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A FIELD-DEPLOYABLE GAMMA-RAY SPECTROMETER UTILIZING XENON AT HIGH PRESSURE

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A FIELD-DEPLOYABLE GAMMA-RAY SPECTROMETER UTILIZING XENON AT HIGH PRESSURE*

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ABSTRACT

Prototype gamma-ray spectrometers utilizing xenon gas at high pressure, suitable for applications in the nuclear safeguards, arms control, and nonproliferation communities, have been developed at Brookhaven National Laboratory (BNL). These spectrometers function as ambient-temperature ionization chambers detecting gamma rays with good efficiency in the energy range 50 keV - 2 MeV, with an energy resolution intermediate between semiconductor (Ge) and scintillation (NaI) spectrometers. They are capable of prolonged, low-power operation without a requirement for cryogenic fluids or other cooling mechanisms, and with the addition of small quantities of ^{3}He gas, can function simultaneously as efficient thermal neutron detectors.

INTRODUCTION

Requirements and Existing Capabilities Practically all of the nuclear materials of importance in the nuclear safeguards, arms control, and nonproliferation regimes emit gamma rays which constitute a characteristic signature for that material, and can be used to detect its presence and determine its properties. So far, two general categories of spectrometers have been available to carry out these measurements:

1. Semiconductor detectors, usually germanium, which have excellent energy resolution (typically better than 2 keV at an energy of 1 MeV) but which must be maintained at a low temperature with the use of liquid nitrogen or mechanical cooling systems.

2. Scintillation detectors, usually sodium iodide (NaI), which have the advantages of portability and ambient-temperature operation, but which suffer the disadvantage of poor energy resolution (for example, typically 10 percent, or 40 keV at an energy of 400 keV).

New and expanded requirements for gamma-ray measurements on nuclear materials point to the need for a detector which can be widely deployed and function independently for protracted periods of time, if necessary, in a wide range of environments, with an energy resolution considerably superior to that of a scintillation spectrometer.

The High Pressure Xenon Ionization Chamber Recently, in the physics research community, a gamma-ray detector capable of meeting these requirements has been developed. It is an ionization chamber which utilizes xenon gas at very high pressure (60 atm). In this device the energy of a gamma ray which has stopped in the xenon is determined by collecting and measuring the number of electrons liberated in the gas by this ionizing event. Because of the high density of the xenon and its high atomic number (Z=54), and its superior energy resolution, its sensitivity for detecting individual gamma rays is in the same range as that of a sodium iodide scintillation spectrometer. Unlike the scintillation spectrometer, the xenon ionization chamber requires very little operating power, and it should possess a fairly wide operating envelope (temperature, pressure, mechanical ruggedness, etc.) so that it is suitable for prolonged, unattended operation in a wide range of environments.

Prior Developments So far, detectors of this type have been developed individually at various laboratories in connection with specific research goals. The original development was carried out at the Moscow Engineering and Physics Institute by V. V. Dmitrenko et al.[1] and produced an ionization chamber which was installed in the "MIR" space station and employed for gamma-ray astronomy research. As of July 1996, this device has been in operation for 5 years with no degradation of its performance. In the United States similar detectors have been fabricated at Yale University by Markey and Levin[2], by Losee and others at the Naval Command, Control, and Ocean Surveillance Center (NRaD), and recently by Bolotnikov at the Marshall Space Flight Center in Huntsville, Alabama. All of these devices have achieved comparable values for the energy resolution, approaching 20 keV for the 661-keV ^{137}Cs gamma ray.

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BASIC PRINCIPLES

Chamber Configuration As an ionization chamber, the spectrometer measures the energy of a gamma ray by collecting and measuring the electric charge (electrons) released in the xenon gas by an interaction of the gamma ray with a xenon atom. While several types of interaction may occur (photoelectric effect, Compton scattering, etc.) only the photoelectric effect, in which the entire energy of the gamma ray is absorbed by one xenon atom, will produce an identifiable, individual spectrum peak. Consequently, the spectrometer efficiency at a given energy will be essentially proportional to the cross section for photoelectric gamma-ray absorption at that energy.

