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Abstract

We have evaluated and compared some of the relevant operating characteristics of NaI and plastic scintillators for use in various safeguards monitoring applications. These include a sensitivity analysis of the two scintillators to various radiation fields and scintillator response as affected by environmental temperature. A comparison of experiment and modeling via the Monte Carlo N-Particle (MCNP) code has been performed to validate our calculational techniques. This then enables complex detector situations to be simulated with increased confidence.

INTRODUCTION

There are many quality publications that evaluate the use of various scintillation crystals for use in portal monitors.[1,2] There are several different operational characteristics not covered in those publications that are important for remote and/or unattended radiation monitoring system-sensor evaluation. Typically the remote or unattended radiation monitoring system is performing detailed radiation identification and tracking nuclear material. The systems usually consist of many different types of radiation sensors and are situated over a large area. Many of the facilities and storage sites covered by these systems are located in remote areas with severe climates and limited human support resources. These properties make it undesirable for inspectors to reside there for extended periods of time to conduct daily inspections or maintenance. For these facilities, it is desirable to develop unattended detection systems that are accurate and reliable. These systems could be physically checked on a regular basis and/or continuously monitored from other locations via network or satellite links. Such a system requires a composite mix of sensors (e.g., radiation, infrared (IR), video, or acoustic) and video cameras that can continuously feed their data into a network which is programmed to provide real-time alarms and selectively store relevant data. The stored data can then be reviewed at periodic intervals for anomalies, which may indicate an undesirable activity has taken place.

The development and deployment of the radiation detectors for use in these safeguard applications are driven by several factors. They include the physical characteristics (size, dimension, radioactivity, and composition) of the source and shielding, radiation signature, methods of physically transporting the materials, cost, sensor reliability, and detection requirements. Different detectors can be employed depending on whether there is a requirement to detect for the presence or radiation; detect a change in the intensity of a radiation field; classify the type of radiation present; identify the material present; and/or quantify the amount of a material present. The nuclear radiation emitted from the nuclear material for these remote radiation monitoring systems are gamma rays and neutrons. The nuclear material can be transported by hand or vehicle with different

levels of shielding. The nuclear detection system has to meet the performance criteria and standards of many organizations, such as the International Atomic Energy Agency (IAEA), Nuclear Regulatory Commission (NRC), Department of Energy (DOE), Department of Defense (DOD), Environmental Protection Agency (EPA), and related foreign organizations.

The radiation detectors in monitoring systems include ^3He tubes, ^{235}U fission chambers, ionization chambers, plastic scintillators, and NaI scintillators. Plastic and NaI scintillators are best suited for lower dose levels of radiation and also perimeter radiation sensors. In terms of a simple pass/fail gross radiation detection system, NaI and plastic scintillators are very similarly suited. NaI scintillators perform significantly better than plastic when region of interest (ROI) information (net counts) is used to specifically detect and trigger on the peak area counts for the different characteristic gamma-radiation energies of specific isotopes. In this paper, we will present results of some measurements that we performed to determine the scintillator characteristics appropriate for various applications.

EXPERIMENT

A Miniature Modular Multichannel Analyzer (M^3CA) was used for all of the spectrum acquisition involved in these measurements. A total of eight scintillators were used for the measurements. Two 10-cm per side cubic NaI, one 10-cm per side cubic plastic of type BC-400, two 2.54-cm thick by 25.4-cm wide by 76.2-cm long plastic of type BC-400, one 5.08-cm diameter by 5.08-cm high right circular cylinder NaI, one 3.7-cm diameter by 3.7-cm diameter plastic of type BC-418, and one 12-cm diameter by 2.54-cm thick right circular cylinder NaI. Not all of the detectors were used for all of the measurements. We have used various quantities of ^{235}U , ^{238}U , ^{137}Cs , and ^{252}Cf to characterize the scintillators' response. NaI response to neutrons was performed using quantities of borated polyethylene surrounding the crystal and then observing the 478-keV gamma ray following neutron capture in the boron.

