,
University of Osaka Prefecture, 1–2 Gakuen-cho, Sakai,
Osaka 593, Japan

A lead concentrate (PbS), which contains from 0.30 to 0.35% of bismuth, has been measured for gamma emitting radionuclides by a Ge detector. The Ge detector was surrounded by the 9.1 kg concentrate, and they were placed in a heavy lead cave. The gamma-ray spectra showed the presence of weak radioactivity of $^{207}\text{Bi}$ ($T_{1/2} = 32.2\text{y}$) at a level of 2.0 mBq/Bi–g. Besides $^{207}\text{Bi}$, the fission product $^{137}\text{Cs}$ ($T_{1/2} = 30.0\text{y}$) was detected in the concentrate. The $^{207}\text{Bi}$ activity may be ascribed to the nuclear explosion tests which terminated by early 1960’s. The concentrate was prepared through the flotation process at a concentrating mill. One of the origin of the contamination could be from water used in the flotation process.

1. INTRODUCTION

Radioactivity in the constitution materials of radiation detector system causes some background signals. The specification of low quantities of radioactivity is important in its application involving radiation sources of low activity, because the magnitude of the background ultimately determines the minimum detectable radiation level. The radioactivity of the constituent materials, which are detector itself, shielding, supports and ancillary equipment, is due not only to natural radioactive elements contained as an impurity but also to artificial ones.

Bismuth germanate (BGO) scintillators show appreciable background from $^{207}\text{Bi}$. Figure 1 shows a background spectrum measured by a 5–cm diameter by 3–cm thick BGO scintillator. Two photopeaks at 569 and 1063 keV are from $^{207}\text{Bi}$, and one at 1460 keV is due to $^{40}\text{K}$ contained in a photomultiplier and surrounding materials. The counting time was 60 ksec. The insert is the decay scheme of $^{207}\text{Bi}$. It decays with a half–life of 32.2 years by electron capture to $^{207m}\text{Pb}$ and $^{207}\text{Pb}$ $^\beta^\text{+}$. The $^{207}\text{Bi}$ nuclide can be found in commercially available bismuth chemicals and metal recently produced. It does not form part of any natural decay chain. It must be a cosmogenic or artificial radioisotope. At present bismuth is manufactured as a by–product of lead smelting and refining. Lewis has suggested that $^{207}\text{Bi}$ is produced by cosmic–ray proton
transmutation of $^{206}\text{Pb}$ in lead contained in the same ore $^2$.

However, our previous paper showed the transmutation of lead isotopes by cosmic-proton bombardment cannot explain $^{207}\text{Bi}$ activity observed in bismuth chemicals and metal $^3$. On the other hand, $^{207}\text{Bi}$ was detected in environmental samples, and was studied in relation to the radionuclides produced by nuclear explosion tests $^4$.$^{10}$. Radioisotopic contamination, such as $^{207}\text{Bi}$ in bismuth chemicals and metal, is more troublesome than radiochemical contamination because decontamination is practically impossible. The aim of this paper is to clarify the origin of the contamination and how to protect bismuth products from the contamination.

2. SAMPLES AND METHODS

Samples used for the measurement of radioactivity are lead concentrates. In the recovery of lead from its ores the metal is first concentrated and then smelted and refined. Lead ores are treated at concentrating mills at or near mines. For sulfide ores (galena) the flotation process is generally used. The lead concentrates then shipped from the concentration mill to the smelter contain 60 % or more lead. Smelting produces in blast furnaces a semifinished product known as base bullion, which contains bismuth and other useful metals. For refining lead an electrolytic method is particularly useful for the bullion containing high concentration of bismuth. In this method pure lead is collected in a cathode, and bismuth is recovered from the lead bullion as anode slime. The electrolytic method is most popular in Japan, where lead concentrates used for
manufacturing lead have been imported from a few countries since 1960's.

The sample (concentrate) was gamma-assayed by a HP Ge detector with a relative efficiency of 25% and a resolution of 1.9 keV at 1332 keV. A schematic diagram of the detector system is shown in fig. 2. The sample was packed in a plastic vessel of 13 x 23 x 16 cm$^3$. One side of the vessel (13 x 23 cm$^2$ face) has a well of 8 cm in diameter and 8 cm in depth, in which the head of the Ge detector is inserted. A packed sample weighed about 9.1 kg. The detector and the sample chamber were placed in a lead cave of 10 cm in thickness. The inside of the cave was lined with 1-cm thick copper plates.

![Fig.2 Schematic diagram of the detector system: lead shield, copper shield, sample chamber, Ge detector.](image)

The gamma-ray spectra of the concentrate were accumulated for $10^3$ ksec. The measurement was made three times with the same sample. Figure 3 is an example of the spectrum showing a number of peaks from radionuclides in the concentrate. The principal sources of activity in the concentrate are the primordial emitters, $^{238}$U, $^{232}$Th, $^{235}$U and their daughters and $^{40}$K.

![Fig.3 Gamma-ray spectrum of the lead concentrate of 9.1 kg. The counting time was 1000 ksec. The inset shows the effective sample weight in the energy region from 300 to 1400 keV.](image)
These peaks from the concentrate were utilized to determine the efficiency of the detector system shown in Fig. 2. Gamma-ray intensity of each peak from the small amount of the concentrate (60 g) placed just ahead of the detector was obtained. The sample was 3.1 cm in diameter and 3.2 cm in thickness. Calibration was made by point–like standard sources such as $^{60}$Co, $^{54}$Mn and $^{137}$Cs. Correction for the effect of self–absorption and bulky sample were made. The intensity was compared with that obtained by the 9.1 kg sample. The inset of Fig. 3 shows effective sample weight for the 9.1 kg sample as a function of gamma-ray energy in the energy region from 300 to 1400 keV. The effective sample weight is defined as the small sample weight multiplied by the ratio of the gamma-ray intensity of the 9.1-kg sample to that of the 60-g sample. Three regions of interest (ROIs) in the present work are indicated with arrows around 569, 662 and 1063 keV in Fig. 3. We anticipated the detection of $^{137}$Cs (662 keV) and $^{207}$Bi (569, 1063 keV) caused by nuclear explosion tests.

3. RESULTS AND DISCUSSION

Figures 4 and 5 are partial gamma–ray spectra showing the 661.6 and 1063 keV regions of interest. These energy regions are associated with detection of $^{137}$Cs and $^{207}$Bi. As an index for detection limit, $2\sigma$ is shown in these figures, where $\sigma$ is the standard deviation of background counts in ROIs. In Fig. 4 a small peak is seen at 662 keV. There are three possible radioactive sources for the peak, which are $^{137}$Cs (661.66 keV), $^{214}$Bi (660.75 keV) and $^{234}$Pa (660.60 keV). A sample, which was produced from another mine, was gamma–assayed with the same detector system. The concentration of $^{238}$U, $^{223}$Th, $^{235}$U and their daughters was same, but no peak was found at 662 keV. We may say that $^{214}$Bi (660.75 keV) and $^{234}$Pa (660.60 keV) do not contribute to the peak at 662–keV in Fig. 4, because $^{234}$Pa and $^{214}$Bi are daughters of $^{238}$U. The peak at 1063 keV in Fig. 5 may be from $^{207}$Bi. Specific activity of $^{207}$Bi was calculated to be from 2.0 to 1.7 mBq/Bi-g according to the concentration of bismuth in the sample. The value agrees with the one in our previous report. (Ingot 3 of reference 3 was made from the concentrate used in the present work).

Global contamination with $^{207}$Bi in environmental samples has been noticed by several authors. They also determined $^{60}$Co, $^{125}$Sb and $^{137}$Cs in the sample. The concentration of these three radionuclides have been compared for source identification. Yoshihara have summarized experimental data and estimated possible routes forming the $^{207}$Bi contamination, in which the radioactivity ratio of $^{207}$Bi/$^{137}$Cs was from 0.0001 to 0.01. A calculated ratio from our data shown in Figs. 4 and 5 is about 0.5. However, this ratio contains a statistical error of about 100 %, so we cannot say about the detail of the nuclear reactions producing $^{207}$Bi.

The concentrate used was made from the ore which extracted from a underground mine.
In the flotation process, finely crushed ore was diluted with about four times as much water and small amounts of organic reagents for making forth. In a flotation cell froth containing most metallic constituent of the ore is formed on the top of the mixture. The forth flows from the flotation cell and is dried to make concentration. The contamination of $^{207}$Bi possibly comes from this process. Water used for the process is considered to be one of the origin of the contamination.

![Figure 4](image1.png)  
**Fig.4** Gamma-ray spectrum of the lead concentrate showing the 661.6 keV region of interest. The counting time was 3000 ksec.

![Figure 5](image2.png)  
**Fig.5** Gamma-ray spectrum of the lead concentrate showing the 1063 keV region of interest. The counting time was 3000 ksec.

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RECENT ADVANCES IN LOW-LEVEL $\beta$-COUNTING
BY LIQUID SCINTILLATION INSTRUMENTATION

Yuriy Verzilov, Fujio Maekawa, Yukiyo Oyama and Hiroshi Maekawa
Japan Atomic Energy Research Institute, Department of Nuclear Engineering

1. INTRODUCTION

Liquid scintillation counting is certainly the most versatile and most frequently applied technique for quantitative determination of radioactivity in inorganic material. The main application of this technique lies in the detection of low energy $\beta$-particles from radionuclides such as $^3$H, $^{14}$C and others which by other methods may be measured only with much lower efficiency and accuracy. In addition, the technique applies for high energetic $\beta$-particles and if radioactive nuclei exhibit $\beta$-emission above 263 keV, Cherenkov response may be used for the measurement of activity by using the liquid scintillation counting equipment.

Due to the basic principle of liquid scintillation and Cherenkov counting it is necessary to bring the radionuclide in close contact with the scintillator molecules or liquid media. It means that solid radiochemicals must be dissolved or radionuclides must be extracted from solid target with subsequent incorporation the radioactive liquid phase into the liquid scintillator or media. The extraction technique is rather difficult for routine use because a complicated procedure is needed. The dissolving technique is less laborious and the same time the figure of merit (FOM) is below, because of higher solubility limitation for the material. The FOM, the product of the sample solubility and the counting efficiency, denotes the capability for measurement of low level activity of radionuclide. Therefore, the dissolving technique was the subject of interest to increase its FOM and to simplify preparation procedure.

This work attempts to solve these problems for measurement $^3$H and $^{32}$P activity. A measurement of tritium activity in materials is desired for neutron experiments such as a fusion reactor blanket experiment$^{(1,2)}$ and a neutron flux monitor with $^6$Li(n,\alpha)$^3$H reaction$^{(3)}$. In such studies, lithium-containing pellets are usually irradiated by neutrons and the tritium produced in them is measured with a liquid scintillation counter. Tritium technique for Li$_2$CO$_3$ is the subject of interest in the present study because this compound is well soluble and easy to use it. The $^{32}$P is important nuclide in the in-system neutron spectrometry due to the following reactions: $^{31}$P(n,$\gamma$)$^{32}$P, $^{32}$S(n,p)$^{32}$P and $^{35}$Cl(n,\alpha)$^{35}$P. For the reasons of similarity with Li$_2$CO$_3$, NH$_4$PH$_2$O$_2$, 

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CH\textsubscript{3}SO\textsubscript{2}CH\textsubscript{3} and NH\textsubscript{4}Cl were selected for measurement of \textsuperscript{32}P activity.

2. TECHNIQUE FOR TRITIUM ACTIVITY MEASUREMENT IN Li\textsubscript{2}CO\textsubscript{3}

2.1. Outline of Conventional Technique

According to the Dierckx's technique\textsuperscript{(2)}, see fig.1, the maximum weight to be solved in solvent is about 0.7 g of Li\textsubscript{2}CO\textsubscript{3} for the 20 ml volume of standard scintillation vial, by diluted acetic acid. The reaction process takes ~1 day. When the solution becomes clear, 0.7 ml of hydrofluoric acid (40\%) is added to precipitate lithium fluoride. This step is necessary to ensure good miscibility with the liquid scintillator which is then added. The vial is closed and centrifuged. When the LiF forms the white layer on the bottom of the vial, the sample is ready for counting.

