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

PRESENTATION TO THE NUCLEAR WASTE TECHNICAL REVIEW BOARD

SUBJECT: SPENT FUEL LEACHING: HOT CELL TESTS

PRESENTER: DR. CHARLES N. WILSON

PRESENTER'S TITLE AND ORGANIZATION: SENIOR RESEARCH SCIENTIST
BATTELLE, PACIFIC NORTHWEST LABORATORY
RICHLAND, WASHINGTON

PRESENTER'S TELEPHONE NUMBER: (509) 376-5354

AUGUST 28-29, 1990

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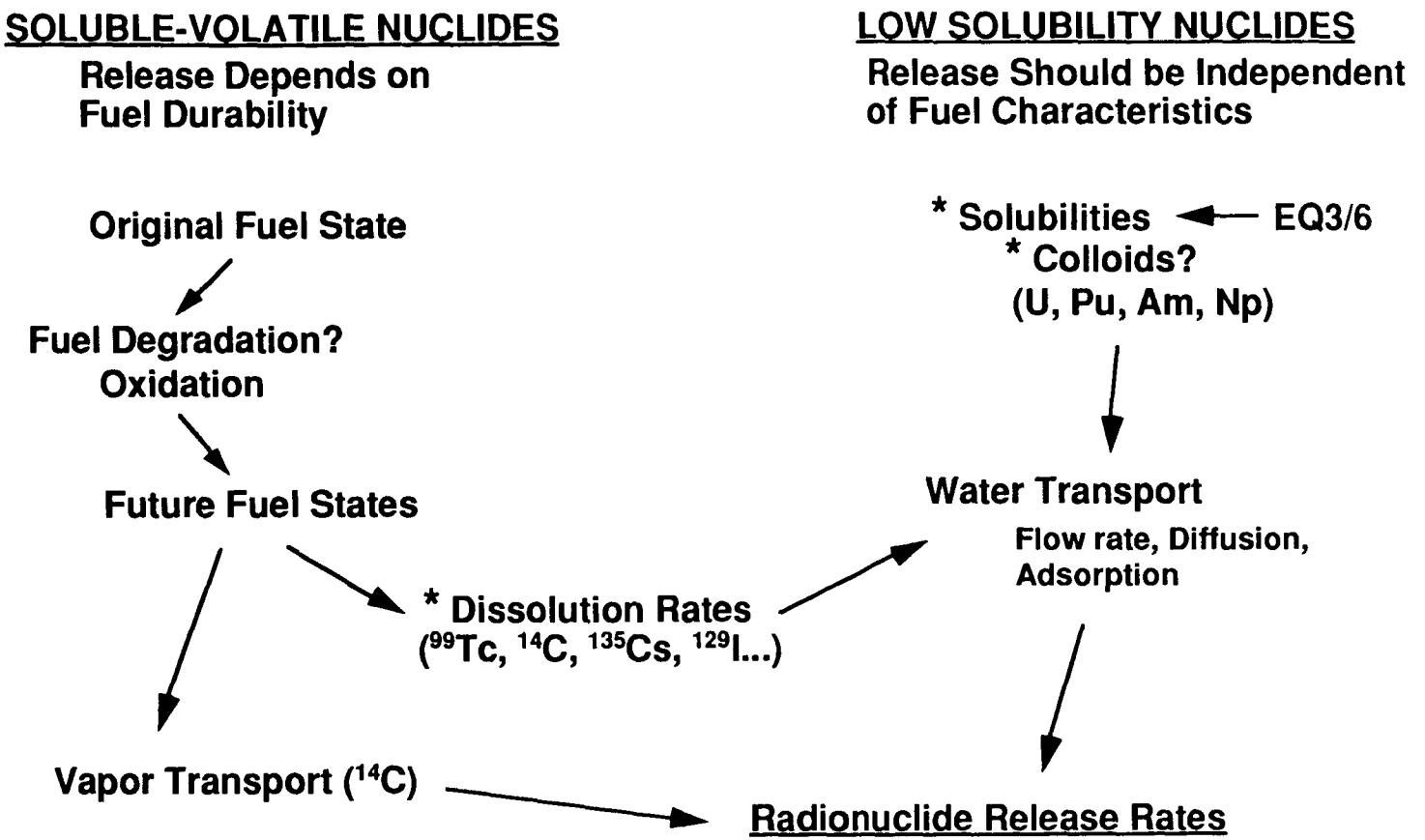
TOPICS DISCUSSED

- **Properties of spent fuel that influence radionuclide release behavior**
- **Methods used for testing release behavior of spent fuel in the laboratory**
  - Semi-static tests
  - Flow-through tests
- **Highlights of laboratory test results**

WHY TEST SPENT FUEL DISSOLUTION BEHAVIOR?

