Absolute ^{22}Na Radioactivity measurement by gamma efficiency variation of $4\pi\beta$-γ coincidence method

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Abstract

The absolute radioactivity of ^{22}Na was obtained by gamma efficiency variation of $4\pi\beta$-γ coincidence method. Some other previous techniques, such as sum peak gate method based on the positron emission rate, relative measurement with calibrated ionization chambers, and gamma spectrometry with a HPGe detector, were also tried to ensure the present result. The results of these methods were in reasonable agreement with the present absolute measurement. The assayed source solution of this experiment was transferred to NBS type ampoules, and sealed ampoules were sent to the SIR (International Reference System) in BIPM, Taiwan and Indonesia for the international comparison.

Introduction

^{22}Na is the positron emitting nuclide and has a simple decay scheme with relatively long half life of 2.6 years, so that it is widely used in many applications. The decay scheme is shown in Figure 1.[1] Both positron and electron capture process decay to an exited state of 1.275 MeV, except for a very weak positron branch which goes directly to the ground state. Although ^{22}Na has such simple decay scheme, absolute activity measurement is rather complicated. It has been pointed out by Campion[2] that a positron emits two 0.511 MeV annihilation radiation and causes a distortion in observed gamma spectrum, so that the simple coincidence equation is no longer valid. The principal formula of $4\pi\beta$-γ coincidence method is given as

$$N_0 = N_\beta N_\gamma / N_c. \quad (1)$$

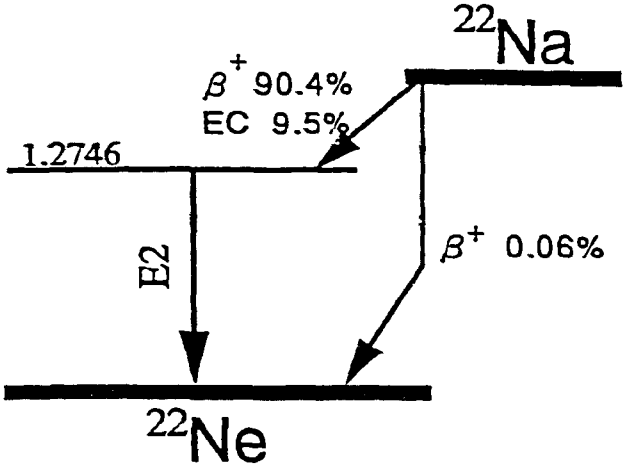

Figure 1 Decay scheme of ^{22}Na.

As for the case of ^{22}Na, positrons are detected with almost 100% of β efficiency, and on the other hand, electron capture events have almost no contribution to the 4πβ detector.

This means the coincidence events will occur only with positron decay. In this case, if gamma gate position is set only to accept the 1.275 MeV peak, some coincidence events of N_c will not be counted because some positron decay events are moved from the 1.275 MeV peak by sum effect.

Since N_c becomes small, apparent activity of N_0 shows larger value to the true activity. On the other hand, if gamma gate is set at the sum peak of 1.786 MeV (1.275 MeV gamma ray + 0.511MeV annihilation radiation), the result of N_0 means just the number of positron decay (about 90% of the total activity). In the previous method, the gamma gate was set at this sum peak and divided apparent N_0 by positron decay probability. However, the accuracy of this measurement has to be depended on the nuclear data of positron emission rate. To solve such problem, we tried gamma efficiency valiation method which is completely independent of the nuclear data.

Measurement of ^{22}Na radioactivity

(i) Coincidence method with gamma efficiency variation

This method changes the gamma efficiency to extrapolate $\varepsilon_\gamma \rightarrow 0$ in the $4\pi\beta$-γ coincidence technique. Figure 2 shows a gamma spectrum measured by the NaI(Tl) scintillator and the gamma gate positions. As shown in the figure, ^{22}Na causes a distortion in observed gamma spectrum by the sum effect of 1.275 MeV gamma rays and 0.511 MeV annihilation radiation. Since the coincidence and sum peak events occur only from positron decay, the electron capture events (which will not cause coincidence) are selectively remained in the 1.275 MeV gamma peak. When the gamma gate position is set at the 1.275 MeV peak, it loses some coincidence events of N_c, so that the apparent N_0 of $N_\beta N_\gamma / N_c$ becomes larger than true value. With the same reason, apparent N_0 becomes smaller when the gamma gate is set to accept the sum events because it will selectively count the coincidence events of N_c. The correction of these effects is rather complicated. It has pointed out by Kawada[3] that the true value of ^{22}Na disintegration rate is given by the extrapolation of the apparent disintegration rate to

