

TECHNICAL LETTER REPORT



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TECHNICAL LETTER REPORT

PRELIMINARY RESULTS FROM ORNL FISSION PRODUCT RELEASE TEST VI-7

M. F. Osborne, R. A. Lorenz, and J. R. Travis

1. INTRODUCTION

Test VI-7, the seventh and final in a series of high-temperature tests of irradiated fuel in a vertical furnace facility, was conducted September 16, 1993. This series of tests, and the previous series of six HI tests, was sponsored by the U.S. Nuclear Regulatory Commission (NRC). The objectives of these experiments have been to provide the reliable data on fission product release and behavior under light-water reactor (LWR) accident conditions that are needed for sequence-dependent analyses and to evaluate and compare these data with results from other laboratories. Release values for the major fission product and fuel elements have been measured, and chemical forms of the released species have been determined where possible. The results, which are used for the development of the release models required for LWR accident analysis, have been reported in data summary reports and in the open literature.^{1,5}

Test VI-7 was designed to investigate the effect of air oxidation, of both the UO_2 fuel and the Zircaloy cladding, on fission product release and behavior. Although most of the HI and VI tests had included steam, no previous test had included air. A moderate concentration of water vapor was included to simulate the conditions in the reactor building during either (1) a loss of water level control during refueling or (2) the late stages of a severe core-melt accident, after vessel penetration. Significant oxidation of the fuel would be expected to enhance the release of all fission products, as well as affect the chemical forms, and thereby the transport characteristics, of many fission products. The characterization of the fuel specimen used in this test was reported previously.⁶

This preliminary report is intended to provide a summary of the early results only; further measurements, analyses, and evaluation/comparison of all data will be more complete and may result in revisions of the current data or changes in interpretations.

2. DESCRIPTION OF TEST AND FACILITY

Test VI-7 was conducted in the vertical fission product release test facility (see Fig. 1).⁷ The fuel specimen was a 15-cm-long section cut from the central region of Monticello fuel rod G5, bundle BND-0304, which was irradiated May 1974 to February 1980 to 40.3 MWd/kg as part of an extended burnup program.⁸ Details of this fuel specimen are summarized in Table 1. Fission product inventories, which are required for determining released fractions, are listed in Table 2. Because of the long decay time, 13 years, direct measurement by gamma spectrometry was limited to the longest-lived fission products, ^{125}Sb , ^{134}Cs , ^{137}Cs , and ^{154}Eu , and ^{60}Co from the cladding. The uniform pretest distributions of these nuclides are shown in Fig. 2.

3. TEST OPERATION

Following assembly of the test apparatus, the fuel specimen was loaded into the furnace, and the system was checked for leakage. The temperature history of the test is shown in Fig. 3. Although the normal heatup rate in the VI tests has been ~ 60 K/min, the rate was reduced in test VI-7 to ~ 25 K/min to allow more cladding oxidation at lower temperatures, and thereby to reduce the danger of explosion of the hydrogen generated by the reaction of water vapor with the Zircaloy cladding. With the exception of a brief power failure early in the test sequence (at ~ 1100 K), the planned temperature history was accomplished. Each of the test temperatures (2000 and 2300 K) was maintained for 20 min. The three test phases, A, B, and C, refer to the periods of fission product



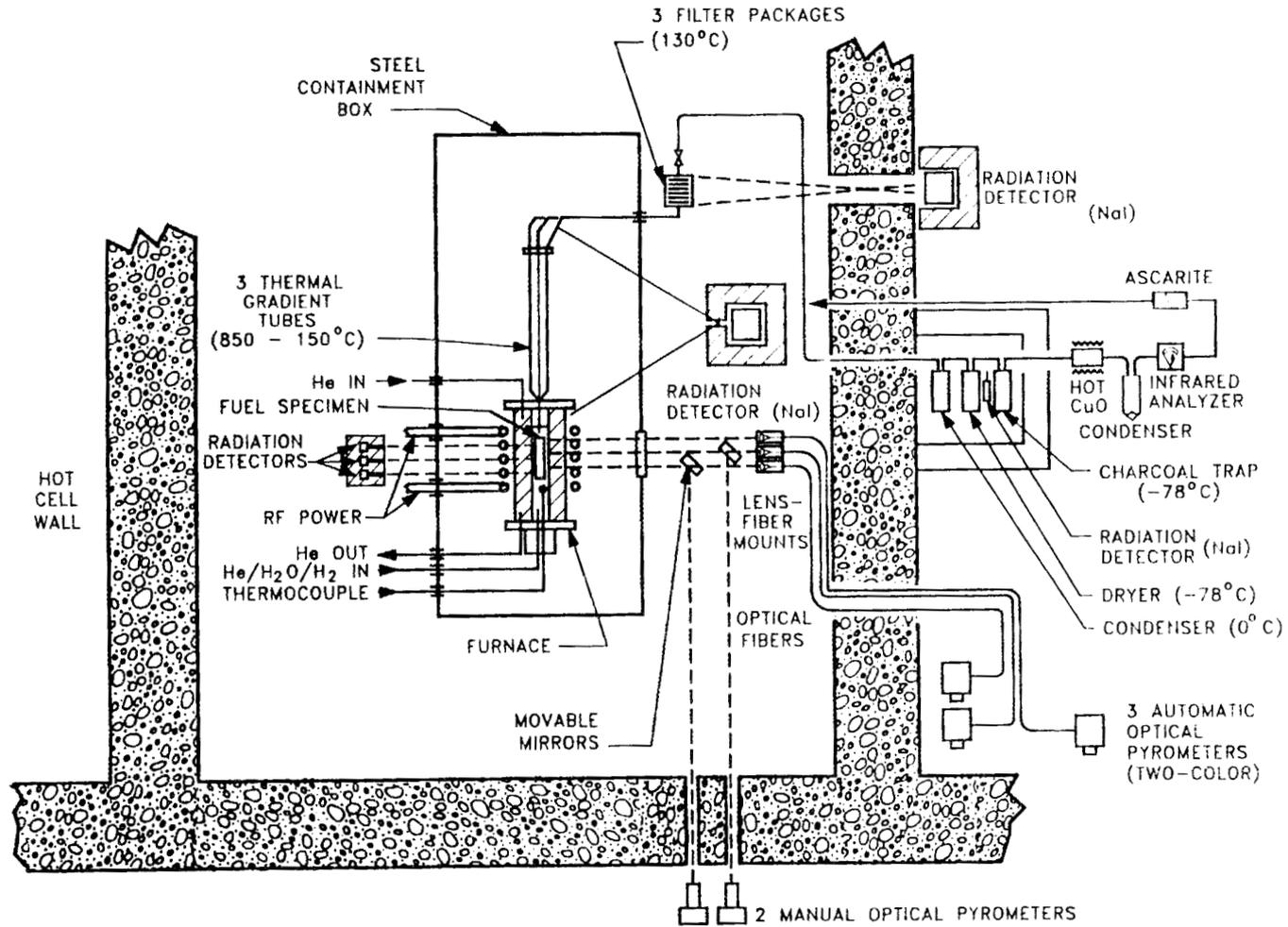


Fig. 1. Vertical fission product release apparatus.

