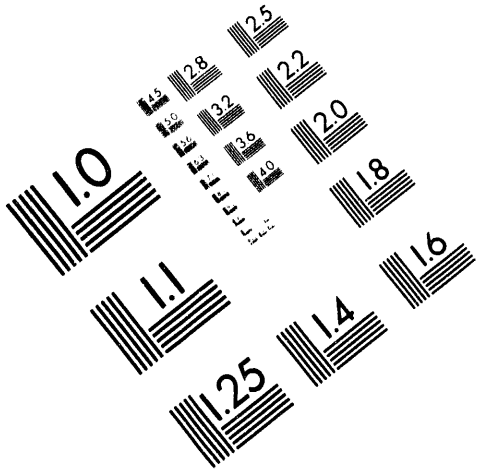

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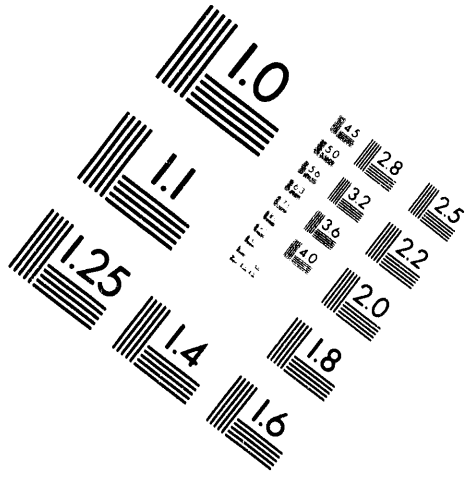

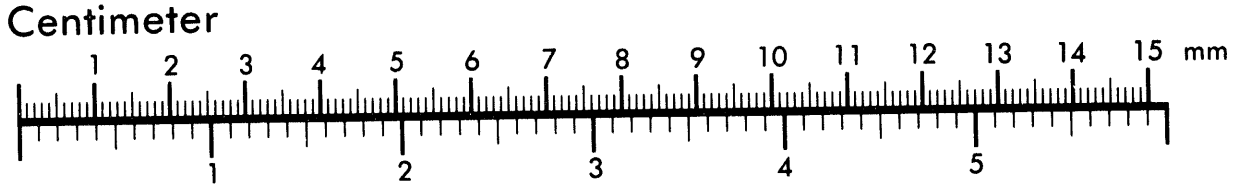

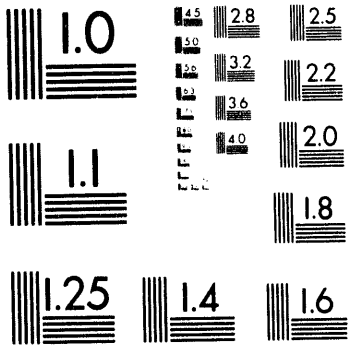

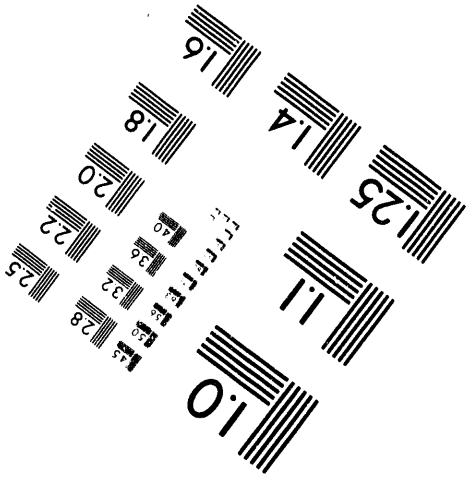

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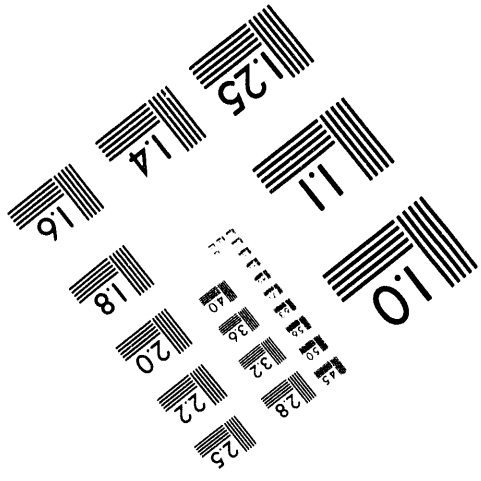

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GAMMA-RAY SPECTROMETER UTILIZING XENON AT HIGH PRESSURE*

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ABSTRACT

A prototype gamma-ray spectrometer utilizing xenon gas near the critical point (166° C, 58 atm) is under development. The spectrometer will function as a room-temperature ionization chamber detecting gamma rays in the energy range 100 keV-2 MeV, with an energy resolution intermediate between semiconductor (Ge) and scintillation (NaI) spectrometers. The energy resolution is superior to that of a NaI scintillation spectrometer by a substantial margin (approximately a factor 5), and accordingly, much more information can be extracted from a given gamma-ray spectrum. Unlike germanium detectors, the spectrometer possesses the capability for sustained operation under ambient temperature conditions without a requirement for liquid nitrogen.

