U.S. DEPARTMENT OF ENERGY
OFFICE OF CIVILIAN RADIOACTIVE WASTE MANAGEMENT

NUCLEAR WASTE TECHNICAL REVIEW BOARD
FULL BOARD MEETING

SUBJECT: GLASS TESTING AND COLLOID EVALUATIONS

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PLAZA SUITE HOTEL • LAS VEGAS, NEVADA
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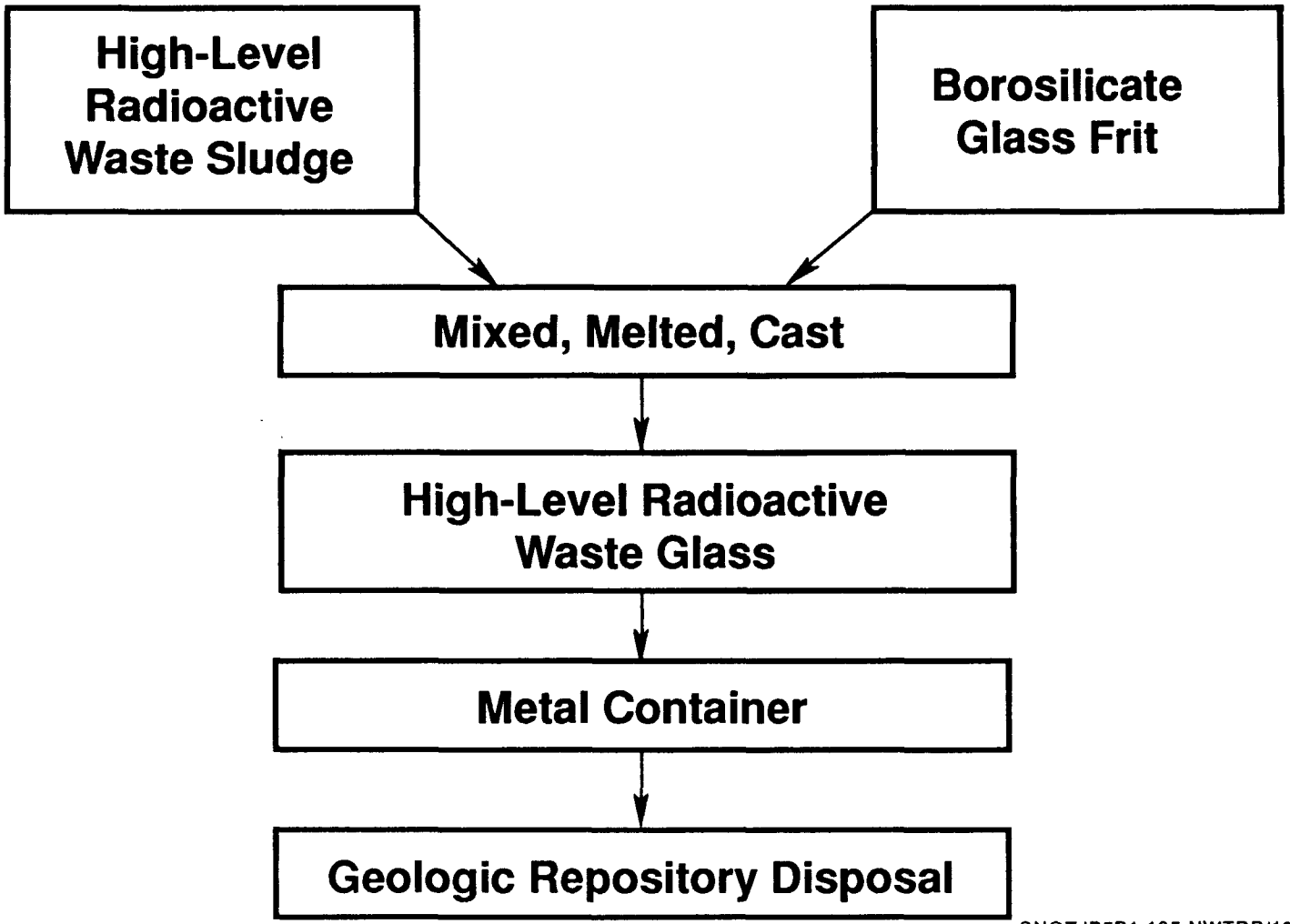
Background