zero efficiency of the gamma counter. The probability of occurring sum events are proportional to square of the efficiency of gamma counter, while the count rate of 1.275 MeV peak is simply proportional to the gamma efficiency, so that the contribution of sum effects become smaller with small gamma efficiency. Variation of gamma efficiency is obtained by changing the distance between the beta counter and the NaI(Tl) gamma detector.

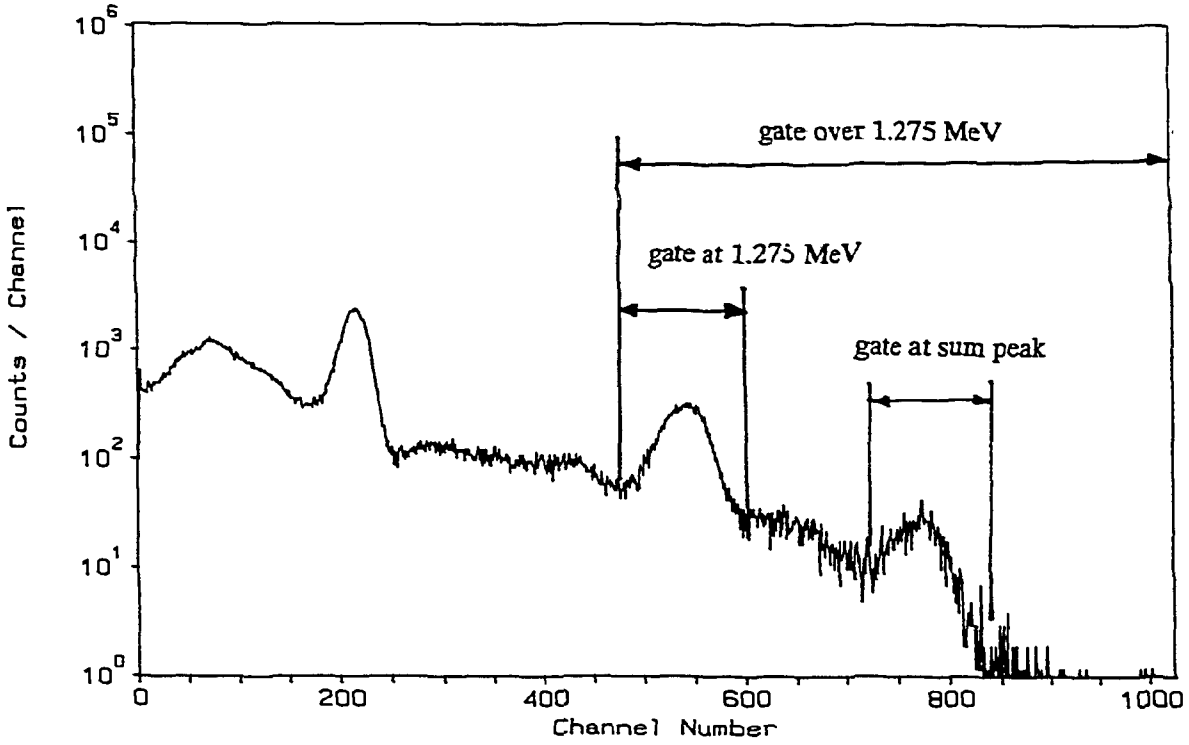

Figure 2 Gamma spectrum of ^{22}Na. Three peaks in the figure are due to 0.511 MeV annihilation, 1.275 MeV gamma ray and sum of these irradiation, respectively.