Table 1. Data for fuel specimen used in Test VI-7

Fuel rod identification	BND-0304 (G-5) Section 3
Bundle type	GE, 8 × 8
Irradiation data	
Period	May 1974 to February 1980
Average burnup of rod	~31.4 MWd/kg
Burnup of test specimen	~40.3 MWd/kg
Fuel rod characteristics (unirradiated)	
Zircaloy-4 cladding	1.243-cm (0.489-in.) OD 1.072-cm (0.422-in.) ID
Initial enrichment	2.87% ²³⁵ U
Initial He fill	Atmospheric pressure
Test specimen characteristics	
Length	15.2 cm (6.0 in.)
Specimen fuel loading	126.0 g UO ₂ (112.3 g U)
Weight of Zircaloy cladding and end caps	44.2 g
Total weight of specimen	170.2 g
Gas release during irradiation	1.6% (from whole rod) 2% (from specimen)

Table 2. Fission product inventories in Test VI-7 fuel specimen

Nuclide	Amount in Test VI-7 fuel specimen (mCi)	
	ORIGEN2	Gamma spectrometry
⁸⁵ Kr	455.5	Not detected
¹⁰⁶ Ru	6.4	Not detected
¹²⁵ Sb	53.9	32.4
¹²⁹ I	0.0044	Not detected
¹³⁴ Cs	241.2	213.3
¹³⁷ Cs	10,163	9,776
¹⁴⁴ Ce	0.5	Not detected
¹⁵⁴ Eu	586.5	271.9
⁶⁰ Co ^a		8.4

⁶⁰Co is not a fission product; however, as an activation product in the Zircaloy cladding, it is a good indicator of neutron dose to the cladding and also to the fuel.

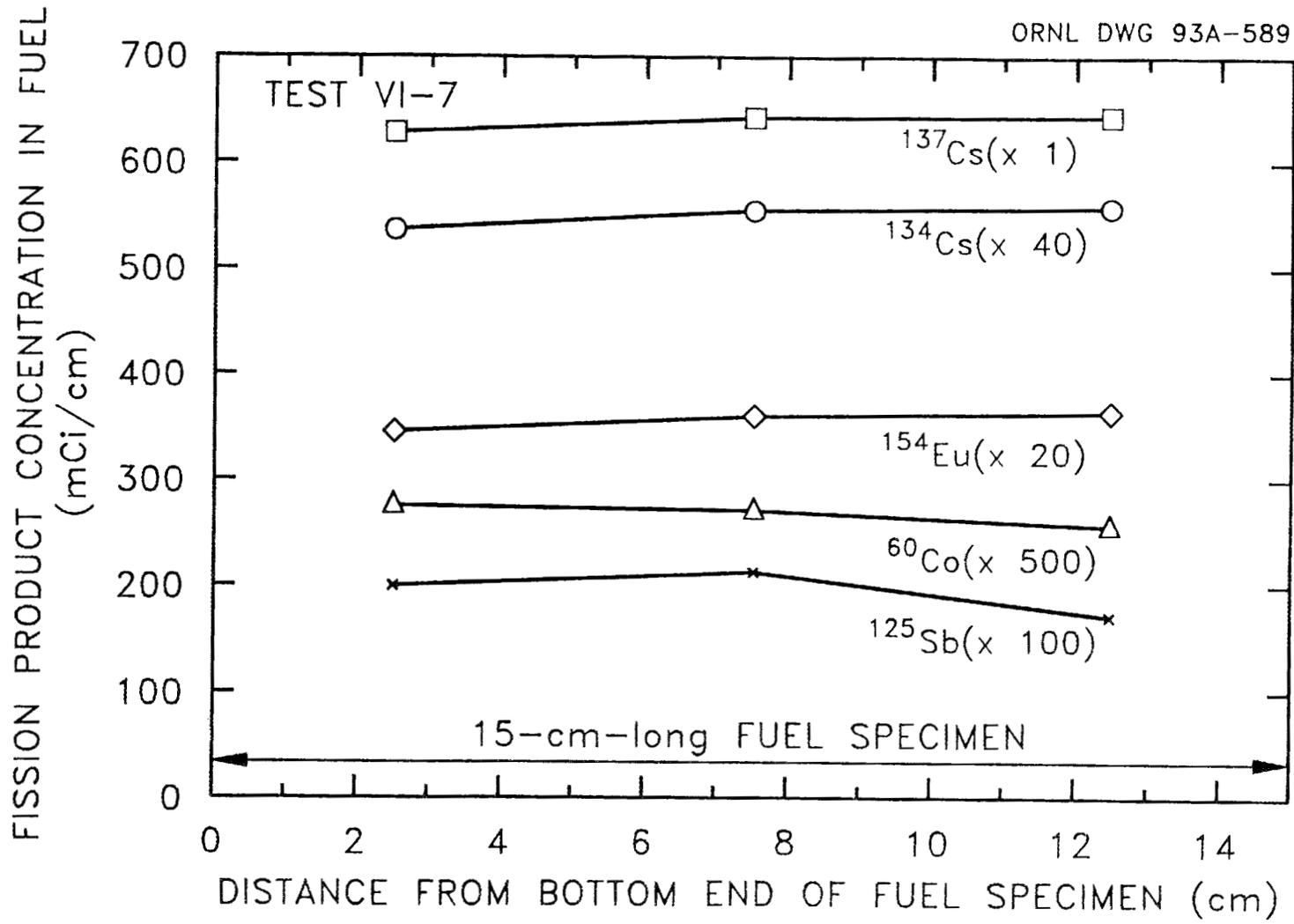


Fig. 2. Axial distribution of ^{134}Cs , ^{137}Cs , ^{125}Sb , ^{154}Eu , and ^{60}Co in Test VI-7 fuel specimen.

