Radionuclide Sensors for Water Monitoring

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Research Objective

Radionuclide contamination in the soil and groundwater at Department of Energy (DOE) sites is a severe problem requiring monitoring and remediation. Radionuclide measurement techniques are needed to monitor surface waters, groundwater, and process waters. Typically, water samples are collected and transported to the analytical laboratory where costly radiochemical analyses are performed. To date, there has been very little development of selective radionuclide sensors for alpha- and beta-emitting radionuclides such as ^{90}Sr, ^{99}Tc, and various actinides of interest.

The objective of this program is to investigate novel sensor concepts and materials for sensitive and selective determination of beta- and alpha-emitting radionuclide contaminants in water. In order to meet the requirements for low-level, isotope-specific detection, the proposed sensors are based on radiometric detection. As a means to address the fundamental challenge of short ranges of beta and alpha particles in water, our overall approach is based on localization of preconcentration/separation chemistries directly on or within the active area of a radioactivity detector, using automated microfluidics for sample manipulation and sensor regeneration or renewal.

The outcome of these investigations will be the knowledge necessary to choose appropriate chemistries for selective preconcentration of radionuclides from environmental samples, new materials that combine chemical selectivity with scintillating properties, new materials that add chemical selectivity to solid-state diode detectors, new preconcentrating column sensors, and improved instrumentation and signal processing for selective radionuclide sensors. New knowledge will provide the basis for designing effective probes and instrumentation for field analytical chemistry.

Research Progress and Implications

This report summarizes work for the first 6 months of a 3-year program. The initial ongoing effort is directed at the investigation of the preconcentration column sensor concepts, development and characterization of the selective scintillating microspheres (SSMs), and development of the new scintillation detection systems. The preconcentrating minicolumn radionuclide sensor is based on the use of dual-functionality bead materials. These materials are designed to incorporate both selective separation chemistry for analyte preconcentration and localization within the detector, and scintillating fluors, so that radioactivity of retained species can be transduced to a measurable light output. To date we have achieved progress in SSM materials prepared by co-immobilization of selective organic extractants and scintillating fluors within inert polymeric beads. Scintillating microspheres selective for Tc, Sr, and actinides were prepared. Using pertechnetate-selective scintillating beads, we have demonstrated the feasibility of a renewable preconcentrating column sensor concept in successful application toward ^{99}Tc determination in contaminated ground water samples from the Hanford site. To avoid sensor material degradation during the regeneration step, the sensor column was renewed via fluidic bead replacement. Using a 50-mL sample volume and 30-minute counting time, the detection limit for ^{99}Tc was 0.37 dpm/mL (9.8 pg/mL). This detection limit is below the maximum permissible drinking water level of 2 dpm/mL. This study is a subject of a recent publication.(1)

In addition, research has commenced on a combined extraction chromatographic and scintillation resin in a solid support for on-line and off-line monitoring of radiostrontium in aqueous solutions.

This extractive scintillator resin has been realized as (a) a mixture of extraction chromatographic resin and granular scintillator, (b) extraction chromatographic material coated on the surface of a scintillating glass and (c) inert polystyrene chromatographic resin impregnated with a proprietary extractant and organic fluors. The extractive scintillator resin has been evaluated for on-line and off-line applications. For on-line measurements, the extractive scintillator resin is used in conjunction with a flow-cell scintillation detection system. For off-line measurements, the resin is used in the same manner as the non-scintillating version, but rather than eluting the activity from the column prior to counting, the scintillating extraction column can be placed in a scintillation vial and counted without the introduction of liquid scintillation cocktail. The absolute detection efficiency for ^{90}Sr beta particles sorbed to the strontium-selective extractive scintillator resin was measured to be approximately 30%. Characterization of the detection efficiency, minimum detectable concentration, selectivity, loading capacity, capacity factor and regeneration capability of the extractive scintillator resin continues.

Actinide SSMs have been developed and characterized to determine feasibility for sequential elution of radioactive species. Tests have indicated that although sequential separation is feasible, available energy resolution information is limited. In addition to the limited energy resolution of the SSM, variable quench associated with different chemical eluants is significant. The absolute detection efficiency of the actinide SSM depends on alpha energy and ranges from ~80% to nearly 100%.(2) Development of a photodiode-based scintillating microsphere detection system continues. The photodiode-based system offers several advantages over commercially available photomultiplier-based systems to include: better pulse shape discrimination capability, higher sensitivity at certain scintillation wavelengths, compactness and reduced power requirements.

Planned Activities

Within the remainder of the first year and during the second year, our activities will be directed at further development of SSM materials for ^{90}Sr and actinide sensing. We will explore SSM materials and sensor concepts based on selective ligands covalently attached to the bead support. Inorganic sorbent and scintillating materials will be evaluated for use with the preconcentrating column sensors. We plan to evaluate the feasibility of selective scintillating sensor concepts in fiber and planar geometries. Investigation of the chemically modified solid-state diode sensor concept will begin during the second year of the program.

Information Access

(1) O.B. Egorov; S.K. Fiskum; M.J. O'Hara; J.W. Grate; "Radionuclide Sensors Based on Chemically Selective Scintillating Microspheres: Renewable Column Sensor for Analysis of ^{99}Tc in Water", *Anal. Chem.* 1999, *71*, 5420-5429.

(2) T.A. DeVol, J.E. Roane, J.M. Williamson, J.M Duffey and J.T. Harvey; "Development of Scintillating Extraction Media for Separation and Measurement of Charged-Particle Emitting Radionuclides in Aqueous Solutions," submitted for publication in *Radioactivity and Radiochemistry*.

(3) T. DeVol, O.B. Egorov, J.E. Roane, A. Paulenova, and J.W. Grate; "Extractive Scintillating Microspheres for ^{99}Tc Quantification in Aqueous Solutions," submitted for presentation to Fifth International Conference on Methods and Application of Radioanalytical Chemistry, Kailua-Kona, HI, April 9-14, 2000.

(4) J.M. Duffey, A. Paulenova, and T.A. DeVol, "Combined Extraction Chromatography and Scintillation Detection for On-line and Off-line Monitoring of Strontium in Aqueous Solutions," submitted for presentation to Fifth International Conference on Methods and Application of Radioanalytical Chemistry, Kailua-Kona, HI, April 9-14, 2000.