The BNL chambers have a parallel-plate configuration, i.e., the gas in the sensitive region is situated between two parallel electrodes, the cathode and anode. To collect the electrons at the anode, an electric field of at least one kilovolt per centimeter (kV/cm) is maintained between the electrodes. This value is chosen so as to provide for efficient charge collection, but not so high as to produce an avalanche in the gas, because statistical fluctuations in the multiplication process lead to a degradation in the energy resolution. In the absence of charge multiplication, the total number of electrons collected in a typical event is relatively small (e.g., 30,000 electrons for the 661-keV gamma ray of ^{137}Cs, and consequently, careful attention must be given to preamplifier design and to minimizing electronic and microphonic noise.

At optimum xenon densities (0.5 - 1.0 g/cm^3), the electron drift velocity is small, typically in the vicinity of 1 mm/μsec. Accordingly, the electron transit time may range up to 50 μsec. For a chamber configured with only a cathode and anode, the detector response would have a correspondingly long rise time, leading to problems with pulse pileup, increased electronic noise, and a position-sensitive signal. To avoid these problems, an additional "Frisch" grid is placed close to the anode. This serves to shield the anode from the electric field of the drifting electron cloud so that no signal appears until the electrons pass through the grid into the small space between the two electrodes, and accordingly, a signal with a short (typically about 10 μsec) rise time ensues. A schematic diagram illustrating this chamber configuration is shown in Figure 1.

Intrinsic Energy Resolution The intrinsic energy resolution of the spectrometer (independent of the electronic noise introduced by the circuitry) is determined by the statistics of the electrons collected for an event of a given energy. If the ionization process were totally random and governed by Poisson statistics, then the variance of the number N of electrons collected would be given by the usual expression

$$\sigma^2 = N.$$

Because the gamma ray deposits a fixed amount of energy in the gas and there is thus a constraint upon the number of electron-ion pairs produced, the variance is reduced. This is expressed by an empirical coefficient, the Fano factor F, defined by

$$F = \frac{\sigma^2_{(observed)}}{N}$$

Experimental Fano factors obtained for xenon gas depend upon the density. They range from the value of 0.13 at one bar (variance approximately 7 times smaller than the Poisson value) to approximately 1 (the Poisson value) at a density of 1 g/cm^3.

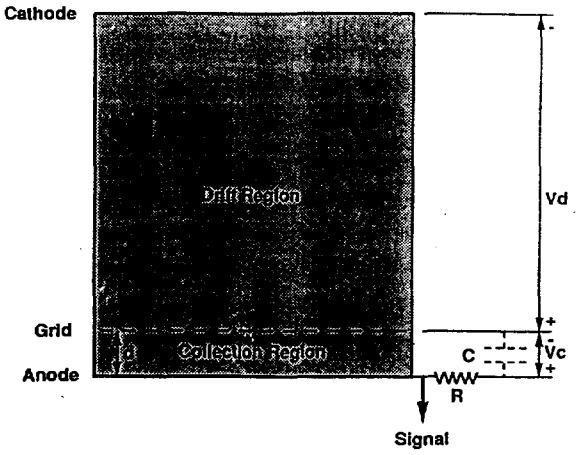

FIGURE 1. SCHEMATIC DESIGN OF A GRIDDED IONIZATION CHAMBER

Gas Purity and Ultra-High Vacuum Requirements Because of the high gas density and low electron velocities, the electrons being collected will undergo many collisions with other particles before reaching the anode. Accordingly, the concentration of electronegative impurities (e.g., O_2, H_2O, halogens, etc.) in the gas must be maintained at extremely low levels, of the order of 10^{-12}, to minimize the rate of attachment of electrons to these impurity molecules. This requires the adoption of rigorous ultra-high vacuum techniques to remove impurities before the chamber is filled with xenon, the use of only metals or ceramics in the fabrication of the ionization chamber, and the thorough purification of the (initially research grade) xenon gas with a hot metal getter or some other method. Since the attachment of electrons to impurity molecules takes place in a three-body collision, the rate is proportional to the square of the gas density, and hence the degradation of the energy resolution is more pronounced for higher densities.

THE BROOKHAVEN SPECTROMETERS

Configuration Two gamma-ray spectrometers utilizing xenon at high pressure have been constructed and put into operation at BNL The first of these (Mark I) was designed as a laboratory prototype device testing over a range operating parameters to provide an understanding of its operating characteristics and to permit the optimization of its performance. The second (MARK II), incorporating some new features derived from experience with the laboratory prototype, has been designed to be field-deployable, i.e., it will be possible to transport this device to various DOE sites, demonstrate its performance, and evaluate its suitability for various applications at the site.

