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OVERVIEW OF EXPERIMENTAL SUPPORT FOR FISSION-PRODUCT TRANSPORT ANALYSES AT OAK RIDGE NATIONAL LABORATORY*

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This paper presents an overview of the experimental work currently in process at Oak Ridge National Laboratory in support of the NRC severe accident source term evaluation work.

FISSION PRODUCT RELEASE FROM FUEL

This program was designed (1) to determine fission product and aerosol release rates from irradiated fuel under accident conditions, (2) to identify the chemical forms of the released material, and (3) to correlate the results with experimental and specimen conditions and with the data from related experiments. Tests are being conducted over a range of fuel operating histories, test times, temperatures, and steam flow rates, so that the data obtained are applicable to a spectrum of accident sequences in both boiling and pressurized water reactors.

The experimental apparatus is constructed utilizing design features from several previous fuel testing programs.[1-3] The induction furnace for heating the fuel specimens to 2000°C in flowing steam required a new design in which a tungsten or graphite susceptor is protected by a blanket of inert gas. Fission product collection and analysis techniques similar to those used by Lorenz et al.[1,4] were expanded to provide more detailed results. The principal analytical techniques are summarized in Table 1; gamma spectrometry is the most versatile and most widely-used of these methods. The configuration of the test apparatus

Table 1. Analytical techniques for fission product release data

Technique	Time	Location	Elements detected
Gamma spectrometry	Pretest, posttest	Fuel specimen	Long-lived, high E, fission products — Cs, Sb, Ru, Eu, Ce
	On-line	Thermal gradient tube, filters, gas traps	Cs, Kr
	Posttest	Furnace components, thermal gradient tube, filters	Cs, Ag, Ru, Ce, Eu
Activation analysis	Posttest	Charcoal, solution from furnace, thermal gradient tube, filters	I, Br
Spark source mass spectrometry	Posttest	Samples from furnace, thermal gradient tube, filters	All

is illustrated in Fig. 1. On-line release data are obtained with NaI(Tℓ) detectors monitoring the thermal gradient tube, the filter package, and the cold charcoal traps. Following the test, quantitative data are obtained via gamma-ray spectrometry of all apparatus components. Additional data are obtained through mass spectrometry and activation analysis of samples from selected locations. Metallographic examination is used for evaluation of test effects on the UO_2 fuel and Zircaloy cladding.

Three tests of PWR fuel have been conducted and evaluated, and the results have been reported in detail.[5-7] Fuel specimen and test operating data for these three tests are presented in Table 2. The temperature and flow histories for a representative test (HI-2) are shown in Fig. 2. The peak in the flow rate out of the system (largely noncondensable hydrogen) at 18 min illustrates the rapid oxidation of the Zircaloy by steam during this period. In test HI-2, both the volume of hydrogen formed and subsequent metallographic examination indicated essentially complete oxidation of the cladding to ZrO_2. The release histories of cesium and krypton in this same test, as functions of time and temperature, are presented in Fig. 3. In this test the 160-g specimen was heated at about 1°/s to the test temperature of 1700°C, which was maintained for 20 min. (The raw data shown in Figs. 2 and 3 require small corrections to both the thermocouple and optical pyrometer values to obtain the true temperature.) The release curves for ^{85}Kr and ^{137}Cs (Fig. 3) show that these nuclides continued to be released from the furnace during cooldown. The release data for all three tests are summarized in Table 3. The iodine data were obtained by activation analysis of the long-lived ^{129}I in leach solutions. Because of the high radiation levels of ^{137}Cs (and also ^{134}Cs), other nuclides (such as ^{125}Sb,

