

Task 36

Evaluation of Cs/Sr Waste Form for Long Term Storage and Disposal

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BACKGROUND

To maximize the utilization of a proposed repository facility, the short-term decay heat generated by high-level radioactive waste must be removed from the waste stream. One proposed advanced fuel cycle strategy calls for the separation of cesium and strontium from used nuclear fuel in order to minimize the short-term heat loading in a repository facility. The separated waste stream will be converted to an aluminosilicate waste form, stored for decay (approximately 300 years), then managed as low-level radioactive waste. The goal is to examine two potential concerns regarding the long-term performance of this proposed cesium/strontium waste form.

To facilitate long-term storage, the disposal containers will need to be able to survive for the entire storage interval. The first aspect of the project will explore the potential interaction of the aluminosilicate waste form with the storage canister materials to determine if there is any corrosion or chemical interaction concerns for the storage of the materials. At the end of the storage interval, most of the cesium (^{137}Cs) in the waste form will have decayed to its daughter, barium (^{137}Ba). While this decay provides a significant reduction in the decay heat generated by the waste form, it poses a new concern. Barium is hazardous, and is identified by the U.S. Environmental Protection Agency as a hazardous constituent under the Resource Conservation and Recovery Act (RCRA). To dispose of any material containing a RCRA-identified constituent, the material must be demonstrated to be durable enough to prevent the release of the hazardous component or must be treated as hazardous waste. For the Cs/Sr waste stream, failure to contain the barium within the waste form would require disposal as a mixed waste stream, greatly increasing the disposal costs. Understanding the potential impacts of radioactive damage, high storage temperatures, and the crystallographic im-

pacts of the decay transmutation itself on the performance of the waste form 300 years from now poses a significant challenge.

RESEARCH OBJECTIVES AND METHODS

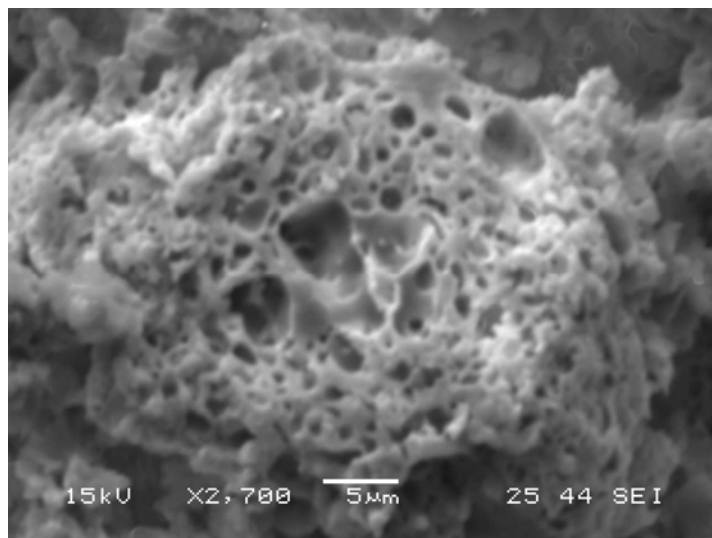
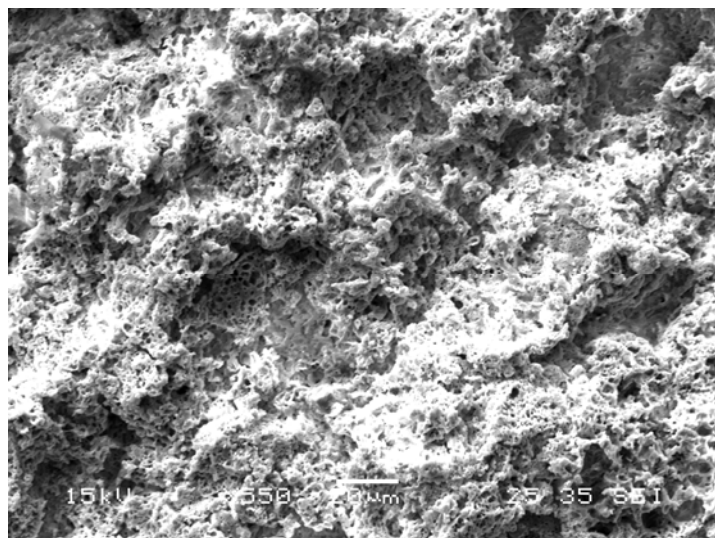
The goal of this task is to develop, characterize, and optimize the proposed aluminosilicate waste form for a separated cesium and strontium waste stream. The research effort at UNLV will be divided into two subtasks:

- Materials compatibility, and
- Waste form optimization and performance.

The materials compatibility subtask will examine the potential for chemical interactions between the waste form material and proposed structural materials for the disposal container (carbon steel, stainless steel, etc.). The waste form performance task will examine the leach resistance of the waste form, with particular attention to barium retention.

The research objectives of this project are:

- To characterize the Cs/Sr-loaded aluminosilicate waste form ceramic,
- To examine the impact of fabrication process parameters on the product waste form,
- To evaluate the potential for chemical interactions between the waste form and container material, and
- To examine the degradation and alteration behavior of the waste form.

