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Organic liquid scintillation detectors for on-the-fly neutron/gamma alarming and radionuclide identification in a pedestrian radiation portal monitor

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ABSTRACT

We present new experimental results from a radiation portal monitor based on the use of organic liquid scintillators. The system was tested as part of a ³He-free radiation portal monitor testing campaign at the European Commission's Joint Research Centre in Ispra, Italy, in February 2014. The radiation portal monitor was subjected to a wide range of test conditions described in ANSI N42.35, including a variety of gamma-ray sources and a 20,000 n/s ²⁵²Cf source. A false alarm test tested whether radiation portal monitors ever alarmed in the presence of only natural background. The University of Michigan Detection for Nuclear Nonproliferation Group's system triggered zero false alarms in 2739 trials. It consistently alarmed on a variety of gamma-ray sources travelling at 1.2 m/s at a 70 cm source to detector distance. The neutron source was detected at speeds up to 3 m/s and in configurations with up to 8 cm of high density polyethylene shielding. The success of on-the-fly radionuclide identification varied with the gamma-ray source measured as well as with which of two radionuclide identification methods was used. Both methods used a least squares comparison between the measured pulse height distributions to library spectra to pick the best match. The methods varied in how the pulse height distributions were modified prior to the least squares comparison. Correct identification rates were as high as 100% for highly enriched uranium, but as low as 50% for ²⁴¹Am. Both radionuclide identification algorithms produced mixed results, but the concept of using liquid scintillation detectors for gamma-ray and neutron alarming in radiation portal monitor was validated.

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1. Introduction

1.1. Motivation

Radiation portal monitor (RPM) systems are installed anywhere an entity wishes to screen passing people, vehicles, or cargo for the presence of radioactive and nuclear material. In their most simple form, RPM systems simply compare measured neutron and gamma-ray count rates to alarm levels set above count rates expected from background radiation.

Naturally occurring radioactive materials (NORM) typically are an insignificant source of neutrons, whereas one special nuclear material (SNM), plutonium, emits copious amounts of fast neutrons. Therefore, any passively measured neutron count rate above the near zero natural neutron background forms one crucial pillar in RPM design [1]. When measuring voluminous objects, such as cargo containers, the ship-effect may cause distortions to the expected neutron background [2].

Historically, the vast majority of RPM systems have used ³He proportional tubes embedded in high-density polyethylene (HDPE) to count fast neutrons upon thermalization in the HDPE moderator. However, the worsening ³He supply crisis has motivated research in alternative neutron detectors for use in future RPM systems [3]. A wide array of alternative neutron detectors have already been considered, including boron coated straws [4], boron tri-fluoride proportional tubes [5] and many others [6].

Polyvinyl toluene (PVT), often simply referred to as plastic scintillator, is typically used for gamma-ray count rate acquisition in RPM systems. Because many NORM sources emit gamma-rays, gamma-ray alarms may be frequent and the RPM may function purely as a primary screening that funnels offenders into a secondary screening using high resolution, handheld detectors that allow for better isotope identification. More advanced spectroscopic portals using NaI inorganic scintillators or cooled HPGe semiconductors combine the detectors. Several spectroscopic RPM systems are







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commercially available, albeit typically for a steeper price than their competitors using relatively inexpensive PVT [7].

The federal government of the United States of America funds two RPM implementation programs. The Department of Energy's National Nuclear Security Administration's Nuclear Smuggling Detection and Deterrence program (formerly Second Line of Defense) has installed over a thousand RPMs at hundreds of locations in dozens of countries as part of two programs. The Megaports Initiative has focused on screening cargo containers at major foreign seaports, while the Core Program has focused mostly on vehicle and pedestrian RPMs at foreign border crossings, airports and other sensitive locations [8]. The TSA VM-250A/VM-250AGN vehicle RPM is widely deployed. These models are composed of two large panel plastic scintillators and four ³He proportional counters per pillar. Plastic scintillator detector signals are fed into a SCA-775 single channel analyzer (SCA). For these systems, a single SCA window is set for the low energy range in which the most dominant gamma-ray emissions are expected for ²³⁵U and ²³⁹Pu. This single window method coupled with PVT's low energy resolution limits the value of the information such RPMs can provide [9,10]. The method does, however, contribute to background rejection as many common NORM y-rays occur at energies above the SNM energy window.

