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Evaluation of Potential Melt-Dilute/Neutron Absorber Systems for DOE SNF Disposal System

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ANS/00 Summary for a Proposed Full Paper

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Introduction

Approximately 50 metric tonnes heavy metal of aluminum-base spent nuclear fuel (Al-SNF) or 30,000 assemblies are being consolidated at the Savannah River Site. The melt-dilute (MD) technology option is being developed to allow ultimate disposal of these fuels in the Monitored Geologic Repository (MGR). Neutron absorbing materials are needed to maintain $k_{eff} < 0.95$ in desired packaging configurations. The aggressive chemical environment in the MGR is expected to lead to the reconfiguration of the contents of the codisposal waste package following waste package failure. This reconfiguration has the potential for increasing the reactivity of the waste package. The reconfiguration and redistribution of materials within the waste package are being investigated in an analytical and experimental program to support the criticality analysis. Further, the incorporation of neutron absorbing materials that will be integral to the MD SNF form is being investigated.

Description

Criticality control in waste packages for disposal at Yucca Mountain is governed by 10CFR60. Currently, the criticality analysis must show no possibility (probability< 10^{-6}) of criticality for 10,000 years following disposal. Therefore, both intact and degraded states of the waste package must be evaluated. The results of preliminary criticality analyses indicate that neutron-absorbing materials will be required to meet criticality requirements. A comprehensive criticality analysis of the waste package is currently underway. This criticality analysis includes geochemical modeling in accordance with the established methodology for conducting criticality analyses.[1]

The efficacy of a specific neutron absorbing material is controlled primarily by the degree to which the material is uniformly distributed in the MD SNF form. Other important properties include the material's susceptibility to selective leaching of the absorber material from the MD SNF form and the susceptibility to enhanced corrosion of the MD SNF from due to galvanic cell formation. Further, it is important to identify the degradation products of the melt-dilute/neutron absorber system and the relative solubility of these products at repository relevant pH values with respect to the fissile constituents of the MD SNF form degradation products.

A review of the natural occurrence of uranium bearing minerals within ore deposits in the western United States and an analysis of thermochemical data and the products formed during laboratory corrosion experiments were performed. The hydrated oxides and silicates of uranium and hydrated aluminum oxides or alumino-silicates are the most likely final degradation products.

Testing is underway to determine the degradation properties of the melt-dilute/neutron absorber systems being considered. These tests include vapor corrosion and aqueous corrosion tests. The tests utilize J-13 and modified J-13 water chemistry to simulate the chemistry effects due to the radiolytic decomposition of water and to the degradation of DHLW glass logs. Also, galvanic corrosion tests will be conducted in the test program. Aqueous corrosion tests include both static and flow tests as described below.

Static tests are being conducted to measure the following properties/behavior of potential melt-dilute/neutron absorber systems: selective leaching, dissolution rates, and corrosion/degradation product formation. Static tests are commonly used for corrosion studies and are generally referred to as immersion or coupon testing. Various analysis techniques must be used in conjunction with the test to measure the desired properties.

Flow testing is being conducted to determine both the dissolution and corrosion characteristics of for inclusion in a codisposal waste package. Flow tests had been used previously to study the dissolution response of commercial spent fuel. Flow test parameters included water composition, temperature, flow cell design, and flow rate. These parameters are expected also to effect both the dissolution rate and corrosion of potential melt-dilute/neutron absorber systems.

The corrosion and stability of potential melt-dilute/neutron absorber systems, which may impact the efficacy of the neutron absorbing material, is being assessed through accelerated electrochemical testing. The electrochemical technique, an accepted standard test method, identifies and characterizes the modes of corrosion for a given material/environment system. The electrochemical tests being used involve controlling a potential applied to a sample and measuring the responding current. The relationship between the current and potential is a function of the active corrosion mechanisms. The electrochemical testing is followed by metallographical analysis of the sample to characterize both the microstructural dependence of corrosion and the morphological changes from corrosion.

Results

The melt-dilute/neutron absorber systems currently being evaluated include melt-dilute plus gadolinium and melt-dilute plus gadolinium and hafnium. These systems have been selected based upon absorption cross-section upon geochemical considerations. Results of this testing program will provide input to the selection of the optimum melt-dilute/neutron absorber system for the disposition of Al-SNF.

[1] *Disposal Criticality Analysis Methodology Topical Report*, YMP/TR-004Q, Rev. 0, US DOE Office of Civilian Radioactive Waste Management, (November 1998).