This technique has the following disadvantages. The pellet solubility in the diluted acetic acid is not so high; about 20 g of Li\textsubscript{2}CO\textsubscript{3} per 100 ml of the solvent. For comparison, 50 g of Li\textsubscript{2}CO\textsubscript{3} can be dissolved in 100 ml concentrated nitric acid. The remaining lithium acetate is liable to make the scintillation solution unstable and to reduce the tritium counting efficiency.

To overcome these disadvantages a new dissolving technique has been developed.

2.2. New proposal of Technique

2.2.1. Experimental procedure

The present sample processing scheme for scintillation counting is shown in Fig.2. It is very simple and does not require to remove lithium salt from the solution. A mixture of two different acids, HNO\textsubscript{3} (61\%) and CH\textsubscript{3}COOH (100\%), strong and weak, has been proposed as a new pellet solvent from the preliminary study and the
following view point.

In order to provide high solubility of lithium salt it is best to use an acid. Since the acid may be the strong quencher, an experiment was carried out to examine the influence of the acids incorporated into the scintillator upon the tritium counting efficiency. The quenching of light output of the cocktail, Clear-sol, was investigated by incorporation of weak and strong acids. The measurements of counting efficiency were carried out in 20 ml volume Teflon vials. The counting samples were obtained by adding acid volumes to 10 ml of cocktail, because variations in scintillation volume gave an insignificant effect on counting efficiency in the range from 8 to 17 ml. Counting efficiency was obtained by using internal standard method. Figure 3 shows the relation of counting efficiency to the sample content for different solvents. For reference, a quenching effect of water is also plotted in the same figure. From the figure it is seen that the strong acid, HNO₃, leads to a rapid decrease in counting efficiency. Therefore, first the amount of the strong acid that is not enough to dissolve the whole pellet should be dropped, and the remained part of the pellet can be dissolved by the acetic acid.

Seven solvent compositions with the different acid-volume ratios were prepared. The volume of each acid for dissolution of 1 g Li₂CO₃ is shown in Fig.4. The first solvent contained only the nitric acid. Its volume was estimated by chemical equation. The others contained the mixture of two acids varying the mixing ratio. These solvents were prepared by gradually reducing the volume of nitric acid and by adding the acetic acid until the pellet was dissolved completely. From the final volume of solvent, the solubility of lithium salts was evaluated.

The counting efficiency for
the mixture of pellet solution and scintillation cocktail, and the compatibility of the solution with the cocktail were examined with respect to the acid-volume ratio and the pellet weight. For this purpose three set of samples with the different pellet weights, 0.4 g, 0.8 g and 1.4 g, were prepared. Each set was obtained by dissolving the Li$_2$CO$_3$ pellet into 10 ml of the cocktail with the acid solvents of seven different acid-volume ratios. After shaking the sample thoroughly, a visual assessment of transparency was made for them.

The comparison was also made in terms of figure of merit, FOM. The FOM denotes capability for measurement of low level tritium activity produced in the pellet and defined as the product of pellet weight and counting efficiency: FOM = $\epsilon$M, where $\epsilon$ is the counting efficiency (%) and M the mass of pellet (g). The FOM and the sample compatibility were used to determine the optimum acid volume ratio. Then the maximum pellet weight to be solved in the 20 ml standard vial for scintillation counting was determined for the solvent with the optimum acid volume ratio. For the latter procedure, the counting efficiency of the pellet solution-cocktail mixture and the compatibility were determined with respect to the dissolved pellet weight.

2.2.2. Results of New Technique

Based on the amounts of acids which were obtained for complete dissolution of the samples, the solubility of Li$_2$CO$_3$ was evaluated. The obtained solubility are shown in Fig.4, according to traditional expression in grams of dissolved substance per 100 ml solvent$^7$. For comparison, the value of Li$_2$CO$_3$ solubility in the diluted acetic acid, i.e., by Dierckx's method, is also plotted in the same figure. From the figure one can see that the influence of solvent composition upon the solubility of Li$_2$CO$_3$ shows a smooth dependence, and a variation in solubility has an insignificant effect on FOM.

From comparison of seven samples with different acid-volume ratios, the effect of the different solvent on counting efficiency was examined. In comparison, the pellet weight was chosen to 0.4 g so as to obtain clear fluid after incorporation the pellet solution into the liquid cocktail. Figure 5 shows that the influence of the acid-volume ratio on the counting efficiency drops rapidly for the nitric acid rich

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Fig.5 Effect of the chemical composition of solvents on pH value, counting efficiency and sample compatibility.
samples because there exists the free nitric acid in samples. This is supported by pH values plotted in the same figure and by the relation between counting efficiency and strong acid content (see Fig.3). On the other hand, in an excess of acetic acid the counting efficiency decreases rather gradually.

The optimum acid-volume ratio was determined by FOM and the sample compatibility in relation to the acid-volume ratio and the pellet weight. A comparison between the two sets of the samples with different weights of 0.8 g and 1.4 g in terms of FOM was shown in Fig.6. The optimum is obtained by the highest FOM value within the clear fluid region due to good compatibility.

The counting efficiency in liquid scintillation counting decreases with increase of weight (impurity quench) and the sample compatibility impairs with increasing amount of the pellet solution in the cocktail. By a series of samples with the optimum chemical compositions for the different pellet weights the maximum weight of the lithium salt that can be solved was determined experimentally. The maximum weight was obtained from the sample which has the highest FOM value within the clear fluid region with good compatibility. Figure 7 shows the results for Clear-sol.

Numerous experiments were carried out to examine the stability of the pellet solution and cocktail mixture, i.e., the change of counting efficiency with time. The stability of the optimized solution-cocktail mixture was excellent and no time-dependence was observed in counting efficiency over one month. Reproducibility of counting characteristics for the replicated mixtures with the same composition was also excellent within the accuracy due to statistics of the counts, in addition to the initial pipetting and the long-term stability of the counting system.
2.2.3. Comparison with the Conventional Techniques

A comparison of the capability for measurement of low-level tritium between the present and the conventional techniques was made by the FOM. The higher FOM denotes the capability for measurement of the lower level tritium activity. The main parameters of the conventional techniques, i.e., Dierckx method, were taken from the table in the reference\(^2\). They are summarized in Table 1 together with the calculated FOM. Though these values are, of course, based on the instruments, the instrument factor of the counting efficiency was assumed to be equal because all the instruments can be considered to be similar. Also there is a ambiguity in the conventional methods listed in the table because they do not all were optimized.

Table 1. Comparison of measuring techniques for \(\text{Li}_2\text{CO}_3\) pellets.

<table>
<thead>
<tr>
<th>Organization &amp; technique</th>
<th>Pellet weight (g)</th>
<th>Counting efficiency (%)</th>
<th>Figure of merit (g-%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Direct incorporation of the pellet solution into the scintillation cocktail</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>CEA/Cadarache Dierckx' (2)</td>
<td>0.7</td>
<td>26.6</td>
<td>18.6</td>
</tr>
<tr>
<td>IGA/EPFL Dierckx' (2)</td>
<td>0.7</td>
<td>20.6</td>
<td>14.4</td>
</tr>
<tr>
<td>ENEA Dierckx' (2)</td>
<td>0.7</td>
<td>22.7</td>
<td>15.9</td>
</tr>
<tr>
<td>Osaka Univ. Dierckx' (2)</td>
<td>0.5</td>
<td>18.6</td>
<td>9.3</td>
</tr>
<tr>
<td>Tokyo Univ. Dierckx' (2)</td>
<td>0.6</td>
<td>29.8</td>
<td>17.9</td>
</tr>
<tr>
<td>JAERI Present tech.</td>
<td>1.4</td>
<td>21.7</td>
<td>30.4</td>
</tr>
<tr>
<td>Incorporation of the pellet solution into the scintillation cocktail after removing lithium salts by cryo-distillation</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>MEPhI Technique (5)</td>
<td>2.0</td>
<td>16.0</td>
<td>32</td>
</tr>
<tr>
<td>JAERI Present tech.</td>
<td>5.7</td>
<td>21.1</td>
<td>120</td>
</tr>
</tbody>
</table>

In contrast to the conventional techniques, all preprocessings of the present one are performed in a standard scintillation vial, without removal lithium salts. Thus, the present...
technique is very simple and convenient, and at the same time it has higher FOM. In addition, the use of cryo-distillation to remove of lithium salts from the solution makes it possible that the FOM increases more. However, the cryo-distillation requires a special equipment and longer preparation time. From Table 1 one can see that the present technique can improve the FOM by a factor of ~ 2, while the technique with the additional process by cryo-distillation does the FOM by a factor of ~ 4.

3. TECHNIQUE FOR PHOSPHORUS-32 ACTIVITY MEASUREMENT IN NH₄Cl

3.1. Outline of Technique

Since a large number of radioactive nuclides exhibit β-emission above 263 keV, the threshold energy for the excitation of Cherenkov radiation in water, Cherenkov response may be used for the measurement of β-activity. Remarkable advantages of this technique are extreme simplicity of sample preparation, the ability to count in aqueous systems without the use of organic fluors. A comparison between the sample processing schemes for Cherenkov radiation counting and liquid scintillation counting is shown in Fig.8. The main disadvantage is low detection efficiency as compared with liquid scintillation counting. All the decay events cannot be detected, because there is a finite number that fall below the Cherenkov threshold. The maximum theoretical detection efficiency in water for ³²P can be calculated by integrating the number of events above 263 keV and comparing this to the total emission. This value is about 86%, and the experimental detection efficiency is about two times less than the theoretical result when the ordinary liquid scintillation counting equipment is used.

In the present study, much attention was given to improve detection efficiency for aqueous solution of NH₄Cl. This improvement is based on some properties of Cherenkov radiation. Classical theory of this phenomenon indicates at first that a significant portion of the emission occurs in the ultraviolet spectral region and at second it has a highly directional light.
If the ultraviolet photons can be converted to the visible region and if these photons could be given an isotropic spacial distribution, the counting efficiency should be distinctly improved. The use of Triton X-100 makes it possible to increase counting efficiency due to wavelength shiftier effect and the use Teflon vial also increases efficiency due to the diffusing effect.

3.1. Efficiency of Cherenkov radiation in relation to vial type and concentration of aqueous Triton X-100

The highly directional nature of Cherenkov emission explains why the typical liquid counter gives lower counting efficiency when used in the coincidence mode. It has the phototubes at angles of 180°. Thus, when directional sensitivity is highest in one direction, the 180° sensitivity is at a minimum. Therefore, it is useful to use the vial with the frost walls.

Cherenkov measurements of ³²P activity were made using three different vials glass, polyethylene and Teflon. Counting efficiency was obtained by adding 1 ml of standard solution of ³²P to a vial containing the aqueous solution of NH₄Cl though variations in solution volume gave an insignificant effect on counting efficiency in the range from 12 to 17 ml. In order to eliminate the effect of the absorption of ³²P onto vial walls, the 60 mg of inactive NH₄PH₂O₂ carrier was dissolved in the 15 ml of water before the dissolution of the sample. The obtained results are shown in Fig.9. From figure one can see that the counting efficiency for Teflon vial is almost 20% higher that of glass vial. For comparison, the instrumental figure of merit, IFOM, is plotted in the same figure. This value denotes capability for measurement of low

![Fig.9 Comparison of Instrument Figure of Merit for measurement of ³²P by using the different types of vial.](image)

![Fig.10 Effect of concentration of aqueous Triton X-100 on ³²P counting efficiency.](image)
level $^{32}$P activity in the aqueous solution and defined as the product of counting efficiency and background level: IFOM = $\varepsilon^2B$, where $\varepsilon$ is the counting efficiency (%), and $B$ the background level (cpm). Figure 9 shows that the instrumental figure of merit for Teflon is almost twice that of glass.

The use of waveshifters in Cherenkov counting continues to be the most important method for increasing total light output. Along the tens of compounds which used as waveshifting agents, Triton X-100 was selected because the aqueous Triton X-100 medium was not affected by solutions of interest. The influence of the concentration of aqueous Triton X-100 on counting efficiency of $^{32}$P is shown in Fig. 10. This dependence has the comparatively smooth maximum. Therefore the optimum concentration of aqueous Triton X-100 was selected within clear fluid region because it is easy to form homogenous samples. Optimum concentration of aqueous Triton X-100 (34%) increases $^{32}$P counting efficiency by a factor about 1.3 as compared to the conventional medium, water.

