Environmental Management Science Program

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Hazardous Gas Production by Alpha Particles in Solid Organic Transuranic Waste Matrices

Dr. Jay A. LaVerne
University of Notre Dame
Notre Dame, Indiana 46556
Phone: 219-631-5563
E-mail: laverne.1@nd.edu

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Research Objective

This project uses fundamental radiation chemical techniques to elucidate the basic processes occurring in the heavy-ion radiolysis of solid hydrocarbon matrices such as polymers and organic resins that are associated with many of the transuranic waste deposits or the transportation of these radionuclides. The environmental management of mixed waste containing transuranic radionuclides is difficult because these nuclides are alpha particle emitters and the energy deposited by the alpha particles causes chemical transformations in the matrices accompanying the waste. Most radiolysis programs focus on conventional radiation such as gamma rays, but the chemical changes induced by alpha particles and other heavy ions are typically very different and product yields can vary by more than an order of magnitude. The objective of this research is to measure the production of gases, especially molecular hydrogen, produced in the proton, helium ion, and carbon ion radiolysis of selected solid organic matrices in order to obtain fundamental mechanistic information on the radiolytic decomposition of these materials. This knowledge can also be used to directly give reasonable estimates of explosive or flammability hazards in the storage or transport of transuranic wastes in order to enhance the safety of DOE sites.

Research Progress and Implications

This report summarizes the work after eight months of a three-year project on determining the production of hazardous gases in transuranic waste. The first stage of the project was to design and build an assembly to irradiate solid organic matrices using accelerated ion beams. It is necessary to measure absolute radiolytic yields, and simulate some of the conditions found in the field. A window assembly was constructed allowing the beam to pass consecutively through a collimator, a vacuum exit window and into the solid sample. The beam is stopped in the sample and the entire end of the assembly is a Faraday cup. Integration of the collected current, in conjunction with particle energy and charge, allows for accurate determination of dose. A stream of gas, such as nitrogen, flows between the sample and the beam exit window to flush away gaseous products that evolve from the sample surface. The gas steam is sampled with a quadrupole mass spectrometer. By monitoring the desired mass peak with the spectrometer it is possible to quantitatively determine a number of different products on line.

The first experiments have focused on the hydrogen evolution in polyethylene irradiated with protons and helium ions. Hydrogen is the main gaseous product formed and there have been some previous studies using gamma rays (1) and low energy (<= 1 MeV) H, He, B, and Ar ions (2). The yield of hydrogen was found to decrease from 0.67 molecules/100 eV energy absorbed with 5 MeV protons to 0.11 with protons of 15 MeV. The protons have a linear energy transfer, LET, of 21 to 10 eV/nm, respectively. Hydrogen yields should increase as the LET increases, but the absolute yields are lower than expected suggesting that other processes are involved in its formation or evolution from the bulk.

Literature values for hydrogen production in gamma radiolysis range from 2.4 to 2.7 molecules/ 100 eV (1), which compares well with our value of 2.7. Previous studies with low energy particles also suggest hydrogen radiation chemical yields of 1 to 3. Further studies with gamma radiolysis have shown that with thick samples of polyethylene a considerable fraction of the hydrogen can be retained in the sample in spite of the large diffusion coefficient (> 10-7 cm2/s) of hydrogen in solid polymers (3). It appears that the retention of gases in heavy ion irradiated material is larger than that

found in gamma radiolysis. The reason for this outcome is unknown. Such a large capture of hydrogen in the bulk can have a large consequence of the management of waste material and will be studied thoroughly.

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Planned Activities

A series of experiments to be performed in the next few months will examine the formation and evolution of hydrogen in thin samples. The information obtained will help determine true hydrogen production yields. Other studies varying sample thickness will help determine hydrogen diffusion and retention in bulk materials. A model of hydrogen diffusion and/or percolation in the bulk from ion tracks will be developed to aid in data interpretation. Different organic matrices, such as polystyrene and various resins, will be examined because of their fundamental and practical importance. Hydrogen production will also be examined under different atmospheric conditions. Oxygen or water vapor instead of nitrogen will simulate more realistic conditions and give fundamental information on possible surface effects on hydrogen formation.

An apparatus for the next generation of experiments is currently being designed and constructed. These experiments will enable the analysis of gaseous products produced form solid matrices in ultra high vacuum. Under these conditions, a number of different products with relatively small yields can be measured simultaneously and correlation made between their yields and incident particle characteristics such as charge, energy, and LET. In the next year, an attempt will be made to examine the bulk material following irradiation. A significant increase in crosslinking is expected. This information will give more understanding of the radiolytic processes involved and enable better prediction of outcomes in systems that are not possible to be examined experimentally.