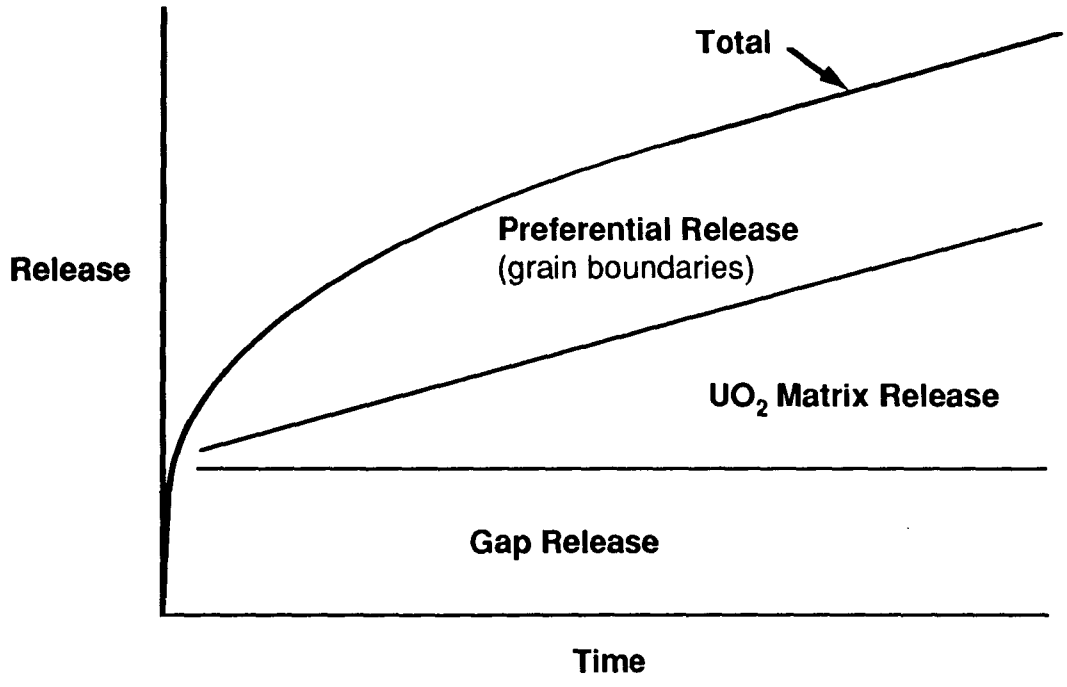
- **Spent fuel will likely be the dominant source** (compared to glass) **of soluble radionuclide** (^{99}Tc, ^{14}C, ^{135}Cs, ^{129}I...) **release**
- **Tests with real fuel specimens are required to obtain needed data such as**
  - — Dissolution rates for soluble radionuclides (initial and continuous)
  - — Identification of secondary phases controlling solubilities
  - — Amounts of radionuclides present as colloids
- **Results provide solubility and secondary phase data for validation of geochemical models such as EQ3/6**

FACTORS AFFECTING RELEASE



* Indicates properties measured in dissolution tests

SOLUBLE RADIONUCLIDE RELEASE



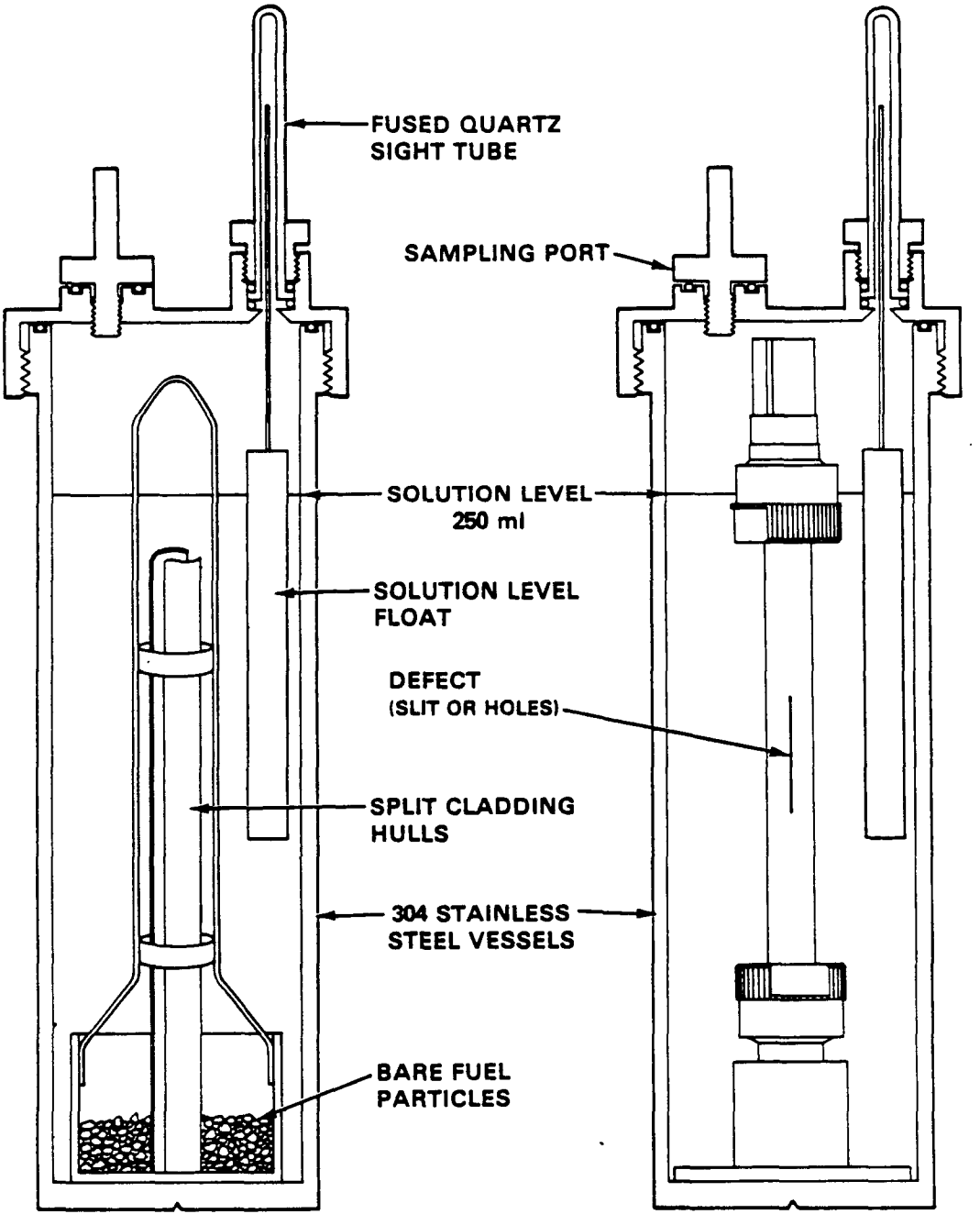
- **Rapid release of "gap inventories" with initial water contact (days)**
- **Preferential release from grain boundaries and other sources of radionuclide concentration (years)**
- **Releases are controlled by matrix dissolution after exposed grain boundaries and other sources of radionuclide concentration become depleted** (assuming fuel is not substantially degraded by oxidation)

TESTING METHODS

- **Semi-Static**
  - Periodic solution samples, sequential test cycles
  - Gives data on steady-state actinide concentrations, secondary phases and soluble radionuclide release ("gap" and continuous) rates
  - Matrix dissolution rates are not measured
  - Three test series conducted during FY-1983 through FY-1987
- **Flow-Through**
  - Matrix dissolution rates can be measured
  - Developmental tests conducted with unirradiated UO_2 during FY-1989 and FY-1990

