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

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BACKGROUND
In order to maximize the utilization of the proposed repository facility, the short-term decay heat generated by high-level waste must be removed from the waste stream. The proposed waste management baseline strategy for the GNEP program calls for the separation of cesium and strontium from used nuclear fuel in order to minimize the short-term heat loading in the 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 of this project is to examine two potential concerns regarding the long-term performance of a proposed cesium/strontium waste form. To facilitate long-term storage, up to 300 years, 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 $^{137}\text{Cs}$ in the waste form will have decayed to its daughter, $^{137}\text{Ba}$. 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 (EPA) 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 impacts 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 research effort at UNLV will be divided into materials compatibility and waste form performance subtasks. 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 subtask will examine the leach resistance of the waste form, with particular attention to barium retention. The performance subtask will also work with the ANL team to develop the cold analog samples for comparison with the aged $^{137}\text{Cs}$-bearing aluminosilicate.

To examine the potential for waste form-storage materials interactions, coupons of potential container materials (carbon steel, 316L stainless steel, Inconel 617, and Zircalloy-2) will be contacted with the Cs/Sr-loaded aluminosilicate waste form material. These coupons will be exposed at fixed temperatures for up to 6 months. The samples will be analyzed for weight loss as well as structural changes to determine if any material-material interactions would be expected for this waste form.

The performance of the waste form will be evaluated using the Toxic Constituent Leaching Protocol (TCLP), established by the EPA as a standard analysis to determine if a material requires disposal as a hazardous waste. The resistance of the waste form to leaching will be examined following the TCLP, with a particular focus on barium retention. Standard dissolution/leaching tests will also be performed for comparison. To evaluate the impact of radiologically produced barium in the waste form, an archived sample of $^{137}\text{Cs}$-bearing aluminosilicate material from ANL will be used to examine the chemical environment of the Ba. Analog cold samples will be prepared at UNLV for comparison and baseline measurements, and experimental protocols will be evaluated with the cold samples to evaluate the feasibility of performing leach testing on the hot, archived sample.

RESEARCH ACCOMPLISHMENTS
This effort represents a new project started in May 2007 with a kick-off meeting between the UNLV researchers and the national program collaborators. Since this meeting, the test plan and sample matrix has been developed, and the procurement for the metal coupons has been initiated.

FUTURE WORK
For the next academic year, the material chemical interaction experiments will be started to explore the corrosion of potential container materials in contact with the waste form. Samples of the waste form will be synthesized using single components of the waste stream to determine the speciation of the Cs, Sr, Y, Ba and Zr within the waste form by X-ray diffraction and electron microscopy.