Glass will be produced at Savannah River, West Valley, and Hanford and disposed of in a geologic repository



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Objective

The objective of the joint testing and modeling program is to evaluate glass reaction under a range of conditions to provide source-term information that can be used for design and risk-assessment activities

Purpose

There are two purposes:

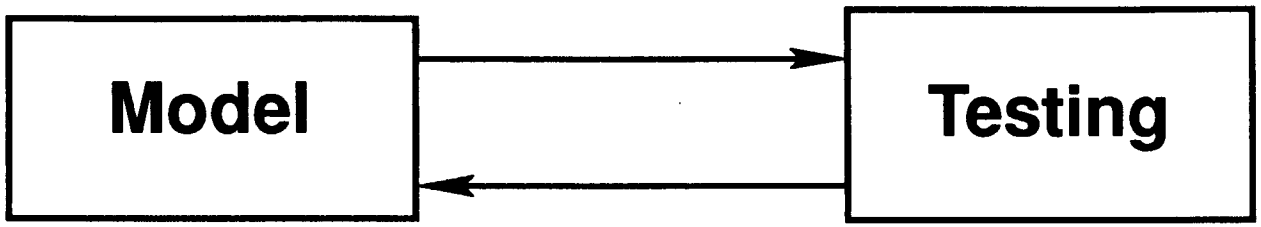
- **Support start-up of the vitrification facilities**
- **Support repository licensing**

Approach

The approach fits well into the American Society of Testing Methods (ASTM) format for prediction of long-term material performance

Identify Materials	Identify Credible Conditions
Glass of varying composition	Unsaturated
• Defense Waste Processing Facility • West Valley	
Continual Refinements	Have to rethink basis premises

Approach



- Must be mechanistically based
- Cannot be empirical extrapolation
- Modeling cannot precede testing

- Response
- Accelerated
- Service condition
- Confirmation
- Validation

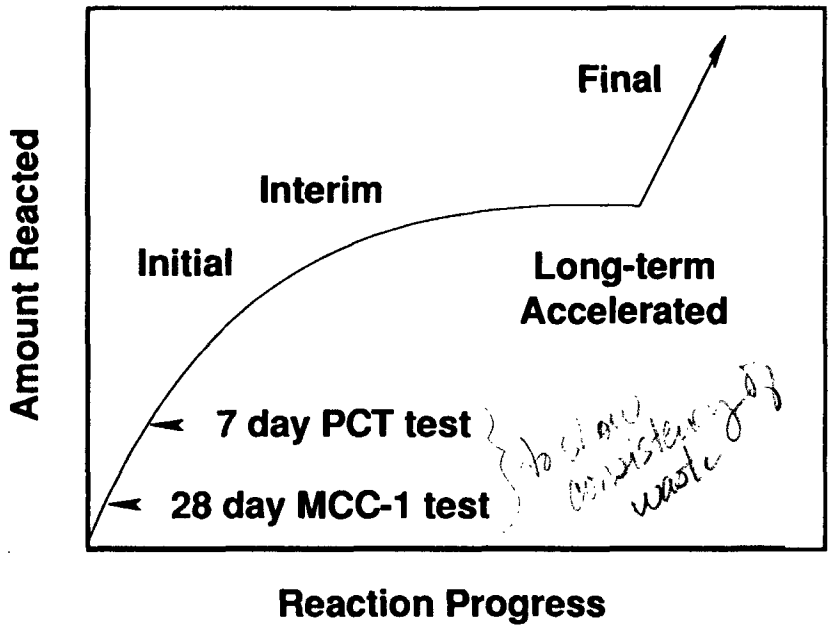
The variability of conditions anticipated for the Yucca Mountain site offers a challenge in designing and performing tests to evaluate waste-form performance

What Does Unsaturated Mean?