3.2. Comparison with Liquid Scintillation Counting Technique

Consequently, the counting efficiency of $^{32}$P in the Teflon vial filled with aqueous Triton X-100 (34%) was 66%, or 1.5 times as large as that in the glass vial filled with pure water. Although this is lower than the efficiency of liquid scintillation counting the FOM are much larger, because the volume of the aqueous sample for counting is much larger. Figure 11 shows comparison of FOM for measurement of $^{32}$P in irradiated pellet of NH$_4$Cl after dissolution pellet in water. From the figure one can see that Cherenkov radiation counting can improve the FOM by a factor of ~ 3.

4. SUMMARY

New binary-acid solvent for dissolving of lithium salt, Li$_2$CO$_3$, was developed. The utilization of combination of the new solvent and the scintillation cocktail containing surfactant created a new efficient technique for measuring the tritium activity in pellets. The developed
technique has high solubility of a sample, good compatibility with scintillation cocktail and small quenching in scintillation counting. This technique is also very simple because all the operations are carried out in a scintillation vial. Therefore, the present method brings excellent improvement for measurement of low level tritium activity produced from lithium containing materials, especially from a $^7\text{Li}$ enriched sample in a weak neutron field. In addition, the method developed here for lithium carbonate can be extended to other lithium salts.

The technique which utilized the principle of direct Cherenkov radiation counting of energetic $\beta$-rays in an aqueous medium is ideal for the measurement $^{32}\text{P}$ activity. It offers a useful and convenient adjunct to the method of liquid scintillation. Compared to liquid scintillation counting, direct Cherenkov radiation counting has particular advantages. Sample preparation is easy, quick and cheap, since no scintillators are used, and chemical quenching does not occur. The use of Teflon vial filled with aqueous Triton X-100 (34%) increases the detection efficiency and also makes this method attractive for the determination of relatively weak $\beta$-emitters.

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A BEAM-ENERGY CHECKING DEVICE FOR ELECTRON BEAMS FROM RADIOTHERAPY ACCELERATORS

Takahiko Aoyama, Hisashi Maekoshi, Masatoshi Tsuzaka and Shuji Koyama

Nagoya University College of Medical Technology
Daikominami, Higashi-ku, Nagoya 461, Japan

Satoru Kondo, Yuichi Aoyama and Shinji Abe

Nagoya University Hospital, Tsurumai-cho, Showa-ku, Nagoya 466, Japan

1. INTRODUCTION

Small sized electron linear accelerators and microtrons have been used for electron beam therapy. Electron beam energy, which is usually between 5 and 20 MeV, must be adjusted exactly to focus the beam on the depth of the cancer in the human body. The incident energy of electron beams from radiotherapy accelerators has been estimated from depth dose measurements in water, where the electron range in water was used as the measure of the electron energy. This is because electron spectroscopy based on the direct energy measurement of a single electron could never be carried out because of the bunching feature of the beam, which consists of a train of short pulses, and a high flux density of electrons on the order of $10^6 \text{ cm}^{-2} \text{ s}^{-1}$. Although the depth dose measurements were adopted as a standard method for the estimation of the beam energy, it was rarely executed since it was time consuming and troublesome. Since beam-energy check should be carried out in routine for quality assurance purposes of the beams, alternative simple methods were studied and developed in recent years.

Johnsen\(^{(1)}\) used a pair of ion chambers and a circular wedge absorber to monitor electron beam energy, where the absorber made of spherical segment of copper was sandwiched between the ion chambers. For this construction of the detector the ratio of the ion current for the downstream chamber to that for the upstream chamber depended on the beam energy alone. Another idea which uses the depth dose concept was introduced by Geske\(^{(2)}\). He used multi-layers of parallel plate ion chambers inserted separately in multi-layers of aluminum-plate absorbers. Using this construction of the detector the sum of charges collected from each chamber at a constant dose was depended on the beam energy alone. The device now commercially available is complex and expensive, and it was found to have nonlinear energy response\(^{(3)}\). On the other hand, Das et al.\(^{(4)}\) used backscattering and transmission of electron beams through a high atomic number material as a measure of beam energy. When a lead plate was sandwiched between a pair of identical ion chambers, current ratio of the chambers depended on the beam energy alone and changed exponentially with beam energy.
The present paper describes the development of a beam-energy checking device based on the Johnsen method. The reason of which is the simplicity and the linear energy response of the device. Although Johnsen used ionization current and integrated charges for output signal of the ion chambers, we used pulse signal derived from the chambers in addition to the average ion current. Pulse characteristics of the electron beams from radiotherapy accelerators could be studied using this pulse readout from the chambers.

2. CONSTRUCTION AND ARRANGEMENT

Fig. 1 shows a schematic diagram of apparatus. The detector part of the device consists of a circular wedge absorber sandwiched between a pair of circular shaped parallel-plate ion chambers. Circular symmetry was adopted for the detector because of the axial symmetry of the beams produced with most radiotherapy accelerators. The detector was set on a Lucite board, a low atomic number material, with a thickness of more than 10 cm - i.e. electron range for 20 MeV electrons in Lucite, and it was irradiated with an electron beam along the detector axis on the center of the top ion chamber from upper side. Irradiation field was limited to 80 mm in diameter by using a metal cylinder attached to the beam outlet of the accelerators.

The ion chambers were composed of two parallel-plate electrodes made of glass-epoxy printed circuit board, 1.6 mm thick, which were separated by 4.8 mm. Effective areas of collecting electrodes were 40 and 80 mm in diameter for top and bottom chambers respectively. The chambers were operated in open air with an applied voltage of 1000 V, since plateau current was observed between 500 - 1500 V at an atmospheric pressure and a room temperature for a constant beam energy and dose rate.

Electronics for pulse processing is shown in fig. 2. Since an electron beam from a radiotherapy accelerator consists of a pulse train, output pulses were derived directly from the chambers besides the average ion current, where the latter was measured using conventional micro-ammeters as shown in fig. 1. In fig. 2, output pulses derived from each chamber were fed through an emitter-follower pre-amplifier, a linear amplifier and a linear gate stretcher to a multi-channel analyzer (MCA) directly or through an analog divider. Pulse shaping time of 10 μsec was used for the Gaussian shaping type of the linear amplifiers whereas 50 μsec decay time was used for the pre-amplifiers. Linear accelerator used in the present experiment, Toshiba Mevatron 77, produced pulse electron beams with a pulse width of 2 μsec and a pulse interval of about 10 msec. Since drift times for electrons and ions in the ion chamber filled with air at an atmospheric pressure were estimated to be less than 0.3 μsec and more than 100 μsec respectively at an applied voltage of 1000 V, pulse height analyzed with the MCA would be due to the charges induced by the drift of electrons and partially by that of ions produced during a unit pulse-beam width of 2 μsec.
Fig. 1. Schematic diagram of the device.

Fig. 2. Block diagram of electronics for pulse processing.

Fig. 3. Typical penetration curve (a) for monoenergetic electrons, and the thickness-radius relation (b) of a circular shaped absorber.
The wedge absorber was made of either aluminum or copper. These absorbers were designed to have axial symmetry with a minimum thickness in the center and with increasing thickness from inside to outside until a diameter of 80 mm. Maximum thickness of the absorbers was adjusted to coincide with the electron range for 21 MeV electrons, i.e. 40 mm for aluminum and 12 mm for copper each.

For the arrangement of figs. 1 and 2 the bottom chamber measures the flux of electrons passing through the wedge and the top chamber serves to normalize the flux to incident beam flux. Since the bottom chamber is exposed only to that area where the thickness of the wedge absorber is less than the electron range, the ratio of the current from the bottom chamber to that from the top chamber would be a measure of the range, hence the energy, of the electron beam. If the area of the bottom chamber that is exposed is designed to be a linear function of the electron beam energy the ratio of the bottom chamber current to that from the top chamber will be linear with energy to a first approximation, which will be confirmed as follows.

Fig. 3(a) shows a typical graph of the number $n$ of monoenergetic electrons penetrating through slabs of varying thickness $x$ of absorbing medium, where $X$ in this figure is the maximum penetration depth. It has been confirmed that the ratio of the depth $X$ to the electron range $R$ calculated using the continuous slowing down approximation showed very little energy dependence with values of $X/R$ of 0.9 for aluminum and 0.8 for copper. The feature of this graph is that the electron number $n$ decreases monotonously with increasing slab thickness $x$ taking the local maximum $N$ at $x=0$ and the local minimum 0 at $x=X$. Since this feature can be expressed most simply by 3rd polynomial function of $x$, relative electron number $n/N$ can be approximated by the function

$$f(x) = \frac{n}{N} = 2 - \frac{x^3}{X^3} - \frac{x^2}{X^2} + 1 \quad (0 \leq x \leq X).$$

Since circular wedge absorbers have axial symmetry, the thickness $x$ of the absorber changes with the radius $r$ as shown in fig. 3(b). Flux density of an electron beam is designed to be uniform in the irradiation field for radiotherapeutic electron beams. Taking $r=r_x$ at $x=X$ we can express the relative intensity of electron beams penetrating the wedge absorber in the form

$$I = \int_0^{r_x} f(x) 2\pi r dr.$$ (2)

If we take cone for the shape of the absorber as the first example, $x=cr$, where $c$ is a constant, and $r_x=X/c$. Substituting the above relation into eq. (1), and calculating eq. (2), we obtain the intensity $I$ proportional to the square of the electron range:
Since the range $R$ is known to be proportional to electron energy $E_e$ above 2 MeV\(^{5}\), energy response of the device would approximate to a square function for the cone type absorber.

As the second example, if we take a parabolic surface of revolution for the shape of the absorber, $x=kr^2$, where $k$ is a constant, and $r_x=\sqrt{X/k}$. Substituting the above relation to eq. (1), and calculating eq. (2), we obtain a linear relation between the intensity $I$ and the electron range $R$:

$$I = \frac{\pi X}{2k} \approx R,$$

and hence the intensity $I$ proportional to the electron beam energy $E_e$. In this case the area of the bottom chamber that is exposed to an electron beam can be confirmed to be a linear function of the electron range and therefore of the beam energy as mentioned before.

To compare the difference of the energy response of the device with that of the shape of the absorbers we designed one cone and two parabola types of wedge absorbers as shown in fig. 4. Absorbers (a) and (b) in fig. 4 were made of aluminum, where (a) was the cone type while (b) was the parabola approximated by cones having two steps of inclination with angles of 45° and 60°. Absorber (c), made of copper, was also the parabola type but approximated by cones with three steps of inclination to improve the approximation.

3. EXPERIMENTAL RESULTS AND DISCUSSION

The ion chamber detector with one of the three different wedge absorbers described above was tested on the energy response for the irradiation of electron beams from the Toshiba Mevatron 77 linear accelerator. The ratio of the ion current from the bottom chamber $I_b$ to that from the top chamber $I_t$ is shown in fig. 5 as a function of electron-beam energy, where curves (a), (b) and (c) in the figure were respectively obtained using the absorbers (a), (b) and (c) in fig. 4. The beam energy was measured using the standard method previously described - i.e. depth dose measurements in water. It is seen from fig. 5 that in the case of the cone type of the absorber (a) nonlinear energy response was obtained at a low electron-beam energy. When the ion chamber was used with the parabola type of the absorbers (b) and (c), energy response became linear at the low beam energy. Best linearity was obtained in the case of (c) over the whole beam-energy range between 5 - 20 MeV, which is consistent with our expectation. It is seen
Fig. 4. Circular wedge absorbers of the cone type (a) and of the parabola types (b) and (c).

Fig. 5. Energy response curves obtained with different designs of wedge absorbers. Curves (a), (b) and (c) were obtained respectively using the absorbers (a), (b) and (c) in fig. 4.

Fig. 6. Pulse height distributions for the pulses from the top ion chamber.
from fig. 5 that the ratio $I_b/I_t$ could be extrapolated to zero at a beam energy of about 2 MeV. The reason of which is due to the stopping of low energy electrons in the glass-epoxy electrodes and Al-foil windows of the chambers. Total thickness of 0.99 g/cm$^3$ of the electrodes and of the windows corresponded to the range of 2 MeV electrons.

For radiotherapy accelerators beam-intensity is controlled to be nearly constant during beam-irradiation by using ion chambers installed in the beam outlet of the accelerators. To investigate the fluctuation of pulse-beam intensity during beam-irradiation, we measured pulse height distributions for the output pulses from the top chamber using the electronics shown in fig. 2. Fig. 6 shows typical results measured for the pulse-beam from the linear accelerator. It is seen from fig. 6 that pulse height fluctuated largely for each electron-beam energy.

