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TECHNICAL LETTER REPORT

David R. Hamrin 6/16/2011
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ABSTRACT

Four tests of fission product release from simulated high-burnup LWR fuel were conducted. In cooperation with the U.S. and ORNL, this fuel material was supplied by the Federal Republic of Germany, with the test results to be shared. The test specimens were 18-cm-long Zircaloy-clad UO_2 rods; the UO_2 contained trace-irradiated fission product simulants. The specimens were heated in flowing steam to temperatures of 1600 to 2400°C for time periods of 1 to 30 min. Fractional releases of 50 to 100% were measured for the relatively volatile elements Ag, Sn, Sb, Te, and Cs; the release of Ru and Eu, however, was <1%. The masses of aerosol collected on the thermal gradient tubes and filters varied between 0.75 and 2.3 g.

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HIGHLIGHTS REPORT FOR FISSION PRODUCT RELEASE TESTS OF
SIMULATED LWR FUEL

M. F. Osborne, J. L. Collins, and R. A. Lorenz

1. INTRODUCTION

Four tests to measure the fission product release from simulated LWR fuel, which was supplied by the Kernforschungszentrum Karlsruhe (Nuclear Research Center) in the Federal Republic of Germany, have been conducted. These tests are a part of the Fission Product Release from LWR Fuel Project which is sponsored at the Oak Ridge National Laboratory (ORNL) by the U.S. Nuclear Regulatory Commission (NRC). As a result of the bilateral agreement between the USNRC and the German counterpart to exchange the results of nuclear safety studies, the Germany laboratory provided this material free of charge for testing at ORNL, with the agreement that all results would be shared equally. This report will summarize the principal results only; all data and conclusions are preliminary and may be revised in the final report of this work. The purpose of this preliminary report is to distribute the principal results as soon as possible to all investigators of severe fuel damage phenomena in the two countries. Some of the results from the first three tests were shared previously.^{1,2} A thorough reporting and interpretation of these tests, along with comparison with related work, will be provided as soon as possible.

2. DESCRIPTION OF TESTS

The test specimens were 18-cm-long by 1.22-cm-diam Zircaloy-clad UO_2 ; each specimen contained 14 fission pellets and two commercial pellets of normal UO_2 (at the ends), totaling ~90.4 g UO_2 and ~38.3 g Zircaloy. The composition of the fission samples is shown in Table 1 and the test construction and conditions are summarized in Table 2.

The shipment of simulant fuel specimens arrived at ORNL on April 17, 1984, and the tests were conducted as rapidly as possible thereafter because we wished to maximize analytical precision for the short-lived tracer nuclides. (The two shortest half-lives were ^{129m}Te at 33.6 d and ^{103}Ru at 39.3 d.) Test HS-4 (Table 2) was delayed because receipt of the required ThO_2 ceramics from Los Alamos National Laboratory was 6 months behind schedule. However, reasonably good data for tracer analysis were obtained in spite of the delay. The tests were conducted in the apparatus shown in Fig. 1.

The test procedure was similar to that used for tests of commercial LWR fuel;³⁻⁷ the major difference was the much lower level of radioactivity in the simulated fuel specimens, so that remote handling of the fuel and fission product collectors was not required. The temperature and fission product release histories for the four tests are

Table 1. Composition of simulant fuel (fissium),
supplied by KfK, Germany

Element/ component	Mass ^a (mg)	Radionuclide	Half-life (d)	Radioactivity per rod (mCi)			
				4/6/84 ^b	5/1/84	6/1/84	7/1/84
<u>Fissium I (volatile tracers, unsintered)</u>							
CsI	242.5	} ¹³⁴ Cs	752	0.133	0.130	0.126	0.123
Cs ₂ CO ₃	1210						
Sb	19.8	¹²⁴ Sb	60.3	0.58	0.44	0.31	0.23
Te ^c	314.1	^{129m} Te	33.6	1.63	0.93	0.49	0.27
Sn ^d	22.4	¹¹³ Sn	115	0.23	0.20	0.16	0.14
Ag	24.1	^{110m} Ag	250	0.25	0.23	0.21	0.20
ZrO ₂	1708						
Mo	1193						
Ru	1491						
Cd	39.4						
BaO	1009						
CeO ₂	1204						
Nd ₂ O ₃	3330						
<u>Fissium II (nonvolatile tracers, sintered at 1500°C)</u>							
Ru	1494	¹⁰³ Ru	39.3	50.0	31.0	18.0	11.0
Eu ₂ O ₃ ^e	2640	¹⁵⁴ Eu	3214	6.75	6.71	6.67	6.62
ZrO ₂	1713						
Mo	1191						
BaO	1025						
CeO ₂	1220						

^a Mixed into UO₂ powder: 290 g in Fissium I and 310 g in Fissium II.

