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THE ZPR-9 AIRBORNE PLUTONIUM MONITORING SYSTEM

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This paper describes an airborne plutonium monitoring system which is installed in the ZPR-9 (Zero Power Reactor No. 9) facility at Argonne National Laboratory. The design and operational experience are discussed. This monitoring system utilizes particle size and density discrimination, alpha particle energy discrimination, and a background-subtraction technique operating in cascade to separate airborne-plutonium activity from other, naturally occurring, airborne activity. Relatively high sensitivity and reliability are achieved.

Introduction

The Zero Power Reactor No. 9 (ZPR-9) facility^{1,2} is operated by Argonne National Laboratory to provide reactor physics data in support of the United States' fast breeder-reactor development program. Plutonium fuel is handled routinely in order to effect changes in the reactor configuration as required by the experimental programs. Since plutonium is a relatively toxic element, considerable care is exercised to assure that operating personnel and the general public are not endangered during these operations.

Although quantities of enriched and depleted uranium are available, the majority of the fuel utilized is an alloy comprised of about 28% by weight of plutonium (Pu), 69.5% by weight of the 238 isotope of uranium (²³⁸U) and 2.5% by weight of molybdenum (Mo). This fuel is contained in 0.015 inch thick hermetically sealed stainless-steel cans which are evacuated and subsequently filled with helium to a pressure of one half of an atmosphere. Loading of the fuel into containers (drawers) for insertion into the reactor matrix is done manually in open-faced hoods. The interior of the hoods is maintained at negative pressure to assure that plutonium leakage from a defective can is contained within the hood system or caught in the high-efficiency filters through which the hood air is exhausted. Visual examinations, size gauging, and monitoring for radioactivity are techniques used to detect an unusual fuel-can condition. Damaged or swollen cans could indicate incipient failure. Abnormal radioactivity indicates a leaky fuel can.

In addition to the precautions taken to assure that no plutonium is released to the environs, radiation surveys are made routinely to assure prompt detection of small quantities of radioactive contaminants if they should be released to the work rooms. Additionally, real-time monitors are utilized to detect airborne-Pu contamination in the hood ducts, in the loading room air, and in the reactor-cell air.

This paper describes the real-time, continuously operating, airborne-plutonium monitoring system which

has been employed at the ZPR-9 facility since its conversion for use with plutonium fuels in 1968.

Statement of Problem

Airborne-Pu monitors detect radiation emitted from the Pu atom. Problems result because the radiation from the permissible concentration of airborne Pu ($\sim 5 \times 10^{-3}$ d/m per l air)^{3,4} is very low compared to background. Background radiations comprised of alpha particles, beta particles, gamma rays, and neutrons exist in the reactor cell and fuel-handling areas, and tend to interfere with the real-time detection of low concentrations of airborne Pu.

Our system, as do most other airborne monitoring systems, detects the α -particles emitted by the decay of ²³⁹Pu and ²³⁸Pu. The alpha particles of interest have energies in the range from 5.1 to 5.5 MeV. With this choice, potential background problems result predominantly from ²³⁴U, ²¹⁸Po, and ²¹⁴Po which emit alpha particles in the range from 4.7 to 7.7 MeV, and neutrons, beta particles, and gamma rays.

The ²³⁴U background results from uranium which is used in reactor-core loadings. The ²²²Rn daughter-product background results from the natural ²²⁶Ra in the massive concrete walls of the reactor cell. A portion of the decay chain resulting in ²²²Rn and daughter products is shown below.

The standards used in evaluating the adequacy of health-protection measures against airborne radioactivity in occupied areas are outlined by the U.S. Atomic Energy Commission.^{3,4} Permissible concentrations of ²²²Rn are 10^5 times those of ²³⁸Pu or ²³⁹Pu, and permissible concentrations of ²³⁴U are 300 times those of ²³⁸Pu or ²³⁹Pu. Therefore relatively large ²³⁴U and particularly ²²²Rn backgrounds are tolerable. Indeed, background concentrations of 10^{-9} $\mu\text{C}/\text{ml}$ of ²²²Rn are normal in the reactor cell and occasionally become significantly higher.

Neutrons, beta particles, and gamma rays exist in varying degrees in different locations and at different times. Each type of radiation is present with a wide range of energies.