Figure 2 shows the top flange and internal structures of the Mark I laboratory prototype spectrometer. These are, starting at the top, the cathode, two field-defining electrodes, Frisch grid, and anode. The active volume of the chamber contained within the electrodes is a cylinder 5.3 cm in length and 7.5 cm diameter, approximately 300 cm^3. The internal structures in the Mark II spectrometer are essentially identical.

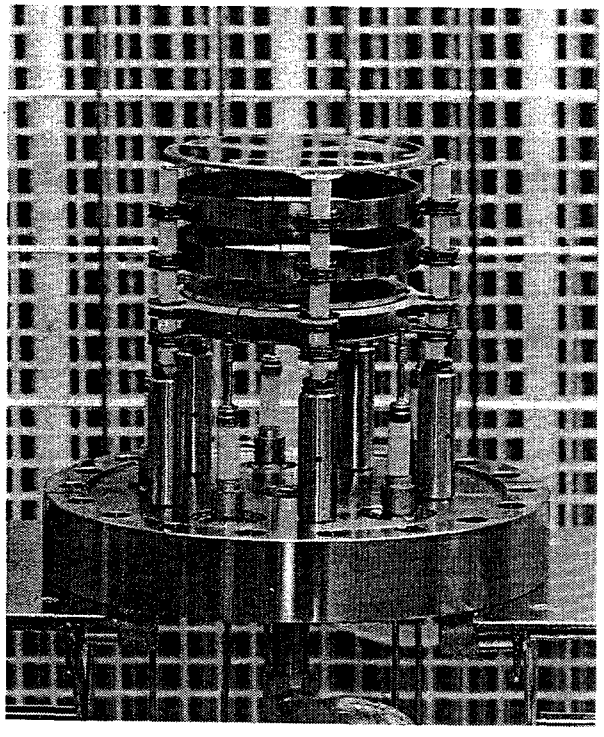

FIGURE 2. INTERNAL STRUCTURES OF THE MARK I LABORATORY PROTOTYPE SPECTROMETER

Figure 3 shows an exterior view of the assembled Mark II spectrometer. Devices attached to the top flange include a vacuum/high pressure valve, a pressure transducer and relief device, and 20-kilovolt (kV) vacuum feedthrus which provide the necessary electrical potentials to the internal electrodes. The chamber is fabricated from a light, high-strength titanium-vanadium-aluminum alloy (Ti_6Al_4V) which combines a high strength/weight ratio with good transmission for gamma rays. The assembled spectrometer, filled with xenon, weighs approximately 10 kg.

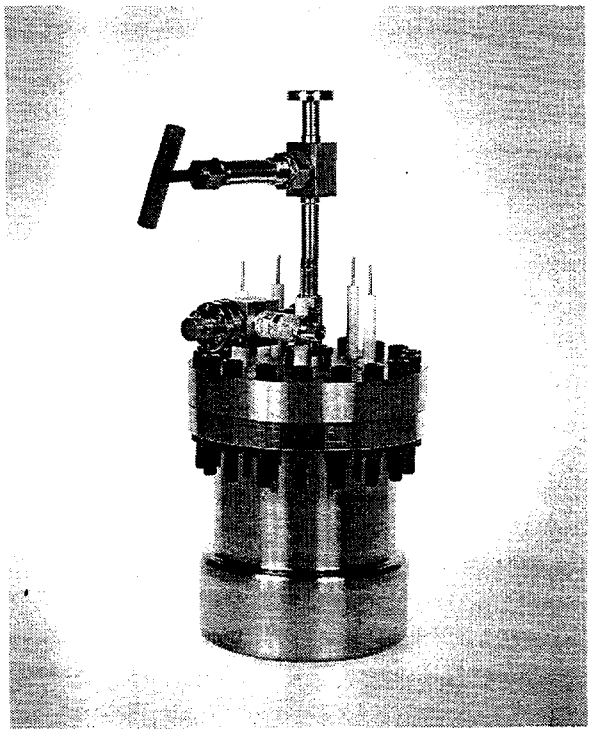

FIGURE 3. ASSEMBLED MARK II GAMMA-RAY SPECTROMETER

Energy Resolution A typical example of the performance of this spectrometer is shown in Figure 4, where the spectrum of a ^{133}Ba source is displayed. This spectrum was obtained for a xenon gas density of approximately 0.4 g/cm^3. The individual ^{133}Ba peaks are seen to be well-resolved; it was possible to obtain accurate areas for each peak. The energy resolution obtained for the 356 keV peak is 15 keV, a three-fold advantage over a scintillation spectrometer. Modest further improvements in the energy resolution are possible with the adoption of ultra-low noise preamplifiers which have been developed at BNL.