Pulse height of the ion-chamber output pulses is proportional to energy loss $\Delta E$ in the chamber by a single pulse electron-beam. Defining $\delta E_i$, the energy loss in the chamber by a single energetic electron and $P$ the number of electrons contained in a single pulse-beam, we can express the energy loss $\Delta E$ in the form

$$\Delta E = \sum_{i=1}^{P} \delta E_i = P \cdot \overline{\delta E}.$$  \hspace{1cm} (5)

Since the single electron energy loss $\delta E_i$ is on the order of 1 keV - too small compared with the incident beam energy - it would largely fluctuate as known as Landau fluctuation$^{(b)}$. A large number of electrons contained in a pulse-beam on the order of $P \sim 10^8$, however, allowed the average energy loss $\overline{\delta E}$ to be nearly constant at each pulse-beam for a given beam energy. Hence the large fluctuation of the pulse height seen in fig. 6 would be due to that of the electron number $P$ contained in a single pulse-beam. The shape and the peak width of the pulse height distributions differed at each measurement for a given beam energy, indicating the instability of the beam intensity.

In fig. 7 another examples are shown in which double peaks and an extremely wide peak width were observed in the pulse height distributions. In fig. 7 also shown are pulse height distributions for the pulses from the bottom chamber. The distribution pattern is seen to coincide with that for the top chamber at a given beam energy.

Fig. 8 shows pulse height distributions for divider output pulses measured using the parabola type absorber (b) in fig. 4 for various electron beam energies. The energy of each electron contained in a single pulse-beam would fluctuate although the energy spectrum could never be measured. Sharp peaks seen in fig. 8 indicate that effective beam energy, which would be close to the average beam energy, was however constant for each electron-beam pulse. This might be due to an excellent constancy or stability of electron-energy spectrum at each beam pulse. Beam energy dependence of peak channels coincided with that of $I_b/I_t$ in fig. 5. Narrow FWHMs of the peaks seen in fig. 8 indicate that the error of the beam energy determination is extremely small within 0.1 MeV.
Fig. 7. Pulse height distributions for the pulses from the top and the bottom chambers. The cone type of the absorber was used in this case.

Fig. 8. Pulse height distributions for divider output pulses.
4. SUMMARY

A simple beam-energy measuring method for therapeutic electron beams was developed by using double ion chambers with a circular wedge absorber. Using this detector and a pulse processing system we studied the intensity fluctuation and the energy stability of electron-beam pulses from a radiotherapy linear accelerator. The results obtained are:

- By designing the shape of the circular wedge absorber to be a parabolic surface of revolution we could obtain a linear response of ion-current and pulse-voltage ratios to the beam energy.
- Pulse beam intensity fluctuated largely, and showed instability.
- Beam energy was, however, highly stable for each pulse.

Unification of the ion-chamber detector with a metal cylinder used for the collimation of electron beams would allow easy attachment and detachment of the detector to the beam outlet of the accelerators. Using the present device we can not only check electron beam energy but inspect the stability of the beam intensity and the beam energy. The present device would be used in routine for quality assurance purposes of therapeutic electron beams.

Acknowledgment

The authors would like to thank to Mr. Hiroshi Yamaguchi of Nagoya University Hospital for the approval to the use of the therapeutic linear accelerator. Thanks are also to Prof. Chizuo Mori of Nagoya University for his kind discussion and suggestion.

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RADIATION INDUCED ELECTRIC CONDUCTIVITY OF ALUMINA INSULATOR AND ITS APPLICATION TO RADIATION DOSIMETRY IN HIGH ENERGY AND HIGH FLUX NEUTRON FIELD

Yukio Oyama, Tetsuya Nakazawa, Kenji Noda and Hiroshi Maekawa
Japan Atomic Energy Research Institute
Tokai-mura, Naka-gun, Ibaraki-ken, 319-11 Japan

1. INTRODUCTION

In a controlled thermo-nuclear fusion reactor, instrumentation for reactor control is one of the issues to be developed. A number of components, such as plasma control magnet system, tritium breeding and energy conversion blanket, are fed back by detection system. Neutron detection is one of such key instrumentations. There have existed many neutron detectors proposed for fusion application, however, most of them are not applicable in fusion reactor, because detector environment in the reactor is too severe due to high neutron flux and high damage rate by high energy neutrons.

In a typical fusion reactor with 3 MW/m$^2$ wall load, neutron flux is expected to be $1.3 \times 10^{14}$ n/cm$^2$/s at the first wall that faced on the plasma region. This produces $2.6 \times 10^{-7}$ dpa/s for Si, i.e., 1 dpa for 2 months. This high damage rate by 14 MeV neutrons does not allow us to use semiconductors and sensors as functional materials for the reactor instrumentation. Neutron flux and spectrum is one of the most important information as a power monitor. Fusion power is basic quantity to evaluate reactor performance such as energy multiplication and tritium breeding. For this purpose, a foil activation system and a fission chamber are possibly considered as candidate detectors. However, these detectors are not satisfactory for requirement of real time response or wide dynamic range.

On the other hand, fusion radiation power can be measured by a calorimetric method in a full power range, but not in a lower range because of low sensitivity. However energy absorption dose is one of possible option for such a monitor, because the measurement of energy absorption in material is equivalent to calorimetric method. This view point reminds us of Solid State Electrical Conductivity Dosimeter, worked by
J.F. Fowler in 1959.\(^{(1)}\) He studied organic insulators for tissue dose together with photo-conductivity of CdSe. The ionizing radiation excites balance electrons to conduction band inside insulators. Concerning that the energy absorption by high energy neutrons contributes mostly to ionization, it is expected that neutron induced ionizing radiation inside insulators can be applied to a detector principle.

Here we tried to investigate alumina ceramics as such an insulator detector which is stronger against neutron radiation than plastics. That is also expected to have wider dynamic range because of higher resistance. The basic performance of radiation induced conductivity of alumina was examined for 14 MeV neutron field. From the results, feasibility for alumina insulator detector is discussed.

2. RADIATION INDUCED ELECTRIC CONDUCTIVITY

Electric conductivity of insulators is usually very low because it is due to ion conductivity. However, in ionizing radiation field, by excitation of balance electrons to conduction band, electric conductivity for insulators becomes larger and then can be written as follows:

\[
\sigma = n_e \cdot e \cdot \mu \quad \text{[ohm}^{-1}\text{cm}^{-1}] \text{, (1)}
\]

where \(n_e\) is the electron density in \([/cm^3]\), \(e\) the electron charge in \([C]\) and \(\mu\) the electron mobility in \([cm^2/volt\text{-sec}]\). The number of excited electrons is balanced by generation rate and disappearance rate;

\[
n_e = f \cdot \tau \text{, (2)}
\]

where \(f\) is the excitation rate of electron-hole pair \([\text{electron-hole pair/s/cm}^3]\) and \(\tau\) the average life \([s]\) of excited electron due to recombination. The relation of excitation rate can be connected to energy absorption dose, \(D\), as follows:

\[
f = \frac{D \text{ (released energy in unit time and unit volume)}}{W \text{ (energy required for production of one electron – hole pair)}} \text{, (4)}
\]

where \(W \equiv 3E_g\) (band gap). The energy absorption dose by neutron is almost equivalent to KERMA (Kinetic Energy Release in Matter) factor of neutron which is connected to total energy of charged particles and recoils produced by neutron nuclear reaction. For low energy neutron, the KERMA factor consists mainly of kinetic energy of recoil atoms by elastic reaction, while, for higher energy, contribution of energy of secondary emitted charged particles increases as shown in Fig.1 for alumina.\(^{(2)}\)
In the case of the absence of electron traps, average life of excited electrons is written as:

\[ \tau = \frac{1}{n_h \cdot v \cdot \sigma} \quad (5) \]

where \( n_h \) is the number of holes, \( v \) the electron velocity and \( \sigma \) the capture cross section of holes for electrons (~ \( 10^{-18} \text{ cm}^2 \)). When \( n_h = n_z = n \), by the eq.(2) and the eq.(5), \( f \approx n^2 \) and \( \sigma \approx \frac{1}{\sqrt{f}} \). Then, from the eq.(4),

\[ \sigma = k \cdot D^\delta \quad (\delta = 0.5) \quad (6) \]

On the other hand, in the case that the number of electron traps (capture centers), \( m \), is larger enough than that of holes, \( n_h \), the number of \( m \) dominates the recombination rate, in stead of \( n_h \). Hence, electrical conductivity is written as:

\[
\sigma = f \cdot \tau = \frac{D/W}{m \cdot v \cdot \sigma_z} = \frac{1}{m \cdot v \cdot \sigma_z \cdot W} \cdot D
\]

\[ = k \cdot D^\delta \quad (\delta = 1) \quad (7) \]

In this case, electrical conductivity under irradiation is in proportion to absorption energy rate. This relation for ceramics is discussed in more detail as material property research(3).

In eq.(7), an assumption that the number of electron traps is larger enough than that of holes may breaks for extremely high dose rate, because a production rate of electron-hole pair is high enough to keep the number of holes for recombination. Thus the dependence of \( \delta \) on the dose rate \( D \) may approach 0.5 with increase of dose rate. At the same time, since the number of \( m \) also may increase by accumulation of point defects with increase of fluence, a critical flux intensity for change of \( \delta \) could be changed with fluence. These conditions limit the dynamic range of the detector.

### 3. SELECTION OF INSULATOR FOR DOSIMETER

According to Fowler(4), a gain factor can be also estimated for alumina insulator dosimeter. The gain factor indicates how much current can the dosimeter observe for the given detector dimension. When \( A, V \) and \( L \) are the detector area, the applied voltage and the detector thickness, the observed current is written as:

\[
i = \frac{A \cdot V \cdot \sigma}{L} = \frac{A \cdot V \cdot \text{nef}^\tau}{L} = \frac{A \cdot V \cdot \text{nef}^\tau}{L} \quad (8)
\]
Substituting the following relations of mobility $\mu$ and electron transit time $T_\nu$ into the above equation (8),

$$\mu = \frac{v}{E} = \frac{v \cdot L}{V}, \quad \text{and} \quad T_\nu = \frac{L}{v \cdot \mu \cdot V},$$

and introducing the following parameters, total excitation $F$ and gain factor $G$,

$$F = f \cdot A \cdot L, \quad \text{and} \quad G = \frac{\tau}{T_\nu},$$

finally, the observed current is presented by:

$$i = e \cdot F \cdot \frac{\tau}{T_\nu} = e \cdot F \cdot G \quad \text{(9)}.$$

This representation shows that, in the case that the average life time of carrier electrons is large or that the mobility is small, the gain factor is large, i.e., observed current is large. However, when the detector applied to high flux, then, high dose field, the high gain factor is not absolutely necessary. In another word, the material with high gain factor has to loose a linear dependence on the dose rate as discussed in Sec. 2.

Assuming that $A=79$ [mm$^2$], $V=10$ [V], $L=0.3$ [mm], $D=1$ [Gy/s] for an alumina detector, and the values of Table 1, an estimation of observed current is less than a hundred nano ampere to the detector. The dose rate of 1 [Gy/s] corresponds to the 14 MeV neutron flux of $1 \times 10^{11}$ [n/cm$^2$-s]. This value is suitable for application of the detector to the flux in fusion reactors where the neutron flux could reach to a thousand times higher. The observed current may reach the several ten mA range for diamond and the sub-nano ampere range for polyethylene, though some parameters, e.g., the carrier life time, are not available.