- **Humid air**
- **Dripping water--intermittent flow**
- **Small amounts of standing water with very slow exchange** static conditions

Approach

Glass reaction occurs in different stages



Source-term information includes

- Glass reaction rate
- Radionuclide release and distribution

Must be evaluated at each stage

Results

The first step in the evaluation process has been to compile information related to glass

"High-Level Nuclear Waste Borosilicate Glass: A Compendium of Characteristics"

- **Production and transportation**
- **Durability (testing)**
- **Modeling**
- **Analogues (natural, historical, commercial)**

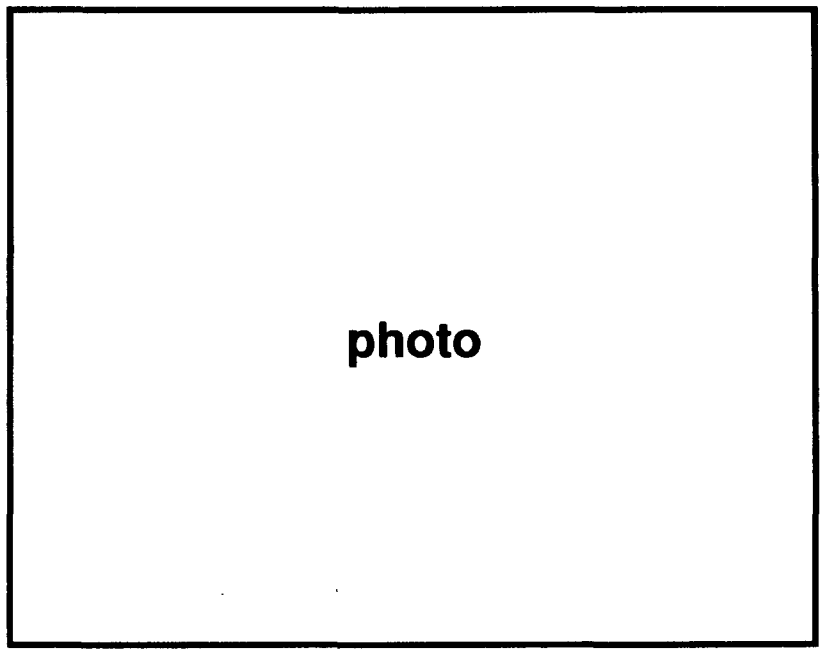
This is a review of world-wide information

Results: Humid Air

In humid air, reaction occurs between a thin film of water and the waste form:

- The water becomes rapidly saturated, with respect to glass components, and secondary phases form. These secondary phases set the "steady-state" concentration levels and drive the reaction
- Temperature, relative humidity, and glass composition are important variables
- As reaction occurs, the glass ages and sorbs additional water from the air
- Eventual contact of aged glass with water provides source-term information

The surface of the vapor phase reacted glass is covered with alteration phases of unique structure and composition



Alteration Products

Reacted Glass

However, under certain circumstances the formation of a stable phase may actually promote reaction