Other Performance Parameters In addition to performance parameters such as the energy resolution and detection efficiency, it is also of interest to determine other parameters such as the electron drift velocity and lifetime in the xenon gas. To

accomplish this, a diagnostic probe which utilizes the 511-keV positron annihilation gamma rays from ^{22}Na was devised. This is shown in Figure 5. A gamma ray emitted in the backward direction from the source passes through a narrow lead collimator to a scintillation detector. This produces a signal which indicates that another 511-keV gamma ray has gone in the opposite direction into the chamber, and provides a start signal indicating the time of this event. In this manner a narrow (2 mm diameter) gamma-ray probe is generated which can be used to determine the drift time required for the electrons to reach the anode and to determine any attenuation of the electron cloud arising from attachment to impurity molecules for any region of the active volume.

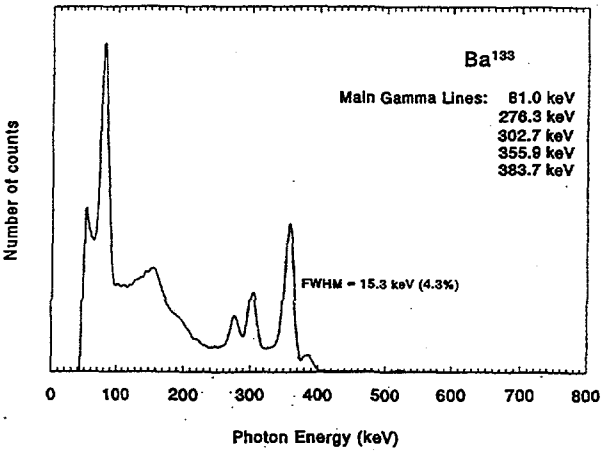

FIGURE 4. GAMMA-RAY SPECTRUM OF A ^{133}Ba SOURCE

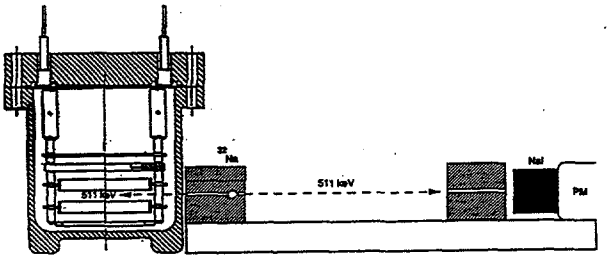

FIGURE 5. DIAGNOSTIC PROBE UTILIZING ANNIHILATION RADIATION FROM ^{22}Na.

Some typical results of this diagnostic approach are shown in Figure 6, where the measured electron drift velocity is shown as a function of the applied electric field. For comparison, some prior results obtained in Russia are also shown.

Light Gas Additives While the electron drift velocity in pure xenon gas is determined by the applied electric field, reaching, typically, a value of 1 mm/μsec for fields of about 1 kV/cm, it is possible to obtain a further increase in the drift velocity with the addition of small percentages of either helium or hydrogen. Previous work[1] has shown that the addition of approximately one percent of hydrogen will lead to a five-fold increase in the drift velocity. This is advantageous both from the standpoint of minimizing losses of electrons from attachment to impurity molecules, and in permitting the spectrometer to function at higher counting rates. If approximately one atmosphere of ^{3}He is added to the xenon, the spectrometer will have a substantial detection efficiency for thermal neutrons, and thus can function simultaneously as a gamma-ray spectrometer and thermal neutron detector. Both of these approaches will be investigated with the Mark I spectrometer at BNL.