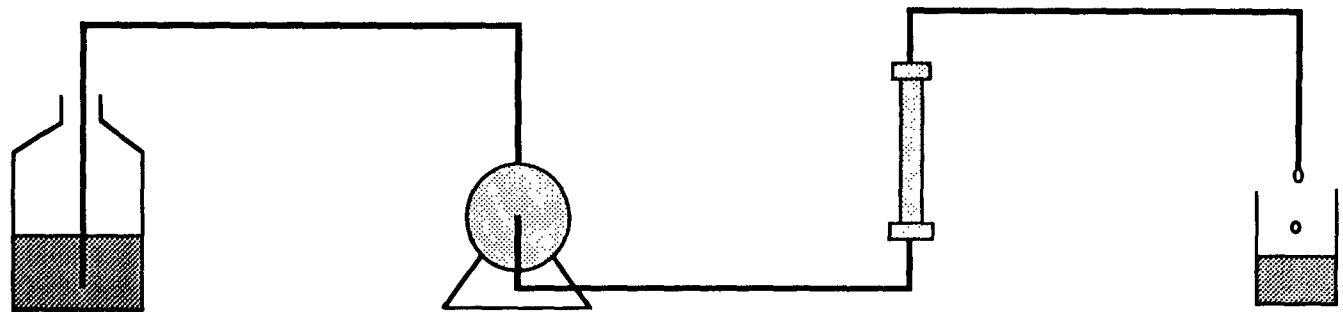
SEMI-STATIC TEST APPARATUS

SERIES 3 TEST CONFIGURATIONS



HEDL 8512-122.1

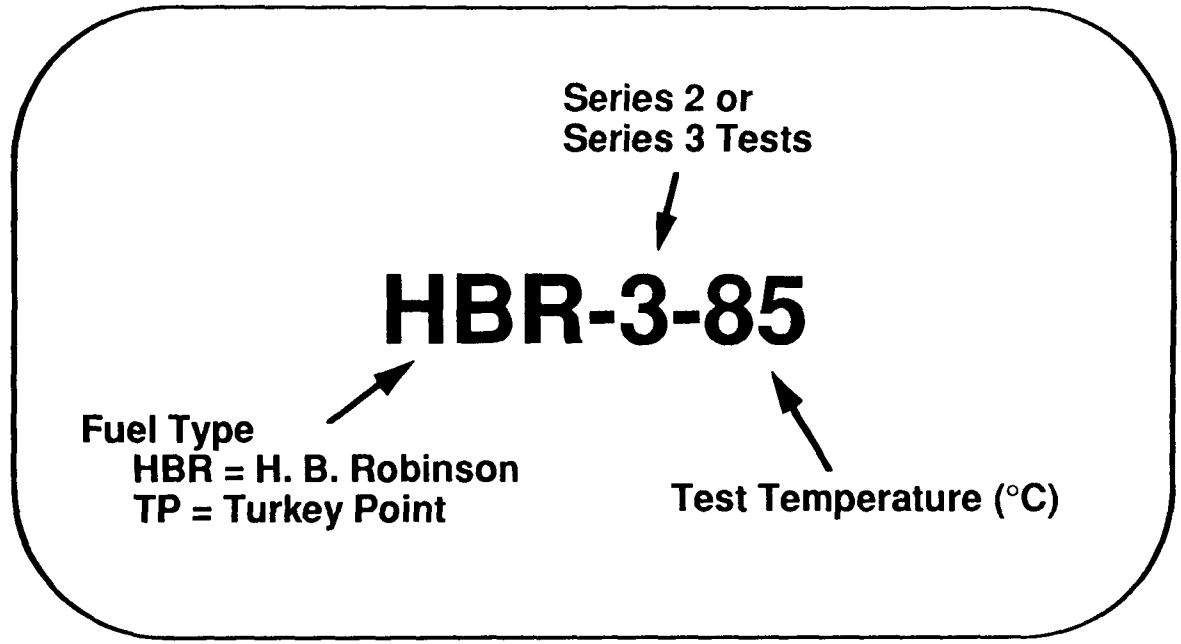
FLOW-THROUGH TESTING



- **OBJECTIVE - To measure uranium and soluble nuclide dissolution in a test where all measured species remain in solution**
- **FLOW RATE - Sufficiently high such that all dissolved uranium remains in solution, but low enough so that soluble nuclides reach measurable concentrations (a difficult compromise)**