The first consideration concerning the use of any radiation detector in a monitoring system is the sensitivity to the environmental conditions that the detector must operate within. There is a considerable amount of misinformation concerning the temperature sensitivity of NaI and the lack of it in plastic. The truth is that NaI and plastic can have very similar temperature response curves that can depend (rather strongly) on the photomultiplier coupled to the scintillator. Figure 1 shows the measured centroid location (half-knee pulse height for plastics) as a function of temperature for several different scintillators. The solid and dashed lines with diamonds compare the results for the acquisition electronics either being maintained at room temperature or at the same temperature as the NaI scintillator crystal. The solid line with square symbols shows the results for a plastic scintillator with the same photomultiplier as the NaI data shown in the lines represented with diamonds. The solid line with circle symbols shows the response of the small plastic scintillator coupled to a fast-timing photomultiplier as used for neutron time-of-flight measurements in a nuclear physics laboratory. This fast-timing scintillator was chosen simply to illustrate the range and variability of photomultiplier/scintillator temperature-dependent response. Throughout the measurements, an electronic pulser was used to determine acquisition electronic drift, none was observed. The sensitivity that NaI has to temperature that should be noted is sensitivity to extremely rapid temperature changes, the manufacturers' stated limit is no greater than 5°C per

hour. The housings used to mount the equipment help protect the crystal from the outside environment and any ambient temperature change.

Fig. 1. Gain vs temperature response for various scintillation crystal types and configurations.

To account for the change in centroid position (gain) as a function of the temperature, a reference peak can be used to maintain stability. The system to maintain constant gain that we use in some of our monitoring systems consists of two parts. The first part is simple but only accurate to within a few percent. The recent time history of the scintillator temperature is stored in the operating system memory and an effective temperature is determined. The effective temperature takes into account the thermal equilibrium time for the scintillator. Once the effective temperature changes beyond a predetermined value, the photomultiplier tube bias voltage is adjusted by the acquisition software to maintain a reference peak centroid within a certain position. The second part of the stabilization routine is a software-monitoring program. The program determines the centroid position for short acquisition times and then shifts the ROI boundaries to account for any slight change in gain occurring in the system.

One of the most significant advantages of using the NaI scintillator is the ability to obtain spectral and isotopic information. This capability also leads to significant improvement in the signal-to-noise ratio when analyzing the data. Figure 2 compares spectra obtained with the large plastic slab detector to that of a 10-cm cubic NaI scintillator for a small ^{235}U (0.27-g foil) source for an acquisition time of 5 s. The line labeled Plastic displays the response of the plastic scintillator, the line labeled NaI displays the response of the NaI scintillator. The dashed line in each case is the respective background measurement for the detector. The signal-to-noise ratio (S/N) values displayed by each curve demonstrates the effectiveness of the net-area activity determination over the gross activity determination. The S/N value for the plastic scintillator used in this figure is the ratio of the gross counts with a source to gross counts with no source. The S/N value for the NaI scintillator used in this figure is the ratio of net counts in the 186-keV ROI to the gross number of counts in the same ROI.

Fig. 2. Comparison of the plastic scintillator with the NaI scintillator pulse height response to a small ^{235}U foil, count time is 5 s. Background shown as the dashed lines for each respective measurement.

For use in a large area, a radiation detector must have uniform area sensitivity. To minimize the insensitive regions, it is best to either place a large slab detector at one end of a room or place a spherical detector in the center of the room. Multiple detectors per room are another more costly option, depending on the volume of the area being monitored it is sometimes necessary. Because radiation sensitivity falls off as $1/r^2$, a centrally placed radiation detector has a more uniform room response than one placed at the edge of a room. Figure 3 demonstrates the angular response of two NaI scintillators. The angular response of a 10-cm cubic crystal is shown in by circle symbols, the response of a 12-cm diameter by 2.54-cm thick right circular cylinder NaI is shown by the triangles. A ^{137}Cs gamma-ray source at 75 cm, as measured from the center of the front face, was used for the measurements. The method of measuring the distance to the crystal accounts for the increase in count rate at back angles, the distance to the geometric center of the crystal was decreasing as the angle approached 90 degrees. The measured data are shown as the points, the solid line is a MCNP calculation. As is obvious from the first principles of geometry, the cubic scintillator displays very little angular bias in radiation sensitivity while the thin scintillator's response falls off at side angles. Also, the measured S/N ratio for the two crystals is very similar.