INTRODUCTION

The isotopes of uranium and plutonium emit gamma rays which afford a unique signature for each particular material, and often, for material in a particular configuration. The detection and measurement of these gamma rays has major potential utility in the areas of nonproliferation, the implementation of arms control agreements, and in nuclear safeguards.

Some specific applications for the detection and measurement of gamma rays are:

•The detection, with appropriate devices and methodology, of kilogram quantities of fissile materials, in particular, plutonium, at distances up to 100 m. In this respect it should be noted that the alternative fissile material, ^{233}U, produced by the reactor irradiation of thorium, emits a penetrating 2.614 MeV gamma ray which provides a characteristic signature for this material.

•The measurement of uranium enrichment, or of the isotopic composition of a plutonium sample. The latter measurement readily distinguishes between plutonium produced as a byproduct of the generation of electric power and that produced specifically for use in nuclear explosive devices.

•The measurement of a specific pattern of gamma-ray energies and intensities from fissile materials in a particular quantity and configuration, most importantly, from a particular type and model of nuclear explosive device. This information can be used to confirm the identity of the device and distinguish it from other categories of devices or from other nuclear materials.

•The remote monitoring of nuclear materials in storage.

*Work performed under the auspices of the U.S. Department of Energy, Contract DE-AC02-76CH00016.

•The identification or confirmatory measurement of a particular item of nuclear material by determining its "gamma-ray fingerprint."

Essential to all of these potential uses is an appropriate gamma-ray spectrometer. So far, two general categories of devices have been available:

1. Semiconductor detectors, usually germanium, which possess excellent energy resolution (typically, better than 2 keV at an energy of 1 MeV), but which must be maintained at a low temperature. At fixed installations, where liquid nitrogen is readily available, these detectors are the device of choice for the gamma-ray spectrometry of nuclear materials.

2. Scintillation detectors, usually sodium iodide (NaI), which have the advantage of robustness and portability, but which suffer the disadvantage of poor energy resolution (for example, typically 10 per cent or 40 keV at 400 keV). Until now, these two categories of device have been the only practical choice for gamma-ray measurements in the field.

New and expanded requirements for gamma-ray measurements on nuclear materials clearly point to the need for a detector which can be widely deployed and function independently for protracted periods of time, with an energy resolution considerably superior to that of a scintillation spectrometer so that gamma-ray spectra from nuclear materials can be acquired and evaluated quantitatively.

Recently, in the physics research community, a gamma-ray detector capable of fulfilling these requirements has emerged. 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 gas is determined by collecting and measuring the amount of charge liberated in the gas by this ionizing event. Because of the high density of the xenon and its high atomic number, the detection efficiency of the device for gamma rays is in the same range as that of a sodium iodide scintillation spectrometer. The energy resolution, typically 15 keV for 661 keV gamma rays, while not approaching that of a germanium detector, is superior to that of sodium iodide by a factor of 4 to 5, sufficient to enable the device to be utilized in most of the applications cited above. In addition, since ionization chambers characteristically require much less power than scintillation detectors, the device is capable of prolonged operation for extended periods of time in remote locations.

So far, gamma-ray detectors utilizing xenon gas at high pressure have been developed within the physics research community only in connection with specific research projects. Although the performance achieved by those already developed clearly points to their wider utility, they are not available on a wider basis. Accordingly, the Office of Nonproliferation and National Security of the Department of Energy has requested Brookhaven National Laboratory (BNL) to proceed with the further development of this type of detector so that it can be deployed and utilized in nonproliferation activities. Starting in March of 1993, BNL's Instrumentation Division has been engaged in the development of a prototype xenon-filled gamma-ray spectrometer. There are three stages to this development project:

1. The spectrometer will serve first as a test bed for establishing performance parameters and for investigating certain research issues.

2. The chamber will be incorporated into a demonstration device which can be utilized at BNL or other sites to establish its capabilities and utility in addressing various problems in the nonproliferation regime.