(ii) Sum peak coincidence method

In this second method, the single channel analyzer of gamma gate is set only to accept the 1.786 MeV sum peak. The counting rates in the individual channel are expressed as

$$N_\beta = N_0[\, p\varepsilon_\beta + (\, 1- p\,)\varepsilon_{EC}],$$
$$N_\gamma = N_0 p\varepsilon_{\gamma(sum)},$$
$$N_c = N_0 p\varepsilon_\beta\varepsilon_{\gamma(sum)}. \qquad (2)$$

Where ε_β and ε_{EC} represent the efficiency of beta counter for positrons and electron capture events, and $\varepsilon_{\gamma(sum)}$ means the efficiency of gamma channel for the sum peak of 1.786 MeV, and p is the number of positrons emitted per disintegration, respectively. For the case of ^{22}Na, energies of X rays and Auger electrons are extremely low, so that

ε_{EC} is negligibly small. In addition, contribution of 1.275 MeV gamma rays to the beta counter is also very small, so that N_β can be treated as $N_0 p \varepsilon_\beta$, and the activity of ^{22}Na will be given directly as

$$N_0 = (N_\beta N_\gamma / N_c)/p. \quad (3)$$

To make sure these assumptions , sources are covered with additional VYNS films to completely stop X rays and Auger electrons, and also the apparent disintegration rate of N_0 were plotted versus $(1-\varepsilon_\beta)/\varepsilon_\beta$ to make extrapolation to $(1-\varepsilon_\beta)/\varepsilon_\beta \rightarrow 0$ which means extrapolate to 100% of the beta efficiency.

(iii) Calibrated ionization chamber

The well type pressurized inoization chambers have been used as the secondary standardizing instrument for gamma emitting nuclides in ETL. These ionization chambers are well calibrated with number of standardized radionuclides which have been absolutely measured by $4\pi\beta$-γ coincidence method. Ionization chambers are coupled with Vibrating Reed Electrometer (VRE) and the stability is checked by measuring the sealed ^{226}Ra source. The total activity of each ^{22}Na ampoule is given as the relative intensity with the electric current from VRE. In this measurement, totally 9 ampoules have been assayed. The stability of such ionization chamber is pretty well, so that many standard laboratories use the similar system for the practical secondary standard. The SIR in BIPM has also a same type ionization chamber of ETL, and up to now, many standard sources have been sent to the SIR in BIPM to establish the international traceability.

(iv) Gamma ray spectrometry

A coaxial n type HPGe detector with beryllium window has been used for the impurity check and secondary standardization in ETL with spectrometric method. The detector has been well calibrated with a series of gamma standard sources. The efficiency curve for point sources at 14 cm distance is shown in the Figure 3. ^{22}Na sources dropped on the plastic disks were measured by this HPGe spectrometer.

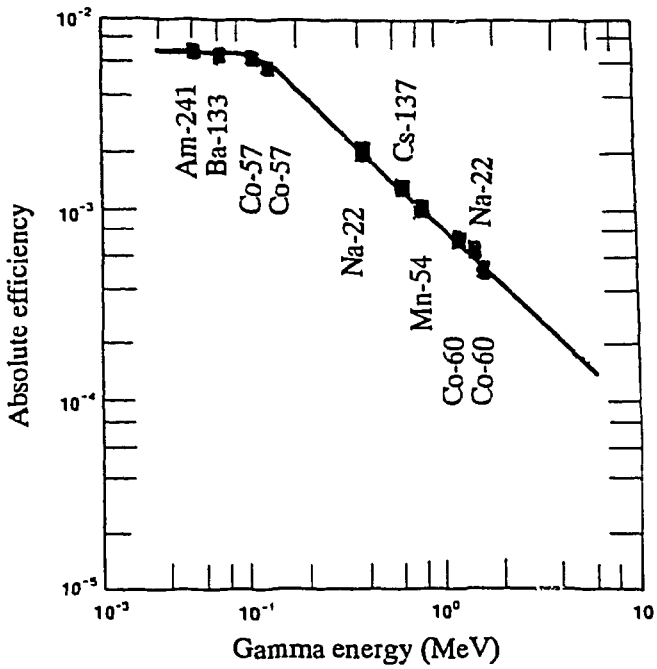

Figure 3 Efficiency curve of HPGe detector at 14 cm position.