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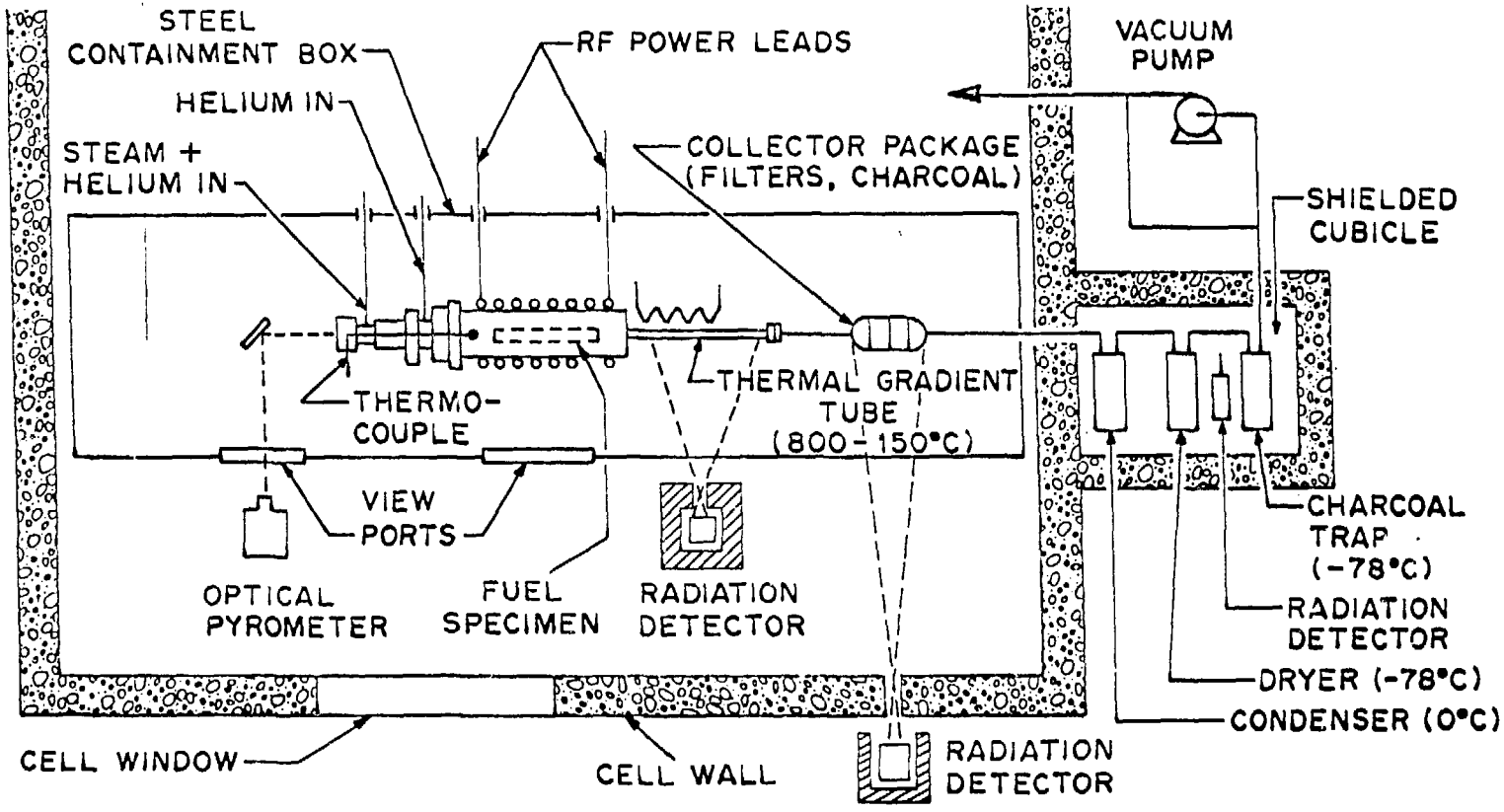

Fig. 1. Fission product release and collection system.