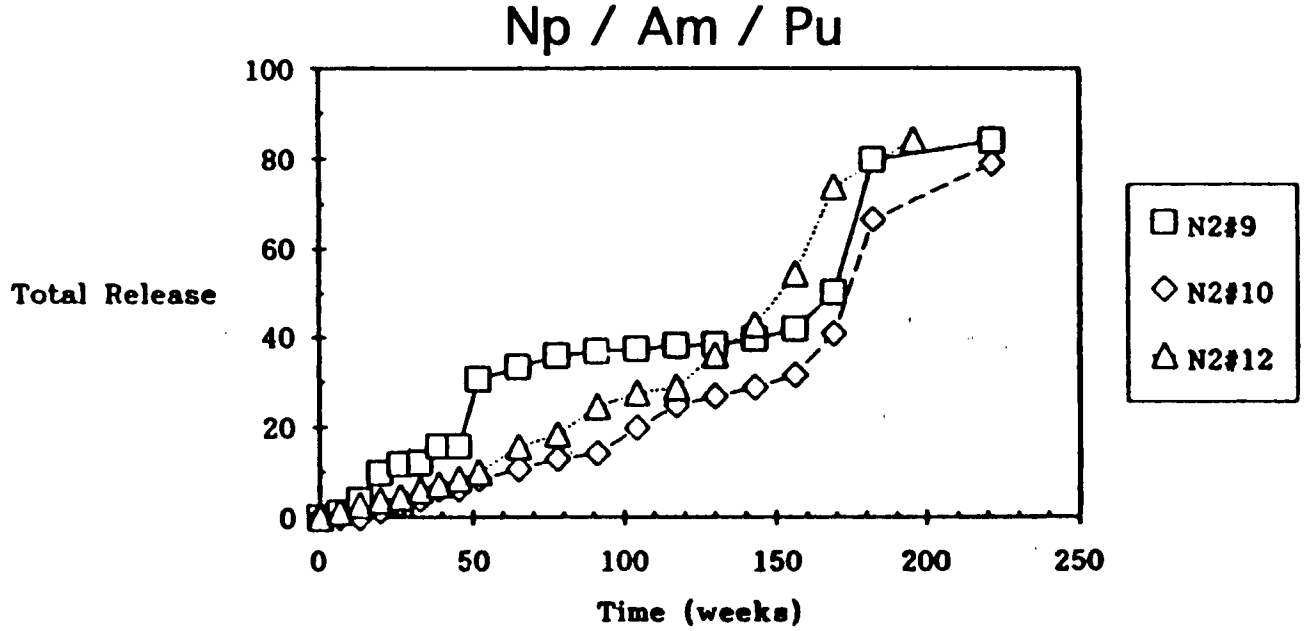
Results: Intermittent or Dripping Water

Tests have been conducted to examine many variables but focus on glass/canister

- **As-cast and aged glass**
- **Actinide-doped and fully radioactive**
- **Varying flow rates**
- **Sensitized stainless steel**
- **Tests in progress for eight years**

Results: Intermittent or Dripping Water

For as-cast, actinide-doped glass, the actinide release over long time-periods proceeds at a fairly continuous rate



- Pu/Am - suspended in solution
- Np - dissolved in solution

Results: Intermittent or Dripping Water

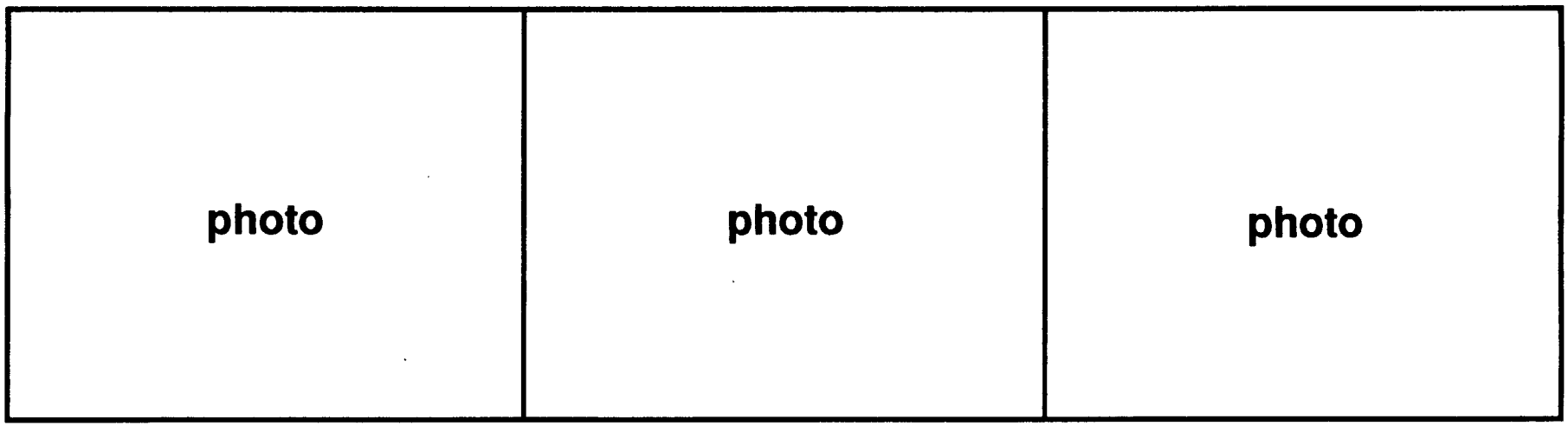
For aged, "fully" radioactive glass, the leachate becomes concentrated in cations and anions leached from the reacted glass, and all the actinides are dissolved in solution

	Solution Composition (ppm)	
	As-Cast	Aged
Li	10	1,000
B	10	20,000
Na	100	40,000
Si	50	1,500
Cl^-	9	200
$SO_4^=$	100	8,000
$PO_4^=$	0	400

Sequential filtering through 30 A filters does not reduce significantly the Pu/Cm in solution

165 Glass

- **The hydrolysis and <u>in situ</u> restructuring now result in a layer not attached to the glass surface**



