

**Environmental Management Science Program**

**Project ID Number 54691**

**Radiation Effects on Materials in the Near-Field of  
a Nuclear Waste Repository**

**Project Summary  
Annual Report**

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February 19, 2000

## 1. Research Objective

Site restoration activities at US DOE facilities and the permanent disposal of nuclear waste generated at DOE facilities require working with and within various types and levels of radiation fields. Once the nuclear waste is incorporated into a final form, radioactive decay will decrease the radiation field over geologic time scales, but the alpha-decay dose for these solids will still reach values as high as  $10^{18}$  alpha-decay events/gm in periods as short as 1,000 years. This dose is well within the range for which important chemical (e.g., increased leach rate) and physical (e.g., volume expansion) changes may occur in crystalline ceramics. Release and sorption of long-lived actinides can also provide a radiation exposure to backfill materials, and changes in important retardation properties (e.g., cation exchange capacity) may occur. The objective of this research program has been to evaluate the long-term radiation effects in materials used in processing high-level nuclear waste or materials in the near-field of a nuclear waste repository.

## 2. Research Progress and Implications

As of February, 2000, ninety percent of the originally proposed work has been completed. Since the project was delayed due to the move of both PI and co-PI from University of New Mexico to University of Michigan in 1997, a no-cost extension to 5/14/2000 has been requested by the PI and approved by DOE.

The long-term radiation effects in concerned materials have been evaluated with accelerated laboratory experiments utilizing energetic electrons and ions, which include *in situ* transmission electron microscopy (TEM) during irradiation using the HVEM/IVEM-Tandem National Facility at Argonne National Laboratory as well as ion exchange studies. The materials studied included: zeolites, layered silicates (smectite clays and mica), as well as crystalline silicotitanate (CST). CST is an important ion exchange material proposed for use in the chemical separation process of high-level liquid radioactive wastes (HLW).

*In situ* TEM has shown that all the studied materials (i.e., zeolites, mica, smectite clays and CST) are susceptible to irradiation-induced solid-state amorphization (i.e., the materials' unique sheet or porous crystalline structure is completely destroyed by irradiation). Amorphization can either be induced by ionization processes (as occurs with  $\beta$ - or  $\gamma$ - irradiation) or by direct displacement damage processes (as by  $\alpha$ -recoils). The critical doses for complete amorphization of these phases are as low as  $<0.1$  displacement per atoms (dpa) or  $10^8$  Gy in ionization energy deposition (a dose equivalent to that expected in a zeolite with 10 wt.% loading of  $^{137}\text{Cs}$  in 400 years). Amorphization may be preceded or accompanied with dehydration, layer spacing reduction and gas bubble formation which may cause significant volume swelling. The critical radiation dose for amorphization or bubble formation in zeolites strongly depends on the size of channels available in the structure for gas release. In the case of zeolites, CST and some layer silicates, radiation effects are significantly enhanced at higher temperatures. Our experiments have shown that amorphization or even partial amorphization will cause a dramatic reduction (up to 95%) in ion exchange and sorption/desorption capacities for radionuclides, such as Cs and Sr. Because the near-field or chemical processing materials (e.g., zeolites or CST) will receive a substantial radiation dose after they have incorporated radionuclides, our results suggest that radiation effects may, in some cases, retard the release rate of sorbed or ion-exchanged radionuclides. The results of these studies have a direct bearing on repository assessment (e.g., the extend to which zeolites can retard the release of radionuclides) and on the technologies used to process high-level liquid wastes (e.g., separation of  $^{137}\text{Cs}$  from HLW using CST at the Savannah River Site).

### 3. Planned Activities

A systematic study on the changes in microstructure and ion exchange capacity of zeolite-NaY after proton beam irradiation is underway and will be completed by the extended ending date of the project. Also, analyses of recently acquired data from clays and CST will also be completed during the next three months. A final report of the project will be submitted within 90 days of the extended ending date of the project.

### 4. Information Access

(A) Web Site: <http://relw.engin.umich.edu>

(B) Publications Generated under the Project:

1. L.M. Wang, S.X. Wang, W.L. Gong and R.C. Ewing, "Temperature dependence of Kr ion-induced amorphization of mica minerals", *Nuclear Instruments and Methods in Physics Research B* 141 (1998) 501-508.
2. L.M. Wang, "Application of advanced transmission electron microscopy techniques in the study of radiation effects in insulators", *Nuclear Instruments and Methods in Physics Research B* 141 (1998) 312-325.
3. L.M. Wang, S.X. Wang and R.C. Ewing, "Radiation effects in zeolite: Relevance to near-field containment", *Proceedings of the 9th Annual International High-Level Radioactive Waste Management Conference* (American Nuclear Society, Las Vegas, NV, May 11-14, 1998), pp. 772-774.
4. L.M. Wang and R.C. Ewing, "Transmission electron microscopy study of radiation effects in materials for nuclear waste disposal", *Electron Microscopy 1998, Proceedings of the 14<sup>th</sup> International Congress on Electron Microscopy* (Cancun, Mexico, Aug 31-Sept. 4, 1998), Edited by H.A. Calderón Benavides and M. José Yacamán, Vol. 2, 825-826.
5. S.X. Wang, L.M. Wang and R.C. Ewing, "Electron irradiation of zeolites", *Materials Research Society Symposia Proceedings* 540 (1999) 361-366.
6. B.X. Gu, S.X. Wang, L.M. Wang, and R.C. Ewing, "Radiation and thermal effects on the structure and ion-exchange/retention capacity of zeolites", *Proceedings of the International Conference on the Future Nuclear Systems—Global'99* (American Nuclear Society, Jackson Hole, Wyoming, August 29-September 3, 1999).
7. B.X. Gu, L.M. Wang and R.C. Ewing, "The effect of amorphization on the Cs ion exchange and retention capacity of zeolite-NaY", *Journal of Nuclear Materials* 278 (2000) 64-72.
8. S.X. Wang, L.M. Wang and R.C. Ewing, "Electron and ion irradiation of zeolites", *Journal of Nuclear Materials* 278 (2000), in press.
9. Binxi Gu, Lumin Wang, Phil A. Simpson, Leah D. Minc and R.C. Ewing, "Radiation and thermal effects in zeolite-NaY", in: *Scientific Basis for Nuclear Waste Management XXIII*, R.W. Smith and D.W. Shoosmith eds. (Material Research Society, Warrendale, PA, 2000), in press.
10. L.M. Wang, B.X. Gu, S.X. Wang and R.C. Ewing, "Radiation effects on materials in the near-field of a nuclear waste repository", *Proceedings of the 24<sup>th</sup> International Symposium on the Scientific Basis for Nuclear Waste Management*, submitted.
11. B.X. Gu, S.X. Wang, L.M. Wang and R.C. Ewing, "The effects of proton beam irradiation on the Cs ion exchange capacity of zeolite-NaY", *Journal of Nuclear Materials*, to be submitted.