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Radiation Effects in Nuclear Waste Materials

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Radiation Effects in Nuclear Waste Materials
(Project Number: 73750)

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Research Objective

The objective of this project is to develop a fundamental understanding of radiation effects in glasses and ceramics, as well as the influence of solid-state radiation effects on aqueous dissolution kinetics, which may impact the performance of nuclear waste forms and stabilized nuclear materials. This work provides the underpinning science to develop improved glass and ceramic waste forms for the immobilization and disposition of high-level tank waste, excess plutonium, plutonium residues and scrap, other actinides, and other nuclear waste streams. Furthermore, this work is developing develop predictive models for the performance of nuclear waste forms and stabilized nuclear materials. Thus, the research performed under this project has significant implications for the immobilization of High-Level Waste (HLW) and Nuclear Materials, two mission areas within the Office of Environmental Management (EM). With regard to the HLW mission, this research will lead to improved understanding of radiation-induced degradation mechanisms and their effects on dissolution kinetics, as well as development of predictive models for waste form performance. In the Nuclear Materials mission, this research will lead to improvements in the understanding of radiation effects on the chemical and structural properties of materials for the stabilization and long-term storage of plutonium, highly-enriched uranium, and other actinides. The research uses plutonium incorporation, ion-beam irradiation, and electron-beam irradiation to simulate the effects of alpha decay and beta decay on relevant glasses and ceramics. The research under this project has the potential to result in improved glass and ceramic materials for the stabilization and immobilization of high-level tank waste, plutonium residues and scraps, surplus weapons plutonium, highly-enriched uranium, other actinides, and other radioactive materials.

Research Progress and Implications

This report summarizes work after 5 years of a 3-year project. This project has been terminated, and no funding was received for work in either Fiscal Year 2004 or Fiscal Year 2005. A small amount of funds have been carried over to complete some critical radiological work that was delayed. Only the progress achieved in Fiscal Year 2005 is reported here. During this past year, three presentations were made and 6 journal articles were published. Several papers are still in press.

Alpha-Decay Effects in Ceramics

Nuclear magnetic resonance (NMR) studies were completed on Pu-containing zircon samples. This work was led by Dr. Ian Farnan (Cambridge University, UK). A triple containment magic-angle spinning rotor insert system has been developed by Dr. Farnan, and a sample handling procedure has been formulated for safely analyzing highly radioactive solids by high resolution solid state NMR. The protocol and containment system have been used for magic angle spinning (MAS) experiments on zircon samples containing 5-10 wt% ^{239}Pu and ^{238}Pu at rotation speeds of 3500 Hz. The work demonstrated MASNMR can be used to measure the amorphous atomic number fractions produced during accelerated internal radioactive decay. This allows detailed characterization of local atomic structure to be determined in other potential ceramic and glass waste forms in which short-lived alpha-emitters are incorporated to study the long-term effects of radiation damage from alpha decay.

This work has provided the first use of MASNMR spectroscopy on samples containing short-lived actinide isotopes, and the preliminary results have been published in the *Review of Scientific Instruments*. More recent results suggest that the radiation-induced amorphous state in ^{238}Pu-containing zircon (4.2 x 10^{19} alpha decays per gram) is different in terms of its local structure than that attained in natural zircons self-damaged by ^{238}U and ^{232}Th decay. The very large difference in dose rates (8 orders of magnitude) may be the origin of this effect. A detailed paper on these results will appear in *Advances in Actinide Science*.

Initial ^{29}Si MASNMR experiments on a 10 wt% ^{239}Pu-containing zircon (23 years old) showed slight evidence for the onset of radiation damage, similar to that observed in natural zircons damaged by the decay of ^{238}U and ^{232}Th. The line width of the previous spectrum was limited by the intrinsic resolution of the experiment, which could possibly obscure a more precise detection of signal from the radiation damaged regions. A final series of experiments were carried out in August 2005 with a significantly higher intrinsic resolution than before. However, the linewidth of the majority (undamaged) peak did not decrease significantly below that observed previously (natural limit reached), and the signal from the damaged regions was similar. The sample was characterized in detail for spin-relaxation properties and quantification of the signal intensities against standards. This allows us to be confident that we observe all the Si atoms (undamaged and damaged) in the sample within an accuracy of 5-10%. The results of these latest studies will be analyzed and prepared for future publication.

Alpha-Decay Effects in Glasses

There has been no new work on alpha-decay effects in glasses.

Simulation of Radiation Effects Using Ion Beams

There has been no new work using ion beams.

Simulation of Radiation Effects using Electron Beams

There has been no new work using electron beams.

Planned Activities

This project has been terminated and no additional work is planned. All critical experimental studies on alpha-decay effects in ceramics were completed in August 2005. Some results have not yet been prepared for publication and, consequently, may never be published.

Information Access

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