Results: Static Tests

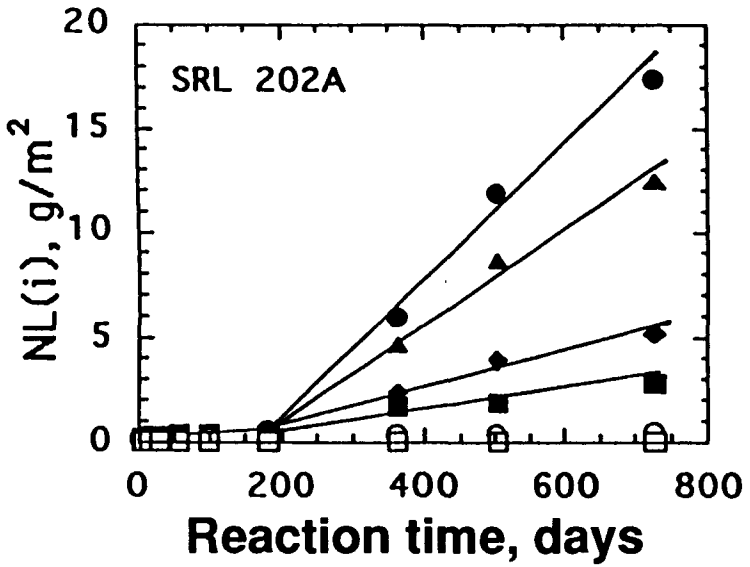
Static tests are used to represent the filled canister reaction mode

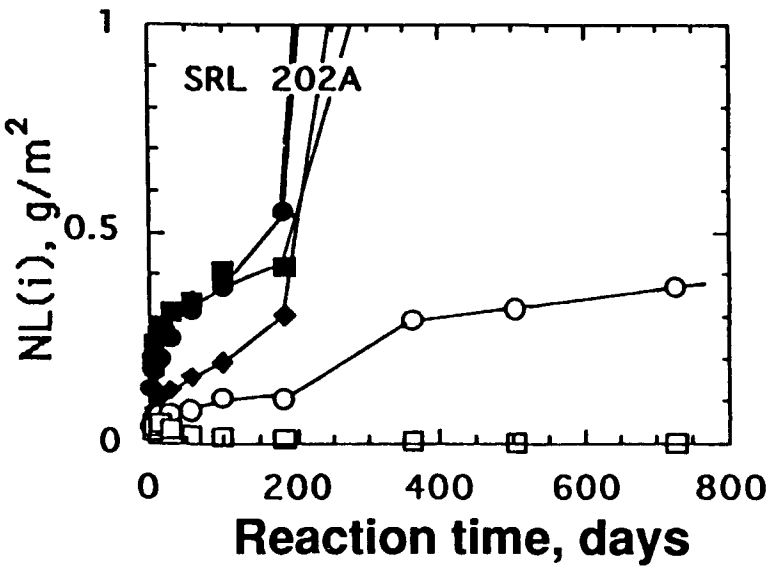
- **Batch tests**
- **Varying SA/V (3040 - 20,000 m^{-1})**
- **90°C**
- **EJ-13 groundwater**
- **Long-term**
- **Full suite of analyses (solution, colloids, layers)**

Results: Static Tests

By performing the tests at high SA/V and long time-periods, all the potential reaction stages are observed

SRL 202 based glass





The final rate can be greater than the forward rate, and within the envelope of acceptable glasses ranges from 0.04 to ~ 1 gm/m^2/day

Results: Static Tests

The final rate is controlled by the leachate pH and the secondary phases that form upon the onset of rate increase

The secondary phases include

- **Amorphous Si**
- **Clinoptilolite**
- **Clay**
- **Weeksite**

202 based glass

Outer layer

Reacted glass

Results: Colloids

To evaluate fully source-term data, the distribution of radionuclides in solution must be known