The second major federal RPM program is coordinated by the Department of Homeland Security's Domestic Nuclear Detection Office (DHS-DNDO) and the United States Customs and Borders Protection (CBP). This collaboration funds and coordinates the installation of RPMs at United States border crossings, domestic seaports, truck weigh stations and entry points, such as major bridges or tunnels, into the largest domestic metropolitan areas. Gamma-ray background from NORM may trigger nuisance alarms in an RPM. The DHS-DNDO RPMs use a discrimination window for the high gamma-ray energies associated with common NORM sources. such as the 1.46 MeV gamma-ray from ⁴⁰K. Alarms with high count rates in this window are categorically rejected as NORM nuisance alarms. Yet this RPM still provides only limited information [11]. Attempts to develop and deploy the Advanced Spectroscopic Portal Monitor were scuttled after poor performance in field tests [12]. Thus in addition to finding a ³He replacement for RPMs, advances in the capabilities of RPMs, like improved energy resolution and spectroscopy, remain highly sought after.

The Detection for Nuclear Nonproliferation Group (DNNG) at the University of Michigan has developed a prototype RPM system based upon the organic liquid scintillation detector material EJ309 [13]. This material possesses fast timing properties and excellent pulse shape discrimination (PSD) capability [14,15] thus making it a good candidate for use in RPM systems. The goal is to demonstrate the use of organic liquid scintillators as a ³He replacement with the added benefit of using the same detector medium for gamma-ray counting and radionuclide identification. Due to the low average atomic number of the scintillator material, incident gamma-rays typically only undergo Compton scattering for most of the gamma-ray energies of interest. Most spectroscopic detector systems use isotope identification mechanisms relying upon the photopeaks created by photoelectric interactions of gamma-rays in the detector. In this paper, we demonstrate two different isotope identification algorithms for liquid organic scintillators.

1.2. Organic liquid scintillation based radiation portal monitor

1.2.1. Hardware design

Fig. 1 shows a photograph of the DNNG RPM design, consisting of a total of eight 7.62 cm diameter by 7.62 cm length EJ-309 detector cells arranged in a four by two array 120 cm from the ground in a custom-designed detector stand. The cost per detector is approximately \$2500 with the most expensive component being the photomultiplier tube. In comparison, due to the ³He supply



Fig. 1. Photograph of the University of Michigan EJ-309 RPM prototype showing the eight 7.62 cm diameter by 7.62 cm length EJ-309 detector cells and the two webcams used as the RPM occupancy sensors.

crisis the cost of a liter of ³He has risen from \$40 to over \$2500 in less than a decade and is expected to increase further. In the United States, the sale of ³He is regulated through an annual federal auction with priority access and price advantages granted to federal users versus foreign/commercial users [16].

The DNNG RPM detector electronics and photomultiplier tubes are powered by two, four-channel CAEN N472 high voltage power supplies [17]. Individual detectors were gain matched by setting voltages such that 80% of the ¹³⁷Cs Compton edge (478 keV) amplitude occurred at a pulse height of 1.80 V for five "high-gain" detectors and 0.63 V for the remaining three "low-gain" detectors. This division into high- and low-gain detectors was necessary for the radionuclide identification algorithm. It allowed one to view complete pulse height distributions (PHDs) from low energy sources, such as ²⁴¹Am and ⁵⁷Co, all the way up to higher energy emitting sources, such as ⁶⁰Co and ⁴⁰K. More detectors were set to high-gain than low-gain to compensate for the larger overlap in PHDs of the higher number of lower energy gamma-ray sources compared to the less numerous and better separated PHDs of the anticipated highenergy gamma-ray sources. Detector pulses were acquired and digitized with a CAEN V1720 digitizer board featuring a 2 V dynamic range, 12 bit resolution, 250 MHz sampling rate, and eight channel inputs [18]. Data is transferred using an optical link to a personal computer running LINUX. Tradeoffs between digitizer dynamic range and resolution also contributed to the need for "high-gain" and "lowgain" detectors for this particular project.

Two commercial off-the-shelf webcams pointing in opposite directions are used as occupancy sensors to trigger a three second data acquisition. Motion detection freeware [19] was used to trigger the data acquisition and analysis scripts when movement was detected in a user-defined image area. The ANSI N42.35-2006 standard outlines in detail the testing conditions to which RPM systems must conform [20]. The standard specifies a source speed of 1.2 m/s passing at 1 m parallel to the single pillar pedestrian RPM system. With that in mind, MCNPX-PoliMi simulations of the RPM system were performed with an ANSI-standard 592 kBg ¹³⁷Cs source [20]. Given this fixed source strength, source to detector orientation, and source speed, various data acquisition times were simulated by representing the moving source as a line source of length corresponding to the fixed source speed multiplied with the measurement time. Fig. 2 shows that the count rate from the Cs source detected in the RPM plateaus after about three seconds. Longer measurement times would only incrementally improve the Cs count rate while the fixed background count rate would simultaneously mean a gradual worsening in signal to noise ratio. Therefore a three second data acquisition window was chosen as the optimal counting time. In order to allow for identical performance of the RPM regardless of whether the source moves "forwards" or "backwards" through the RPM, the triggering masks were designed to trigger the three second data acquisition at 180 cm to the left or right of the RPM [21].

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