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Project Title: **Ultrahigh Sensitivity Heavy Noble Gas Detectors for Long-Term Monitoring and for Monitoring Air**

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Ultrahigh Sensitivity Heavy Noble Gas Detectors for Long-Term Monitoring and for Monitoring Air DOE Award No. DE-FG07-97ER62521

Annual Report
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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 was assembled to complete this detector development project. Effective 1/4/99, the UC PI (John Valentine) became an Associate Professor in the Nuclear and Radiological Engineering Program of the George W. Woodruff School of Mechanical Engineering at the Georgia Institute of Technology. Consequently, this project was transferred to Georgia Tech (GT) with the PI. UC funding extended to 1/31/99 and GT funding became active 4/26/99. Due to this transfer, we will refer to the research team as the GT/UC/ANL Team for this Annual Report. Subsequently, we will use GT/ANL Team. 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 15, 1999, the GT/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 while the PI was at the University of Cincinnati.

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 implication 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.

To complement ANL simulations, GT/UC has developed a concentration system simulation program based on mass transfer first principles. The simulation determines optimum air and oil flow rates, absorption tower height and diameter, as well as the need for parallel and/or series absorption tower stages, based on a user prescribed output concentration and rate that is required by the detection system. The simulation can also be used to predict output concentration based on absorption tower specifications and flow rates. This simulation will also provide a valuable tool for optimizing future concentration designs.

In addition, GT/UC 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 was 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.

The GT/UC/ANL Team is continuing 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 were conducted January 25, 1999 at the University of Cincinnati to evaluate this complication using air that was spiked with samples of enriched Xe and Kr that were passed through an oil absorber and desorber. Use of isotopically enriched noble gas enables us to distinguish between our tag gas and entrained natural- abundance Xe and Kr from the air. Mass spectrometric analyses of these samples at ANL has indicated that the degree of noble gas concentration vs. the degree of air-constituent concentration is more promising than it was at one time believed to be. However, these experiments did not provide conclusive results, thus additional series of similar experiments are planned for the coming year. Based on future results and the previously described simulation program, we will be able to predict what a realistic fieldable system will require (i.e., size and number of absorption towers, air and oil flow rates, series and parallel unit requirements, etc.).

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. Based on the consistency of preliminary detector simulation results compared with experimental results, we have used the simulations to help design a second generation of detectors. These newer detectors more than double the efficiency of detecting certain heavy noble gas radioisotopes and thus bring us closer to realizing fieldable systems.

To accurately detect and quantify some of the radioactive heavy noble gas isotopes, a detection scheme which distinguishes between alpha and beta particles, as well as recording gamma-ray spectra, results 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 GT/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 were evaluated for use with NaI(Tl) or some other gamma-ray detector. Specifically, plastic and liquid scintillators and PIPS semiconductor detectors were simulated

and experimentally investigated. To date, plastic scintillators and PIPS have been used with NaI(Tl) with superior timing performance as compared with the gas proportional detectors. Pulse shape analysis was evaluated as a means of discriminating between alpha and beta particles for the plastic scintillator and shown to provide improved performance.

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), PIPS/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.

In summary, the GT/UC/ANL Team has made significant progress toward achieving the goals of this project. While fieldable systems have not been realized to date, all of the pieces are now in place to start going from lab-scale to field-scale systems primarily due to the achievements thus far. When such fieldable systems are available, DOE EM activities will be significantly enhanced in the air monitoring and long-term monitoring areas. There also exists a significant opportunity for this technology to be applied in the spent fuel characterization area. We have begun to pursue defining what will be required for this technology to be successfully implemented to characterize DOE spent fuel, as well as spent fuel from the Former Soviet Union.

Planned Activities

The GT/ANL Team will focus on two major areas over the next year. First, we will focus on further characterizing the absorption and degassing mechanisms associated with the fluid transfer concentration system. This effort will consist of conducting experiments with spiked non-radioactive krypton and xenon samples and subsequently analyzing the results using mass spectrometry. In addition, a new facility has been constructed at Georgia Tech that will allow more extensive characterization of the fluid transfer concentration systems. A 3 foot deep by 6 feet wide by 7 feet tall walk-in hood has been constructed that will provide a facility in which absorption tower sub-systems and degassing sub-systems can be tested. Due to the size of this facility, much larger sub-systems can be constructed and tested (at UC we were limited to about 20 inches in height, whereas the new facility could easily accommodate sub-systems of 6 feet in height). This capability will facilitate our effort to characterize the absorption and degassing mechanisms and improve our capability to predict the behavior of fieldable systems. Finally, a more sophisticated controls system has been implemented in the new fluid transfer concentration system at Georgia Tech. Thus, more extensive and systematic series of experiments can be conducted to characterize the absorption and degassing mechanisms, as well as optimize the fluid transfer concentration system for fieldable implementation. The second major focus area for the next year consists of further development of application specific detector and pulse processing techniques. While we have developed significant expertise and knowledge in this area to date, the effort will become more focussed and application specific. In the final year of the project, the GT/ANL Team will integrate these two efforts to develop a fieldable system (or systems) consisting of an optimized fluid transfer concentration system and an optimized detection system.

Information Access

The GT/UC/ANL Team presented papers at the *1998 Symposium on Radiation Detection and Measurements* and at the *1998 IEEE Nuclear Science Symposium* describing the small prototype fluid transfer system and its performance and evaluating detectors and pulse processing techniques, respectively. The former paper has been published in *Nuclear Instruments and Methods in Physics Research, Section A*, Volume 422, pp. 820-825, 1999. The latter paper has been accepted for publication in *IEEE Transactions on Nuclear Science*, and is expected to appear in Volume 46, No. 3, 1999. We have also had a paper describing progress over the past year accepted for presentation at the *1999 IEEE Nuclear Science Symposium*.