^b At shipment from KfK, April 6, 1984.

^c 98.21% ¹²⁸Te, 0.96% ¹³⁰Te, 0.67% ¹²⁶Te, supplied by ORNL.

^d 80.04% ¹¹²Sn, 4.85% ¹¹⁶Sn, 4.72% ¹²⁰Sn, 4.67% ¹¹⁸Sn, etc., supplied by ORNL.

^e 98.76% ¹⁵³Eu, 1.24% ¹⁵¹Eu, supplied by ORNL.

Table 2. Data for tests of simulant fuel

	Test No.			
	HS-1	HS-2	HS-3	HS-4
<u>Fuel specimens</u>				
Identification No. ^a	1	6	2	8
Fissium type ^b	I	II	I	II
Added material, ^{c,d} type	SST			SST; Ag
Added material, mass (g)	9.1			9.1; 1.9
Furnace ceramics	ZrO ₂	ThO ₂	ZrO ₂	ThO ₂
<u>Test conditions</u>				
Date conducted	5/4/84	6/11/84	6/29/84	10/18/84
Temperature (°C)				
Phase A	1600	2000	2000	2000
Phase B	1900	2400	2000	2400
Time (min)				
Phase A	15	10	1	5
Phase B	30	10	20	15
Heatup rate (°/s)				
Phase A	1.1	1.6	1.1	1.3
Phase B	1.9	1.3		1.9
Gas flow rate (L/min)				
Steam	1.45	1.34	1.35	1.30
Helium	0.50	0.49	0.50	0.51
Aerosol sampler operation	Phase B	Phase B	Phase B	Phase A

^aAssigned at KFK during fabrication.

^bType I contained ^{110m}Ag, ¹¹³Sn, ¹²⁴Sb, ^{129m}Te, and ¹³⁴Cs tracers, was not sintered. Type II contained ¹⁰³Ru and ¹⁵⁴Eu tracers, was sintered at 1500°C during fabrication.

^cSST ≡ stainless steel rod, 3.18-mm diam × 14.6-cm long, containing ⁵¹Cr, ⁵⁴Mn, ⁵⁹Fe, and ⁶⁰Co tracer.

^dAg ≡ Ag-In-Cd-Sn control alloy, contained in SST tube (2.0 g), containing ^{110m}Ag tracer.

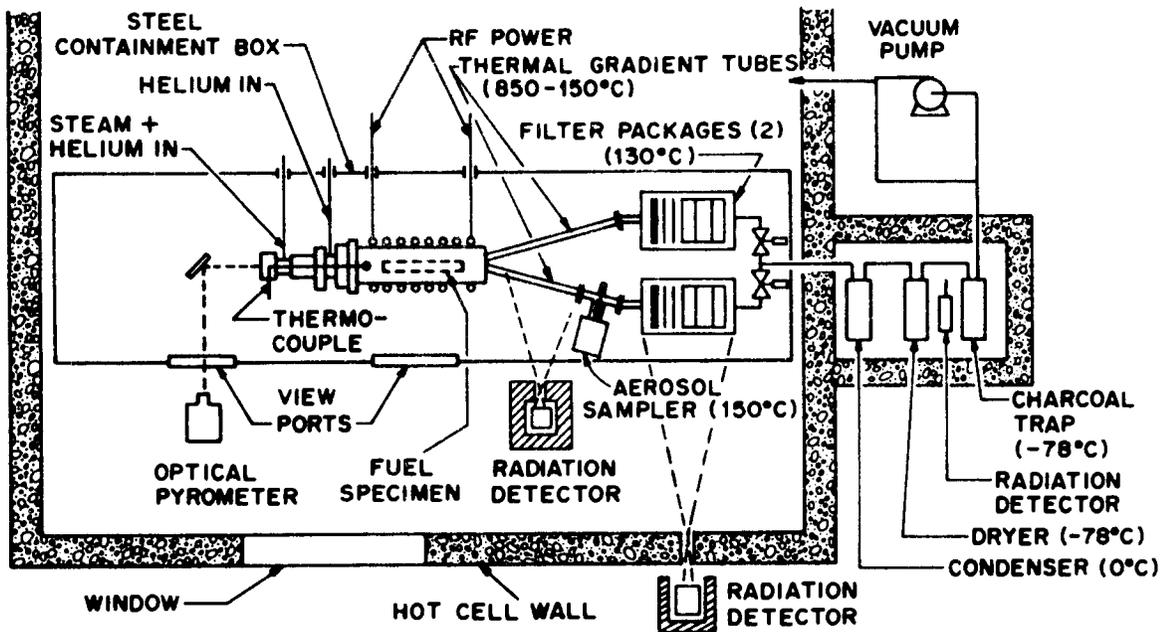


Fig. 1. Fission product release and collection system.