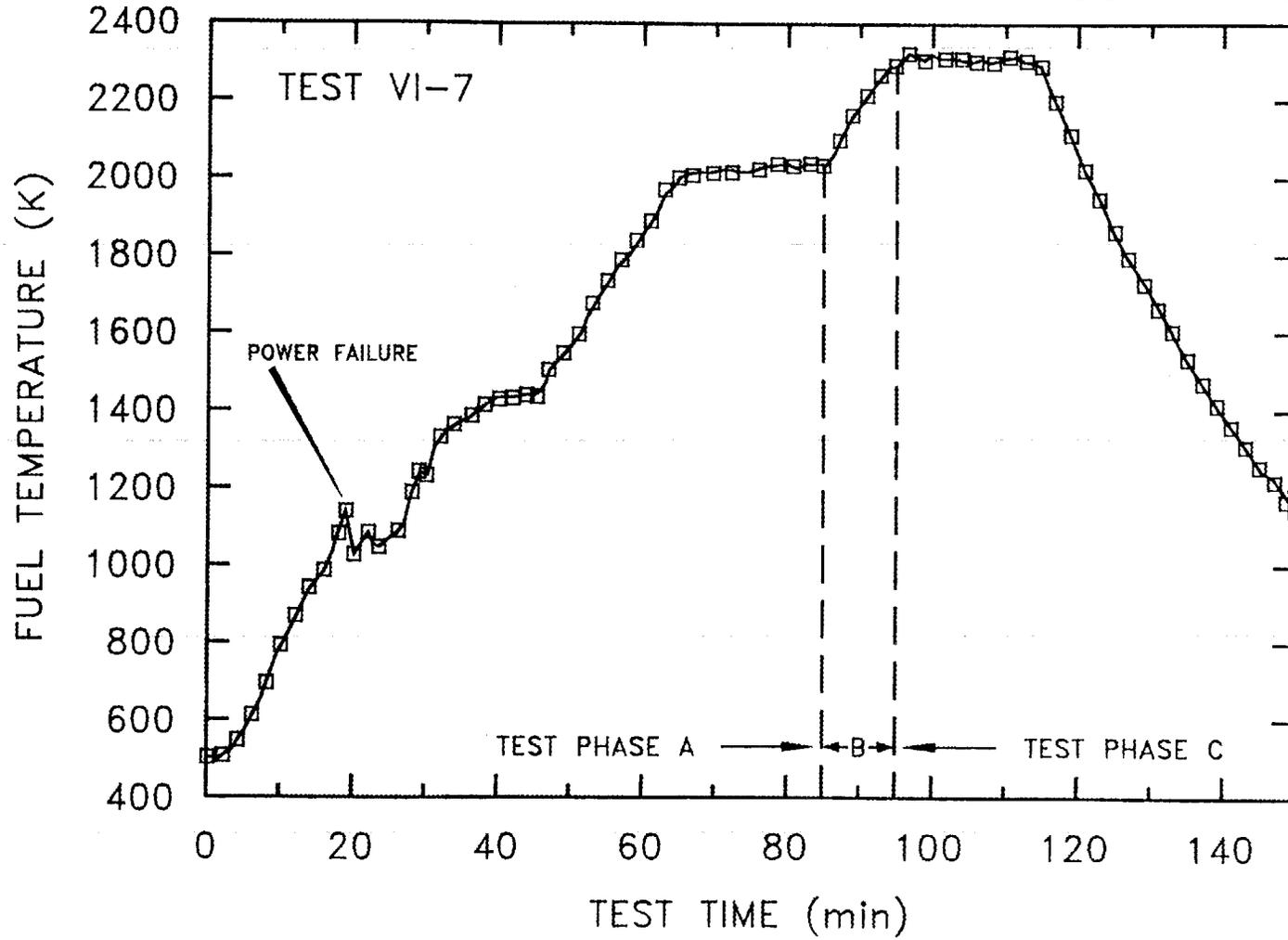


Fig. 3. Temperature history of test VI-7, showing test phases A, B, and C.

collection in the three sequentially operated collection trains. The temperature and gas flow data are summarized in Table 3, and significant events during the test are listed in the test chronology (Table 4).

The test atmosphere was a mixture of 1 L/min air and 0.5 L/min helium, as shown in Fig. 4. During the heatup to 2000 K, the air was saturated with water vapor at 303 K (30°C) to allow only limited hydrogen generation during cladding oxidation. Upon reaching 2000 K, the steam generator temperature was increased to 323 K (50°C) to provide the desired concentration of water vapor for the high-temperature phases of the test.

As in previous tests, a small amount of Kr release was observed early in the heating cycle (at ~600 to 700 K); this Kr is believed to be gas released from the fuel during irradiation and adsorbed on fuel and cladding surfaces until its desorption during the test heatup. As indicated in Fig. 5, significant Kr release as a result of diffusion from the UO₂ began at ~1630 K (1357°C), and cesium was detected on the thermal gradient tube (TGT) at a slightly lower temperature, ~1500 K (1227°C). While the release of Kr appeared to cease at the end of the 2300 K (Phase C) test period, Cs collection on both TGT C and filter C continued for ~10 min as the fuel cooled.

4. TEST DISASSEMBLY AND EXAMINATION

After completion of the test, the containment box was opened, and the radiation in the apparatus was mapped with the in-cell ion chamber for comparison with previous tests. Then the top flange and adjacent ceramic components were removed from the furnace, allowing inspection of the interior. The top end cap and the cladding of the fuel specimen appeared to be heavily oxidized, but the specimen remained vertical and generally intact. The furnace cavity was slowly filled with epoxy resin to keep the fuel specimen in the as-tested orientation during subsequent handling.

Posttest examinations included extensive gamma spectrometry of all components: the fuel specimen, the platinum TGT liners, the filters, furnace ceramics, and TGT-to-filter connecting tubes. Further work will include opening, sectioning, and leaching of the platinum TGT liners and filter components to facilitate analyses for Sr, Mo, Ru, Te, I, Ba, U, and Pu.