Background

The types of colloids include

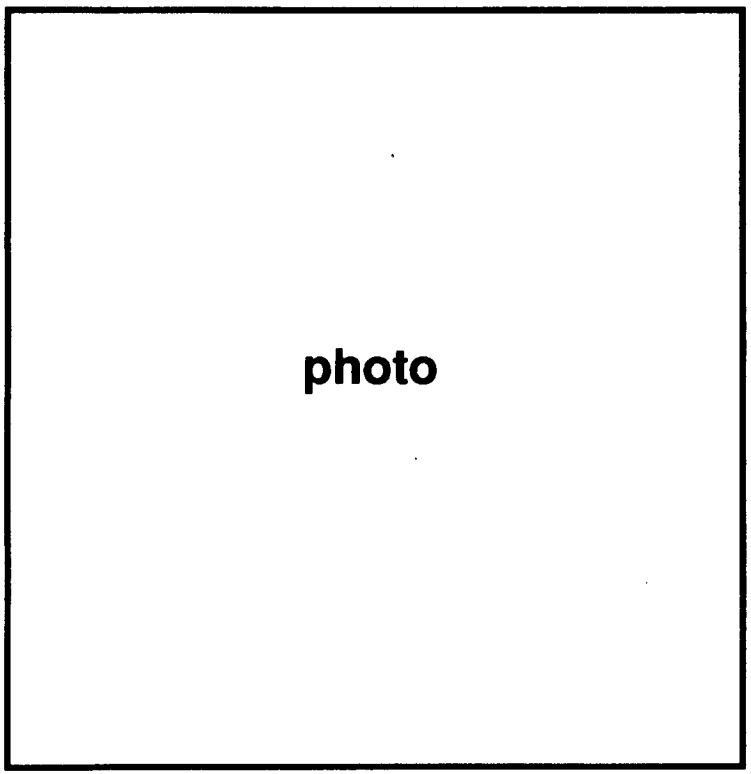
- **Radiocolloids (hydrolysis)**
- **Pseudocolloids (sorption)**
- **Primary colloids (directly from waste)**

Colloids

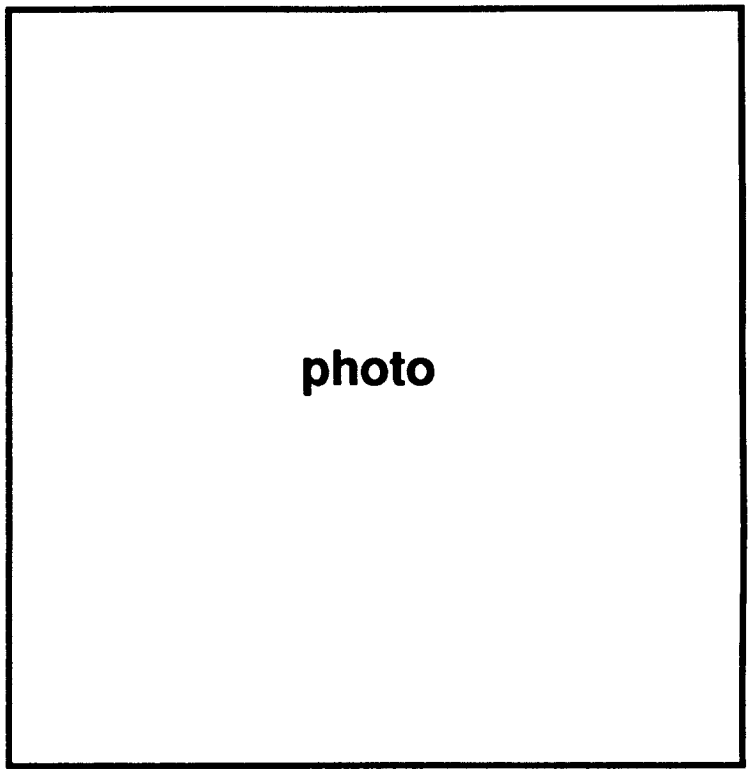
Objectives

- Determine whether radionuclide-containing colloids are formed in waste-form reactions
- Characterize any colloidal material observed
- Characterize the transport behavior

The Nature and Size Distribution of Actinide-Bearing Phase in Solution is Dependent upon Glass and Test Condition