shown in Fig. 2-5. These temperature data are uncorrected; based on calibration testing, the true maximum temperatures were $\sim 100^\circ\text{C}$ higher than those indicated by the optical pyrometer. Although the pressure drop through the apparatus increased with aerosol deposition on the filters, the gas flow rates during these four tests were relatively constant.

3. TEST RESULTS

All four test specimens were heavily oxidized and, as evidenced by fractures, appeared to be quite brittle. The specimens in tests HS-2 and HS-4 had melted sufficiently to attack the ThO_2 boat and ThO_2 furnace tube. As a result, the entire assembly was fused together; the upper part of the furnace tube was broken away to permit inspection of the interior. Much of the Zircaloy cladding in test HS-2 and HS-4 had melted, but in tests HS-1 and HS-3, clad melting was not apparent. The stainless steel and Ag-In-Cd-Sn additives in tests HS-1 and HS-4 had melted completely, apparently contributing to the fuel/cladding interaction in the latter test.

3.1 RELEASE FROM TYPE I FISSIUM SPECIMENS

The fission product data from gamma spectrometric analysis of tests HS-1 and HS-3 are summarized in Tables 3 and 4. The results of these tests are quite similar: $>90\%$ of the Ag, Sn, Te, and Cs was released from the fuel in each case, and the antimony release (from the fuel) was 54% and 91% in HS-1 and HS-3 respectively. Although essentially all of the Te and Cs that was released from the fuel escaped from the furnace, large fractions of the released Ag, Sn, and Sb were retained on the furnace ceramics. Independent measurements of the amounts of these elements in the fuel specimen before and after the tests were in reasonably good agreement. The values in Tables 3 and 4 exceeding 100% provide an indication of the precision of our results.

3.2 RELEASE FROM TYPE II FISSIUM SPECIMENS

The type II fission specimens, which were heated in tests HS-2 and HS-4, did not contain any of the more volatile elements, either as radioactive or nonradioactive species (see Table 1). If these elements had been included during fuel fabrication, large fractions of Cs, Sb, Te, Sn, and Ag would have escaped during sintering. The tracer nuclides in the type II fission were ^{103}Ru and ^{154}Eu . In addition, irradiated stainless steel, containing ^{51}Cr , ^{54}Mn , ^{59}Fe , and ^{60}Co , and irradiated Ag-In-Cd-Sn control alloy, containing $^{110\text{m}}\text{Ag}$, were included with the fuel specimen (Table 1) to investigate the release of these materials during test HS-4.

The release data for tests HS-2 and HS-4 are shown in Tables 5 and 6. As expected, very small fractions ($\ll 1\%$) of the ^{103}Ru and ^{154}Eu were released from the fuel, even at $\sim 2400^\circ\text{C}$. The significant