5. PRELIMINARY RESULTS OF TEST

The fuel specimen—furnace tube assembly was measured by gamma spectrometry, both in successive 1-cm lengths unshielded and as the entire assembly shielded through a 2.54-cm-thick lead slab, to determine the distribution and the total inventory of fission products remaining after the test. The posttest distributions of the fission products ¹³⁴Cs, ¹³⁷Cs, ¹²⁵Sb, and ¹⁵⁴Eu in the fuel, and of ⁶⁰Co in the cladding, are shown in Fig. 6. It is apparent that the releases of both Cs and Sb were greater at the bottom (inlet) end of the fuel specimen than at the top. Whether this axial release gradient was a result of an axial oxidation gradient along the fuel will be investigated.

A summary of the fission product release data obtained by gamma spectrometry is presented in Table 5. As would be expected from previous results, krypton and cesium release were highest during Phase A, but most of the antimony release was delayed until Phase C, with almost 50% of the released antimony being retained in the furnace. Trace amounts of ¹⁵⁴Eu were detected also, primarily in the outlet end of the furnace.

The ¹³⁷Cs profile in each of the TGT liners is shown in Fig. 7. The maximum concentration (peak) was at 17 to 22 cm, which corresponds to deposition temperatures of ~773 to 673 K (500 to 400°C) in all cases. Two additional high, narrow peaks were present nearer the entrance to the TGT A liner. In the case of TGT C, much lower concentrations of ¹²⁵Sb and ⁶⁰Co were found also, as shown in

Table 3. Operating data for Test VI-7

Specimen temperature	
At start of heatup ramp, K	~ 500
During first plateau to check pyrometers, K	1430
Phase A heatup rate to 2000 K, K/min	23
During 20-min Phase A plateau, average, K	2025
Phase B heatup rate to 2300 K, K/min	26
During 20-min Phase C plateau, average, K	2307
Cooldown rate, K/min	33
Time above 2000 K, min	57
Nominal gas flow rate (L/min at 20°C and 1 bar)^{a,b}	
<u>During Phase A heatup to 2000 K:</u>	
Air to fuel specimen, saturated with water at 30°C	1.8
Helium to susceptor (and fuel specimen)	0.4
Helium to thermowell (and fuel specimen)	0.1
Recirculation/purification system	1.5
<u>During 2000 and 2300 K test period:</u>	
Air to fuel specimen, saturated with water at 50°C	1.0
Helium to susceptor (and fuel specimen)	0.4
Helium to thermowell (and fuel specimen)	0.1
Recirculation/purification system	1.5

^aMeasured by mass flowmeters.

^bAbsolute pressure in furnace was 0.09925 MPa (744.4 mm Hg).

Table 4. Chronology of Test VI-7, conducted September 16, 1993
 Fuel specimen loaded into furnace September 15, 1993

Event/observation	Clock	Time	Temperature at test midpoint of fuel (K, corrected)
	(h)	(min)	
Vacuum and pressure tests	930		
Complete alarm checks	1000		
Begin system preheat	1020		RT ^a
Begin furnace preheat, with gas flow to furnace	1130		RT ^a
Stable flow and temperature	1230		480 ^b
<u>Test Phase A:</u>			
Start ramp to ~1600 K, at ~30 K/min	1300	0	470 ^b
Heatup based on thermocouple	1320	20	960 ^b
Begin accurate pyrometer measurement	1335	35	1305
Reached stable plateau	1339	39	1424
Resume ramp to 2000 K	1345	45	1434
Kr release observed	1352	52	1634
Cs detected on TGT	1358	58	1804
Reached 2000 K plateau, increase steam generator to 50°C	1405	65	2000
After 20 min at 2000 K, end Phase A	1425	85	2033
<u>Test Phase B:</u>			
Begin Phase B, heat to 2300 K at ~30 K/min	1425	85	2033
End Phase B, at 2300 K	1435	95	2292
<u>Test Phase C:</u>			
Begin Phase C, 20 min at 2300 K	1435	95	2292
Rapid Cs deposition on filters	1437	97	2322
End 2300 K plateau, reduce power to cool at ~50 K/min	1455	115	2292
Power off	1509	129	1724
Cooling at ~30 K/min	1520	140	1385
Air flow ended, helium continued	1527	147	1214
Last pyrometer measurements	1530	150	1146
End Phase C, He flow reduced	1631	211	~500

^aRT = room temperature.

^bBased on thermocouple measurement.