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Design Goals

Our intent was to make the ZPR-9 airborne-Pu monitoring system as sensitive as practicable, consistent with the requirement that it be virtually free of false-alarm signals. The detection of airborne Pu, before exposure to the guideline limit for one week, was considered mandatory. It was clear that existing airborne-Pu monitors were not adequate for our situation because of their inherent sensitivity to background radiations which would result in intolerable false-alarm rates. False-alarm rates in excess of one or two per year per unit are highly undesirable.

References 3 and 4 indicate that the appropriate guide for the maximum-permissible concentration of ^{239}Pu and ^{238}Pu , assuming for the sake of conservatism that it is in a body-soluble form, is 2×10^{-12} $\mu\text{C}/\text{ml}$ in areas where personnel work for forty hours. Using this criterion, personnel could be exposed to $(40)(2 \times 10^{-12})$ $\mu\text{C}\cdot\text{hr}/\text{ml}$ each week. It is convenient to refer to this value as 40 RCG-hr (Radiation Concentration Guide Hours).

System Performance

The ZPR-9 airborne-Pu monitors are capable of detecting airborne-Pu doses released by mechanical disruption of Pu or its oxide of the order of 1 RCG-hr in the presence of normal backgrounds in the ZPR-9 facilities. The monitors, as utilized, are insensitive to ^{234}U , and are not affected significantly by the neutron, gamma-ray, or beta-particle radiations existing in the reactor cell and work areas. Each unit is set to alarm if airborne-plutonium exposures exceed 10 RCG-hr of soluble Pu. The monitoring systems are, except during periods of very high radon background, free of false alarms. Occasionally a detector failure results in a false alarm.

There are nine monitors in use on a continuous basis. The present false-alarm rate is less than one per month for all of the monitors.

System Description

In order to obtain the greatest possible discrimination against the naturally-occurring radon daughter-product backgrounds, it was considered essential first to separate as much of the radon-daughter-product-bearing dust as possible from the collected Pu sample. Following Tait,⁵ Collins,⁶ and Lapsley,⁷ this is accomplished using an impactor. Separation is based on the observation that 90% of the radon daughter-product activity is associated with dust particles which are less than 0.035 microns in diameter.⁸ By contrast, most of the plutonium activity dislodged by mechanical action is associated with particles which are larger than 1 micron in diameter. The velocity of the sampled air is adjusted to permit the small lighter particles to negotiate a sharp turn in the impactor while the large, heavier particles are impacted onto a detector surface. When particles are collected by the impactation process the collection efficiency is proportional to ρd^2 , where ρ is the particle density and d is particle diameter.⁹ Thus the impactor can differentiate between the larger and denser Pu particles and the smaller lighter radon-bearing dust particles.

Plutonium is impacted directly onto the surface of a passivated-surface, diffused-junction, silicone detector in order to permit immediate detection of Pu activity, and to permit discrimination between Pu alpha particles and radon-daughter product alpha particles on the basis of energy. Figure 1 shows the impactor and

detector assembly. A thin coating of silicone grease is used on the detector surface to ensure particle adhesion after impact. The grease is mixed with benzene (10 g of silicone grease per liter of benzene) and 50 λ of the liquid is applied to the crystal. The benzene evaporates quickly, leaving a thin coating of silicone grease on the detector. Tests have shown only a slight shift (40 keV) in the Pu spectrum due to this coating.

The detector presently in use is the Simtec 2002 which has an active surface area of 200 mm^2 . (A Harshaw Model DJC-400-50-100 detector is presently under evaluation as a potential replacement, since Simtec is no longer in business.) The impactor collection efficiency for U_3O_8 , density 7.3 gm/cc , was measured to be greater than 55%. The efficiency for PuO_2 , density 11.5 gm/cc , has not been measured but also is expected to be greater than 55%. Measurements indicate that the collection efficiency for radon daughter products is between 1 and 2%.

Under normal conditions the detectors are operated continuously for eight hours each day. Dust collected during such a period of operation results in a spectral shift of about 55 keV. Therefore the detectors are cleaned and greased each morning. Calibration tests are also performed each morning.