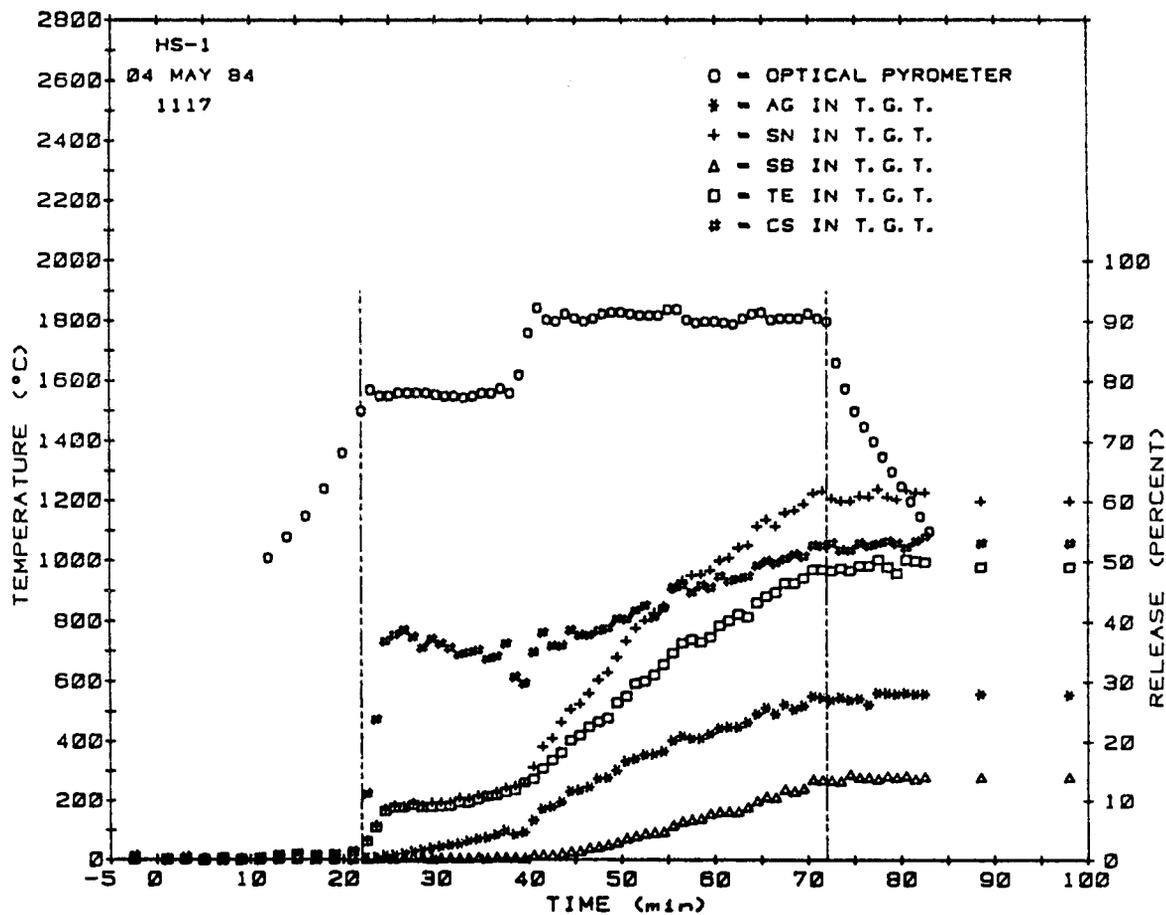


Fig. 2. Temperature and fission product release history for test HS-1.