Results of the measurements

Figure 4 shows the results of the gamma efficiency variation method. As described before, each line shows the typical slope and focuses at 525.5 kBq/g within 0.2% disagreement.

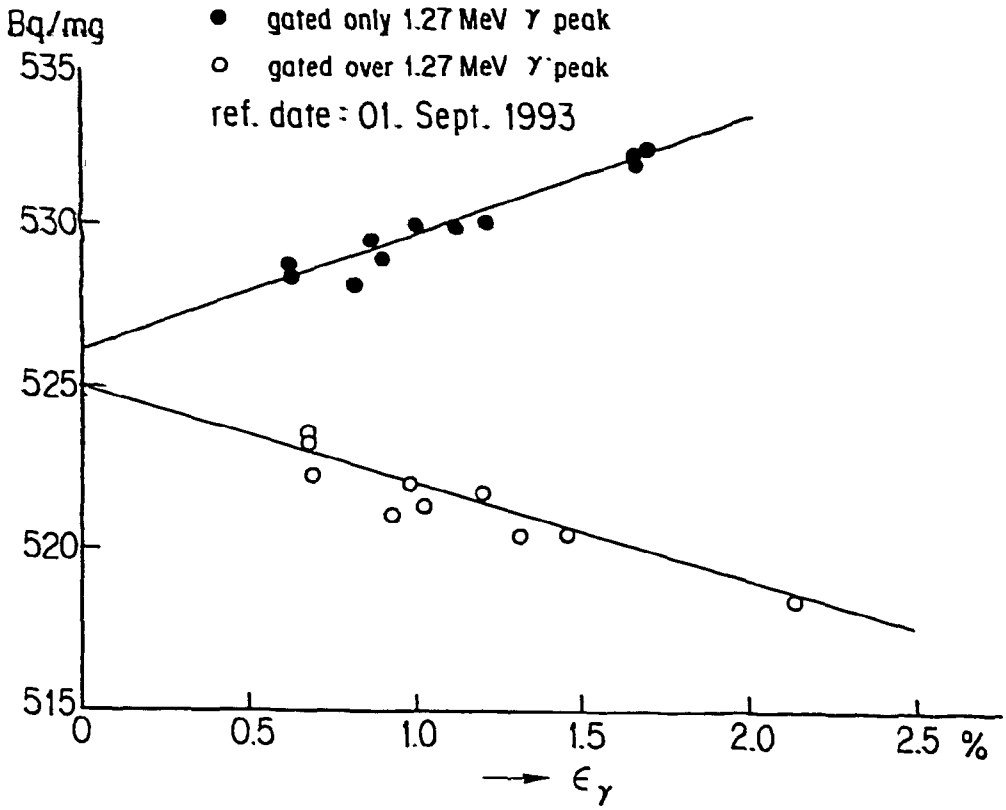

Figure 4 Results of gamma efficiency variation method. The upper curve shows the results from 1.275 MeV gate position, and lower curve shows the results from the gate over the 1.275 MeV peak as shown in the figure 2.

The result from the sum peak gate method is shown in the figure 5 together with all the present results. In this measurement, sources were covered additional VYNS film to stop X rays and Auger electrons from electron capture decay. Apparent disintegration rates of N_0, which were obtained from the formula (3) with the p value evaluated as 0.904[1], were plotted with $(1-\varepsilon_\beta)/\varepsilon_\beta$. As shown in this figure, the slope of extrapolation to $(1-\varepsilon_\beta)/\varepsilon_\beta \rightarrow 0$ is very flat which means the sensitivity of beta counter to the gamma events are very small. The result of this method is 524.7 kBq/g, which is in very good agreement with the gamma efficiency variation method within 0.15%.