3. A fully field-deployable gamma-ray spectrometer with the capability to function for protracted periods of time under a wide variety of conditions will be fabricated.

BASIC PRINCIPLES

The ionization chamber has a parallel plate design, i.e., the gas in the sensitive region is situated between two parallel electrodes maintained at a potential difference in excess of 1 kV/cm. When an ionizing event occurs in the sensitive region creating a cloud of electron-ion pairs, the electrons are collected at the anode and provide a signal whose amplitude is a measure of the gamma-ray energy.

Because of the high gas density (0.5 to 1.5 g/cm^3, the drift velocity of the electrons is small and typically, they require 50 to 100 μsec to traverse the length of the chamber. The long signal rise times that result are undesirable in terms of pulse pileup, electronic noise, and a dependence of the signal upon the point of origin of the ionizing event in the chamber. To produce a shorter rise time and minimize these effects, an additional grid (Frisch grid) is placed near the anode. The effect of the Frisch grid is to shield the anode from the field of the drifting electron cloud, so that the signal does not begin until the electrons have entered the collection region between the grid and anode. This type of configuration is illustrated in the schematic diagram of Figure 1. Ionization chambers employed for high resolution spectroscopy customarily have this design.

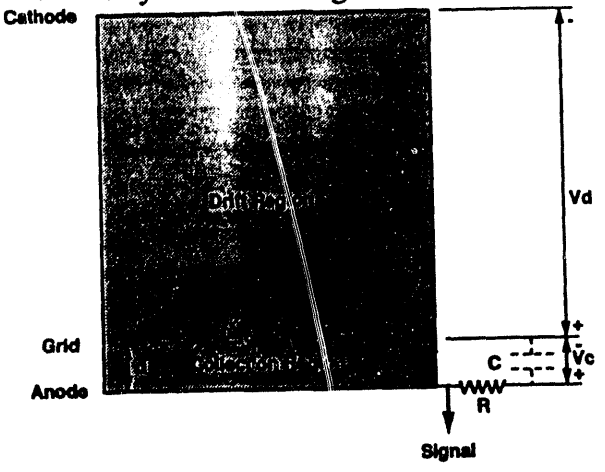

Figure 1. Schematic diagram of Gridded Ionization Chamber.

The average energy required to create a single electron ion pair in gaseous xenon is comparatively small (21.9 eV), so that the number of such pairs created will be large for typical gamma rays of interest (for example, N = 19,000 electron-ion pairs will be produced by the 414 keV gamma ray of ^{239}Pu). If the ionization process were totally random and governed by Poisson statistics, then the statistical variation in the number N would be given by the usual Poisson expression

$$\sigma^2 = N$$

where σ^2 is the variance of N. For an ionizing event in which a fixed quantity of energy is deposited in the gas, however, such as the total absorption of the 414 keV gamma ray of ^{239}Pu, the deposition of this fixed energy places a constraint on the number N of pairs produced, so it will not typically obey Poisson statistics. Since, for gaseous xenon, approximately half of the energy absorbed creates ionization, and the remainder is taken up in other modes of excitation in the gas, with an inherent randomness, there will be some residual variability in the quantity of charge produced in a given event. This is customarily quantified by an empirical coefficient, the Fano factor[1] F, defined by

$$F = \frac{\sigma^2\,(\text{Observed})}{\sigma^2\,(\text{Poisson})} = \frac{\sigma^2\,(\text{Observed})}{N}$$

For many radiation detectors, the Fano factor is considerably less than unity, and the energy resolution consequently better than what would be obtained if the charge generated obeyed Poisson statistics.

For gaseous xenon at a pressure of 1 atm the Fano factor has been determined to be 0.13. Measurements of the energy resolution obtained for ionization chambers filled with xenon at higher pressures indicate the Fano factor increases steadily with pressure approaching unity at the critical pressure[2,3].

Electrons drifting through xenon at high pressure will undergo many collisions with gas atoms, and several different interactions may occur which will influence the performance of an ionization chamber. These are, in particular, electron-ion recombination

and the attachment of electrons to impurity molecules. To minimize these effects, it is necessary, in the construction of a high-performance chamber, to maintain an extremely low level of electronegative molecules in the xenon gas, typically at a concentration of less than 10^{-10}. To achieve this requirement it is necessary to evacuate the system to the vicinity of 10^{-10} Torr and to achieve outgassing rates of 10^{-12} Torr liter/sec, and carefully purify the xenon charge with the use of a metallic "getter" at a high temperature.