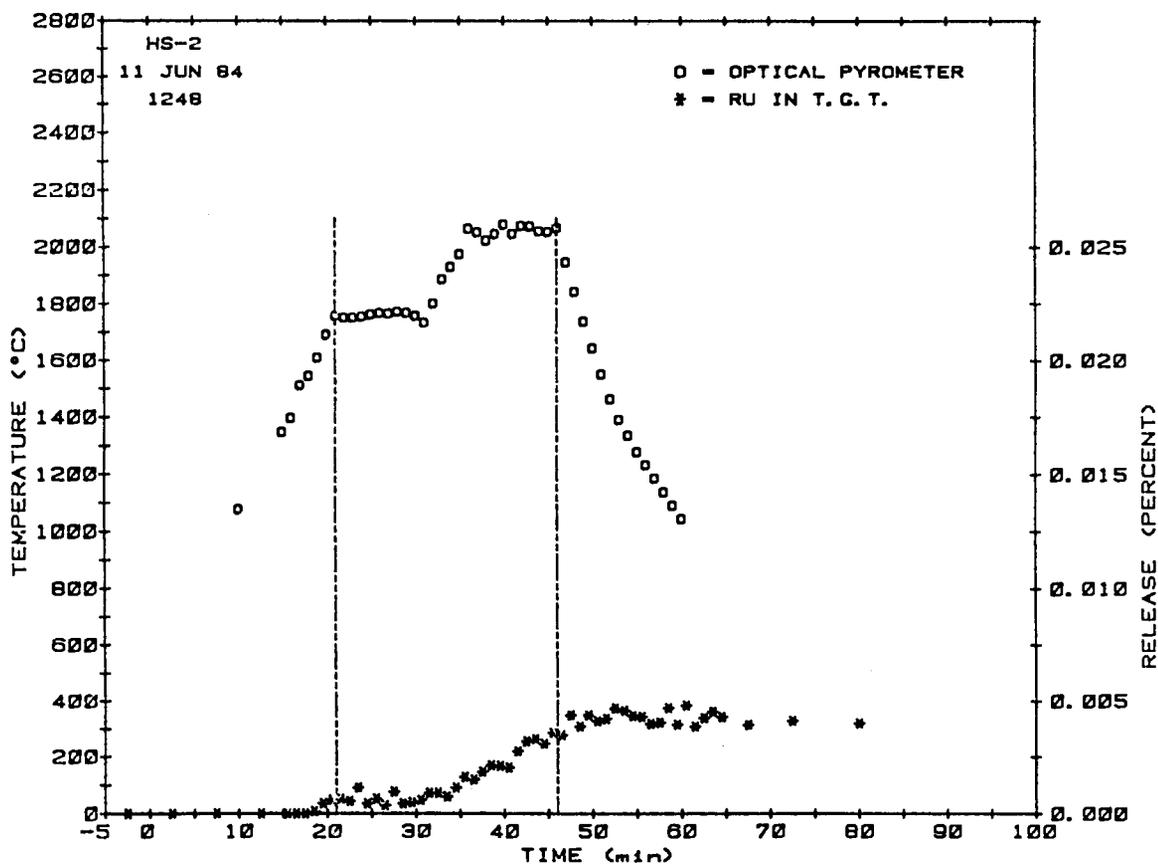


Fig. 3. Temperature and fission product release history for test HS-2.

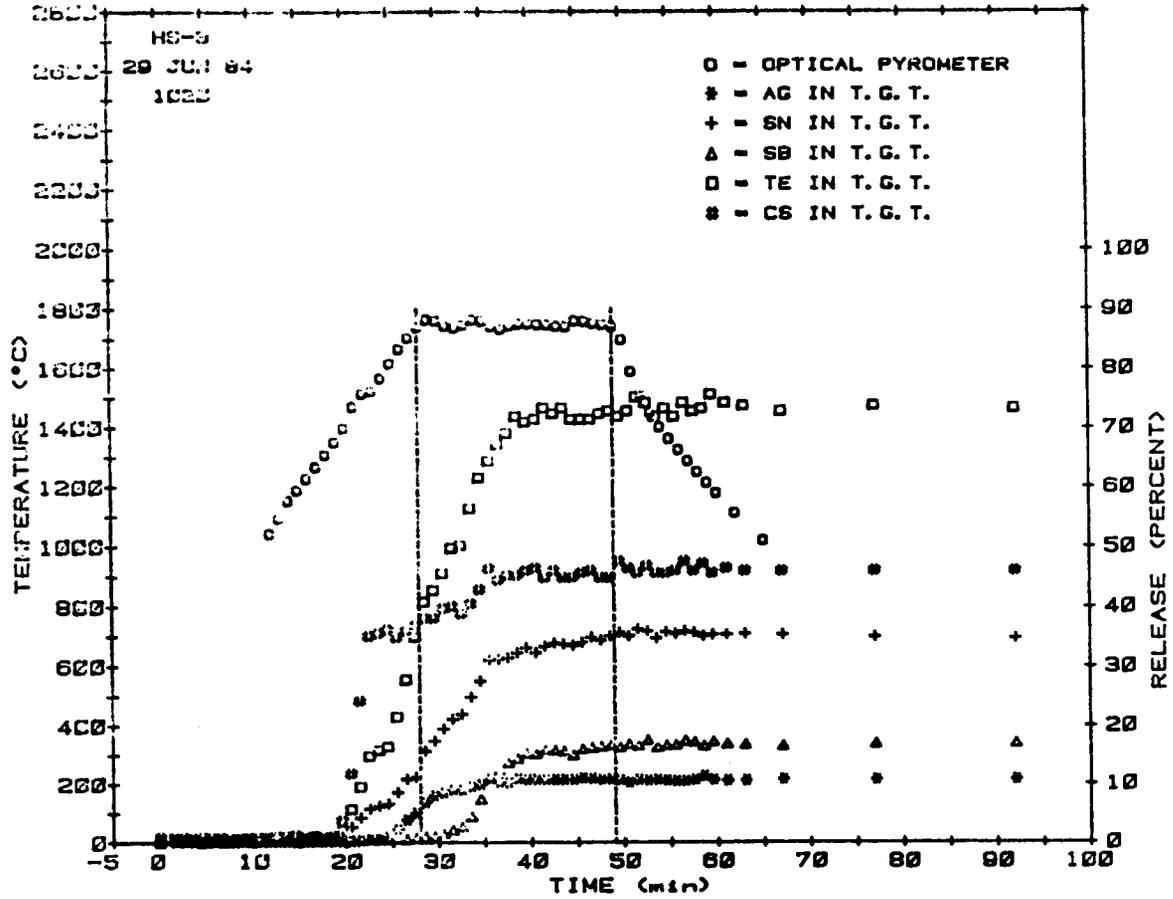


Fig. 4. Temperature and fission product release history for test HS-3.