Table 2. Fuel specimen and test operating data

	Test		
	HI-1	HI-2	HI-3
Fuel specimen data			
Length (cm)	20.3	20.3	20.3
Mass (g)	168.0[a]	166.0[a]	166.9[a]
Reactor	H. B. Robinson	H. B. Robinson	H. B. Robinson
Burnup (MWd/kg)	28.0	28.0	25.2
Krypton release during irradiation (%)	~0.35	~0.35	~0.35
Test operating data			
Heatup rate (°C/min)	72	75	125
Maximum temperature (°C)	1400	1700	2000
Time at test temperature (min)	30.0	20.0	20.0
Average helium flow rate (L/min)[b]	0.436[c]	0.334[c]	0.302
Average steam flow rate into furnace (L/min)	1.01	0.99	0.366
H_2 generated (L)	12.17	13.54	4.92

[a]Including 30.7-g Zircaloy cladding and end caps.
[b]At STP.
[c]Argon was substituted for helium in tests HI-1 and HI-2.

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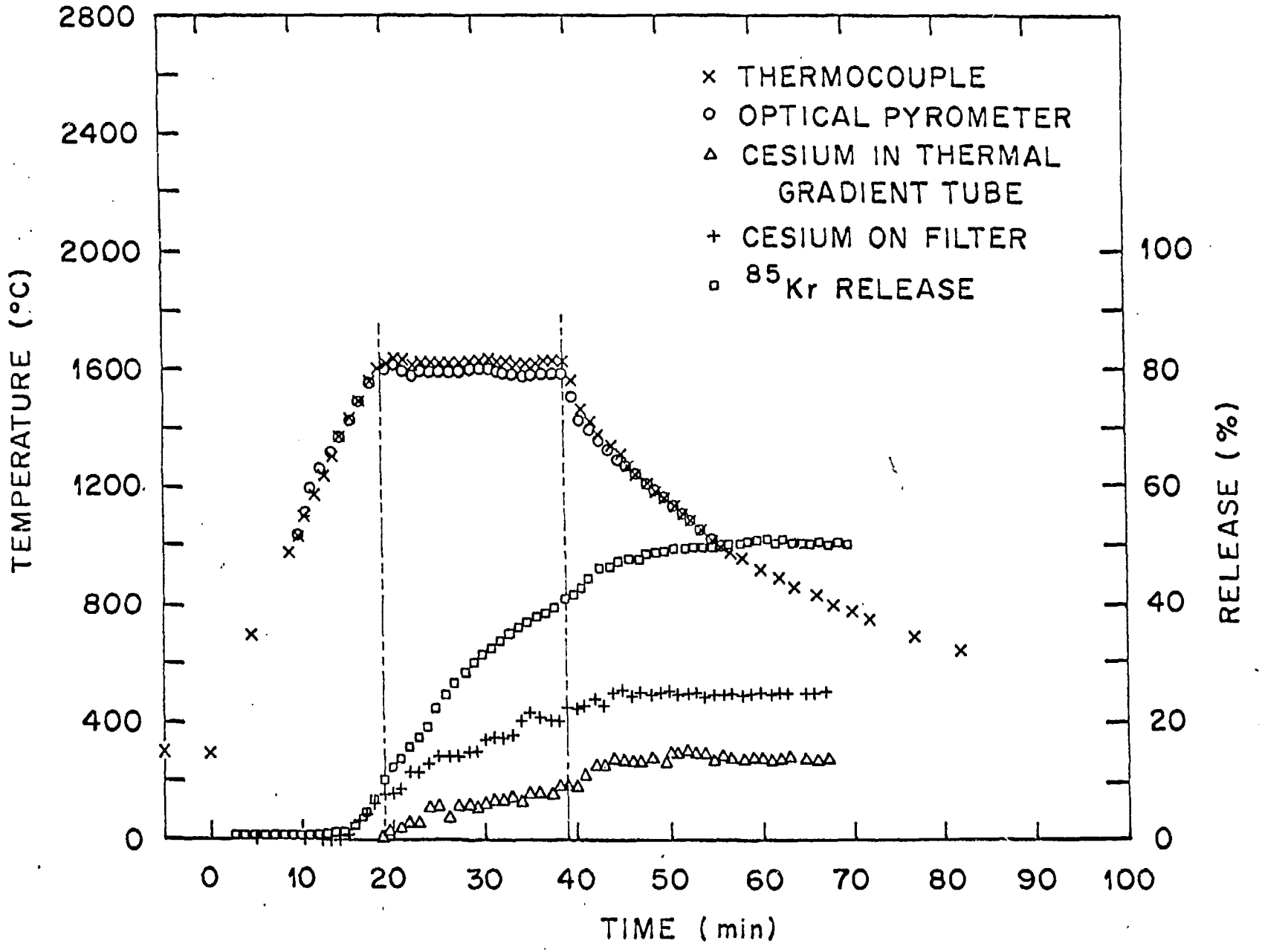