**Table 1: Electrical Properties of Candidates of Solid State Conductivity Dosimeters**

<table>
<thead>
<tr>
<th></th>
<th>Conductivity [Ohm$^{-1}$cm$^{-1}$]</th>
<th>Band Gap [eV]</th>
<th>Mobility [cm$^2$/Vs]</th>
<th>Carrier Lifetime [sec]</th>
</tr>
</thead>
<tbody>
<tr>
<td>CdS</td>
<td>$10^{-9}$-$10^{-12}$</td>
<td>2.4</td>
<td>210$^{*1}$</td>
<td>20</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>$10^{-2}$-$10^{-3}$</td>
</tr>
<tr>
<td>Diamond</td>
<td>$10^{-12}$-$10^{-13}$</td>
<td>7.2</td>
<td>1800$^{*1}$</td>
<td>1200</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>?</td>
</tr>
<tr>
<td>Polyethylene</td>
<td>$10^{-16}$-$10^{-20}$</td>
<td>3</td>
<td>1 ?</td>
<td>1 ?</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>$10^{-9}$</td>
</tr>
<tr>
<td>Alumina</td>
<td>$&lt; 10^{-16}$</td>
<td>8.3</td>
<td>22</td>
<td>?</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>$&lt; 10^{-9}$ ?</td>
</tr>
</tbody>
</table>

$^{*1}$: Ref(5)
4. MEASUREMENT OF ELECTRICAL RESISTIVITY UNDER 14 MEV NEUTRON IRRADIATION USING FNS

Electrical conductivity of alumina under 14 MeV neutron irradiation was examined by neutron source of FNS (6). In accelerator D-T neutron source FNS, ratio of absorption dose rates of gamma-rays to that of neutrons is less than 10%, and more than 80% of neutrons are 14 MeV neutrons. Maximum 14 MeV neutron flux of $2 \times 10^{11} \text{n/cm}^2 \text{s}$, corresponding to 2.4 [Gy/s], can be obtained at 1 cm distance forward from the target. The measurement of resistivity for an alumina foil was performed using the HP4339A high resistance meter by three-electrodes geometry with a guard ring as shown in Fig. 2. The alumina sample was a pure alumina crystal, 12 mm in diameter and 0.3 mm in thickness. The alumina sample was set in the vacuum chamber to eliminate parasitic current due to ionization of air and placed in front of the rotating target as shown in Fig. 3. The beam current of the deuteron accelerator was changed from 1 mA to 30 mA, corresponding to the dose from 0.01 to 1 [Gy/s]. The resistivity was promptly changed when neutrons were injected as shown in Fig. 4. Time response was governed by time constant of measurement device, but shorter than 1 sec. From the observed conductivity, i.e., the current of 1 nA for the dose rate of 1 [Gy/s], a carrier lifetime might be the order of $10^{-11}$ s for alumina.

The measured conductivity in the range of $10^{-2}-10^{0}$ [Gy/s] is plotted in Fig. 5 together with the other experimental data for various radiation fields. The result shows good dependence that $\delta=0.96$. In comparison with the other data, the dose dependence $\delta$ is almost the same as that of X-ray. For fission reactor at high temperature, the figure shows rather higher dose dependence. From the figure, it is anticipated that the dose dependence $\delta$ is independent of the kind of radiation and much depends on the temperature. The former feature means that this detector works as a calorimeter in a mixed radiation field, and the latter requires careful examination of temperature and high flux effects on the dose dependence.

5. CONCLUDING REMARKS

The possibility of use of alumina insulator for neutron detector in high energy and high flux neutron radiation field was investigated. The old work of solid state conductivity dosimeter was recalled in relation to the recent work of radiation induced conductivity phenomena for alumina. Because the ceramic insulators has intrinsically
radiation hardness, alumina is considered to be candidate. The measurement of neutron dose dependence on electrical conductivity of alumina showed good property. However, for practical application the further examinations including temperature, fluence and high flux effects are necessary together with practical design of the detector.

References


![Fig. 1 Neutron KERMA factor for alumina](image-url)
Volume resistivity is defined as the resistance per unit volume.

\[ \rho_v = \frac{S}{l} R_v \]

where,

- \( \rho_v \): Volume resistivity [\( \Omega \text{-mm} \)]
- \( S \): Cross section [mm\(^2\)]
- \( l \): Length [mm]
- \( R_v \): Indicated volume resistance [\( \Omega \)]

Fig. 2 Volume conductivity measurement by three-electrode geometry with a guard ring
Irradiation Equipment for Resistivity Measurement

Fig. 3 Experimental setup of D-T neutron irradiation
Fig. 4 Time response of the observed resistivity for pulsed neutron irradiation
Fig. 5 Measured conductivity as a function of neutron dose together with the other experiments
NEUTRON SPECTRUM MEASUREMENT
IN THE ENERGY REGION OF eV
WITH THE SLOWING DOWN TIME METHOD

Fujio Maekawa and Yukio Oyama
Department of Reactor Engineering, Japan Atomic Energy Research Institute
Tokai-mura, Ibaraki-ken, 319-11 Japan

1. INTRODUCTION

In terms of neutron spectrum measurement in the energy region of eV, no general-purpose method has been established so far. The time-of-flight (TOF) method, the multi-foil activation (MFA) method and the multi-spherical Bonner counter (MSB) method have been applied for the spectrum measurement in the energy range, however, these techniques have their own weak points and restrictions for the measurement. On the other hand, the slowing down time (SDT) method was proposed by Bergman, et al. in 1955 and the principle of the method has been applied for the so-called “lead slowing down spectrometer.” In this study, the SDT method was investigated as another possibility of measuring method of neutron spectrum in the energy region of eV.

2. SLOWING DOWN TIME METHOD

When pulsed neutrons are injected into a large medium, the neutrons gradually slow down with passage of time. Assuming an ideal case for the method, i.e., (1) an elastic scattering cross section of the material is independent from neutron energy without any resonance structures, (2) the other reaction cross sections are much smaller than the elastic one and (3) a mass number of composing nuclei of the medium are large, the asymptotic shape of neutron spectrum at each moment approaches a Gaussian distribution. In an ideal case, according to the theory of neutron slowing down in a heavy medium, the mean neutron energy at time t, $E_{\text{ideal}}(t)$, can be approximately represented by

$$E_{\text{ideal}}(t) = \frac{K}{t^2}$$

and the energy resolution $(\Delta E / E)_{\text{ideal}}$ in full width at half maximum is given in percentage by
The relation (1) states that one observes neutrons gathered around energy $E(t)$ at the time $t$, and spread of the neutron energies is given by Eq. (2). It is easily recognized from Eq. (2) that the spread of neutron energies is smaller in a medium of heavier nuclei such as lead.

The theory of neutron slowing down does not completely stand up any longer for non-ideal cases. However, the SDT method can be applied to the neutron spectrum measurement because only the feature that neutrons gradually slow down with passage of time is necessary for that purpose and this feature is always true except the case that there exist neutron up-scatterings in the thermal energy region. The theory describing Eqs. (1) and (2) for the ideal case is also useful to roughly grasp characteristics of slowing down neutrons in a realistic medium.

\begin{equation}
(\Delta E / E)^{\text{ideal}} = 235.5 \sqrt{\frac{8}{3A}}
\end{equation}

\(A:\) mass of nuclei of the medium
\(K:\) constant.

The relation (1) states that one observes neutrons gathered around energy $E(t)$ at the time $t$, and spread of the neutron energies is given by Eq. (2). It is easily recognized from Eq. (2) that the spread of neutron energies is smaller in a medium of heavier nuclei such as lead.

The theory of neutron slowing down does not completely stand up any longer for non-ideal cases. However, the SDT method can be applied to the neutron spectrum measurement because only the feature that neutrons gradually slow down with passage of time is necessary for that purpose and this feature is always true except the case that there exist neutron up-scatterings in the thermal energy region. The theory describing Eqs. (1) and (2) for the ideal case is also useful to roughly grasp characteristics of slowing down neutrons in a realistic medium.

3. EXPERIMENT

A BF3 gas proportional counter was selected for a neutron detector because of simple $1/v$ dependence and well known cross section of the $^{10}\text{B}(n,\alpha)^{7}\text{Li}$ reaction, and the property of superior neutron-gamma separation. The 96% boron-10 enriched BF3 gas of 71.5 kPa was contained in an aluminum casing of 14 mm in outer diameter and 99 mm in effective length. The counter efficiency was calibrated in a standard neutron field inside a graphite pile, and the effective number of boron-10 atoms in the counter was determined as $2.18 \times 10^{20}$ with accuracy of 3%.

The experiment was conducted at the Fusion Neutronics Source (FNS) facility \(^2\) in Japan Atomic Energy Research Institute. Neutron spectra were measured in three kinds of shielding experimental assemblies for fusion reactor development, that is, an iron assembly \(^3\), an SS316 assembly \(^5\) and an SS316/water layered assembly \(^6\). The iron assembly was a cylinder of 1000 mm in diameter and 950 mm in thickness. The SS316/water assembly is illustrated in Fig. 1. The SS316 assembly had almost the same dimensions as the SS316/water assembly but the pure SS316 was used instead of SS316/water layers. The experimental assemblies had several measurement channels as shown in Fig. 1, and the BF3 counter was inserted into one of the channels.

Pulsed D-T neutrons, typically, 1 µs in duration and 200 µs in intervals, were injected into the experimental assembly, and time-spectra of the $^{10}\text{B}(n,\alpha)$ reaction events were recorded by employing the electronic circuit shown in Fig. 2. In order to calibrate energy scale of the
measured time-spectra, the conventional resonance filter method\(^{(1)}\), which was frequently utilized for the lead slowing down spectrometers, was adopted. Five resonance filters of Mn, Co, In, W and Au, were used. The energy range of the resonance peaks ranged between 1.46 eV of In and 336 eV of Mn. The BF3 counter was covered with one of the filters, and time-spectra of the \(^{10}\)B(n,\(\alpha\)) reaction rate were measured in just the same way as without the filters. Typical measuring time and neutron yield for the individual runs are 30 - 60 minutes and \(10^{12}\) neutrons, respectively.

4. DATA PROCESSING

Figure 3 shows a typical example of the measured time-spectra with and without the cobalt filter in the SS316 assembly. A small dip is seen around 900 channels on the time-spectrum with the cobalt filter. The dip is formed by the prominent resonance peak of cobalt at 132 eV. Both the spectra were normalized by total numbers of neutrons generated during the measurements, and the spectrum with the cobalt filter was divided by that without the filter. The result, Fig. 4, clearly represents the small dip observed in Fig. 3. The dip was fitted with a Gaussian distribution to determine the center of it; 5.00 \(\mu\)s in the case of Fig. 4. Accordingly the slowing down time of the source 14 MeV neutrons to 132 eV was found to be 5.00 \(\mu\)s. The same procedures were repeated for all the time-spectra measured with the resonance filters, and slowing down times were determined. All the experimentally obtained slowing down times for the SS316 assembly are plotted on Fig. 5 with the calculated calibration curve by the Monte Carlo transport code MCNP. As for the SS316 assembly, agreements between the experiment and calculation are very good as within 4 % except the case of the tungsten filter.

The measured time-spectrum of \(^{10}\)B(n,\(\alpha\))\(^{7}\)Li reaction rate \(C(t)\) in counts/\(\mu\)s is converted into energy-dependent reaction rate \(C(E)\) in counts/eV by using the calibration curve, \(E(t)\), which is experimentally validated, as,

\[
C(E) = C(t) \cdot \left( \frac{dE(t)}{dt} \right)^{-1}. \tag{3}
\]

Neutron energy-spectra were obtained by the next formula.

\[
\phi(E) = \frac{C(E)}{\sigma(E) \cdot N \cdot Y_n \cdot f_{ss}(E) \cdot f_{\sigma}(E)} \tag{4}
\]

where,

\[
\phi(E) : \text{Energy-spectrum [n / eV / Source Neutron]}, \quad \sigma(E) : \text{Cross section of the }^{10}\text{B(n,}\alpha)\text{ reaction taken from JENDL-3.1,} \\
N : \text{Number of effective boron-10 atoms in the BF3 counter,}
\]
Neutron source yield, $Y_n$; 
Self-shielding correction factor of the counter, $f_{ss}(\bar{E})$; 
Correction factor for effective $^{10}$B(n,$\alpha$) cross section, $f_{\sigma}(\bar{E})$.

Since a neutron spectrum at time $t$ has a broad energy distribution centered at $\bar{E}$, spectrum weighted effective $^{10}$B(n,$\alpha$) cross section differs from the cross section at $\bar{E}$ due to energy dependence of the cross section. Hence the correction factor $f_{\sigma}(\bar{E})$, which is a ratio of both effective and real $^{10}$B(n,$\alpha$) cross section, was introduced with a help of the calculated spectrum $\phi(E,t)$ by MCNP, as

$$f_{\sigma}(\bar{E}) = \frac{\int \sigma(E) \cdot \phi(E,t) \cdot dE / \int \phi(E,t) \cdot dE}{\sigma(\bar{E})}. \quad (5)$$

The correction factors for the iron and SS316 assembly below 1 keV are 1.02 - 1.05 while those for the SS316/water assembly range between 1.3 and 1.5, due to worse energy resolution comparing with the other two assemblies.