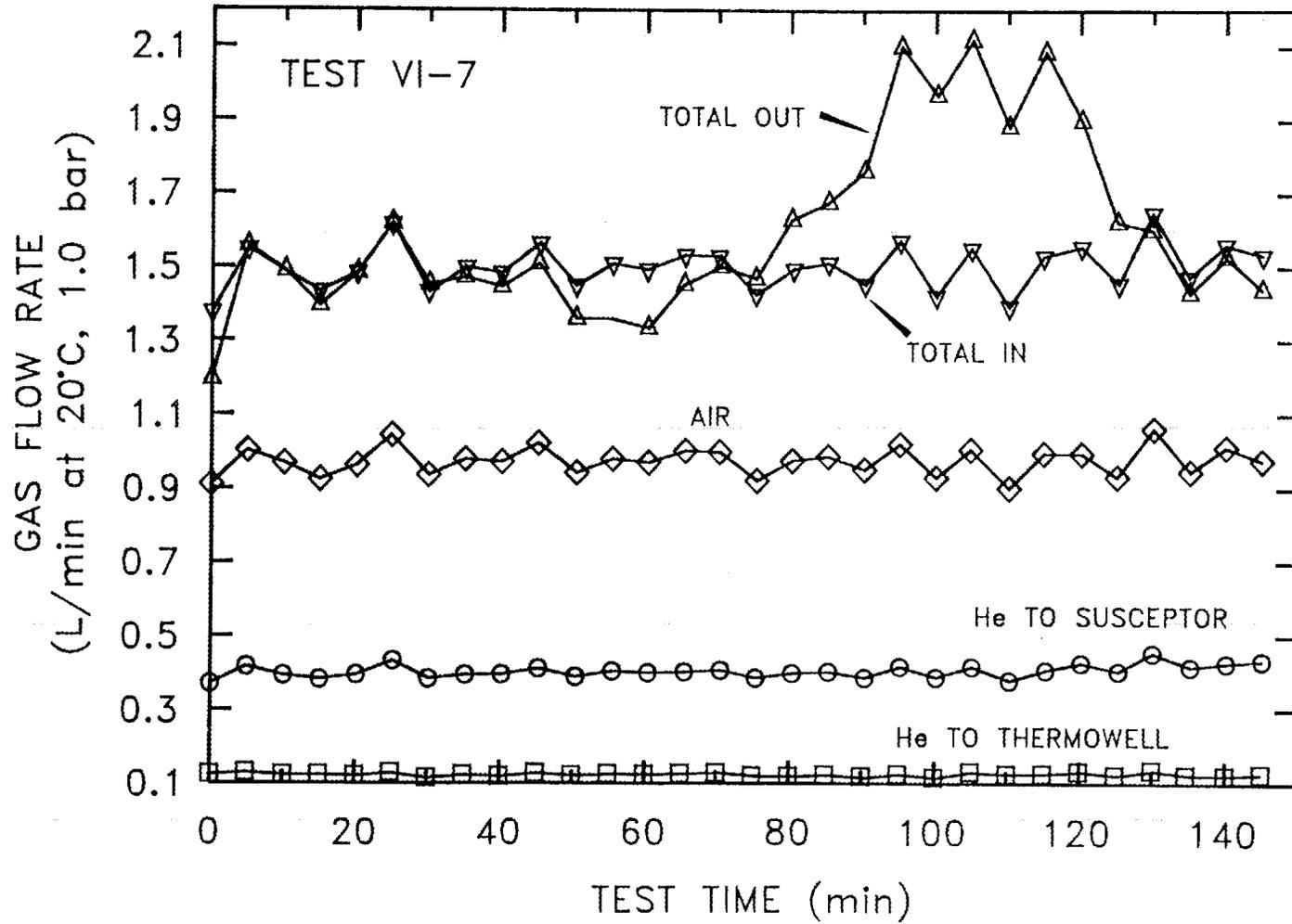


Fig. 4. Gas flow history in Test VI-7.

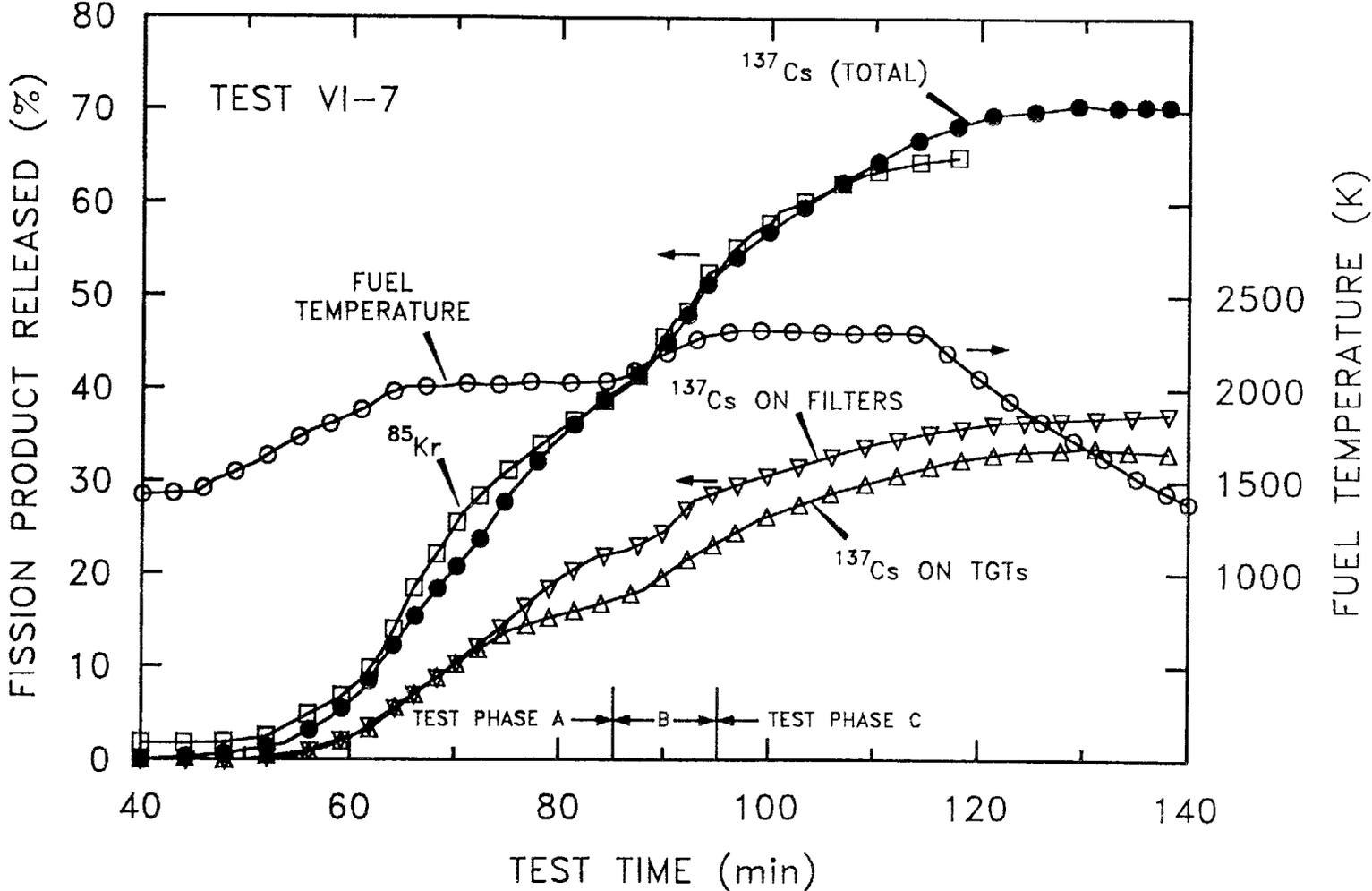


Fig. 5. Release behavior of Kr and Cs in Test VI-7; Cs collected on TGTs was primarily vapor, and Cs collected on filters was primarily aerosol.