SEMI-STATIC TEST IDENTIFICATION FOR FOLLOWING SLIDES

- HBR-2-25
- TP-2-25
- HBR-3-25
- HBR-3-85
- TP-3-85



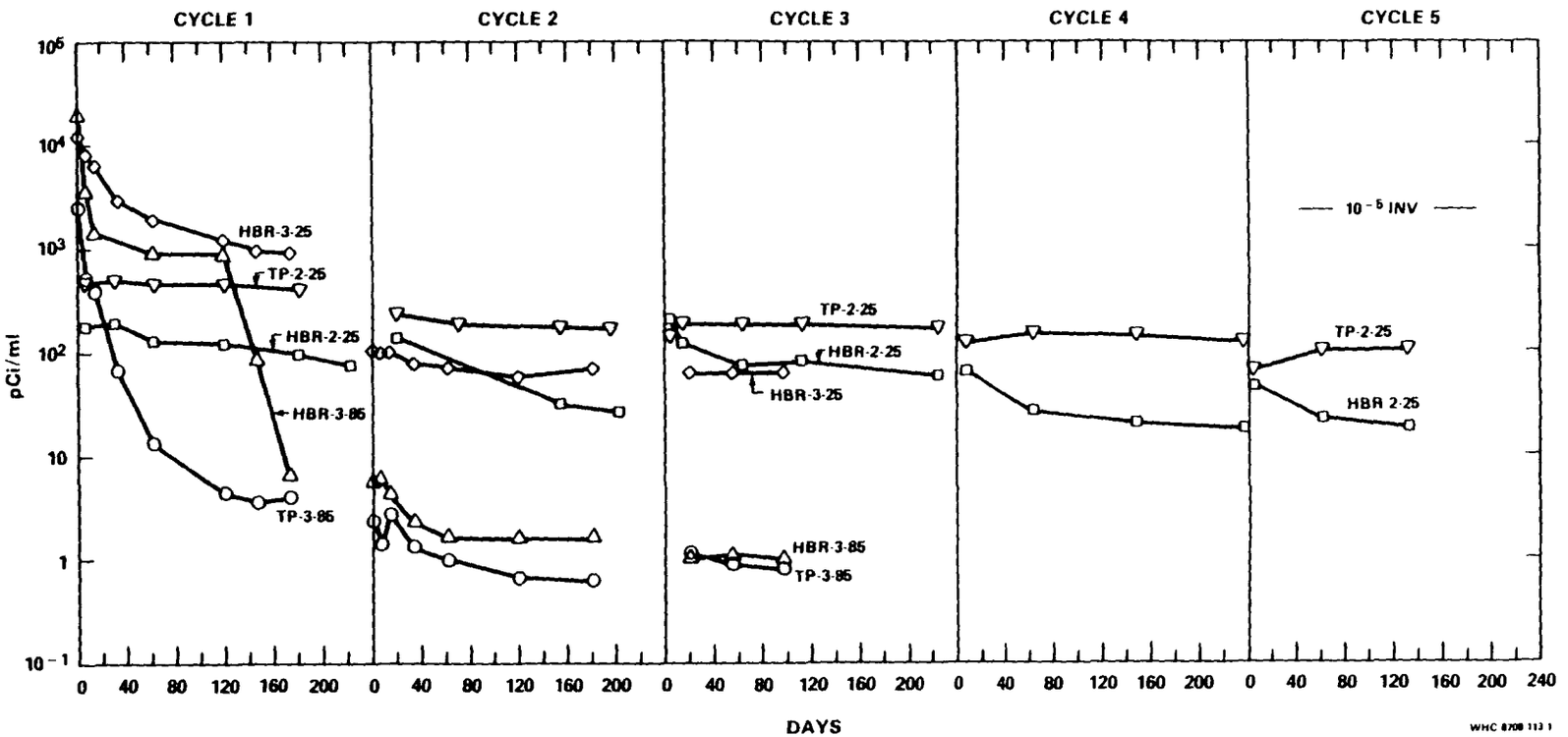
Series 2 -- J-13 water, unsealed glass vessels
Series 3 -- J-13 water, sealed stainless steel vessels

ACTINIDE RESULTS

Semi-Static Tests - U, Np, Pu, Am, Cm

- **Actinides rapidly reach steady-state concentrations**
  - Suggests that actinide release will be solubility limited and not depend on particular characteristics of the spent fuel
  - However, more data is needed to assess the effects of colloids
- **Sample filtration results suggest actinides were present as colloids (particularly Am & Cm in 25°C tests)**
- **Actinide concentrations tended to be lower at 85°C than at 25°C**
  - Kinetic factors appear to favor precipitation of secondary phases over colloid formation at the higher temperature
- **Order of temperature and filtration effects: Np < U < Pu < Am & Cm**

Pu-239 + 240 ACTIVITY IN 0.4 μm FILTERED SOLUTION SAMPLES



Actinide Annual Releases as Fractions of 1000-Year Inventories Based on HBR-3-25 Test Data

Actinide	Log (M)	Log (Release)
U	-5.9	-8.6
Np	-8.9	-8.8
Pu	-8.4	-9.0
Am	-9.8	-9.1

Based on approximate steady-state concentrations measured in 0.4 μm filtered samples during Cycles 2 & 3 of HBR-3-25 test

Calculated annual releases assume water flow rate of 20 L/yr per waste package containing 3140 kg of 33,000 MWd/MTM burnup fuel

Concentrations of Low-Solubility Nuclides will be Controlled by Secondary Phases

Uranophane [$CaO \cdot 2UO_3 \cdot 2SiO_2 \cdot 6H_2O$] Crystals Formed on Fuel Surface During HBR-3-85 Test

10 μm

SOLUBLE RADIONUCLIDE RELEASES MEASURED IN SEMI-STATIC TESTS*

- **^{99}Tc, ^{137}Cs, ^{90}Sr and ^{129}I release rates**
  (inventory fraction per year)