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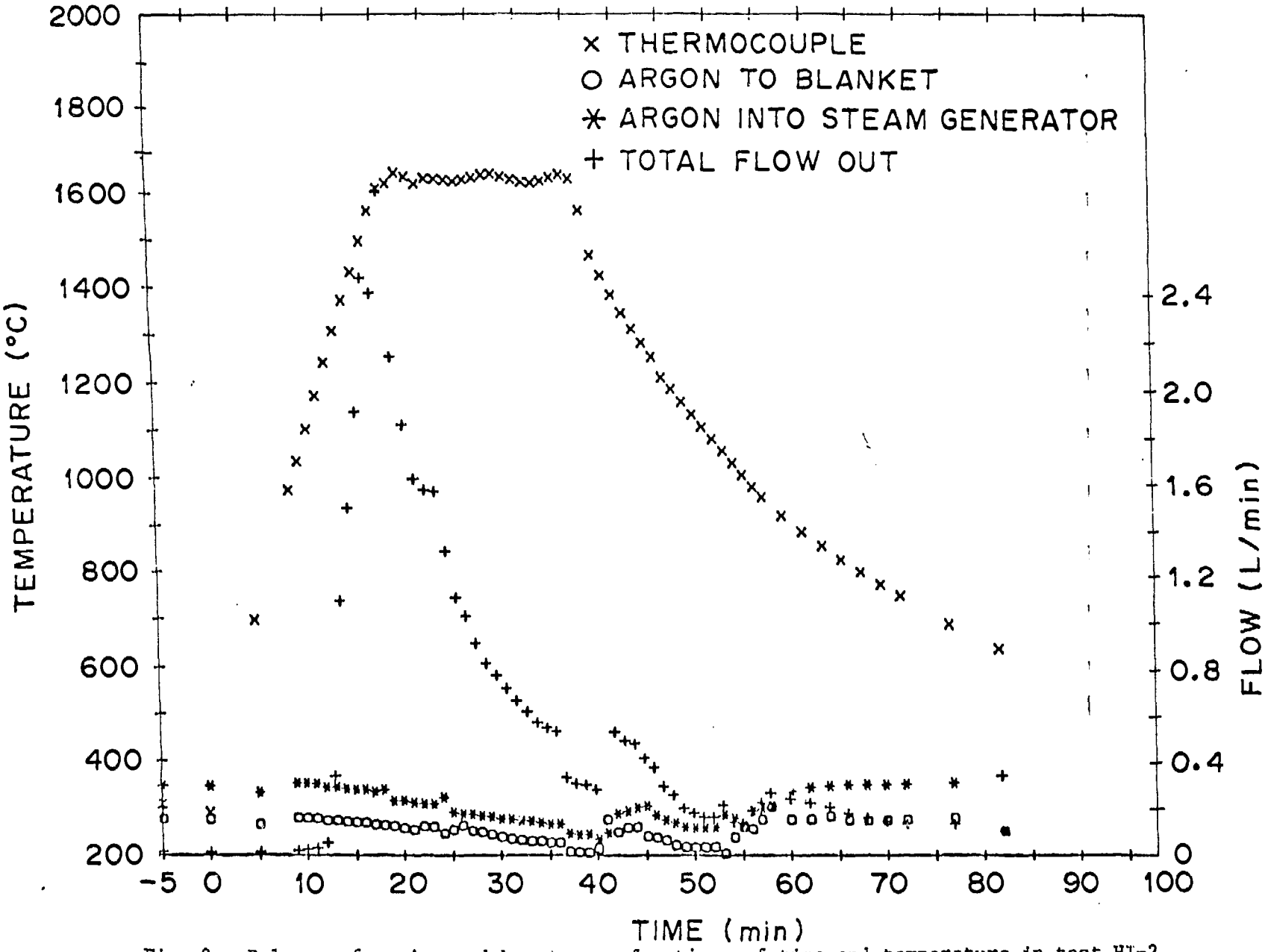