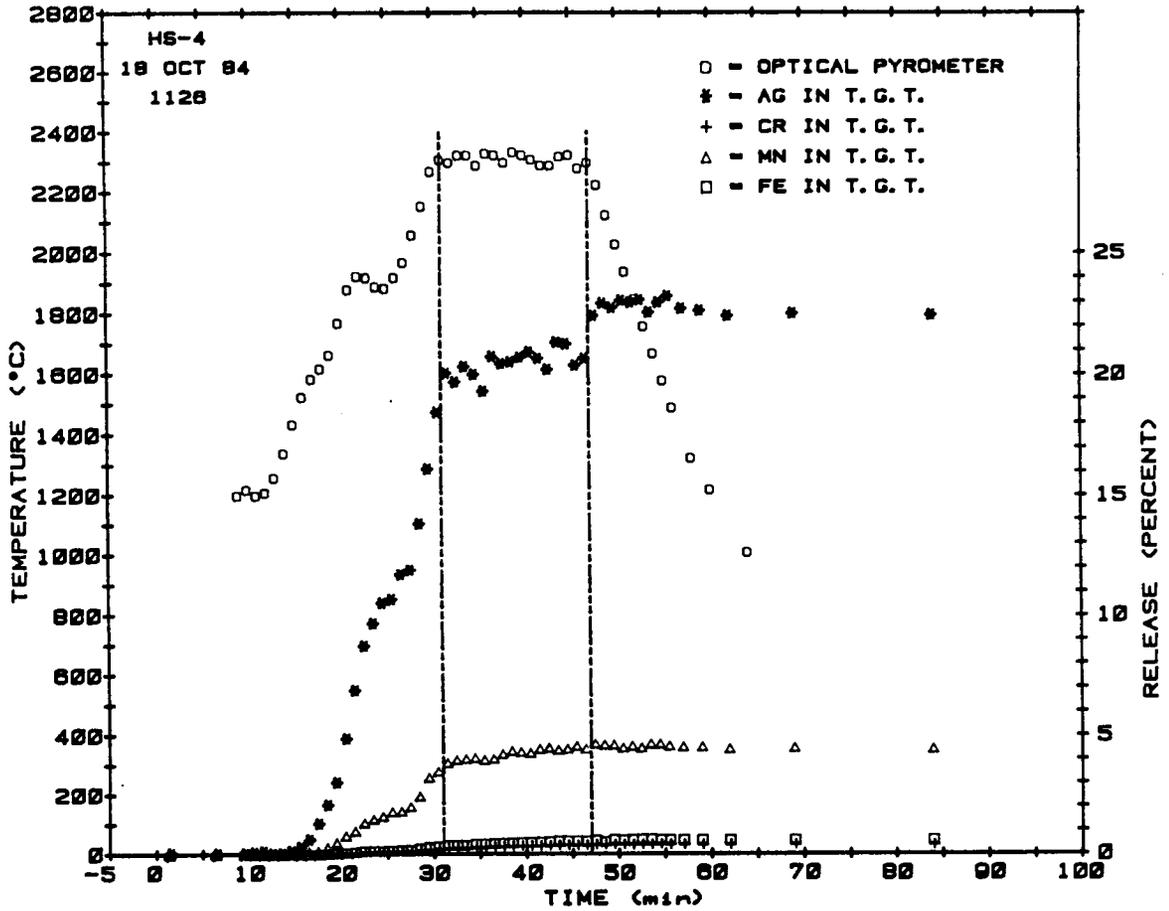


Fig. 5. Temperature and fission product release history for test HS-4.