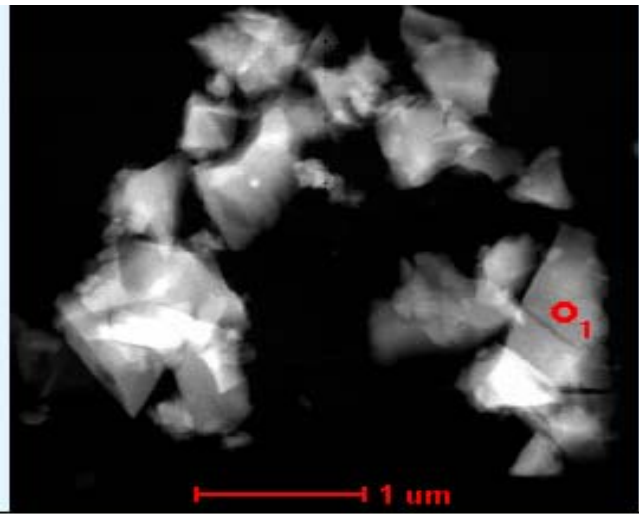


Cs/Sr/Ba-loaded waste form matrix at 550X (a) and 2700X (b) magnification.

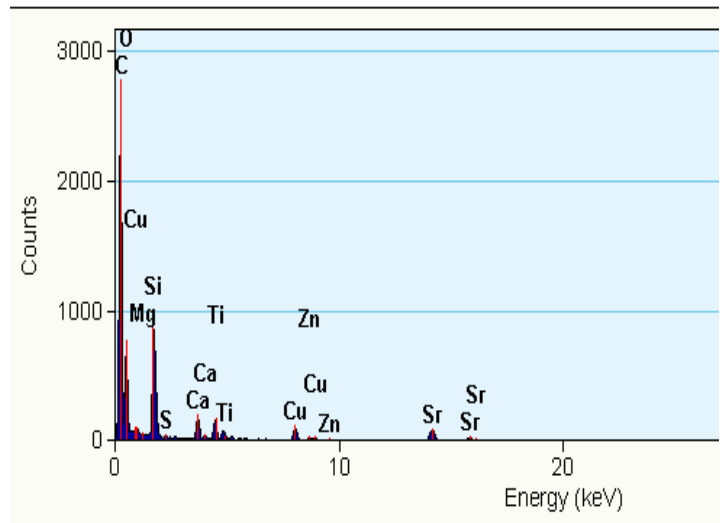
RESEARCH ACCOMPLISHMENTS

To optimize the Cs/Sr waste form, the impact of fabrication process parameters on the final product had to be evaluated. Initial experiments examined the impact of sintering temperatures on the waste form. Thermogravimetric analysis (TGA) identified the temperatures where sorbed and interstitial water is driven from the waste former matrix, as well as the temperatures where the bentonite waste former material undergoes a phase transition to a glass, establishing an upper temperature limit for the fabrication of the waste form. The impact of sintering time and temperature on the waste form and on the starting materials was examined by X-ray Diffraction (XRD) analysis. Significant differences in the crystallinity as well as phase composition of the Cs/Sr-loaded waste form were observed from increasing the sintering temperature from 800 °C to 1000 °C, with additional changes observed when the sintering time at temperature was increased to over 20 hours. Characterization of the product waste forms, including identification of all the composite phases, will continue through the end of the project.

Characterization of the product morphology by scanning electron microscopy showed that the current batch processing method (in which the bentonite clay is loaded with Cs and Sr in solution, dried, then sintered) yields a highly porous material with a sponge-like morphology. Coupled with the results of the TGA analysis, this is likely due to the release of interstitial or matrix water during the early stages of the sintering step. Electron microprobe analysis (EMPA) showed a uniform distribution of Cs, Sr, and Ba throughout the sample matrix, at least at the micron scale.



(a)



(b)

TEM image of Cs/Sr/Ba-loaded Waste Form Matrix (a) with elemental analysis of indicated spot (b).

Research Staff

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ACADEMIC YEAR HIGHLIGHTS

- ◆ Identified glass transition temperature for bentonite waste former materials to establish waste form fabrication process limits.
- ◆ Identified and characterized barium- and strontium-bearing phases in sintered waste form matrix.

Further analysis by transmission electron microscopy (TEM) indicated, however, that the waste stream components are actually segregated into discrete phases, and that these discrete, sub-micron particles are distributed throughout the matrix. Initial characterization has identified separate Sr- and Ba-containing aluminosilicate phases in the host matrix. The Cs-bearing phase (or phases) in the matrix have not been identified and confirmed at this time.

FUTURE WORK

With the change in national program direction towards a combined fission product waste stream, work on the waste forms for separated cesium and strontium has been reduced to a lower priority. The work at UNLV will focus primarily on wrapping up the characterization of the waste matrices so that they could be pursued in the future if program directions change, focusing on the identification of cesium-bearing phases within the waste form matrix.