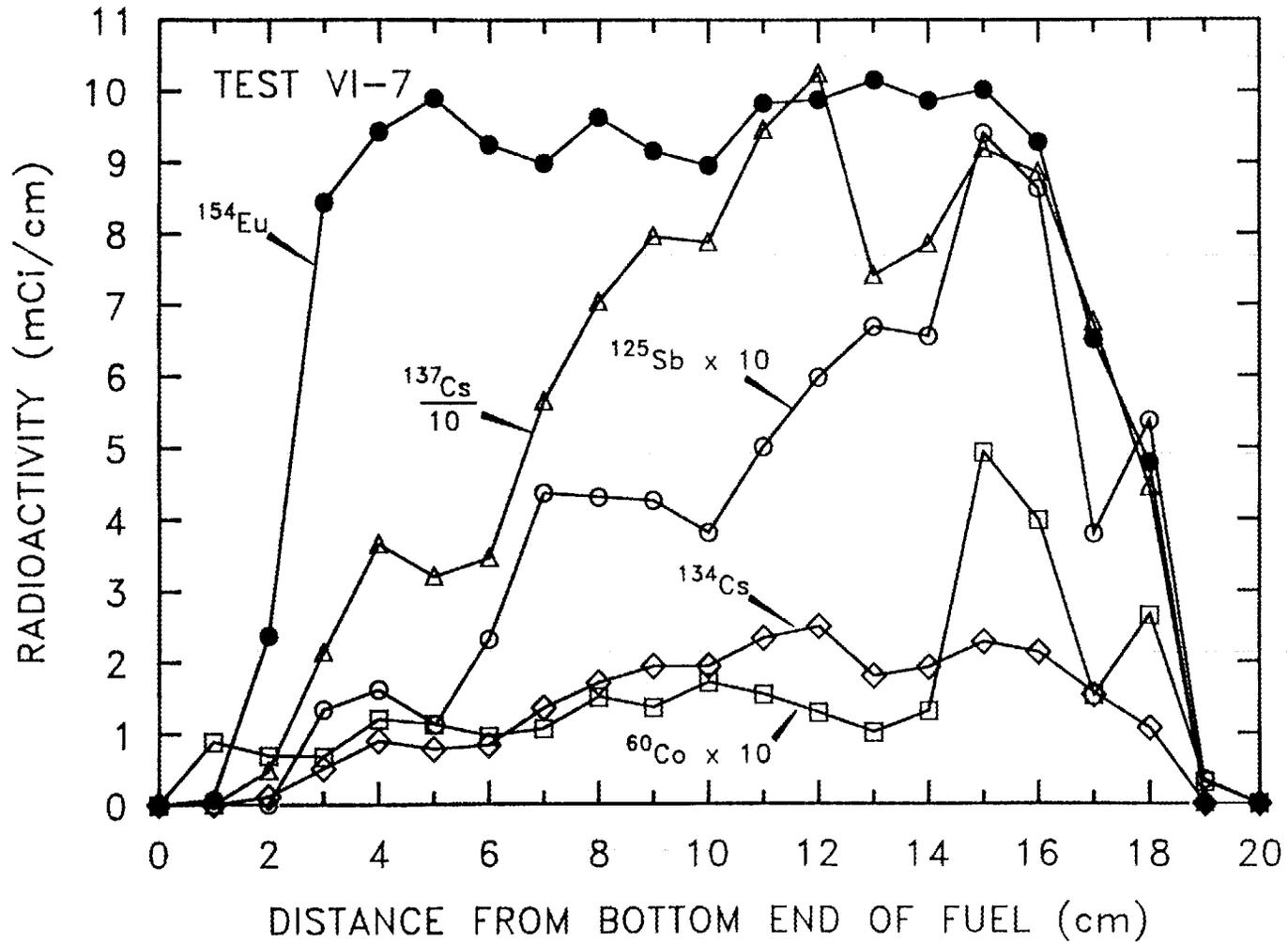


Fig. 6. Distribution of radionuclides in fuel specimen after Test VI-7. Note lower concentrations of ^{137}Cs and ^{125}Sb near bottom (gas inlet) end of fuel.

Table 5. Summary of fission product release data for Test VI-7
(As determined by gamma spectrometry)

Component/ collector	Operating time (at T 2000 K)	Percentage of fission product inventory released during each test phase ^a		
	(min)	⁸⁵ Kr	¹²⁵ Sb	¹³⁷ Cs
Furnace	57	0	25.3	7.3
Train A	20			
TGT			0	15.7
Filters			0	21.0
Total		39	0	36.7
Train B	10			
TGT			0	4.2
Filters			4.8	6.4
Total		14	4.8	10.6
Train C	27			
TGT			6.3	8.0
Filters			15.2	8.8
Total		12	21.5	16.8
Total for test	57	65	51.5	71.4

^aInventories based on fuel analysis and ORIGEN2 calculations.