Table 3. Summary of release data for test HS-1

Component or collector	Temperature (°C)	Fission products found ^{a,b} (%) ^c				
		^{110m} Ag	¹¹³ Sn	¹²⁴ Sb	^{129m} Te	¹³⁴ Cs
Furnace	1000-1900	46.67	16.25	25.32	1.09	12.43
Train No. 1 (heatup + 15 min at 1600°C)						
T.G. tube	150-850	5.63	7.94	2.18	6.17	47.77
Filters	200	1.20	0.16	0.15	2.88	40.82
Total		6.83	8.10	2.33	9.05	88.59
Train No. 2 (30 min at 1900°C + cooldown)						
T.G. tube	150-850	22.48	51.77	12.15	42.70	5.80
Aerosol sampler	200	0.27	0.21	0.19	0.52	0.12
Filters	200	21.90	16.66	13.55	40.94	5.95
Total		44.65	68.64	25.89	84.16	11.87
Total released from furnace		51.48	76.74	28.22	93.21	100.46
Total released from fuel		98.15	92.99	54.54	94.30	112.9
Total remaining in fuel		7.9	0.0	48.3	7.1	0.0
Apparent release based on pre- and posttest fuel specimen analysis		92.1	100	51.7	92.9	100

^aBased on dominant gamma rays: 885 keV for ^{110m}Ag, 392 keV for ¹¹³Sn, 1691 keV for ¹²⁴Sb, 460 keV for ^{129m}Te, and 796 keV for ¹³⁴Cs.

^bWith corrections for attenuation in glass, quartz, ZrO₂, platinum, and stainless steel where appropriate; no self-shielding corrections for fuel counting.

^cPercent released based on analysis of a 5-g standard sample.

Table 4. Summary of release data for test HS-3

Component or collector	Temperature (°C)	Fission products found ^{a,b} (%) ^c				
		^{110m} Ag	¹¹³ Sn	¹²⁴ Sb	^{129m} Te	¹³⁴ Cs
Furnace	1000-2000	50.70	37.11	32.40	0.0	9.65
Train No. 1 (heatup + 1 min at 2000°C)						
T.G. tube	150-850	2.86	5.15	2.59	5.55	41.81
Filters	150	9.51	0.51	0.012	6.33	50.28
Total		12.37	5.66	2.60	11.88	92.09
Train No. 2 (20 min at 2000°C + cooldown)						
T.G. tube	150-850	7.85	29.80	13.81	66.73	4.20
Aerosol sampler	140	0.240	0.348	0.347	0.194	0.061
Filters	150	24.75	38.59	41.94	24.36	5.77
Total		32.84	68.74	56.10	91.28	10.04
Total released from furnace		45.22	74.40	58.70	103.16	102.13
Total released from fuel		95.92	111.51	91.10	103.16	111.78
Total remaining in fuel		2.02	7.74	10.21	2.55	0.0
Apparent release based on pre- and posttest fuel specimen analysis		98.0	92.3	89.8	97.4	100

^a Based on dominant gamma rays: 885 keV for ^{110m}Ag, 392 keV for ¹¹³Sn, 1691 keV for ¹²⁴Sb, 460 keV for ^{129m}Te, and 796 keV for ¹³⁴Cs.

^b With corrections for attenuation in glass, quartz, ZrO₂, platinum, and stainless steel where appropriate; no self-shielding corrections for fuel counting.

^c Percent released based on analysis of a 5-g standard sample.

Table 5. Summary of release data for test HS-2

Collector or component	Temperature	Fission products found ^a (%) ^b	
		¹⁰³ Ru	¹⁵⁴ Eu
Furnace	~1000-2400	0.576	0.00577
Train No. 1 (heatup plus 10 min at 2000°C)			
T.G. tube	150-850	0.00106	0
Filters	140	0.00013	0.00041
Total		0.00119	0.00041
Train No. 2 (10 min at 2400°C plus cooldown)			
T.G. tube	150-850	0.00297	0.00171
Aerosol sampler	150	0.00106	0.00030
Filters	140	0.00327	0.00165
Total		0.00729	0.00366
Total released from furnace		0.00848	0.00407
Total released from fuel		0.584	0.00984

^aWith corrections for attenuation in glass, quartz, ZrO₂, platinum, and stainless steel, where appropriate.

^bPercent released based on analysis of a 5-g standard sample for the dominant gamma rays: 497 keV for ¹⁰³Ru and 1274 keV for ¹⁵⁴Eu.