Fig. 3. Release of cesium and krypton as functions of time and temperature in test HI-2.

Table 3. Extent and distribution of fission product release from three tests of H. B. Robinson fuel

Test component or collector	Temperature or range (°C)	Fraction of fuel inventory found (%)				
		^{85}Kr	^{137}Cs	^{129}I	^{125}Sb	^{110m}Ag
Test HI-1 (30 min at 1400°C in steam at 1.0 L/min)						
Furnace	1400–900	0	0.79	0.016	0.0011	0
Thermal gradient tube[a]	800–130	0	0.58	0.83	~0.017[b]	0
Filters	~130	0	0.38	1.18	0[c]	0
Hot charcoal	~130	0	0	0.010	0	0
Cold charcoal	−78	2.83	0	0	0	0
Totals		2.83	1.75	2.04	0.018	0
Test HI-2 (20 min at 1700°C in steam at 1.0 L/min)						
Furnace[a]	1700–1000	0	8.82	~0.14	0.68	0
Thermal gradient tube	1000–150	0	15.5	16.8	~0.85[b]	1.86
Filters	~150	0	26.2	35.9	0.005	0.26
Hot charcoal	~150	0	10^{-6}	0.187	0	0
Cold charcoal	−78	51.5	0	0	0	0
Totals		51.5	50.5	53.0	1.53	2.12
Test HI-3 (20 min at 2000°C in steam at 0.36 L/min)						
Furnace[d]	2000–1000	0	13.5	0.60	10^{-3}	0.015
Thermal gradient tube	900–140	0	15.7	12.4	0	0
Filters	~140	0	28.5	22.4	0	0
Hot charcoal	~140	0	10^{-7}	10^{-3}	0	0
Cold charcoal	−196	59	0	0	0	0
Totals		59	57.7	35.4	10^{-3}	0.015

[a]In addition, particles of fuel and/or cladding recovered from the furnace contained significant amounts of ^{137}Cs, ^{125}Sb, ^{106}Ru, and ^{60}Co.

[b]Measured only after >90% of the Cs activity had been removed by leaching.

[c]Not detected because of high Cs activity, possibly as high as 0.08%.

[d]Fuel specimen could not be removed from ZrO_2 furnace tube and end plug. Because some released material remained with fuel and could not be measured, these values represent minima only.

^{110m}Ag, and ^{154}Eu) could not be measured until after most of the cesium had been chemically removed. As shown in Table 3, most of the material released from the furnace was collected either on the platinum-lined thermal gradient tube or on the fiberglass filters.

These three tests have provided data on fission product release under idealized LWR accident conditions in a temperature range (1400-2000°C) where only limited data previously existed. In Fig. 4 our release rate data are compared with curves from a recent NRC-sponsored review of available fission product release data. Only the curves for the more volatile elements are shown. Although the iodine release in test HI-3 was low, the other data points for Kr, I, and Cs fall reasonably close to the corresponding curve. The limited data for antimony and silver release fall below the curves in Fig. 4 by varying amounts; since we could not analyze for these elements at all apparatus locations, low values were not unexpected. The on-line release rate data for krypton and cesium are consistent with current understanding of the temperature dependent release mechanisms.