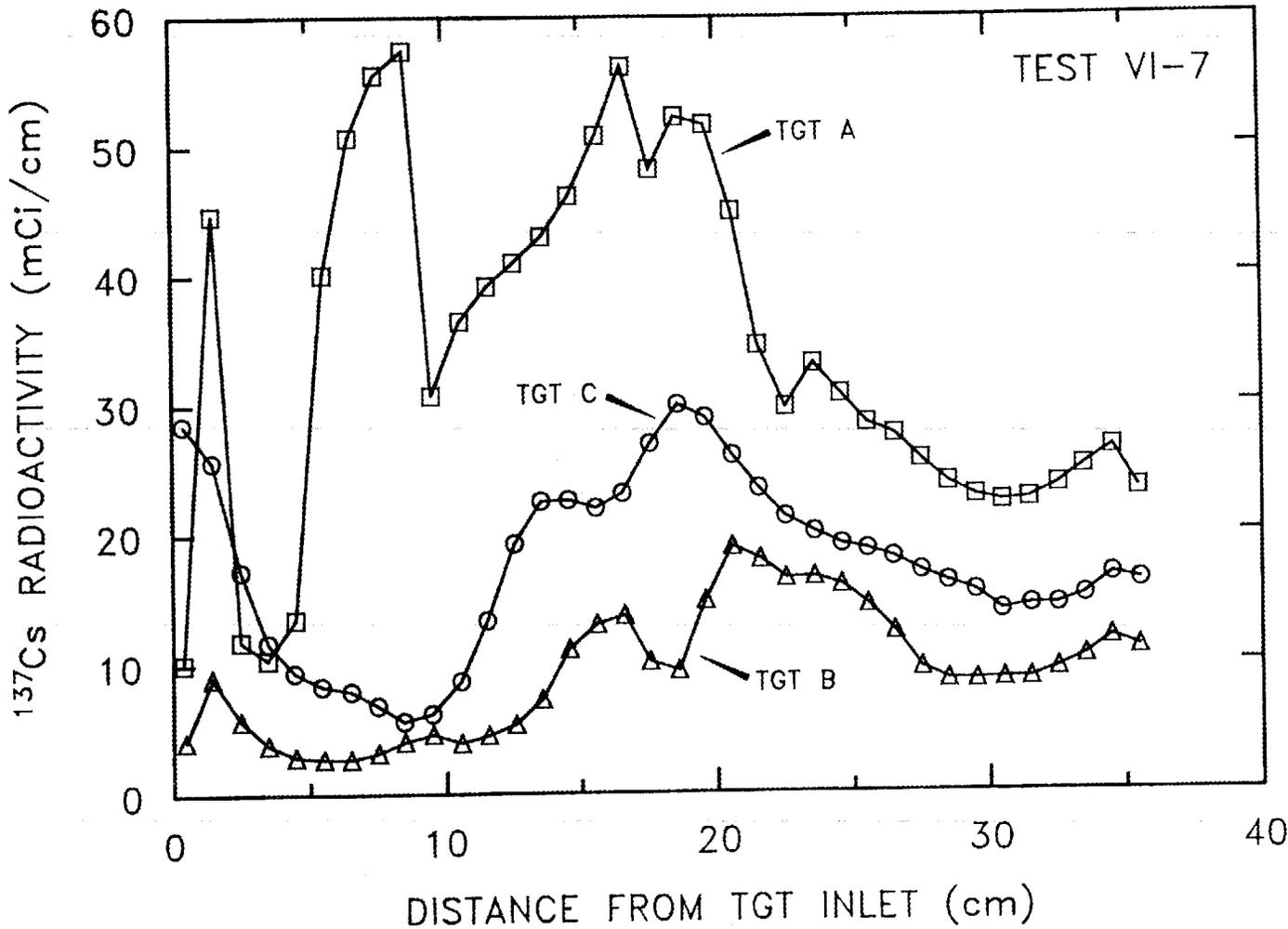


Fig. 7. Distribution of ¹³⁷Cs in TGT liners of Test VI-7.

Fig. 8. Each of the TGT liners and filters was weighed to determine the mass of material collected. These data are summarized in Table 6. As observed in most previous tests, the greatest mass release occurred during Phase C, the highest temperature period.

As an aid in understanding any release results related to oxidation of the Zircaloy cladding, a calculation of cladding oxidation vs test time (and therefore temperature) was carried out using the simple model reported by Yamashita.⁹ In this model, oxidation of 11 cladding nodes and the 2 end caps was considered, and the results are displayed in Figs. 9 and 10. In the oxidation profiles at test times from 60 to 85 min shown in Fig. 9, the progress of the oxidation front along the cladding is illustrated. In Fig. 10, it should be noted that, although complete cladding oxidation was predicted by a test time of 87 min, complete oxidation of the thicker end caps was not predicted until a test time of 98 min.

6. SUMMARY AND CONCLUSIONS

In Test VI-7, a 15-cm-long section of a Monticello (BWR) fuel rod was heated for successive 20-min periods at 2000 and 2300 K in an oxidizing atmosphere, damp air + helium. Although the Zircaloy cladding was heavily oxidized, the specimen remained vertical and essentially intact after the test. According to pretest and posttest gamma spectrometry measurements, the fractional release values from the fuel were 65% for ⁸⁵Kr, 63% for ¹²⁵Sb, 71% for ¹³⁷Cs and ¹³⁴Cs, and <0.05% for ¹⁵⁴Eu. In addition, 35% of the ⁶⁰Co was released from the Zircaloy cladding. Total mass releases of all materials to the thermal gradient tubes and filters were 0.261 g, 0.091 g, and 0.536 g, during test phases A, B, and C, respectively; the total mass release was 0.888 g. The release values for non-gamma-emitting species will be measured by other techniques which require radiochemical methods.

The limited results obtained to date indicate that the test was successfully conducted and that there were no major effects of the air atmosphere. Fuel behavior was similar to that observed in steam atmosphere tests. However, a small amount of ¹²⁵Sb was found on the charcoal cartridges designed to collect volatile forms of iodine, indicating that a small fraction of the released Sb was oxidized to a volatile form that could penetrate the filters at ~120°C, an effect never seen in previous tests in steam.

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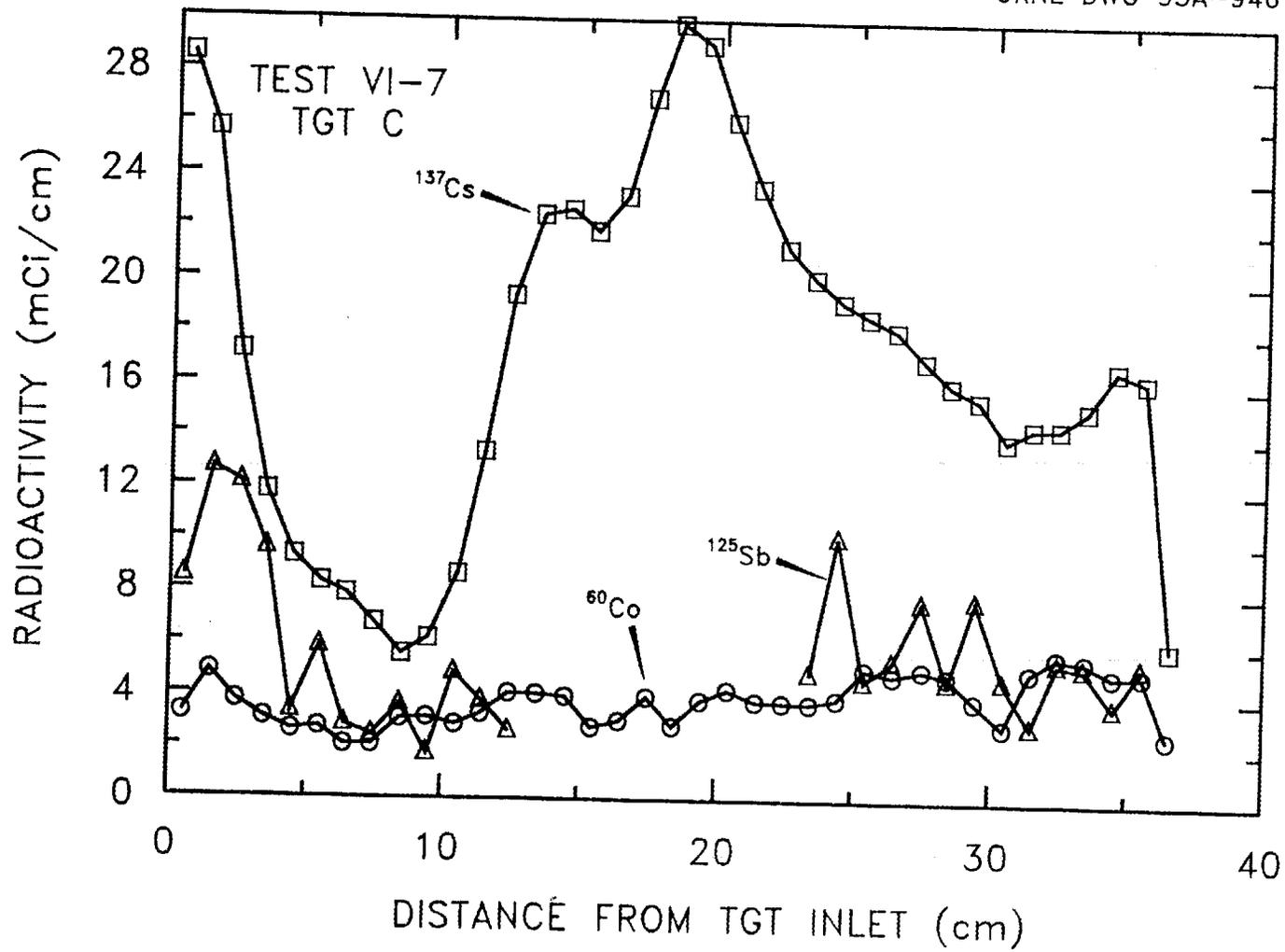