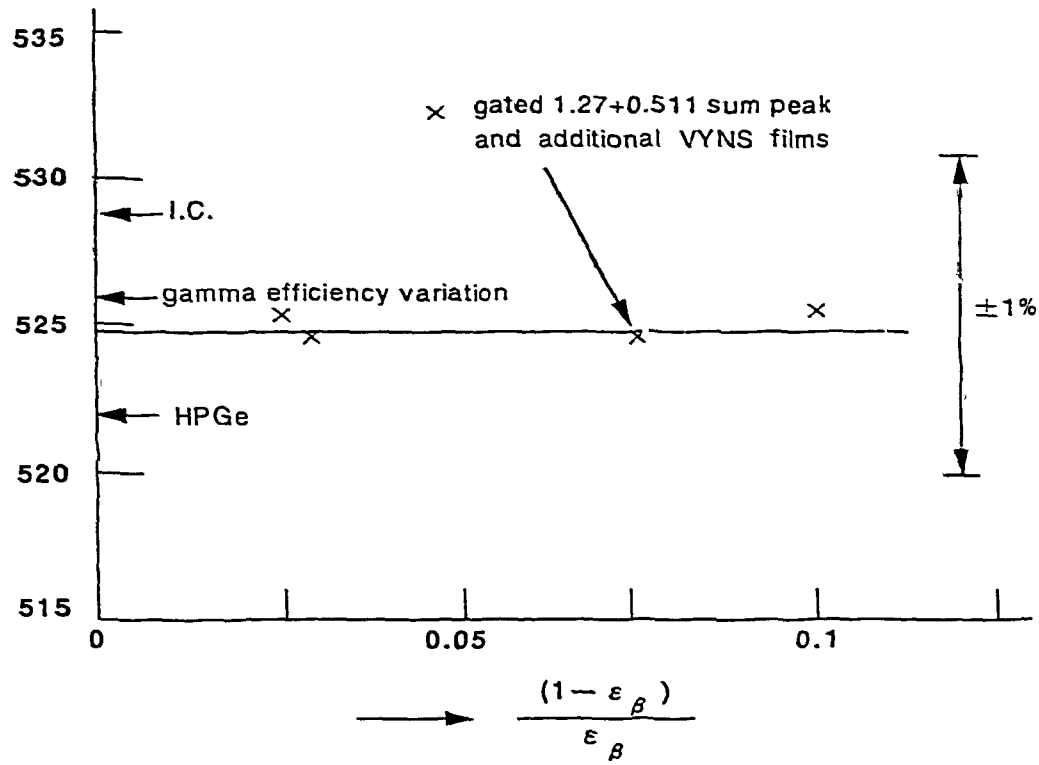

Figure 5 Results from sum peak method compared with other present results

The results of relative measurements are also in good agreement. Totally 9 ampoules have been measured with the calibrated ionization chamber and the averaged value of 529.5 kBq/g is slightly higher to that of coincidence method, however it is still within 1% difference.

The gamma sources of 5 plastic disks were measured by the HPGe detector at 14 cm distance from the surface of the detector. Each sample was measured 3600 seconds, and activities were derived from the efficiency curve of the figure 3. The result of this HPGe spectroscopy method is also in good agreement as shown in the figure 5.

Conclusively, the radioactivity measurements on ^{22}Na have been successively done. Especially the absolute measurements of gamma efficiency variation and sum peak gate methods are in very good agreement. The final value of these measurements, 525.2 ± 2.6 kBq/g has been derived from these result. The results from calibrated ionization chamber and HPGe detector are also in good agreement within 1%.

Registration to the SIR

One of the NBS type ampoule was sent to the SIR in BIPM to ensure the present results and contribute to establish the international traceability. The SIR has a pressurized ionization chamber and sealed ^{226}Ra source, which is the same system of

secondary standardizing system for gamma emitting nuclide in ETL. Many national laboratories, which have the role of standardization, send gamma emitting nuclides to the SIR. The SIR measures these sources and registers the equivalent values to produce same electric current with the ^{226}Ra source. These values are registered and all the national laboratory can access this reference system. Figure 6 shows the present result which has been registered to the SIR. The agreement is very good and the ionization chambers of ETL are re-calibrated with this result, and domestic traceability is also maintained by sending the JRIA type ampoule sources.