Neutrons in thermal energy region form an equilibrium spectrum not only by down-scatterings but also by up-scatterings. The mean energy of thermal neutrons is constant and does not have a one to one correspondence to the slowing down time. Thus the SDT method cannot be applied to measure neutron spectra in the thermal energy region. However, a total thermal flux instead of an energy-spectrum can be derived by extending the SDT method. The $^{10}$B(n,$\alpha$) reaction events observed after the time when mean neutron energy reached to the upper limit of thermal energy, defined as 0.322 eV in this study, are regarded as caused by thermal neutrons. The total thermal flux, $\phi^{th}$, is obtained with the formula,

$$\phi^{th} = \frac{C}{\sigma^{th} \cdot N \cdot Y_n \cdot f_{ss}^{th} \cdot f_{\sigma}^{th}} \quad (6)$$

where

- $C$: Counts after mean neutron energy reached to 0.322 eV
- $\sigma^{th}$: Thermal cross section of the $^{10}$B(n,$\alpha$) reaction, 3837 b
- $f_{ss}^{th}$: Self shielding correction factor of the counter for thermal neutrons
- $f_{\sigma}^{th}$: Correction factor for effective $^{10}$B(n,$\alpha$) cross section.

4. RESULTS AND DISCUSSIONS

The neutron spectra at 210 and 810 mm depths in the iron assembly measured by the SDT method (below 3 keV) are shown in Fig. 6. In the figure, the spectra measured by proton...
recoil gas proportional counters (PRC) by Konno, et al. (above 3 keV) and those by the MCNP calculation are also plotted. Typical assigned errors of the measured spectra are 5 - 10 %. The spectra by the SDT method agree very well with the calculated ones. This figure obviously demonstrates that the SDT method is very useful to measure neutron spectra in the energy region of eV which is below the lowest detection energy of the PRCs.

Figure 7 shows neutron spectra in the SS316 and SS316/water assemblies measured by the SDT method and calculated by MCNP. Typical assigned errors of the measured spectra for the SS316 and SS316/water assemblies are 5 - 10 % and 6 - 30 %, respectively. Agreements between the measurements and calculations are also good for both assemblies.

In the spectra for the SS316 assembly, two broad dips are found around 40 and 300 eV. They are attributed to the dips formed by the large resonance peaks at 45 eV of Mo and 336 eV of Mn. Although both Mo and Mn are contained only about 1 atomic percentage in the SS316, the effects of their large resonance peaks are clearly seen in the measured spectrum. This fact suggests that the measured spectra by the SDT method are so reliable that such faint dips in the spectra can be identified.

In the spectra for the SS316/water assembly, a bump of thermal flux is represented in the measured spectra as similar to the calculation. Although errors of the measured spectrum are larger in the higher energy part, the result demonstrates that neutron spectrum can be measured even in the assembly which contains light atoms such as hydrogen by applying the SDT method.

The $^{197}$Au(n,$\gamma$) reaction rates are also measured along with the neutron spectra. Since cross section of the reaction has the giant resonance peak at 4.9 eV, most of the reactions are caused by neutrons between 1 - 10 eV. The foil activation technique has been already firmly established, and assigned errors for the measured reaction rates are as small as 3 - 5 %. Hence in order to verify the present measurements, the integral fluxes between 1 - 10 eV were compared with the $^{197}$Au(n,$\gamma$) reaction rates. Calculated to experimental value (C/E) for both the integral flux and reaction rate is served for the comparisons regarding the calculation as a reference value, because both measured quantities are not directly comparable in themselves. The C/E curves as a function of measuring position in the SS316 and SS316/water assemblies are shown in Fig. 8. It is seen in the figure that trends of the C/E curves for the integral flux and $^{197}$Au(n,$\gamma$) are similar each other for both assemblies. The C/E values of both measured quantities also agree within about 10 %. These facts tell us that the measured spectra are consistent with the $^{197}$Au(n,$\gamma$) reaction rate, and the consistency is the proof of validity of the present spectrum measurements.

This study clarify features of the SDT method as a measurement technique of neutron spectrum in the energy region of eV. The measuring positions should be inside bulk assemblies.
This imposes a restriction on the measurement, but the in-situ measurement is effective to reduce background neutrons. Although the SDT method requires an appropriate pulsed neutron source and the quality of the measured spectra highly depends on the surrounding medium, there are many advantages in the SDT method comparing with the TOF, MFA and MBS methods: (1) high sensitivity, (2) non-oscillating property of the spectrum if the counting statistics are enough, (3) high accuracy and (4) medium energy resolution.

ACKNOWLEDGMENT

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References

Fig. 1  Experimental assembly of the SS316/water layered shield.

Fig. 2  Electronic circuit employed to measure neutron spectra with the SDT method.
Fig. 3  Measured time-spectra with and without the cobalt filter at the 533 mm position in the SS316 assembly.

Fig. 4  Ratio of the measured time-spectra with and without the cobalt filter (black diamonds) and the fitted curve.
Fig. 5  Experimentally obtained slowing down times by the resonance filter technique for the SS316 assembly (open diamonds) with the calculated calibration curve.

Fig. 6  Neutron spectra at 210 and 810 mm depths in the iron assembly measured by the SDT method (below 3 keV) by proton recoil gas proportional counters by Konno, et al.\(^{(4)}\) (above 3 keV) in comparisons with those by the MCNP calculation.
Fig. 7 Measured neutron spectra for the SS316 (black diamonds) and SS316/water (open diamonds) assemblies with the calculated ones by MCNP (histograms of solid and broken lines).

Fig. 8 Comparison of the integral flux between 1 - 10 eV in the SS316 and SS316/water assemblies by the present spectrum measurement and the $^{197}$Au(n,γ) reaction rates in the calculated to experimental values.
ENERGY RESPONSE OF A FAST-THERMAL NEUTRON SIMULTANEOUS COUNTING DETECTOR

Makoto Hashimoto, Tatsuo Torii, Hideki Ando
Oarai Engineering Center, PNC
4002 Narita, Oarai, Ibaraki-ken, 311-13, Japan

Taichi Michikawa
Tsukuba Lab. JREC
5159-1, Oazakurihara, Tsukuba, Ibaraki-ken, 305, Japan

1. INTRODUCTION

A method of measuring the impurities of nuclear material samples using a pyrochemical multiple counter has been developed by Langner and others\(^1\). The pyrochemical multiple counter consists of many thermal neutron detecting tubes arranged in four concentric rings within a polyethylene moderator, and a nuclear material sample is set in the center of the counter. Each ring of tubes has a different energy response owing to the difference of the effective moderator thickness, and each ring gives a different count with the same sample. The stopping power of the moderator varies with the incident neutron energy, and the differences in the counts from the various rings are related to the energy. The average energy of neutrons from a nuclear material sample varies with the impurities of the sample, therefore it is expected that the ratios of counts from the four rings will indicate the impurity concentration of the sample, and this was confirmed by experimentation.

Langner and others used a multiple counter, but this method can be applied to two detectors which possess different energy responses and are in the same condition. In this paper, which investigates the applicability of this method to a fast-thermal neutron simultaneous counting detector which possesses two kinds of energy response in a single detector, we present the results of an experimental examination of the energy response of this detector to neutrons at energy of several MeV.

2. FAST-THERMAL NEUTRON SIMULTANEOUS COUNTING DETECTOR

The fast-thermal neutron simultaneous counting detector is a kind of spherical BF\(_3\) proportional counter of 2.5" diameter, the inside of the detector wall is coated with polypropylene\(^2\)(See Fig.1). The detector functions as a BF\(_3\) proportional counter which has high
sensitivity to thermal neutrons, and it also functions as a recoil proton proportional counter for fast neutrons because of the polypropylene coating inside the wall. It detects these reactions simultaneously, so it has an acceptable response to the range of neutron energies from thermal to fast, in spite of having thinner moderator than existing detectors with a wide range of sensitivity. Furthermore, it is expected that the absorbed energies from thermal and fast neutrons will be distinct, because the Q-value of the $^{10}$B(n, $\alpha$) reaction is much larger than the energy absorption from fast neutrons due to the small stopping power of BF$_3$ gas to protons and the small volume of the sensitive region. So, these reactions can be detected independently by pulse height discrimination.

Figure 2 shows the result of calculations of the energy response of this detector using the ANISN code. The energy response of each reaction varies sharply in MeV region. So, it is expected that the ratio of the counts from the two reactions will also be sensitive to variations in the neutron energy.

3. ENERGY RESPONSE

Figure 3 shows an example of the pulse height spectrum of this detector in an ordinary neutron field. The spectrum consists of two elements and Fig.4 and 5 show those elements. Fig.4 shows a spectrum with fast neutrons: the detector is irradiated with 1.5MeV monoenergetic neutrons without moderator. Fig.5 shows a spectrum with thermal neutrons: the detector is irradiated by $^{252}$Cf within a graphite pile.

From such spectra, we could calculate the ratio of counts from the two reactions as mentioned the present section. But, to simplify the data treatment, we divided the pulse height spectrum into two regions. The upper region is simply composed of (n, $\alpha$) reaction counts and the lower region is composed of (n, $\alpha$) reaction counts and recoil reaction counts; we defined the ratio of the count for the lower region (A) to that of upper region (B) as the "spectrum index"(See Fig.3). We made experimental measurement of the relation between spectrum index and neutron energy. The experiments were carried with neutrons of RI source ($^{252}$Cf, $^{241}$Am-Be, $^{239}$Pu-Be), and also with monoenergetic neutrons at three energy ranges (around 1.5MeV, around 3.0MeV, around 5.0MeV) induced by an accelerator. During the experiments with monoenergetic neutrons, we varied the neutron emission angle stepwise to change the neutron energy by several tens of keV.

Figure 6 shows the spectrum indices with monoenergetic neutrons. With neutrons around 3.0MeV and 5.0MeV, the spectrum index shows a clear variation with the neutron energy for changes of several tens of keV. Fig.7 shows the spectrum indices with neutrons from RI sources, the spectrum index shows a clear variation with the dose equivalent average energy. It is noted that, for $^{241}$Am-Be and $^{239}$Pu-Be, the spectrum indices are different, and this indicates that the dose equivalent average energies of the sources are different. It is considered that this difference is caused by the small variation of $\alpha$-energy between $^{241}$Am and $^{239}$Pu. The $\alpha$-energy variation...
must cause the neutron average energy different(3)(4). However, the neutron average energy variation is too small to be detected by existing detectors, so it has been assumed that the dose equivalent average energies of $^{241}$Am-Be and $^{239}$Pu-Be are same. In conclusion, this detector is very sensitive to the variations of the average neutron energy.

Figure 8 shows the spectrum indices calculated from the results of the energy response calculations. The theoretical spectrum indices vary greatly with neutron energy, and this is confirmed by the experiment. From the energy response calculations, the counts of (n, $\alpha$) reaction and recoil reaction both vary with neutron energy, but these counts also depend on the source intensity and it is difficult to extract the neutron energy information without knowing the incident neutron flux level. In the meantime, the spectrum index is not affected with the source intensity. This is the advantage to estimate the neutron energy with the fast-thermal neutron simultaneous counting detector.

4. CONCLUSIONS

The spectrum index of the fast-thermal neutron simultaneous counting detector is very sensitive to variations in neutron energy in the MeV region. From this result, this detector is expected to be applicable to the evaluation of energy information not only for neutrons from fissionable materials but also for neutrons from transuranic nuclides or (\(\alpha,n\)) reactions.

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Fig. 1 Schematic Diagram of the Fast-Thermal Neutron Simultaneous Counting Detector

Fig. 2 Calculated Energy Response
counts

\[ \frac{\text{Count } "A"}{\text{Count } "B"} \]

Fig. 3 Pulse Height Spectrum of the Detector Irradiated in an Ordinary Neutron Field

Fig. 4 The Pulse Height Spectrum of the Detector Irradiated with 1.5MeV Monoenergetic Neutrons without Moderator

Fig. 5 The Pulse Height Spectrum of the Detector Irradiated with 252Cf within a Graphite Pile
Fig. 6 Spectrum Indices with Monoenergetic Neutrons

*The experimental conditions of fig(a), (b) and (c) are different, so the spectrum indexes are not standardized between them.