CORE MELT PROGRAM

In this program, a 1-kg portion of an LWR core is heated to melting. Core samples have included simulated PWR and BWR fuel, PWR control alloy, and some fission product simulants. A specially designed furnace inductively heats, first, the metallic portions of the sample and, then, the fuel material. A steam/hydrogen environment is provided by the in-flowing steam and by reaction with the cladding to form hydrogen.

The material condensed on vessel surfaces, the deposit on the filter, and the melt residue are analyzed for amount and composition. To date, information has been obtained on the nature of control-rod failure and on possible interactions of control-rod material with cladding. In addition, it has been

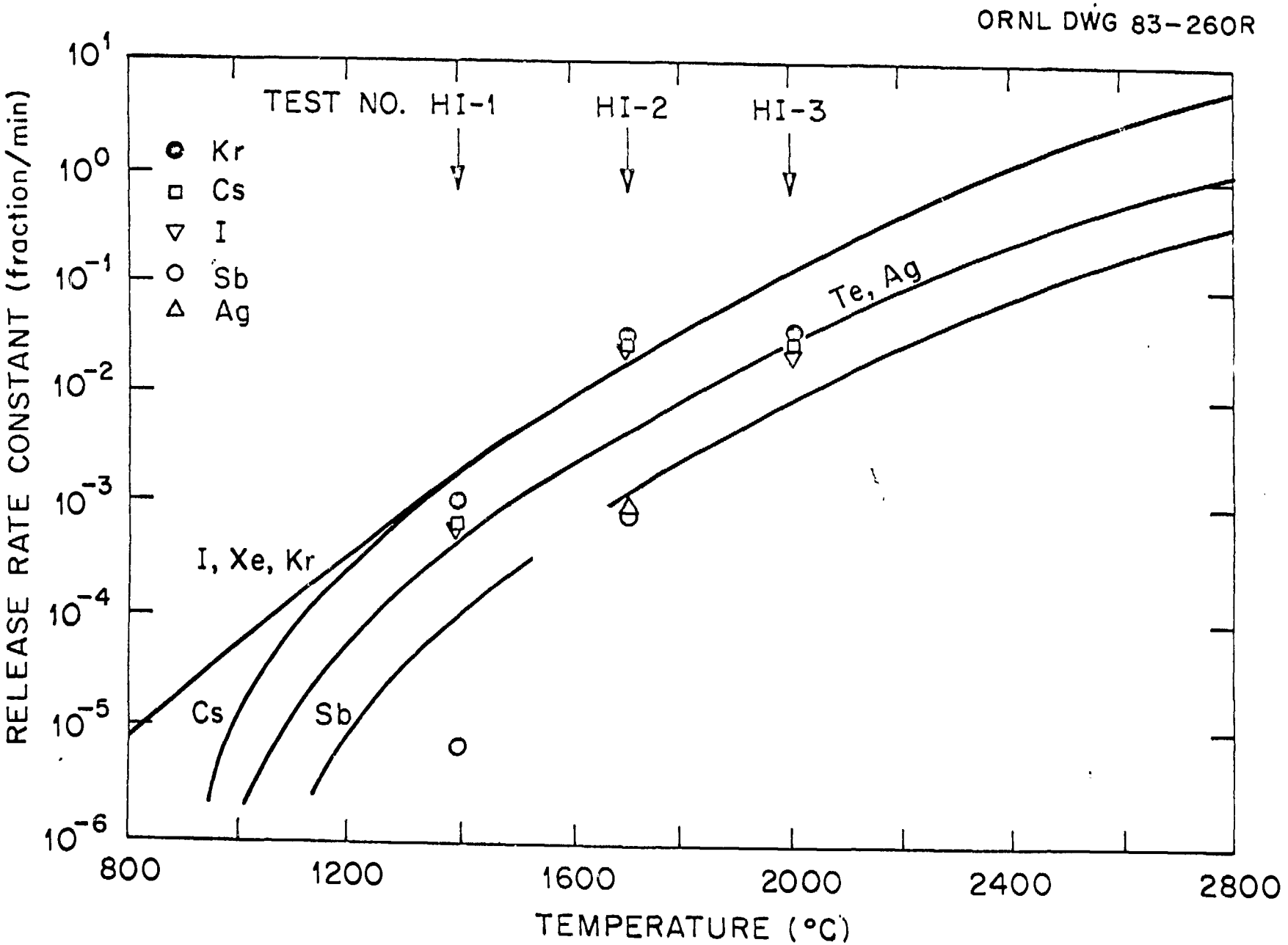

Fig. 4. Release rate data from tests HI-2, HI-2, and HI-3, compared to curves from NUREG-0772.

observed that tellurium has a strong tendency to alloy with cladding or steel materials, if such opportunities arise.

FISSION PRODUCT VAPOR DEPOSITION ON AEROSOLS

The transport of fission products in both the primary vessel and in secondary containment is related to aerosol behavior. In this experimental program, the nature and rate of fission product vapor interaction with aerosols have been studied.

Two experimental techniques (both using radiotracers) are being investigated. The first is a flowing system in which simulated aerosol and fission product vapors are fed into a pipe maintained at temperatures up to ~700°C, and the relative deposition of the vapor between the pipe surface and the aerosol is measured.

A second experimental technique employs a static system for determining the chemi-sorptive capacities of a range of typical aerosols with fission product vapors. The sorptive capacities of nonfuel aerosols (which will be generated by a plasma torch) for fission product vapors will be measured by radiotracer methods.