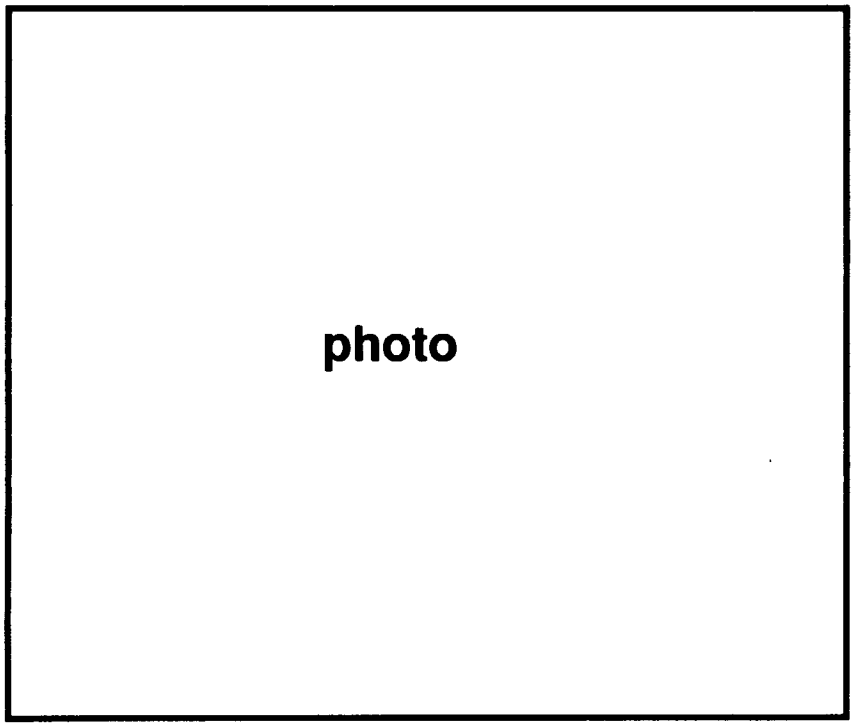


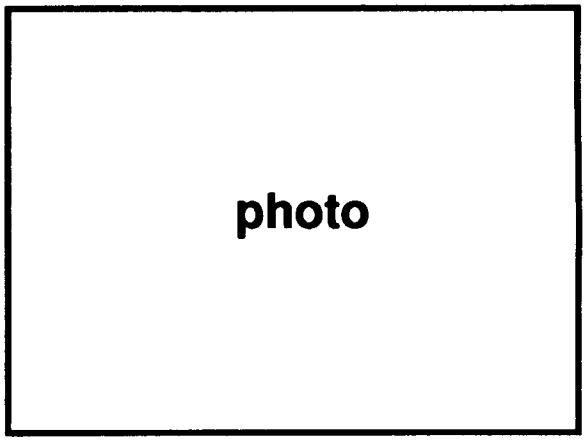
From solution

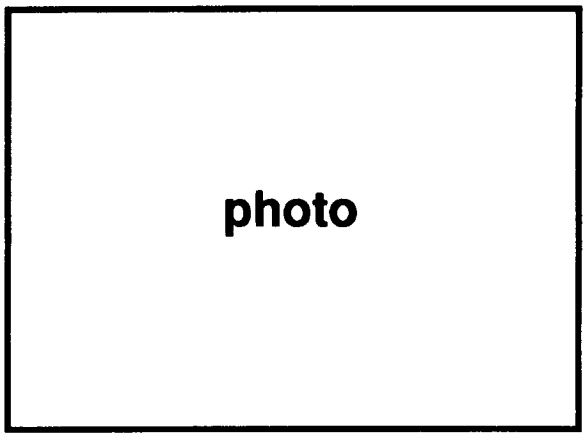


From glass

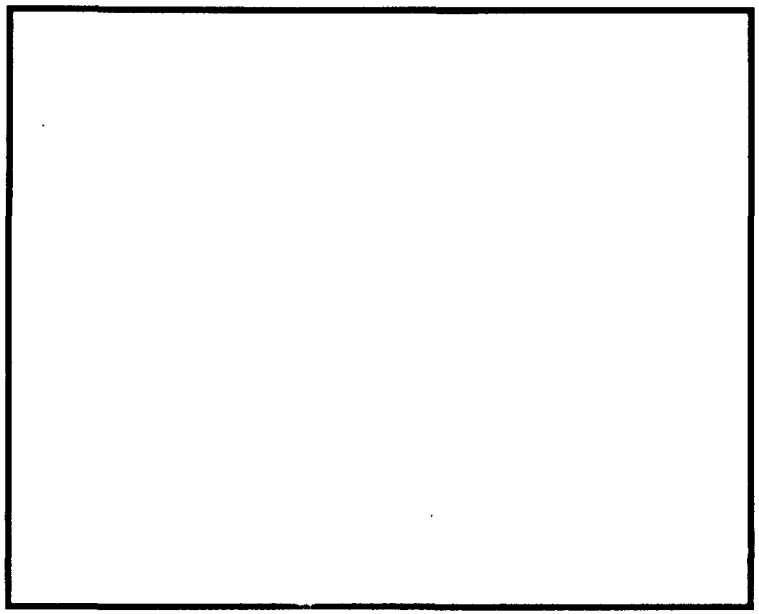
The Isolation and Identification of Actinide-Bearing Phases in Solution is Essential