Fig. 7 Spectrum Indices with RI Neutrons

Fig. 8. Calculated Spectrum Index
A NEW APPROACH FOR RADIATION INVERSE-PROBLEMS
BASED ONLY ON THE BAYES' THEORY

S. Iwasaki

Department of Nuclear Engineering
Tohoku University
Aramaki-Aza-Aoba, Aobaku, Sendai 980-77, Japan
iwa@minel.nucle.tohoku.ac.jp

1. INTRODUCTION

A new method is proposed in this paper for various radiation inverse-problems, specifically for the spectrum unfolding. This method is based only on the Bayes' theorem; it is quite different approach from the conventional matrix-inversion type ones. This method has the following characteristics; the spectrum inference is made in the iterative fashion by applying the Bayes's theory repeatedly as getting a pulse height value for each count by a detector; it provides intrinsically non-negative radiation spectrum because the flux is regarded as the probability; the resultant spectra do not oscillate because of condition of the inference process satisfying the maximum information-entropy; and finally the algorism is quite simple and vary easy to calculate being enough to be applied in the real time analysis mode. In the following chapters, the new method is described in the example case of fast neutron detection

2. PRINCIPLE OF THE METHOD

First, in the proposed method the neutron flux \( p(E_i) \) (\( i=1, n \)) is regarded as a probability of an event of neutron measured having \( E_i \); \( E_i \) is one of the \( n \) partitions of a space \( \Omega \) of a neutron event, where \( E_i \) is an energy of incident neutron into the detector; these partitions \( E_i \) (\( i=1, n \)) are mutually exclusive and exhaustive; thus, \( p(E_i)>0 \) and \( \sum p(E_i)=1 \). Another partitions \( H_j \) (\( i=1, m \)) are defined in the same space \( \Omega \); \( H_j \) is a pulse height value which a neutron produces when it is detected; \( H_j \) (\( i=1, m \)) are also mutually exclusive and exhaustive.

The Bayes theorem is then given as (1), for an event detected with the pulse height \( H_j \),

\[
P(E_i|H_j) = \frac{P(H_j|E_i)P(E_i)}{\sum_{i=1,n} P(H_j|E_i)P(E_i)}. \tag{1}
\]
where $P(\bullet)$ and $P(\bullet\mid\bullet)$ denote ordinary probability and conditional probability, respectively. $P(E_i)$ is called a priori probability, or simply prior, an assumed probability of an event having $E_i$ from the knowledge on hand, before the pulse height value of the neutron is not obtained. The conditional probability $P(H_j|E_i)$ is usually called likelihood, and can be regarded in the present case as the detector response (function) which gives the probability of a radiation whose pulse-height is $H_j$ with the condition of the energy of the radiation is known as $E_i$. The left side conditional probability $P(E_i|H_j)$ called a posteriori probability, or simply posterior is the probability of a radiation event $E_i$ with the condition of the pulse height of the radiation is known as $H_j$.

In the Bayesian statistics, the above formula is aggressively used as a statistical inference; for the next event, the posterior can be used as the new prior for the next event. However, this simple cyclic use of the Bayes' formula is not adopted in the present unfolding method, as shown below.

In Fig 1, the flow diagram of the process of the present method is shown. First, the prior should be assumed, usually posed as a constant, or uniform distribution as the noninformative prior. If we have some information on the source spectrum, we can adopt this as the prior. We assume the uniform distribution from zero energy (usually) as the lower limit of the energy range considered to the maximum energy of the range depending on the problems; this is essentially the maximum energy at which the detector response is given or known. If we take a bin width being equal for all $E_{i_0}$, $P(E_{i}) = 1/n$, where $n$ is the number of energy bins. The uniform prior, however, is not important because the present method is not sensitive to the prior values after sufficient number of events.

Next the pulse height value $H_j$ from the detector for each event is given, then, the likelihood is calculated using the response data matrix $P(H_j|E_i)$. After normalization of the denominator of the equation [1], the normalized likelihood values are accumulated. This accumulation is essential point of the present method, instead of the cyclic use of the Bayes' formula called 'simple method'. It was found that the simple method worked well only in the case of the single hypothesis $i=1$, i.e., neutrons with a mono-energy at $E_i=E_{i_0}$; while in the multi-hypothesis case, or neutrons which had broad energy distribution, the simple method failed; probabilities for all hypotheses $P(E_i)$, $i=1,n$ finally became zero since at least one event with $P(H_j|E_i)=0.0$ surely happened for every hypothesis after a certain number of events. The accumulation of the a posteriori probability every time can resolve this difficulty.

The accumulated posterior is normalized to one every time, and replace the prior for the next event (count). After getting sufficient large number of total counts of $N$, the posterior converges to asymptotic one; this can be adopt as the estimation of the spectrum. In the above accumulation, sometimes it is desirable to use a multiplier $a$ for the posterior probability as shown in the diagram. The coefficient $\alpha$ or $\alpha'$ is a small positive number, which is an adjustable parameter to accelerate the convergence or to depress the temporal oscillation of the estimated probabilities, respectively, depending on the problems. An analysis code based on this method called UnBay has been produced.
constraint, the present method has provided intrinsically non-negative, stable spectrum, and no numerically ill-condition. The negative flux unstable spectra and numerical ill-condition have been frequently observed in the conventional matrix-inversion type approaches. These are probably due to the statistical error of the observed data or caused by use of the biased response data, and its stabilization or regularization is one of the most important task. The instability of the results also depends on the degree of singularity of the response matrix. The maximum entropy principle has been understood as one of the most important and effective constraint in the various area of the inverse problems (2).

Besides above mentioned simple example, more realistic examples have already been successfully tested, such as, neutron spectrum unfolding for the another spectra data observed by an NE213, and the elemental analysis problems for the x-ray data in PIXE, which will be reported in detail elsewhere (3).

The present method can be applicable to broad inverse problems where the response of the event is known and many events are expected to be available in one experimental run, not only in the spectrum analysis (energy, time, position, etc.) but also in the radiation application, i.e., radiation imaging, such as, radiography and computed-tomography, etc.

References

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Fig. 1 Flow diagram of spectrum unfolding process on the basis of the Bayes' theory.
Fig. 2 Pulse height data at the accumulated counts of 3, 23, 235, 2350, 23570, and 235000, respectively.

Fig. 3 Unfolded spectra corresponding to the accumulated counts shown in Fig. 2, as well as the true spectrum and prior.
1. INTRODUCTION

Various kinds of analysis programs for X-ray spectra have been developed for the purposes of the elemental analysis usually based on the linear or nonlinear least squares method to fit the peaks. Online or automatic analysis mode is, however, usually not adopted because the existing of the heavily overlapping peaks to be separated and of ambiguity of the continuous background shape necessitate analysts to be of deep experience, and prevent those kinds of analysis mode. Therefore, most manufactures have concentrated their efforts into development of user friendly systems or environments with excellent user-interfaces between the analyst and system.

A new analysis technique for spectra of gamma- and X-rays based on linear associative neural network (LANN) has been developed by the present authors (1-3). The fundamental characteristics of the network technique proved to be feasible to the analysis with respects to the qualification and speed. The technique was applied actually to the spectrum analysis of PIXE experiment and showed good performance (3). Name of the pattern analysis came from the fact that we treated a set of reference spectra of each element as the spectrum pattern and fitted the reference spectra to the objective spectra. In this technique, X-ray peak, e.g., Kα or Kβ is not dealt with separately, as done in the conventional peak analysis technique.

In the network analysis for the PIXE spectra, a set of reference spectra of each single element should be prepared for the network learning so as to have suitable weight values for each network unit; actually these are the same with the elements of the Moore-Penrose pseudo-inverse of a matrix whose column vectors are consists of the reference spectra (1). One of the key points of the pattern analysis based on the neural network is how to get proper reference spectra. The best way is the measurement using a given detector system with a suitable set of single elemental specimens as done in an example(3),
because we are almost free from various ambiguities appearing in the model calculation as mentioned below. Actually, it is not impossible to measure all data for the single elements included, e.g. in bio-samples of about 40 because such standard samples are available now. In practice, it is also useful to obtain the relative reference spectra using a model calculation because the number of elements is quite large, and the measurements are tedious. The model calculation approach is, on one hand, versatile, and adaptable for any measuring system and condition, while it needs a set of reliable model parameters, such as X-ray fluorescence yields, ratio data for $X_p/X_{\alpha}$, relative intensities of L-subshell lines, ionization cross sections for each X-ray lines (K, L and M), and detector efficiencies including the effect of the geometry and absorber, etc.

In the present study, a semi theoretical model based approach is taken. In this approach, the model calculation provides the first order approximation of the reference spectra, and then these will be revised or some of them will be replaced by the experimental data if we find a significant disagreement between the calculation and observation. First, the measurement of experimental spectra for some single element samples has been carried out. These spectra could be used to deduce the model parameters which are dependent on the measurement system, and also used to validate the calculated reference spectra. Objective spectra to be analyzed by the network with above reference data are also measured in the same system.

2. Preparation of Reference Spectra

2.1. Experimental spectra measurement

Measurement of the reference spectra has been made at Nishina Memorial Cyclotron Center (NMCC) of Japan Radioisotope Association in Takizawa, for the samples which were prepared by dropping of solution with a pipet of 5-\mu cc on thin polypropylene films from twenty single elemental samples for the atomic absorption spectrometry: 13 elements of Al, Ca, Cr, Mn, Fe, Ni, Zn, Se, Y, Nb, Cd, Sn, Pb, in HNO$_3$ solution and 7 elements of K, Sc, Co, Cu, As, Ag, and In in HCl solution. The samples have been dried by an infra-red lamp. Among them, seven samples were mounted on a Mylar target holder, and set in the vacuum chamber of the PIXE course of a Baby Cyclotron accelerator. The target holder was tilted by 45 deg. with respect to the ion beam axis, and samples were in the backward side of the film; therefore the beam hit the sample atoms after through the baking film. An accelerated proton beam of about few n-amps at 2.9 MeV impinged on the samples, and induced X-rays were observed by a Si(Li) detector (with the energy resolution of 170eV for $^{56}$Fe X-rays) at 90 deg. with respect to the beam. at 7 cm from the sample position. A Bremsstrahlung absorber of a thickness of 100 \mu m was inserted between the sample and detector. Some samples contained the impurity of chlorine which was a residual of the HCl solution.

The normalized backing film's spectrum was subtracted from each elemental spectrum; thus the
residuals of the spectra were the characteristic X-ray's peaks and small amount of continuous component probably due to the Bremsstrahlung in the elements themselves.

In addition to the above standard samples, we measured a spectrum for a mixed elemental sample called INTB1 which contains the following 12 elements, Ag, Ba, Be, Cd, Co, Cr, Cu, Mn, Ni, Pb, V, and Zn.

2.2. Semi theoretical model based approach

The model calculation were carried out using the X-ray emission related data given in the literature (4). Shape of single peak for a Si(Li) detector can not been described by a Gaussian function, but necessitates some additional terms, such as exponential tails, and step function in the lower energy side of the peak, etc (5). It is also known that the escape peaks appear for some elements. In this paper, however, we did not adopt the detailed model for simplicity in the preliminary stage of the approach by the model calculation.

From the set of the measured spectra data for each elements, we can obtain other model parameters for the calculation which are specific to the measurement system, such as, peak position vs. channel, peak width vs channel, and relative efficiency of the detection system. Each single elemental spectrum has been analyzed by the code SAPIX (6), and peak channels and peak widths were deduced. These data have been fitted with the linear and quadratic functions of channels, respectively. The data of the relative detector efficiency curve was given by Sera, et al. (7). In the model calculation, we included the K and L X-rays, but not M X-rays (e.g., for Pb). The intensity of the spectra were normalized by the Kα-peak area. An example of the model calculation for iron is demonstrated in the Fig. 1, where the experimental data are compared with the calculated one. The iron sample was taken from HCl solution, and the chlorine peaks are also seen. Two K X-ray lines for iron are well described by the present model calculation. The reference spectra for each element were generated by replacing the peak part with the model calculation and chlorine peaks if existed were just removed.