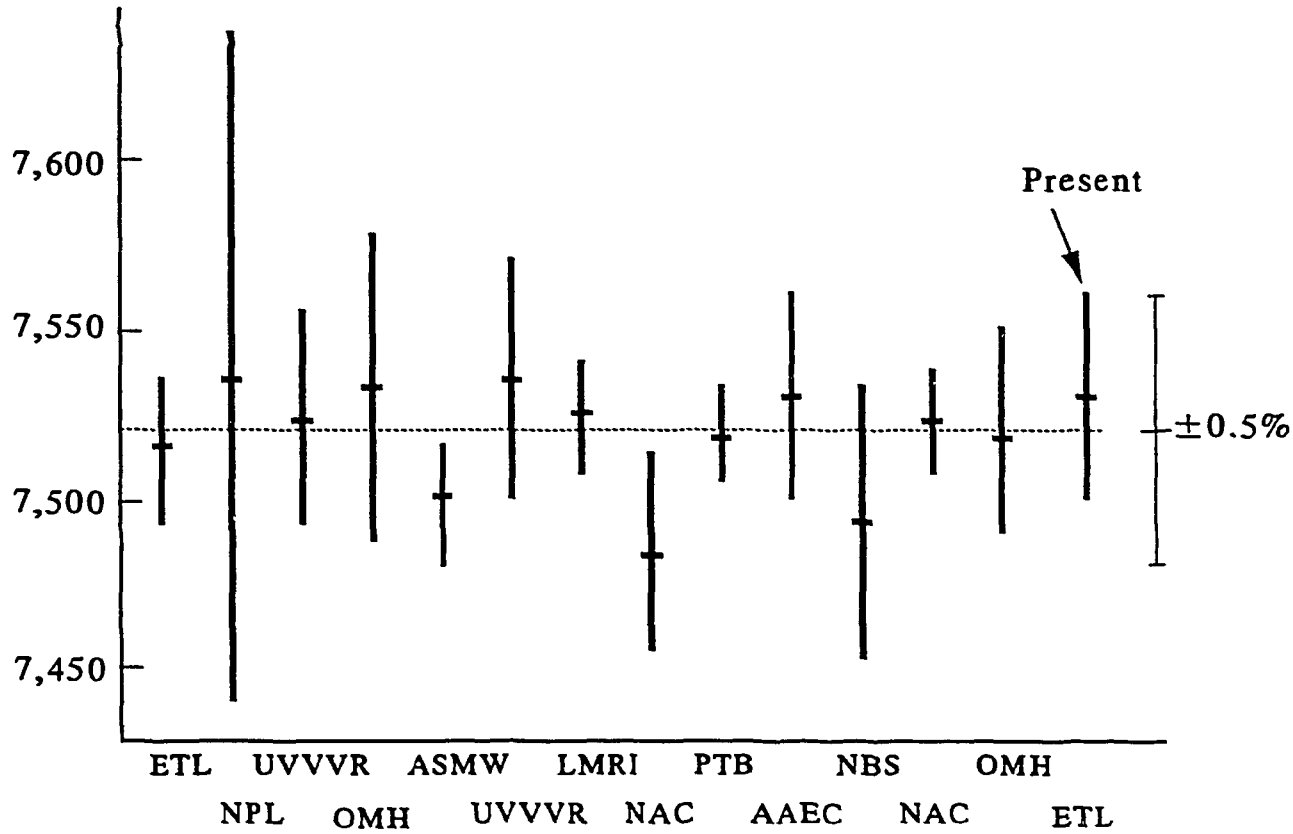

Figure 6 Result of registered values to the SIR.

In this figure, acronyms of laboratory and registered years are as follows;
ETL (Electrotechnical Laboratory, Japan, 1976),
NPL (National Physical Laboratory, England, 1976),
UVVVR (now is IIR, Inspectorate for Ionizing Radiation, Czech Republic, 1977 and 1979),
OMH (National Office of Mesures, Hungary, 1977 and 1985),
ASMW (now is in PTB, East Germany, 1977),
LMRI (now is LPRI, Laboratorie Primarie des Rayonnements Ionisants, France, 1979),
NAC (National Accelerator Center, South Africa, 1980),
PTB (Physicalish Technishe Bundesanstalt, Germany, 1980),
AAEC (Australian Atomic Energy Commission, Australia, 1981),
NBS (now is NIST, National Institute of Standards and Technology, U.S.A., 1983)

References

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3) Y. Kawada : Extended application and improvement of $4\pi\beta$-γ coincidence method in the standardization of radionuclides, ETL, Japan, 730 (1972).