

Environmental Management Science Program

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Ultrahigh Sensitivity Heavy Noble Gas Detectors for Long-Term Monitoring and Monitoring Air

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Research Objective

The primary objective of this research project is to develop heavy noble gas (krypton, xenon, and radon) detectors for: 1) long-term monitoring of transuranic waste, spent fuel, and other uranium and thorium bearing wastes, and 2) alpha particle air monitors that discriminate between radon emissions and other alpha emitters. A University of Cincinnati/Argonne National Laboratory (UC/ANL) Team has been assembled to complete this detector development project. DOE needs that are addressed by this project include improved long-term monitoring capability and improved air monitoring capability during remedial activities. Successful development and implementation of the proposed detection systems could significantly improve current capabilities with relatively simple and inexpensive equipment.

Research Progress and Implications

As of June 1, 1998, the UC/ANL Team has: 1) made significant progress toward characterizing the fluid transfer process which is the basis for this detector development project and 2) evaluated several radiation detectors and several potential pulse processing schemes. The following discussion describes the progress made during the first year of this project and the implications of this progress.

ANL first developed the fluid transfer process through which heavy noble gases in the atmosphere are preferentially absorbed by certain organic fluids (corn oil has been used most extensively in this project) and can subsequently be degassed by adding a small amount of energy. It is this fluid transfer process that allows the heavy noble gases to be concentrated for enhanced detection. Due to ANL's extensive experience in characterizing this process, this part of the Team is continuing to take the lead in this area. Toward this end a set of experiments was conducted to measure the pressure drop for 3-in-1 oil using structured packing material and comparing with the data for corn oil. The motivation for testing with 3-in-1 oil is that for long-term deployment of our ultrasensitive detectors, untreated corn oil may oxidize with time, lowering the absorption coefficient and degrading the energy utilization (because of increased viscosity caused by oxidation-induced polymerization of oil molecules). For long-term, field-deployed monitoring systems, there may be advantages to using a fluid with an extremely low oxidation potential. One implications that can be drawn from these experiments is that the pressure losses when using structured packing materials are not only small, but are independent of fluid flow rate for both 3-in-1 oil and for corn oil. This important finding will be leveraged in future prototype designs. We also intend to use results from these experiments as input into ANL's ISGA FORTRAN design code for packed column scrubbers to optimize future prototypes.

In addition, UC has assembled and characterized a small prototype fluid transfer system to use as an input to the detector and pulse processing scheme development components of this project. This prototype system is being used in a charcoal-filtered hood to minimize atmospheric emissions and is consequently limited by the size of the hood. Nonetheless, the system behaves as a larger system would, but with much lower efficiencies. Thus, results must be appropriately scaled to realistic system dimensions and efficiencies.

A significant part of scaling prototype results consists of accounting for limited geometry and performance of radiation detectors used in these experiments. Simulations have been developed to model the behavior of detectors and pulse processing schemes used in these laboratory prototype

experiments. Once the simulations are shown to accurately predict lab experiments, they can be used to optimize geometries and performance in these optimized geometries.

To accurately detect and quantify the radioactive heavy noble gas isotopes, a detection scheme which distinguishes between alpha and beta particles, as well as recording gamma-ray spectra, will result in the best system sensitivity. To accomplish this discrimination, potential detector candidates typically consist of one detector for the alpha and beta particles and a separate detector for the gamma rays. A gas proportional detector coupled with a NaI(Tl) scintillation detector is one such promising detector combination. The UC/ANL Team has carefully evaluated a gas proportional detector for use as the alpha/beta detector based on pulse height discrimination. Results indicate that while this proposed scheme has potential for good performance, use with more sophisticated coincidence timing pulse processing schemes which are necessary to accurately quantify isotopic concentrations is not optimal due to poor gas proportional detector timing resolution.

Other potential alpha/beta detectors are being evaluated for use with NaI(Tl) or some other gamma-ray detector. Specifically, plastic and liquid scintillators are being simulated and experimentally investigated. To date, plastic scintillators have been used with NaI(Tl) with superior timing performance as compared with the gas proportional detectors. Pulse shape analysis is being evaluated as a means of discriminating between alpha and beta particles.

One example of the significance of simulating detector performance for guidance is the proposed use of CsI(Tl) flow-cell detectors to accomplish the alpha/beta discrimination. Through radiation transport simulations, it was found that such detectors would need to be prohibitively thin in order to allow gamma-ray emissions to reach the NaI(Tl) detector. Consequently, this type of detection scheme is not currently being pursued further.

Finally, beta/gamma coincidence techniques have been evaluated as a means of enhancing system sensitivity. By only recording decays in which two or more emissions (typically a beta particle or conversion electron and a gamma ray) are detected in coincidence, background counts can be significantly reduced. Such coincidence pulse processing techniques have been evaluated for use with the gas proportional detector/NaI(Tl), plastic scintillator/NaI(Tl), and through simulation the CsI(Tl) flow-cell detector/NaI(Tl) systems. Triple coincidence techniques have also been studied for use with those isotopes that decay by emitting a beta particle followed by a gamma ray and subsequently a conversion electron. Results indicate that background can be virtually eliminated in certain cases.

Planned Activities

In the coming year the UC/ANL Team will continue to characterize and optimize the absorption and degassing of the fluid transfer system in an effort to arrive at a realistic system for field use. One potential complication associated with the fluid transfer is the extent to which air is concentrated by the oil. The simplest way to explore this issue quantitatively is via stable gas mass spectrometry. Experiments are planned for evaluating this complication using air that is spiked with samples of enriched Xe and Kr that will be passed through an oil absorber and de-sorber. Use of isotopically enriched noble gas will enable us to distinguish between our tag gas and entrained natural-abundance Xe and Kr from the air. Mass spectrometric analyses will enable us to accurately quantify the degree of noble gas concentration vs. the degree of air-constituent concentration. The Team will continue to develop simulations of the fluid transfer system, as well as proposed detectors. These simulations will be used to guide further system optimization. In addition, the Team will continue to evaluate detectors and detection schemes for integration with the fluid transfer system. This detector evaluation effort will include the gas proportional/NaI(Tl), plastic scintillator/NaI(Tl), and liquid scintillator/NaI(Tl), as well as any additional detectors that are identified. Along these lines, we will reconsider the use of CdZnTe as a gamma-ray detector to replace NaI(Tl). With the significant recent advances in spectroscopy quality CdZnTe detectors, it may become more favorable to use these detectors coupled with charged particle detectors for air monitoring. With respect to evaluating detection schemes, pulse processing techniques which reduce minimum detectable activities and simultaneously facilitate isotopic analysis will be developed and optimized.

Other Access To Information

The UC/ANL Team recently presented a paper at the 1998 Symposium on Radiation Detection and Measurements describing the small prototype fluid transfer system and its performance. This paper has been accepted for publication in Nuclear Instruments and Methods in Physics Research, Section A and should appear in this journal in late 1998 or early 1999.