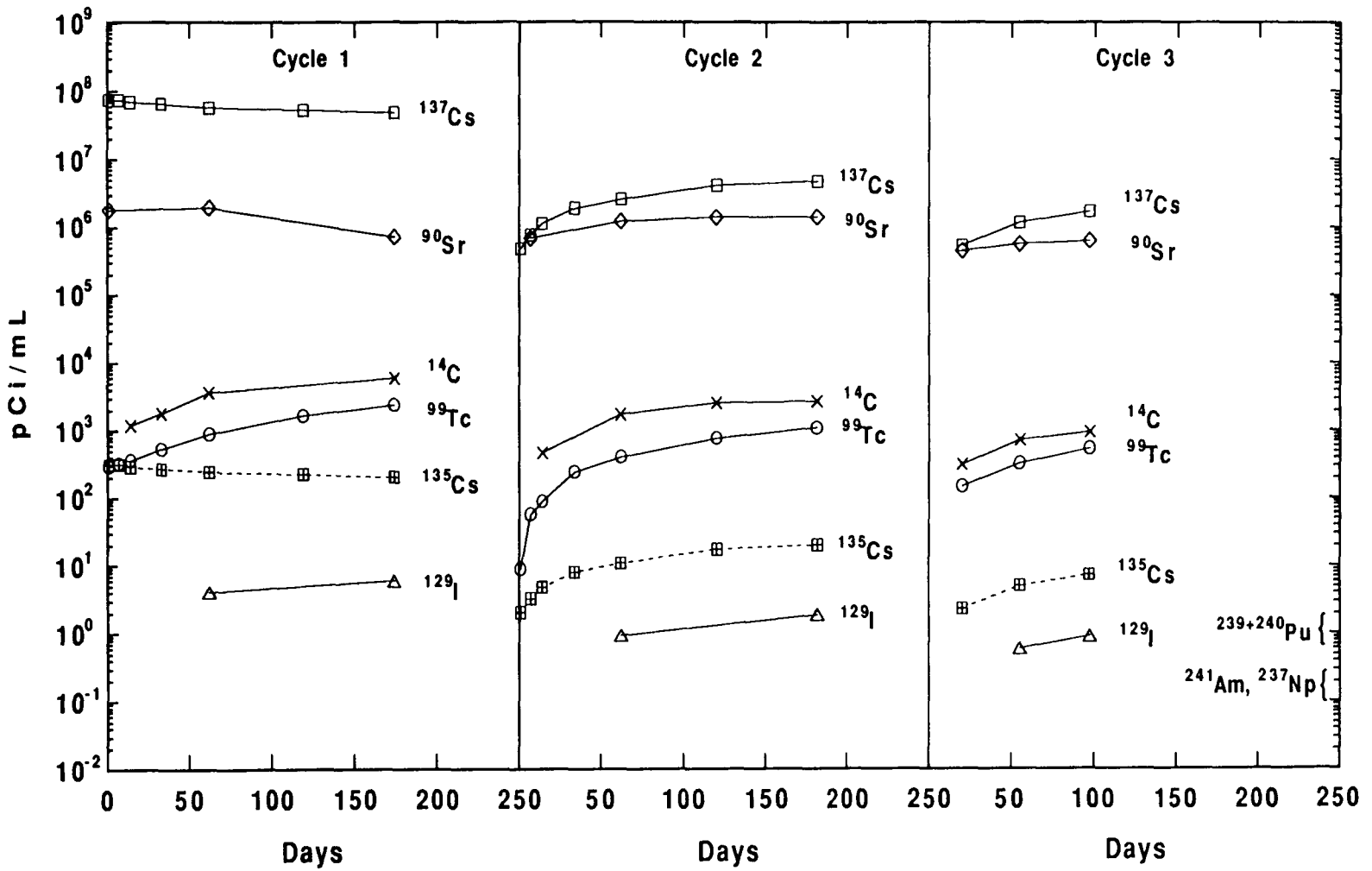
  5×10^{-5} to 2.5×10^{-4} at 25°C

  3×10^{-4} to 1.2×10^{-3} at 85°C

- **^{14}C**
  - **~ 1% of specimen inventory released in first year**
  - **Release from fuel** (matrix, gap and grain boundaries) **was much greater than from cladding exterior**
  - **Released as CO_2 from unsealed vessels**

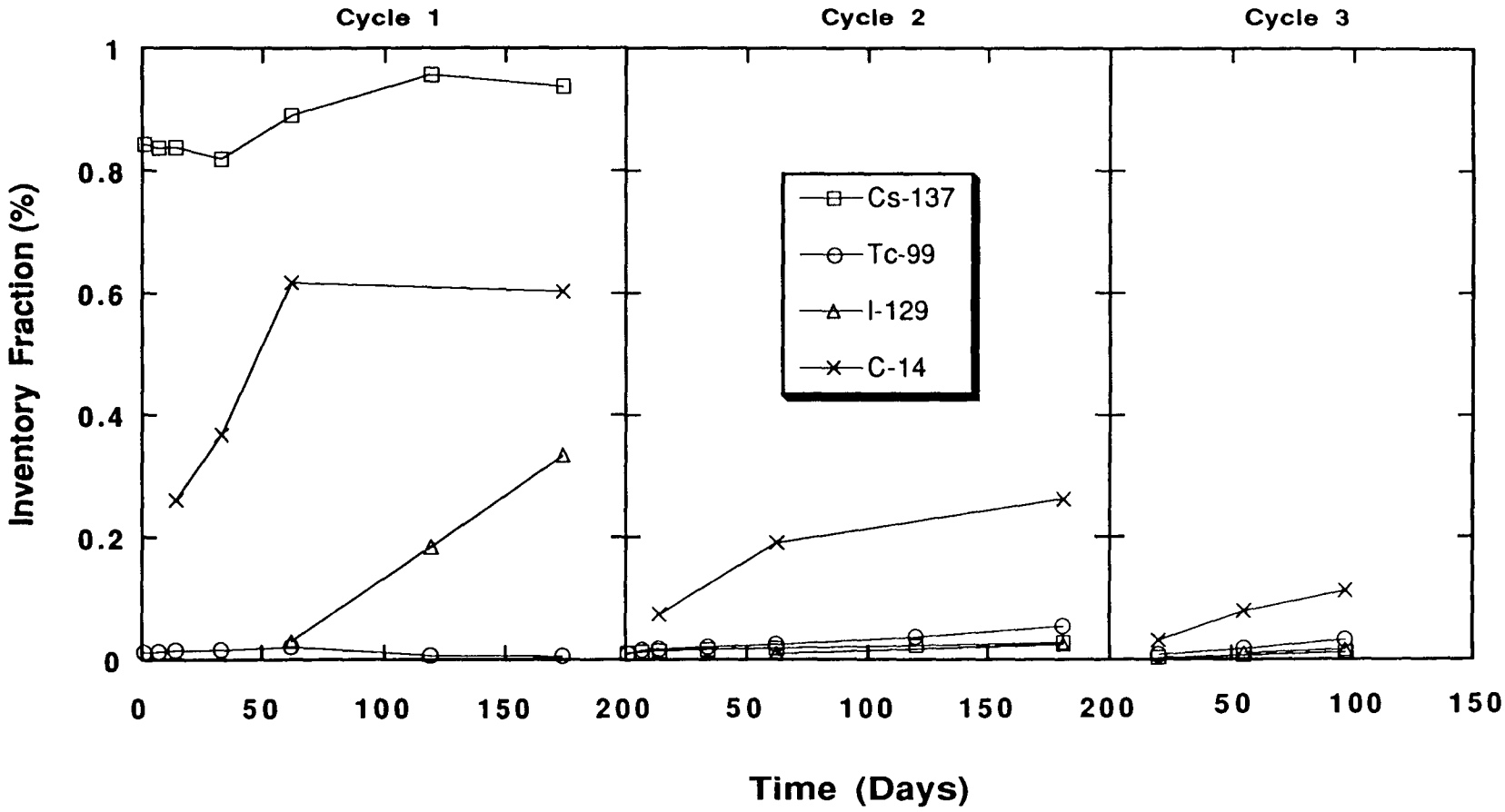
*** Tests with as-irradiated fuel particles with geometric surface area ~2.5 cm^2/g**