Although most gaseous impurities will degrade the performance of the chamber, it has been found that the addition of small admixtures of either hydrogen or helium (approximately 1%) to the xenon has a pronounced effect on the electron drift velocity, increasing its value by as much as a factor of five[3]. This is highly desirable in its effect on the signal rise time and other performance parameters.

PRIOR WORK

Ionization chambers utilizing xenon gas at high pressure have been developed and put into operation at three different laboratories.

In the United States the definitive work on the development of these devices has been carried out at Yale University by J. Markey and C. Levin[2]. In this work extensive investigations were carried out of the effect of the chamber configuration and various operating parameters, including gas pressure, on the chamber performance. For a chamber operating at high pressure and gas density (62 atm, 1.4 g/cm^3) the intrinsic energy resolution arising from the statistics of charge collection was 16 keV full width half maximum (FWHM) at an energy of 570 keV and 20 keV FWHM at 976 keV.

Several different gamma-ray spectrometers employing xenon gas at high pressure have been developed by a research group at the Moscow Engineering and Physics Institute led by V. V. Dmitrenko. In 1989 this group developed a double, parallel-plate chamber similar in concept to the Yale design[3]. When filled with xenon at a density of 0.6 g/cm^3 and with a small admixture of hydrogen added to improve the drift velocity, an energy resolution of 17 keV FWHM was obtained at an energy of 1.33 MeV. This spectrometer was installed in the "MIR" space station and employed for gamma-ray astronomy research. At this time it has been in operation for three years with no degradation of its performance.

J. Losee and coworkers at the Naval Command, Control, and Ocean Surveillance Center (NRaD) in San Diego have recently developed a high-pressure, xenon-filled gamma-ray spectrometer similar in concept to the Yale and Moscow devices but differing from them in certain important design features[4]. In initial tests, the performance of this device is comparable to that of the other spectrometers.

THE BROOKHAVEN SPECTROMETER

The principal features of the ionization chamber which has been designed and fabricated at BNL are shown in Figure 2. It is similar to the designs of Markey and Levin in that the active volume is a right circular cylinder defined by the cathode (C) and anode (A) at the ends and by two circular, conducting rings (E) along its length. A Frisch grid (G) is situated one centimeter from the anode. The active volume is 300 cm^3, comparable to that of a 3 x 3-inch scintillation detector.

The chamber is designed to operate at pressures up to 60 atm and ambient temperature over a range of gas densities from 0.5 to 1.5 g/cm^3.

The pressure vessel, shown in Figure 3 has been fabricated from a light, high-strength titanium-vanadium-aluminum alloy (Ti_6Al_4V). This alloy combines a high strength/weight ratio with good transmission for gamma rays.

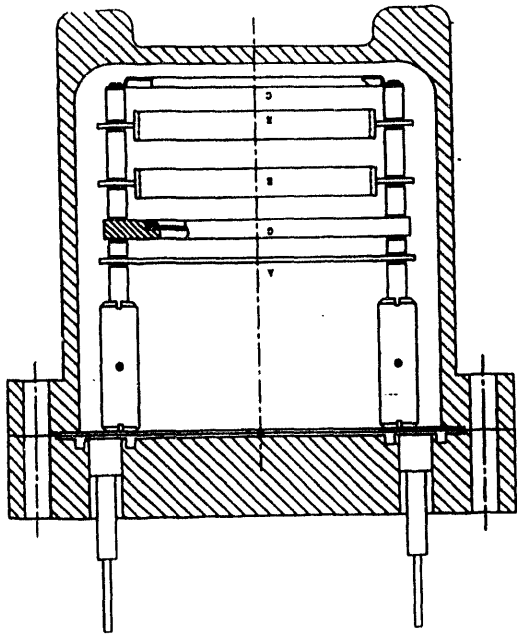

Figure 2. BNL Ionization Chamber Pressure Vessel and Internal Structures.

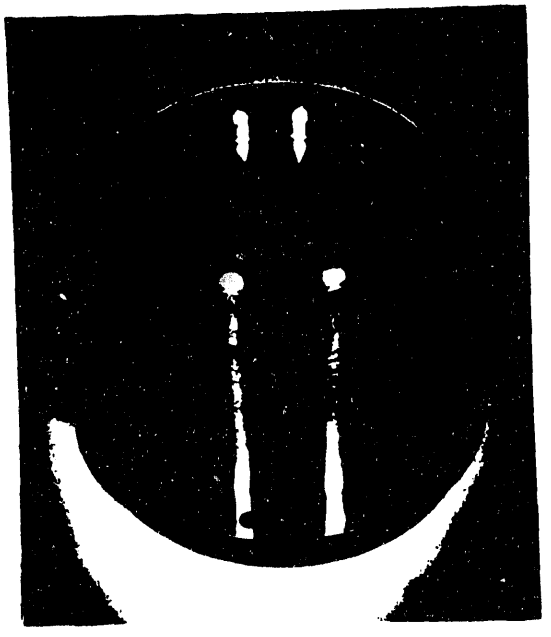

Figure 3. BNL Ionization Chamber. Pressure Vessel.