3. RESULTS

Thus, a neural network which had 1024 input units corresponding to the total PHA channels of the spectra and 20 output units for all elements considered was trained with the reference spectra. The objective spectrum of the INTB1 solution as shown in Fig. 2 was then fed to the network. The validity of the reference spectra are shown by comparison of the reconstructed spectrum (denoted by projection 1 in Fig. 2) from the output of the network using the reference spectra with the objective spectrum; the former spectrum is mathematically called orthogonal projection of the objective spectrum to the reference space, which is spanned by the reference spectra (vectors). These two spectra should agree well with each other, if the reference spectra are valid and the objective spectrum is a linear combination of...
the reference spectra. No significant difference is found between them, except for the K-lines of Ag and Cd, as shown in the inset of Fig. 2. When we replace these elements' reference spectra by the measured ones, the resultant projection spectrum (denoted by the projection 2 in Fig. 2) shows improved agreement for the case of Ag, but still not for Cd. There might be some reasons of the disagreement in the calculated spectra for the two elements; one is due to the unreliable data base for the ratio of L and K X-ray yield for heavier elements, the other is due to the non-linear effect, such as pile up happened in the measurement of the reference spectra for the In data.

4. CONCLUSIONS

Complete agreement could not be obtained between the calculated and measured spectra for the elements in a wide range of atomic number. This is partly due to the inadequacy of the data base for the X-ray emission calculation, and partly due to the possibly existing non-linear effect in the measurement spectra. In spite of such simple model calculation, however, generally good agreement between the calculated and measured spectra showed that the approach based on more realistic model calculation for the reference spectra would be promising. It is also necessary to prepare the Bremsstrahlung components in respective samples in addition to the peak components, which have been treated experimentally in the present work. This approach could solve the ambiguity of the unknown shape of the continuous components under the peaks which is inevitably associated to the conventional peak fitting analyses.

References

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Fig. 1 Companions of measured reference spectrum for iron elements with that of model calculation.
Fig. 2. Companions of the spectrum for the multi-elemental standard sample INBTL1 with the constructed spectrum (denoted as projection 1) based on the calculated reference spectra, and with the same spectrum but based on reference spectra partly replaced by the experimental ones (projection 2).
APPLICATION OF PATTERN ANALYSIS TECHNIQUE TO ROOM-TEMPERATURE OPERATIONAL TYPE X-RAY DETECTORS WITH POOR RESOLUTIONS

K. Yoshizaki, H. Fukuda, S. Iwasaki, and M. Kitamura

Department of Nuclear Engineering, 2Cyclotron Radio-isotope Center, Tohoku University, Sendai 980-77, Japan

1. INTRODUCTION

In recent years, the energy dispersion type X-ray elemental analysis, such as XRF and PIXE, has widely been used in various fields. Commonly Si(Li) detectors have been used for X-ray detection in analysis equipments. This situation is mainly due to its high quality in the energy resolution and thus high capability of elemental analysis even if it should be operated or maintained in an low temperature condition by liquid nitrogen. Recently, there have been some new type X-ray detectors developed. Especially Si-PIN photodiode detector with Peltier cooler shows relatively high performance in resolution\(^1\); a model with a resolution of about 600eV is now commercially available and a very recent model shows the resolution as good as 250eV; this is less than double of that of Si(Li) detector of the highest class.

These detectors have high potentiality for replacing the Si(Li) detectors in near future if its lower performance of the spectrum analysis could be solved, because these detectors are almost a tenth of the Si(Li) in cost and is operable in room temperature. Pattern analysis method (1-4) which have been developed by the present authors showed high analysis capability for poor resolution spectra of gamma-rays measured by a NaI(Tl) detector (1-2). In this paper, we study its applicability to the analysis of X-ray spectra by such poorer resolution detectors for the purpose of the elemental analysis.

We tested this technique for the calculated spectra by a simple model; we consider only the characteristic X-ray peaks and not the continuous components from various Bremsstrahlung processes. Although the latter components are commonly observed in the XRF or PIXE spectra, it is not directly related to the performance of the elemental separation. It is also known that the profile function for a single X-ray line is not described by a pure Gaussian but needs some additional components, such as exponential tails and step function(5). Such detailed model of the peak shape could not alter the situation of the present test. Thus we applied the simple Gaussian function model to the case of SUS316

* e.g., found in catalogue of AMPTEK Inc., Bedford, U.S.A.
metal sample. SUS316 samples contain some neighboring elements in the periodic table with various concentrations as shown in Table 1 (note: molybdenum (3%) was not included in the present calculation). In this case, quantification of Mn element is of prime important because Mn's K X-ray energies are very nearby those of the neighboring elements: \( K_\alpha \) and \( K_B \) of Mn are close to \( K_B \) of Cr and \( K_\alpha \) of Fe, respectively. In both cases, the energies are too closely spaced to be correctly analyzed by the conventional double peak fitting technique(3). Manganese was understood as one of the difficult element in the commonly occurring samples(7). Difficulty of Mn quantification also arises from its low concentration in the sample. Therefore the SUS316 is a good test sample for the analysis performance.

2. PATTERN ANALYSIS TECHNIQUE

The principle of the pattern analysis method based on the neural network technique is described briefly here. The linear associative neural network is able to realize direct mapping of the objective pulse height spectra to elemental concentration of specimens without any iterative calculation because the network is pre-trained to the reference spectra of the single elements. Function of the network is linear transfer of a vector in the space of spectra, linear combination of \( x_i \) to another vector space of elemental concentration, \( y_i \). This is carried out by multiplying the weight coefficient matrix \( W \), whose elements have been gained by the learning phase before the analysis and stored in the network, to \( x \) as expressed in a matrix form, \( y = Wx \). The elements of the matrix \( W \) are obtained so as to satisfy the condition of the least-squares: \( \parallel Y-WX \parallel ^2 = \text{min} \). through the learning where matrix \( Y \) is consists of \( y \), and usually a unit matrix, \( E \) (this means that the response of the network is unit vector if the input is one of the reference spectra). Thus, the matrix \( W \) is the Moore-Penrose pseudo-inverse matrix of \( X \): \( X^+ \) whose column vectors consists of the reference spectra (1-3).

Pattern analysis means that we deal with a set of reference spectra of each elements as the spectrum pattern and fit the reference spectra to the objective spectra by the linear least squares sense. In this technique, respective X-ray peak, e.g., \( K_\alpha \) and \( K_B \) is not treated separately, as done by the conventional peak analysis technique implemented in computer of all X-ray analysis equipments.

3. APPLICATION TO POOR RESOLUTION X-RAY DETECTORS

The basic data for the model calculation of the reference spectra for the content elements in SUS316 were taken from the tables in the appendices of the reference(5). In the calculation, we considered the range of the resolution of X-ray detectors, actually the FWHM in the Gaussian peak as follows: the lowest end of the range is 100eV, which is better than that of the existing Si(Li) detector of the highest quality, and the upper end of 1000eV, which is worse than that of the existing Si-PIN photo
diode detector as mentioned above. Each elemental response of the detector has been generated by a Monte-Carlo simulation calculation. Counts of $K_{\alpha}$ peak of each element were 100000. The objective spectra for SUS316 were also calculated independently to the above reference spectra. In this calculation, counts of the strongest peak of iron $K_{\alpha}$ were accumulated to $5 \times 10^5$. In Fig. 1 the calculated objective spectra are shown for the cases of five FWHMs by 200eV step. Above 600eV we cannot identify in sight the peak around 4.9 keV which contains Cr $K_{\beta}$ and Mn $K_{\alpha}$ lines.

Pseudo-inverse of the $X$, namely $X^+$ was calculated by a pure mathematical algorithm using the each calculated reference spectra, not by the learning the network as done before. Essentially these two methods give the same results. The pattern analysis results for the objective SUS spectra is obtained just multiplying $X^+$ to the spectra. The objective spectra were also analyzed by the conventional peak fitting method. Both results are summarized in Fig.2 and 3, respectively. The ordinate gives the relative values of the output to the true concentration values. In Fig.2 the pattern analysis technique shows remarkable results; almost all outputs are unity within the statistical uncertainties, and only two results for Mn above 800-eV resolution are a little below by only 5% than unity. The conventional peak fitting technique gives reasonable results only for three abundant elements Cr, Fe and Ni below 500eV but not Mn; above 500eV all results scatter or fluctuate and are unreliable; in this case the fitting failed due to the ill-condition of the problem. This ill-condition was partly solved if we set a constraint to the ratio of $K_{\beta}$ to $K_{\alpha}$ for each element as the known values, as shown in Fig 4. However, the fitting failed again in particular for Mn in the broader width cases.

4. DISCUSSION

As shown above, the pattern analysis method showed good resistivity for the poor-resolution spectra of the X-rays. This can be understood that we deal with two or more X-ray peaks of a single element altogether as a fixed pattern and mutual independence of the reference spectra is relatively high even in the worse resolution case.

The pattern analysis method assume the linearity condition; nonlinear effects such as pileup effect, gain shift, or change of the measurement condition cause unreliable results. If we are taking care about the maintaining the linearity condition in the actual measurement, it can be said that the pattern analysis method enable us use such lower resolution X-ray detectors could be used in various analysis fields and application areas.

References


(4) H. Fukuda, et al.. "Elemental Response Functions for Pattern Analysis in PIXE Spectra", in this proceedings.


<table>
<thead>
<tr>
<th>element</th>
<th>X-ray line energy</th>
<th>average concentration in SUS316b) (%)</th>
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<tr>
<td></td>
<td>keV</td>
<td>K_β /K_α ^a)</td>
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<tr>
<td></td>
<td>K_α</td>
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a) Taken from Ref. (5).
b) Mo (usually contained about 3%) is not included in this table and present model calculations.

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Fig. 1 Calculated objective spectra to be analyzed for the cases of five FWHMs by 200eV step.

Fig. 2 Pattern analysis results for the SUS spectra.
Fig. 3 Analysis results for SUS spectra by the conventional peak fitting method.

Fig. 4 Analysis results for SUS spectra by the conventional peak fitting method with a constraint to the ratios of $K_B$ to $K_a$ (see text).
List of Participants

T.Saitoh, (Seikei University)
A.Zakou, (Free Electron Laser Research Institute)
C.Mori, (Faculty of Engineering, Nagoya University)
M.Nishiza, (Japan Atomic Energy Research Institute)
K.Oda, (Kobe University of Mercantile Marine)
N.Izumi, (Institute of Laser Engineering, Osaka University)
H.Ono, (Seiko EG&G)
T.Iguchi, (Nuclear Engineering Research Laboratory, The University of Tokyo)
K.Tamura, (Aloka Co. Ltd.)
M.Miyajima, (Radiation Safety Control Center, National Laboratory for High Energy Physics)
M.Katagiri, (Japan Atomic Energy Research Institute)
N.Shipoguchi, (School of Science and Engineering, Waseda University)
S.Nagashima, (School of Science and Engineering, Waseda University)
M.Hashimoto, (O-ari Engineering Center, PNC)
H.Zenba, (School of Science and Engineering, Waseda University)
K.Fukuda, (Research Institute for Advanced Science and Technology, University of Osaka Prefecture)
M.Nakazawa, (Faculty of Engineering, The University of Tokyo)
Y.Amemiya, (Photon Factory, National Laboratory for High Energy Physics)
M.Takebe, (Faculty of Engineering, Tohoku University)
N.Ikuta, (Faculty of Engineering, Tokushima University)
K.Sato, (Shimizu Corporation)
T.Suzuki, (Radiation Safety Control Center, National Laboratory for High Energy Physics)
A.G.Prokopets, (Grad. University for Advanced Studies)
M.Okanishi, (School of Science and Engineering, Waseda University)
T.Aoyama, (College of Medical Technology, Nagoya University)
J.Kawarabayashi, (Faculty of Engineering, The University of Tokyo)
T.Shouji, (Tohoku Institute of Technology)
K.Kondoh, (Radiation Safety Control Center, National Laboratory for High Energy Physics)
Yu.G.Zdesenko, (Institute for Nuclear Research in Ukraine)
M.Ukibe, (Faculty of Engineering, The University of Tokyo)
N.Yamanaka, (Faculty of Engineering, The University of Tokyo)
K.Itoh, (Photon Factory, National Laboratory for High Energy Physics)
H.Takahashi, (Faculty of Engineering, The University of Tokyo)
Y.Fukushima, (Department of Physics, National Laboratory for High Energy Physics)
H.Harada, (Faculty of Engineering, Tottori University)
K.Terai, (The Shimane Prefectural Institute for Public Health and Environmental Science)
S.Ban, (Radiation Safety Control Center, National Laboratory for High Energy Physics)
N.Ujiie, (Department of Physics, National Laboratory for High Energy Physics)