RADIONUCLIDE ACTIVITIES MEASURED IN SOLUTION DURING THE TP-3-85 TEST



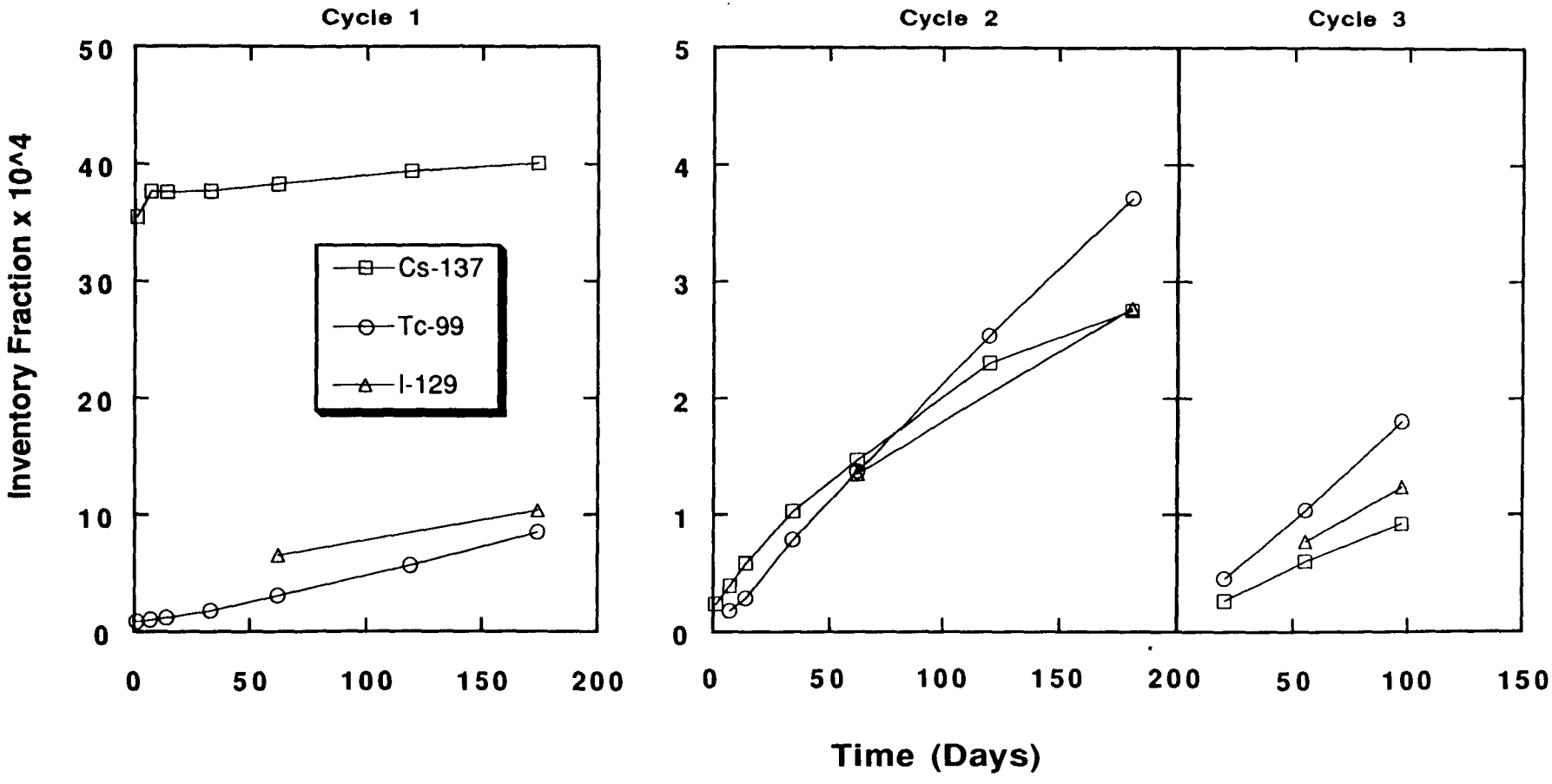
FRACTION IN SOLUTION

HBR-3-85 Test



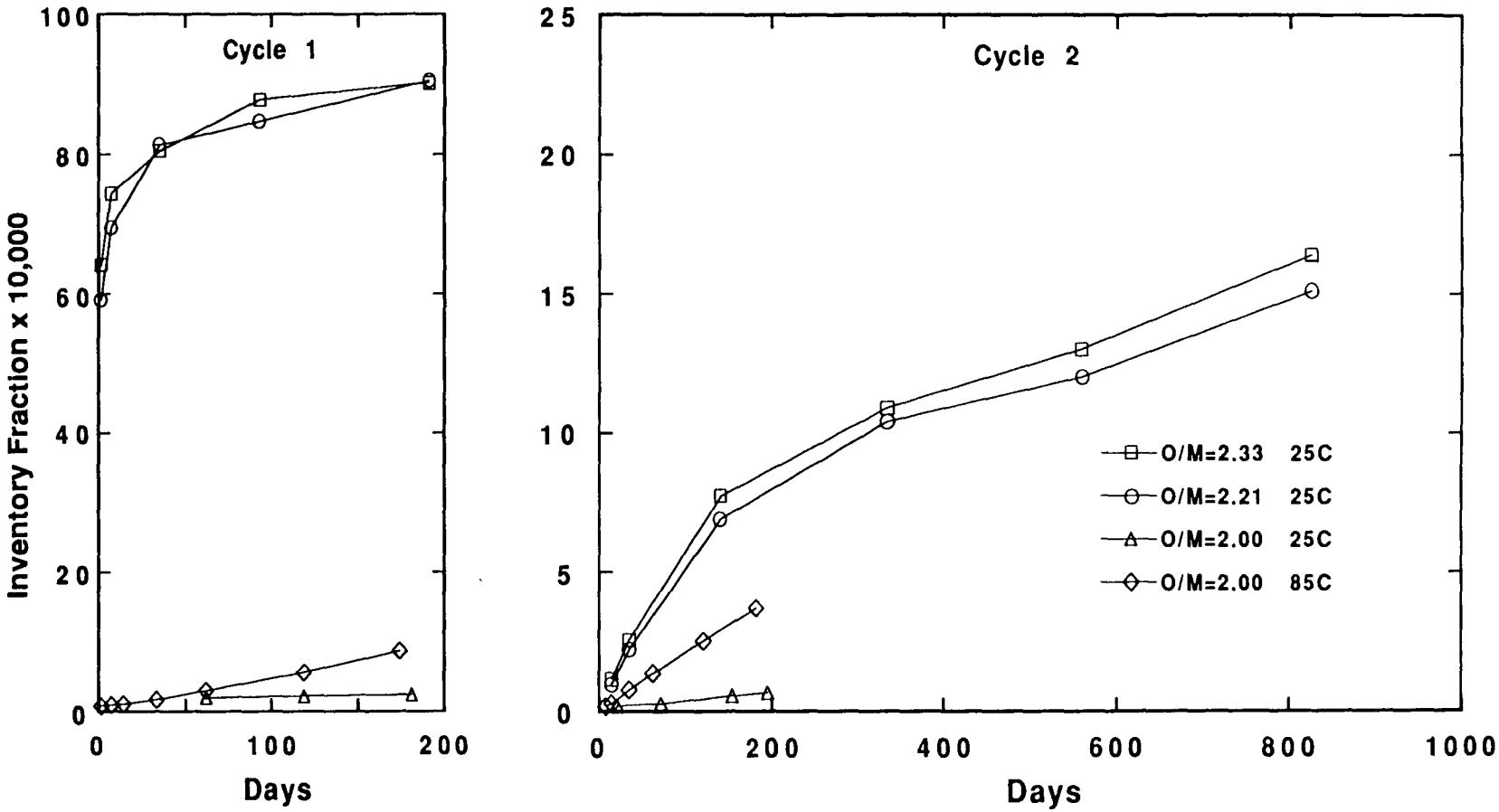