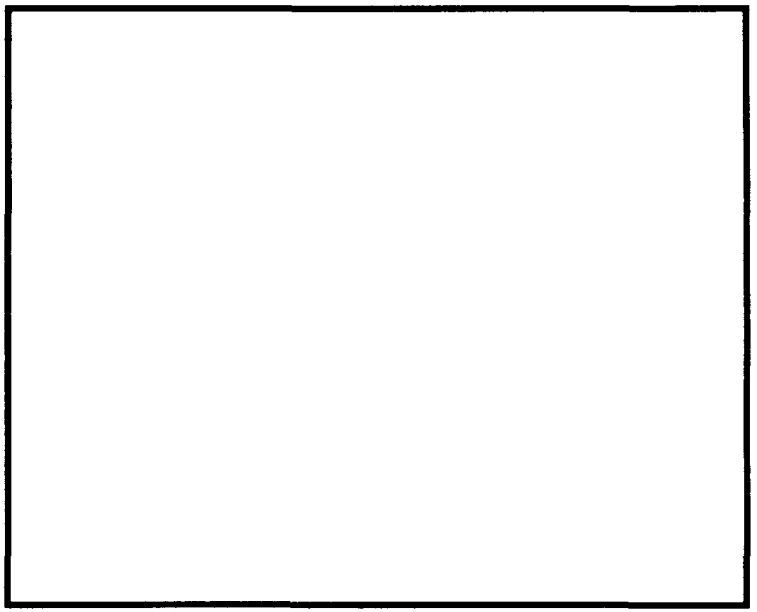


Spallation of Glass During the Drip Test Results in Increased Release



Mag = 400X

Spallation of reacted layer exposing base glass



Mag = 10,000X

Precipitation of clay onto newly exposed base glass

Colloids: Summary

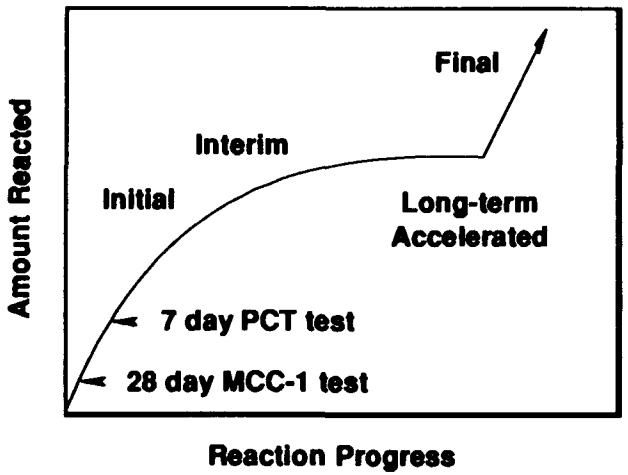
As the waste form reacts, it strongly influences the distribution of radionuclides to solution. It is possible to identify the colloidal species, the size distribution, and the radionuclide content

- **Primary colloids form directly from glass reaction due to spallation of material from the glass, contain concentrated Pu/Am phases, and remain suspended in solution**

- **Pseudocolloids form as glass dissolution products nucleate in solution (J-13 > DIW). The distribution in solution depends on the ionic strength of the leachate**

Concluding Remarks

In terms of the reaction progress diagram, the source-term information depends where one is located on the diagram. This position depends on the glass/water contact conditions



- **Humid air ages glass and affects subsequent radionuclide release**
- **Intermittent contact yields constant release rate with Pu/Am suspended in solution (as-cast) or dissolved (aged)**
- **Static yields final reaction rate after extended reaction time with actinides likely retained in the reacted glass**