TRAP-MELT VERIFICATION PROGRAM

This program focuses on aerosol deposition rates and transport in the reactor vessel during LWR core-melt accidents. Specifically, the objectives are (1) to measure aerosol deposition and resuspension rates in idealized geometry using representative aerosols, contact times, and lift-off velocities; and (2) to compare test results with values obtained by using current computer models such as TRAP-MELT and QUICK.

Two types of aerosol deposition rate experiments are currently in progress. In the first type, a plasma torch is used to generate aerosols into a vertical

pipe 12 in. in diameter by 12 ft. high. Aerosol plateout, settling, and airborne concentration level are measured as a function of air flow rate, wall temperature gradient, and solids mass-input rate. Two tests (a zinc aerosol and an iron oxide aerosol) have currently been completed. A second series of experiments is currently in progress to investigate the degree of aerosol resuspension that may occur as a result of high gas velocities.

NUCLEAR SAFETY PILOT PLANT (NSPP)

This facility is currently dedicated to developing an expanded data base on the behavior of aerosols generated during a severe accident. The facility consists of a 38.3-M^3 insulated vessel with provisions for (1) aerosol injection by a plasma torch, (2) steam injection for simulation of the environment expected in the containment vessel for some accident sequences, (3) on-line aerosol sampling for determination of suspended concentration vs time, and (4) fallout and plateout surfaces used in posttest examination.

The principal objective of the current test series is to determine the effect of steam condensation on typical aerosols produced by an overheated core and by core melt/concrete interaction. Two oxides, Fe_2O_3 and U_3O_8, and powdered, nuclear-grade concrete are being used to simulate these aerosols.