FRACTION IN SOLUTION

TP-3-85 Test



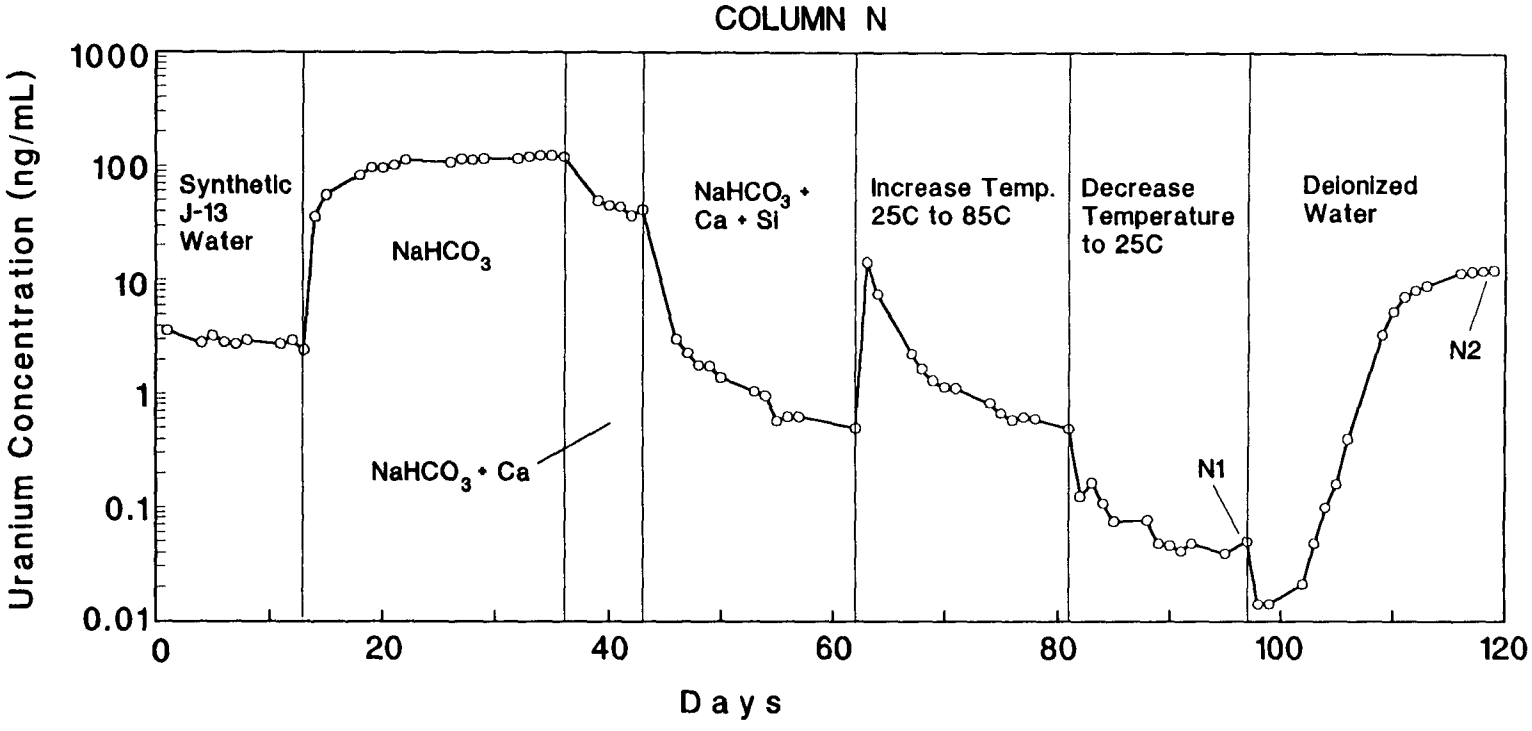
^{99}Tc Measured in Solution

Effects of Temperature and Oxidation (O/M)