Table 6. Summary of release data for test HS-4

Component or collector	Temperature (°C)	Nuclide found ^{a,b} (%) ^c					
		¹⁹³ Ru	¹⁵⁴ Eu	^{110m} Ag	⁵¹ Cr	⁵⁴ Mn	⁵⁹ Fe
Furnace ^d	500-2500	0.111	0.125	39.2	19.4	43.4	18.1
Train No. 1 (heatup plus 5 min at 2000°C)							
T.G. tube	150-850	0	0	14.6	0.18	3.08	0.27
Aerosol sampler	150	0	0	1.1	0	0.34	0
Filters	130	0	5E-4	15.1	0.14	3.71	0.17
Total		0	5E-4	30.8	0.32	7.13	0.44
Train No. 2 (15 min at 2400°C plus cooldown)							
T.G. tube	150-850	0	0	12.0	0.27	2.11	0.40
Filters	130	0	0	18.9	0.46	4.22	0.98
Total		0	0	30.9	0.73	6.33	1.44
Total released from furnace		0	5E-4	61.7	1.05	13.5	1.88
Total released from fuel region		0.11	0.13	100	20.5	56.9	20.0

^a Based on dominant gamma rays: 497 keV for ¹¹³Ru, 1274 keV for ¹⁵⁴Eu, 885 keV for ^{110m}Ag, 320 keV for ⁵¹Cr, 835 keV for ⁵⁴Mn, and 1099 keV for ⁵⁹Fe.

^b With corrections for attenuation in glass, quartz, ZrO₂, platinum, and stainless steel, where appropriate.

^c Percent released based on analysis of a 5-g standard sample.

^d Preliminary values based on incomplete analyses of furnace components.

deposit of ^{103}Ru (0.576% of inventory) found near the furnace exit in test HS-2 (Table 5) appears to be an anomaly; it was not confirmed in test HS-4.

3.3 MASSES OF MATERIAL COLLECTED ON THE THERMAL GRADIENT TUBE (TGT) AND FILTERS

Significant masses of material were deposited on the platinum TGTs and the glass fiber filters in all four tests. These data are summarized in Table 7. Based on preliminary analyses, the deposited material was a mixture of the fission product tracers from the fuel and structural and impurity elements from the furnace. As would be expected, these data show that the mass of released material increased with test temperature.

4. SUMMARY AND CONCLUSIONS

Because the objective of this report is to provide only a summary of the principal findings of the tests, and because all analyses have not been completed, any conclusions must be considered as preliminary. Specific interpretation of the test results will be included in the final report on these tests of simulated LWR fuel. The following statements, however, appear justified at this time.

1. The furnace and dual collection trains, which were revisions of previously used 2000°C furnace and single collection train, operated successfully, verifying the use of these test components for similar tests up to 2400°C with highly irradiated LWR fuel.
2. Large fractions, 50 to 100%, of the relatively volatile tracers in the Fissium I fuel ($^{110\text{m}}\text{Ag}$, ^{113}Sn , ^{124}Sb , $^{129\text{m}}\text{Te}$, and ^{134}Cs) were released from the fuel in both tests (HS-1 and HS-3).
3. Very small fractions, <1% of the ^{103}Ru and ^{154}Eu tracers were released from the Fissium II fuel in tests HS-2 and HS-4.
4. With the exception of $^{129\text{m}}\text{Te}$, significant fractions (10 to 50%) of the nuclides released from the fuel deposited in the outlet end of the furnace, primarily on ZrO_2 ceramics; almost all (~99%) of the released Te reached the fission product collection system.
5. The mass of material collected on the TGT and filters increased with test temperature, from 0.75 g in test HS-1 to 2.27 g in test HS-4.
6. The Zircaloy cladding on specimens in tests HS-1 and HS-3 was essentially completely oxidized to ZrO_2 , but remained largely intact. The Zircaloy in the higher temperature tests, HS-2 and HS-4, had melted to a significant extent, and had oxidized as well.

Table 7. Material collected on TGT and filters

Test No.	Temperature (°C)	Time (min)	Mass collected (g)		
			Thermal gradient tube	Filters	Total
HS-1a	1600	15	0.151	0.166	0.317
HS-1b	1900	30	0.176	0.252	0.428
Total			0.327	0.418	0.745
HS-2a	2000	10	0.105 ^a	0.134	0.239
HS-2b	2400	10	0.348 ^a	0.579	0.927
Total			0.453	0.713	1.166
HS-3a	2000	1	0.127 ^a	0.258	0.385
HS-3b	2000	20	0.174 ^a	0.326	0.500
Total			0.301	0.584	0.885
HS-4a	2000	5	0.480	0.574	1.054
HS-4b	2400	15	0.342	0.873	1.215
Total			0.822	1.447	2.269

^a Small pieces were lost from the high temperature ends of the platinum thermal gradient tubes; true values probably should be ~0.020 g higher.

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