Monitoring fresh fuel movement within a reactor facility is an example of the way to use an NaI-based volume radiation monitor. The important information to be obtained and questions to be answered are when is nuclear material moving, what is the enrichment or type, and is there any other material moving besides the permitted item? Monitoring systems consist of several layers that permit radiation, video, motion, and mechanical switches to be interconnected. This approach provides redundancy to the monitoring system. As an example of the radiation monitoring aspect of the system, we have measured the movement of various enrichment fresh fuel assemblies in a reactor through a fixed reversible geometry. Figure 4 shows the time history of the gross radiation

Fig. 3. Angular response for a 10-cm cubic NaI (in circles) and a 2.54-cm thick by 12-cm diameter NaI (in triangles). The respective solid lines are MCNP calculations.

count rate of the four different types of assemblies, depleted uranium (DU), 17%, 21% and 26% respectively. This information can be used to trigger a video camera for visual confirmation of the activity. Gross radiation counts cannot differentiate between the 21% and 26% assemblies. Figure 5 shows the analysis of the ratio of the ^{235}U and ^{238}U ROI for each of the assemblies. The dramatic increase in sensitivity to type of assembly is evident over the NaI gamma-ray gross counting rate figure. As a second exercise of this type of monitoring system, three sources, ^{235}U , ^{137}Cs , and ^{238}U , were moved relative to a 10-cm cubic NaI and the ROI net peak area for each of the isotopes was recorded in 1-s increments. Figure 6 shows the results of this measurement. There is interference between the ^{137}Cs and ^{238}U ROI due to the 700-keV complex of the ^{238}U and the low resolution of the NaI (7.5% full width at half maximum [FWHM] at 662 keV). The ^{137}Cs source was moved halfway through the ^{235}U source movement time period. This short acquisition time software is not able to operate independently in a remote monitoring system. However, by coupling this software within the framework of the entire monitoring system, it can provide real-time alarms on specific ratios of the ROI and gross count rate information. It will provide a facility operator or an inspector with additional information for periodic review.

CONCLUSION

While space constraints limit an exhaustive comparison of detector characteristics, previous publications do cover many aspects of the performance characteristics of NaI and plastic scintillators, especially for portal monitor applications. The main point to gain from this paper is that one type of scintillator or another does not have all encompassing beneficial attributes for use in any radiation monitoring application. We have used more NaI crystals in our applications for the following reasons: size constraints, NaI is much more efficient than similar volume plastic, spectral analysis capability, and the benefit of net peak area analysis to gross count rate analysis greatly decreases the minimum detectable activity. The spectral acquisition capability of NaI coupled with appropriate software provides isotope or material type identification capabilities.

Fig. 4. Time history plot of gross count rate data in a cubic NaI crystal for various enrichment reactor assemblies moving through a fixed reversible geometry.

Fig. 5. Analysis of net ROI peak areas for the assembly data shown in Fig. 4. An increased differentiation capability among assembly types is evident.

Fig. 6. Time history plot of three different source motions relative to a single NaI detector.

REFERENCES

- [1] W. H. Chambers, et.al., "Portal Monitor for Diversion Safeguards," Los Alamos Scientific Laboratory Report LA-5681 (December 1974).
- [2] P. E. Fehlau, "An Applications Guide to Pedestrian SNM Monitors," Los Alamos National Laboratory report LA-10633-MS (February 1986).

For the interested reader, numerous other references are contained within these two publications.

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