Electronics

A block diagram of the system is shown in Fig. 2. The detector signal is converted to a voltage pulse by a charge-sensitive preamplifier located near the impactor. The output of this preamplifier is then transmitted to the system electronics, which is housed in Nuclear Instrument Modules located in the reactor control room. One module contains a variable-gain pulse amplifier to allow the system to be calibrated at 0.5 V per MeV. This pulse amplifier drives two, independent single-channel analyzers (SCA). One analyzer selects pulses with energies between 5.0 and 5.4 MeV and outputs a logical pulse to the Pu scaler for every event falling within this window. The second SCA selects pulses with energies between 5.5 and 7.0 MeV and outputs a logical pulse to the radon-channel scaler and to a Pu subtraction network when an event falls within its window. The subtraction network will be discussed later. Fig. 3 is a normalized plot of counts per unit energy as a function of energy for alpha particles from ^{222}Rn and its daughter products, ^{239}Pu and ^{238}Pu , and ^{234}U . High resolution is not obtained because the silicone detector has an energy resolution of about 300 keV (FWHM). However, it is clear that the Pu channel can be made to reject most of the ^{234}U counts and most of the Rn counts.

After the impactor has discriminated against radon on the basis of particle size and density, and the pulse-height analyzer has discriminated against radon on the basis of energy, there is still some nominal spillover of radon counts into the Pu channel. A subtraction system, in the form of an up-down scaler in the Pu channel, is used to compensate for this small but significant error in recorded Pu counts. The smallest ratio of counts in the Pu channel to counts in the Rn channel, as determined over a long period of time, is used as a basis for subtracting one count from the Pu channel for every "n" counts in the Rn channel. A conservative value for "n" of 10 was selected to assure that it is not possible to subtract more counts from the Pu channel than actually spill into it from the radon channel. The subtraction circuit is disabled when the counter reaches 0000 counts. This prevents an additional down pulse from lowering the counter to a 9999 count which would then cause an alarm.

The output of the counter module is delivered to a digital to analog converter. A signal proportional to the log of the output of the converter is used to drive local and remote readout meters each of which presents a four-decade display. The log output also drives the trip circuit.

System Operation

The counts in each channel are summed for a period of 15 minutes. At the end of this time the system is reset and begins a new cycle. At the instant the number of counts in the Pu channel exceed the number corresponding to a dose of 10 RCG-hr an alarm is sounded. This can be a fraction of a second after a new cycle is initiated if a high airborne-Pu level is present, or later in the cycle if the airborne-Pu level is lower. The number of Pu counts in the Pu channel is determined by Eq. 1 for a delta-function release of Pu distributed uniformly throughout the room. (The Pu is assumed to remain airborne during the sampling interval.)

$$C = \sum k_i N_i V T^2 \quad (1)$$

The summation is over all Pu isotopes. The quantity k_i is a system constant. N_i is the density of type i Pu atoms in the atmosphere, V is the volumetric air-sampling rate, and T is elapsed time from the instant of Pu release.

System Experience

During the five years of plutonium use at ZPR-9 there have been three very small plutonium releases, due to minute leaks in the fuel can welds, within the loading hoods. In two of the three releases, the airborne-plutonium monitor in the hood system alarmed at the 10 RCG-hr exposure. In one of these two releases a room monitor sampling the air at the outside surface of the hood recorded a 1 RCG-hr exposure. In the third instance the amount of plutonium released was extremely small and did not become airborne, and therefore was not detected by the hood-system monitors. Smears of the fuel-can surface detected this contamination. In all cases except one, no plutonium could be detected outside of the hoods using direct instrument-survey methods and smears. In the one case, there was about 5000 dpm in one small spot on the floor directly in front of the hood. Therefore a very small quantity of Pu would be expected to be airborne outside of the hood.

A fourth situation involved a Pu storage container which was very slightly contaminated with plutonium. When this container was brought into the work area and set down, several airborne plutonium monitors alarmed. Each indicated an exposure of about 20 RCG-hrs. Direct surveys and smears did not detect contamination on any surfaces other than on the container and on the floor beneath the container. A smear of the floor beneath the container showed a contamination level of 30 dpm.

The ZPR-9 airborne-plutonium monitor is an effective instrument for real-time airborne-plutonium detection in areas where relatively high radon backgrounds occur.

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Fig. 1. Annular Impactor used with ZPR-9 Airborne-Pu Monitoring System.

Fig. 2. Electronics used with ZPR-9 Airborne-Pu Monitoring System.

Fig. 3. Spectral Response of Detector used with ZPR-9 Airborne-Pu Monitoring System.