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Dr. R. Weller
Office of Nuclear Safety and Safeguards
Nuclear Regulatory Commission
WASHINGTON, D.C. 20555
U.S.A.

Dear Dr. Weller:

As we discussed on the phone recently, here is a program description for our work on spent fuel. I hope you will forgive the informal nature of the description, which was cribbed from various internal documents; our formal program document is too out of date to be of much use to you. As you can see, our studies cover a broad range of applied and underlying research related to spent fuel and UO_2.

We would be pleased to have you come and visit Whiteshell Laboratories at your convenience so that you can look at our work first hand. Please give me a call if you are interested, or if you require further publications or other information.

Yours sincerely,

L. Johnson

L.H. Johnson, Manager
Fuel Waste Technology Branch

c: C.A. McDovall
D.W. Shoesmith

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SPENT FUEL STUDIES PROGRAM

OBJECTIVE

The objective of the spent fuel studies program is to develop a model to describe radionuclide release from spent fuel under disposal conditions. Two main areas of experimental work contribute to this objective. The first involves studies of UO_2 dissolution, including the effects of ionizing radiation. The second is studies of fission product leaching and matrix dissolution of spent fuel.

BACKGROUND

A model for the dissolution of spent fuel in a disposal vault has been developed and implemented in SYVAC (the Systems Variability Analysis Code). The model assumes that short-term release of fission products such as ^{137}Cs, ^{129}I, ^{99}Tc and ^{14}C from the fuel sheath gap and grain boundaries will dominate the source term. In the longer term, dissolution of the spent fuel matrix, which is assumed to be relatively stable under the chemically reducing conditions of the disposal vault, is expected to occur via mass transport control. Studies of UO_2 dissolution in the presence of alpha-radiolysis of water are focusing on examining this assumption and developing the basis for a kinetic model, which has been described recently by Shoesmith and Sunder (1991).

SPENT FUEL LEACHING AND DISSOLUTION STUDIES

Gap Inventories

Studies of the leaching of ^{137}Cs and ^{129}I from spent fuel and the correlation with measured fission gas release and those calculated with the ELESIM gas release code are complete. Preliminary results on ^{14}C release have been recently reported (Stroes-Gascoyne et al., in press). Work on ^{14}C release, including measurement of the total ^{14}C content in spent fuel, is continuing.

Grain Boundary Inventories and Release Kinetics

Leaching of air-oxidized fuel specimens to determine ^{137}Cs, ^{90}Sr and ^{99}Tc inventories at grain boundaries suggest that these inventories are smaller than predicted using the ELESIM code. Further studies of air-oxidized powders and unoxidized crushed samples will be carried out using flow-through columns. These studies are being complemented with XPS studies of grain surfaces of spent fuel samples.

Matrix Dissolution Studies

A range of studies of spent fuel dissolution are being performed, including long-term (up to 15 years) static tests in oxidizing groundwaters at 25°C, long-term (2 year) static tests at 100°C in reducing groundwaters and flow-through tests at 25°C in oxidizing and reducing solutions. The rates measured in these experiments will be compared to those estimated with the electrochemical model of Shoesmith and Sunder (1991).

ORIGEN-S Validation

The ORIGEN-S code has been used to predict radionuclide inventories in spent fuel. A Pickering fuel element was dissolved in HNO_3 and the solutions were analyzed for 20 radionuclides of concern in waste management. Agreement between measured and predicted inventories was generally within 10%.

UO_2 DISSOLUTION STUDIES

Electrochemistry of UO_2

Studies of the mechanism of oxygen reduction on electrodes fabricated from standard unirradiated UO_2 are in progress. Experiments will focus on determining the values of parameters; (i) to specify a mathematical relationship for O_2 reduction; (ii) to extrapolate this expression to determine corrosion rates under anticipated vault redox conditions. The key parameters are the slope of the log (current) vs. voltage relationship and the reaction order with respect to the oxygen concentration. The impact of the degree of oxidation of UO_2 (extent of corrosion) and the concentration of the groundwater constituent carbonate on these parameters will also be characterized. Some data will be collected in the reference groundwater.

The kinetics of oxygen reduction will be investigated on two different specimens of SIMFUEL, one simulating 3 at.% and the other 6 at.% burnup. The results of these experiments will assist us to determine the impact of UO_2 properties on the cathodic processes likely to drive the oxidative dissolution of UO_2. They constitute a step in our attempts to determine the applicability of our experimental results to the interpretation of the behaviour of used fuel.

A series of electrochemical experiments will be performed to determine the steady-state dissolution currents on various specimens of UO_2. These results are required to improve the reliability of procedures for determining the oxidative dissolution rates for UO_2. This year we hope to finish experiments on standard unirradiated CANDU specimens.