The internal structures of the ionization chamber are shown in Figure 4. The chamber is designed to operate at an overall potential difference of 12 kV between the cathode and anode with a 5 kV grid-anode potential difference. To provide a grid structure which is stable mechanically and insensitive to microphonic noise, the grid has been fabricated from an 0.005 inch thick wafer of silicon with the use of semiconductor production techniques .

In all devices employed for radiation spectrometry, the achievable energy resolution is limited not only by the statistics of charge collection but by the system electronic noise as well. Recently, a novel, charge-sensitive preamplifier has been developed at BNL whose performance is characterized by an extremely low level of electronic noise[5]. Accordingly, this device will be employed to process the signals from the BNL xenon gamma-ray spectrometer. In conventional charge-sensitive preamplifiers a feedback loop consisting of a high-value resistor and capacitor in parallel is employed to provide a return path for the detector leakage and signal current. The resistor in this circuit is a major source of electronic noise. The BNL design takes a novel approach in eliminating this feedback loop, and instead provides an alternative return path for the accumulated charge. While conventional charge-sensitive preamplifiers characteristically have noise levels in the vicinity of 200 electrons rms, an equivalent noise level of only 20 electrons rms has been achieved with this design.

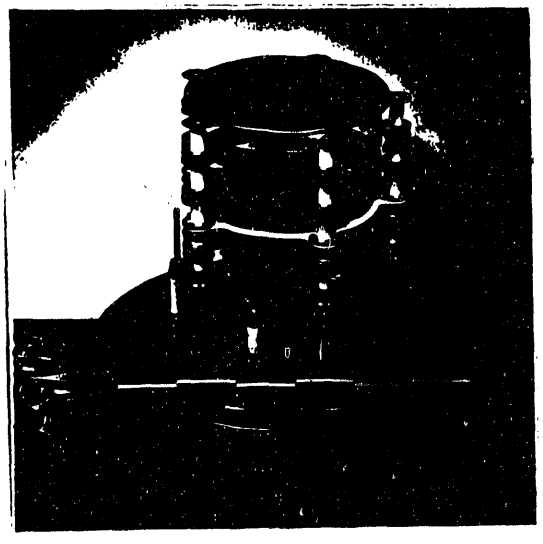

Figure 4. BNL Ionization Chamber. Internal Structure.

CONCLUSIONS

A gamma-ray spectrometer employing xenon gas at high pressure has been designed and fabricated at BNL. A sequence of tests is under way, which will establish the spectrometer's performance as a function of gas density and other parameters and which will be aimed at achieving a more complete understanding of the mechanisms underlying the performance. Figure 5 illustrates the range of the energy resolution that can reasonably be expected to be achieved, as a function of gamma-ray energy, based on existing experience with other devices of this type. The intrinsic energy resolution is shown for two values of the Fano factor: F = 0.13 (1 atm) and F = 1 (60 atm), and the total line width attainable when the intrinsic width and 6 keV electronic noise are added in quadrature. For comparison, the energy resolution attainable for a sodium iodide (NaI) scintillator is shown.

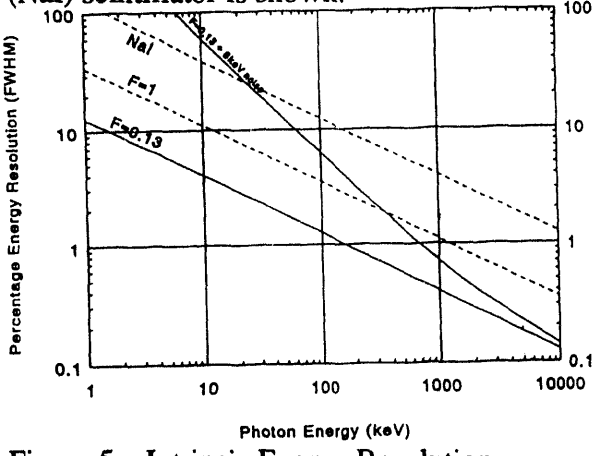

Figure 5. Intrinsic Energy Resolution Versus Gamma-Ray Energy.

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