FLOW-THROUGH TEST WITH UO_2

EFFECTS OF WATER COMPOSITION AND TEMPERATURE

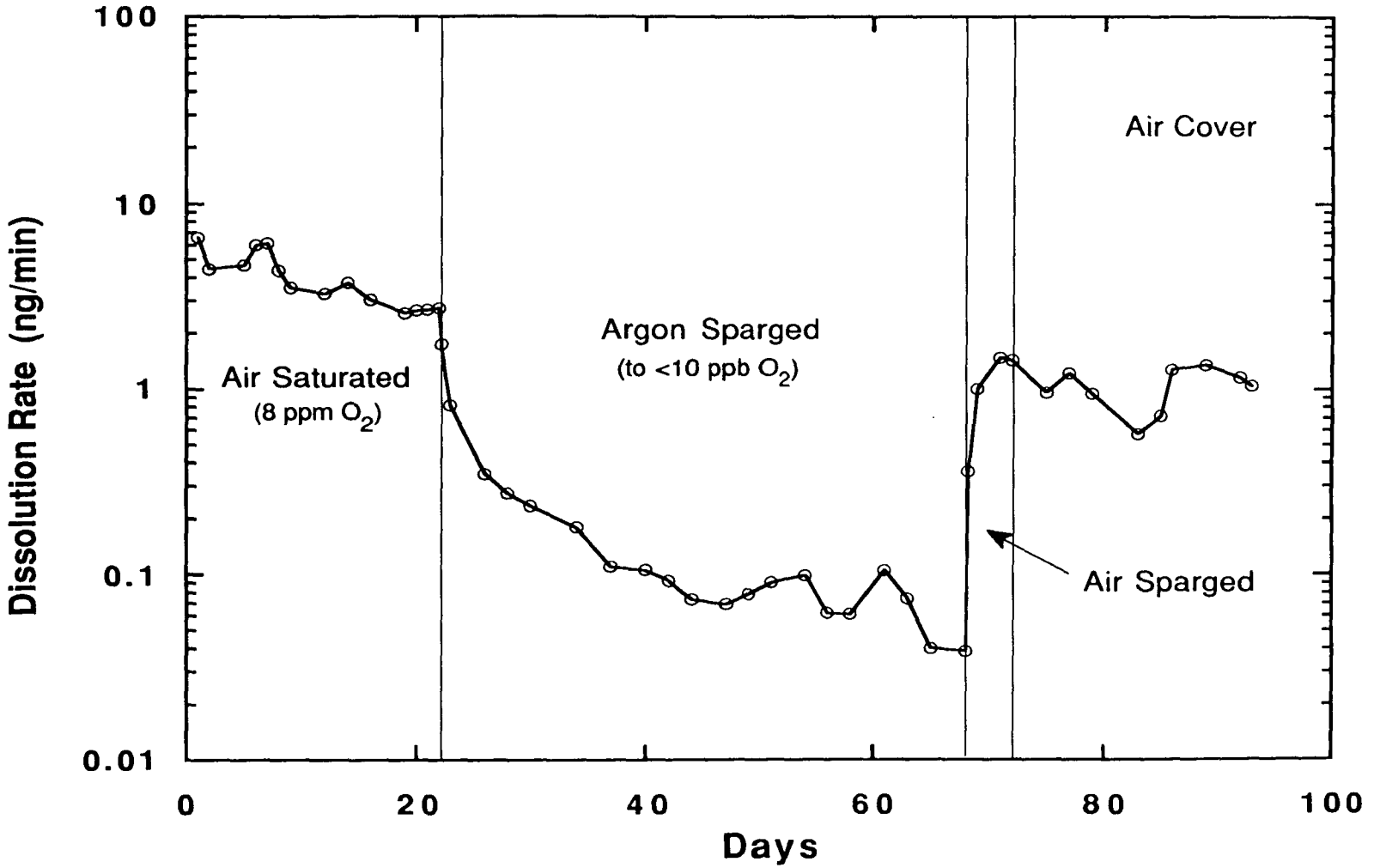


NOTES: Auger microprobe examination of particle at N1 indicated 5 nm Ca-Si-U surface layer which was partially redissolved from particle examined at time N2

Constant Flow Rate, 0.2 mL/min

FLOW-THROUGH TEST WITH UO_2

Deionized Water, 25 °C



SUMMARY

- **Actinide releases appear to be solubility limited**

- **Soluble nuclide releases will be complicated to model**
  - Fuel is nonhomogeneous - gap, grain boundary and matrix components of release
  - Fuel degradation - state of fuel and surface area change with time

- **Soluble nuclide releases measured in semi-static tests**
  - ^{137}Cs and ^{14}C; ~ 1% of inventory in first year
  - ^{99}Tc, ^{137}Cs, ^{90}Sr and ^{129}I; ~10^{-4} to 10^{-3} of inventory per year in later test cycles

- **Additional information needs**
  - Radionuclide distributions in spent fuel, particularly ^{14}C
  - Dissolution behavior of oxidized fuel and other fuel types
  - Effects of colloids
  - Effects of water conditions on matrix dissolution rates
  - Time-dependent model for exposed fuel surface area

REFERENCES

- **Semi-static and flow-through test methods**
  - Wilson C. N. and W. J. Gray. 1989. "Measurement of Soluble Nuclide Dissolution Rates From Spent Fuel." Scientific Basis for Nuclear Waste Management XIII. Materials Research Society Symposium Proceedings, 176:489-498.
- **Comparison of EQ3/6 results with results from laboratory tests**
  - Wilson, C. N. and C. J. Bruton. 1989. "Studies on Spent Fuel Dissolution Under Yucca Mountain Repository Conditions." Ceramic Transactions, 9:423-441. (also UCRL-100223.)
- **Semi-static test results**
  - Wilson, C. N. 1990. Results from NNWSI Series 3 Spent Fuel Dissolution Tests. PNL-7170, Pacific Northwest Laboratory, Richland Washington.
- **Flow-through test results**
  - Wilson C. N. and W. J. Gray. 1990. "Effects of Water Composition on the Dissolution Rate of UO_2 Under Oxidizing Conditions." High Level Radioactive Waste Management, pp 1431-6. (Proceedings of topical meeting, Las Vegas, NV, April 8-12, 1990)