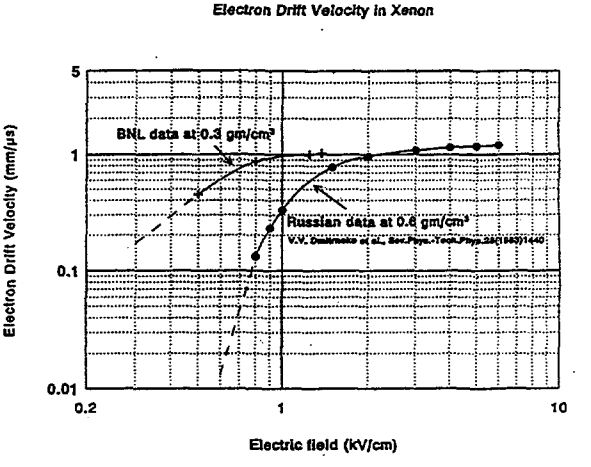

FIGURE 6. ELECTRON DRIFT VELOCITY IN XENON AS A FUNCTION OF APPLIED ELECTRIC FIELD

Integration with the MIMS Network The DOE's Office of On-Site Monitoring, in the Office of Research and Development of the Office of National Security and Nonproliferation has established the Modular Integrated Monitoring System (MIMS) program to develop, evaluate, and demonstrate the use of secure, unattended, ground-based monitoring systems to detect activities associated with the proliferation of weapons of mass destruction. As part of its tasking for the development of the xenon gamma-ray spectrometer, BNL was requested to provide for the integration of the spectrometer into the MIMS network.

Under the MIMS program, Sandia National Laboratories (SNL) has developed a communication network based on Lonworks technology, including a data logging computer and a SNL-developed interface. At BNL hardware was acquired and software created to acquire and process data from the spectrometer and transmit it to the MIMS network. The software enables the detector to operate independently, commencing operation and acquiring data in response

to an external alarm or signal. The information transmitted to the MIMS network consists of a ten-channel "packet" which contains normalized peak areas of principal gamma rays of plutonium, americium, and uranium. Data of this type have been acquired with the spectrometer, processed, and transmitted to SNL.

Field Deployability The Mark II spectrometer is currently being incorporated into a "package" which includes the spectrometer, preamplifier, power supplies, etc., and which provides a dry, hermetic enclosure for the high voltage circuitry. This, together with a Canberra Industries "Inspector" portable multi-channel analyzer and IBM Thinkpad laptop computer constitute a complete, portable spectrometry system, weighing less than 20 kg, which can be transported to DOE sites for demonstration and evaluation of its performance in potential applications. To satisfy regulations concerning the shipment of pressure vessels, the system will be transported in a second, larger "overpack" designed to contain the xenon gas at a pressure of 200 psi in the event that the gas is vented, for any reason, from the spectrometer chamber. This approach will permit the shipment of the spectrometer by any means, including transportation on passenger aircraft.

Potential Uses The particular features of the spectrometer suggest a number of potential uses:

- At distances ranging up to 100 meters, the sensitivity and energy resolution of the spectrometer are sufficient for the detection, in a few minutes, of an object containing several kilograms of plutonium. Furthermore, the energy resolution is sufficient so that by means of a comparison of individual gamma-ray peaks, the isotopic composition of the material can be established, i.e., weapons grade plutonium can be distinguished from that produced in a civilian power reactor.

- While existing SNM portal monitors include a radiation detector with sensitivity greater than that of the high pressure xenon-filled ionization chamber, they usually have little, if any, spectroscopic capability. At a facility where various types of material may exit the site, the spectrometer, employed together with a portal monitor, could determine quickly, after an alarm, what type of material generated the alarm, e.g., plutonium, uranium, fission products, etc.

- Experience with the BNL CIVET (Controlled Intrusiveness Verification Technology) concept and equipment has demonstrated that each individual type or model of nuclear explosive device emits a unique gamma-ray "fingerprint" which serves to identify that device and distinguish it from other devices or items containing fissile materials. Similar fingerprints exist for other items containing nuclear materials. In many instances this may serve to satisfy the DOE requirement for a confirmatory measurement on plutonium (and in favorable cases, high-enriched uranium). Ordinarily, a high purity germanium (HPGe) gamma-ray spectrometer would be employed for this purpose, but under conditions where this is not convenient or feasible, for example, at remote locations, the xenon gamma-ray spectrometer will suffice for this purpose.

- An item of current concern is the continuous monitoring of a number of items stored in a vault or other storage location. The sensitivity and specificity of the xenon gamma-ray spectrometer for individual gamma rays is such that it should be able to generate an alarm in a few minutes if one out of a substantial number of items is removed from its normal, designated location. The superior energy resolution precludes "spoofing" of the system with the use of other radioactive materials.

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