The NSPP facility diagramed schematically in Fig. 5 includes a test containment vessel, aerosol generating equipment, analytical sampling and system parameter measuring equipment, and an in-vessel liquid spray decontamination system. The NSPP vessel is a stainless steel cylinder with dished ends having a diameter of 3.05 m, a total height of 5.49 m, and a volume of 38.3 m^3. The floor area is 7.7 m^2 and the internal surface area (including top, bottom, and structural items) is 68.9 m^2. The equipment for the measurement of aerosol parameters includes filter samplers for measuring the aerosol mass concentration,

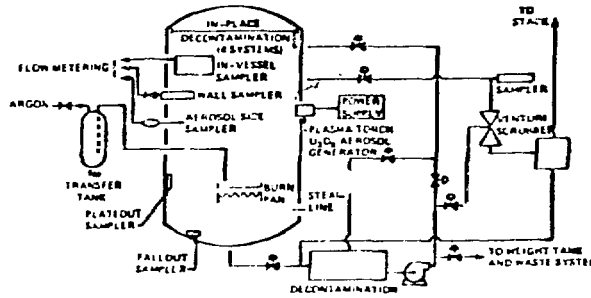

Fig. 5. Nuclear Safety Pilot Plant (NSPP) Facility schematic.

coupon samplers for aerosol fallout and plateout measurements, cascade impactors and a centrifuge sampler for determining the aerodynamic particle size distribution of the aerosol, and devices for collecting samples for electron microscopy. System parameters measured are moisture content of the vessel atmosphere, steam condensation rates on the vessel wall, vessel atmosphere temperature, wall temperature gradients, and vessel pressure.

Five experiments were conducted to investigate the behavior of U_3O_8 aerosols in a steam environment. The vessel was heated and the steam environment was produced by injecting steam for a period of about 1 h. When the desired temperature was reached, the steam injection rate was reduced and the accumulated steam condensate was drained from the vessel. Steam injection at the low rate was continued for either 2 or 6 h to replace steam losses to the vessel walls. U_3O_8 aerosol was generated and introduced into this quasi-steady state environment. In the first three experiments, an aerosol concentration gradient was noted during the early stages of the experiment; the aerosol mass concentration was greater by a factor of 2-3 in the upper region of the vessel than in the lower region. A small fan-mixer was installed near the bottom of the vessel and utilized during the last two experiments. The initial aerosol mass concentration was varied from about 5 to 26 $\mu g/cm^3$ over the five experiments. After the first hour, the aerodynamic behavior of U_3O_8 aerosol was essentially the same in all five experiments; the rate of aerosol disappearance from the vessel atmosphere was very similar. Scanning electron microscopy showed the aerosol to be in the form of spherical clumps of particles much the same as noted during the humid atmosphere experiments.

Comparison of the aerosol behavior in the three environments reveals that the rate of aerosol disappearance from the steam environment is larger than in the dry or moist environments. The times required for 90% of the maximum aerosol mass to disappear from the NSPP vessel atmosphere in these three experiments

are approximately: 100 min in the dry environment; 107 min for the moist environment; and 75 min in the steam environment. The times required for 99% of the aerosol mass to disappear would be approximately 340 min, 270 min, and 115 min, respectively.

It is clear that under the influence of a condensing steam environment, as would be present in LWR containment during and following an accident, the U_3O_8 aerosol behaves in a manner different from that in a dry or moist environment. This change in behavior could be the result of several factors, or combinations of these factors: a change in aerosol morphology enhancing gravitational settling; influence of steam flux toward the walls enhancing plateout; thermal factors influencing particle agglomeration; or currently unrecognized influences of steam condensation.

POST-ACCIDENT IODINE AND TELLURIUM CHEMISTRY

Numerous LWR accident sequences involve interaction of various types of water bodies with gases containing fission product vapors. The principal objective of our iodine chemistry work is to determine iodine volatility from water solutions for the various realistic conditions that would exist in a severe accident. We are taking two approaches toward this goal. In the first, we are making direct measurements of volatility for a range of iodine concentrations (10^{-4} to 10^{-6} M) a range of pH conditions (6 to 9), both buffered with boric acid and unbuffered. In addition, tests are under way to determine the effects of radiation and oxidizing or reducing impurities on iodine volatility.

The second approach, which is more fundamental, is needed for the interpretation of the volatility measurements. This work employs solution spectrophotometry to examine the rate of production or disappearance of the iodine species I^-, I_3^-, I_2, IO_3^-, and possibly HOI.

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