

Studies

Commercial Spent Fuel Tests at ANL

Presented to: CLST Appendix 7 Meeting

Presented by: James Cunnane Argonne National Laboratory



U.S. Department of Energy Office of Civilian Radioactive Waste Management

July 7-8, 1999

Overview

- Test Types:
 - Unsaturated Testing on Spent Fuel Pellet Fragments and UO₂
 - Unsaturated Testing on Segments of Clad Fuel Rods
- For Each Test Type:
 - Test Configuration and Operation
 - Objectives and Scope
 - Results

Spent Fuel and UO₂ Tests

- Unsaturated "Drip" Tests on Spent Fuel Pellet Fragments
- High Surface Area to Solution Volume Batch Tests
- Unirradiated UO₂ "Drip" Tests

Schematic - Spent Fuel Unsaturated Tests

5



High Solid-Surface-Area-to-Solution-Volume Batch Tests

Batch Experiments

High S/V (~5000 m-1)

- Small Solution Volumes (0.5 mL)
- Crushed solids (0.2 g)

Analyze

- Solution
 - » changes in water film
- Solids
 - » reacted UO_2 or fuel
 - » alteration products



Objectives

- Examine the corrosion behavior of spent UO₂ fuels upon extended exposure to repository-relevant conditions
- The tests are designed to:
 - Determine the alteration modes
 - Determine the alteration products
 - Assess how the alteration modes and products influence radionuclide release

Scope and Status

- Drip tests:
 - Three fuel types; ATM-103 and 106 (~6y); ATM-109(~1y)
 - Three groundwater contact modes (HDR, LDR, Humid Air)
- PDT tests:
 - ATM-103 fuels
- Drip tests on unirradiated UO₂ (~14y)

ATM-103 High-Drip-Rate Test







Interval Release Fractions - Tc, U, Np, Pu



Interval Release Rates - Tc, I, Cs, Sr

High-Drip-Rate





Low-Drip-Rate





"Normalized" Rates of Release High-Drip-Rate Tests



ATM-103



ATM-106

Pu Distribution in High-Drip Tests

 The Pu in the leachate in the high-drip-rate tests was primarily associated with colloids or was sorbed on the stainless steel vessel's walls





Corrosion Layers



. ·

High Drip



General Corrosion

Vapor



High Drip



Low Drip



High Drip



Grain Boundary Corrosion



ATM-103 High Drip-Rate

Experimental Paragenesis



Drip Tests Reproduce Natural Alteration

Drip Tests (UO₂)

Nopal I (uraninite)



M&O Graphics Presentations/Appendix 7/YMCunnane1-070799.ppt 17

Np(V) in Dehydrated Schoepite

UO3•0.9H2O

$$(UO_2)O_{0.1}(OH)_{1.8}$$

$$(NpO_2)^+ + H^+ \leftrightarrow (UO_2)^{2+}$$







PDTs and Unsaturated Tests a Comparison

PDTs

Reactive surface area

» 2.5 to 9.5 x 10⁻³ m²

Alteration phases

- » Uranyl oxy-hydroxide
- » Uranyl silicates

Test Design

- » Small-scale batch tests
- » Simple vessel design

Unsaturated Tests

Reactive surface area » 1.7 x 10⁻³ m²

Alteration phases

- » Uranyl oxy-hydroxides
- » Uranyl silicates

Test Design

- » Flow-type tests
- » Complex vessel design & Drip apparatus

ATM-103 Fractional Tc Release PDT and HDR





Solution Compositions for UDTs (UO₂ and SNF)



Clad Fuel Tests

- Four Vapor Tests (ATM103; 3.5 inch segments; 175°C and 100% RH)
 - Capped at each end with Ti Swagelok fittings
 - 1/16-inch defect in cladding
- Five Drip Tests (2,3,4 inch ATM103, 2 inch ATM106, and 3 inch ATM109)
 - 0.75 mL of EJ13 injected twice a week

Clad Fuel Drip Tests



Clad Fuel Vapor Integrity Tests: 152-d Reaction



Pressure Transducer Output: ATM103



Summary

- General and grain boundary corrosion
- Incongruent release of radioelements
- U alteration phases consistent with observations of uraninite alteration
- "Normalized" corrosion rates (based on Tc) are higher in drip tests than in PDTs
- Solution U concentrations indicate solution saturation
- Clad fuel vapor and drip tests started