Fig. 8. Distributions of ^{137}Cs , ^{125}Sb , and ^{60}Co in TGT C liner of Test VI-7.

Table 6. Vapor and aerosol deposits in Test VI-7

	Weight of deposits (g) ^a			
	Train A	Train B	Train C	Total
TGT	0.108	0.012	0.206	0.326
Filters				
Prefilter 1	0.149	0.075	0.326	0.550
Prefilter 2	0.004	0.004	0.004	0.012
HEPAs	0.000	0.000	0.000	0.000
Total filters	0.153	0.079	0.330	0.562
Total deposits (TGTs and filters)	0.261	0.091	0.536	0.888

^aPrecision = ± 0.003 g.

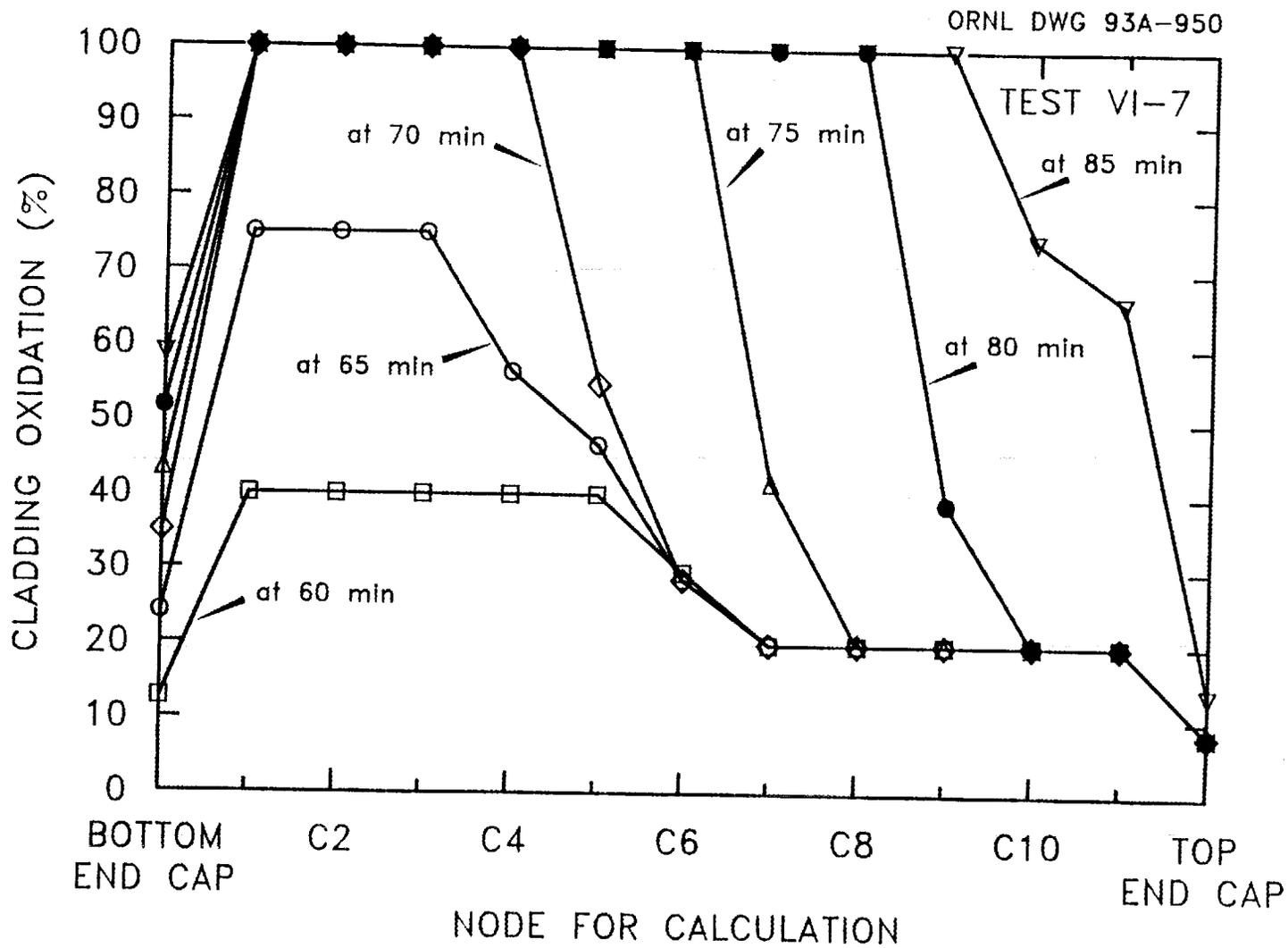


Fig. 9. Successive oxidation profiles at test times of 60 to 85 min as calculated for Test VI-7.

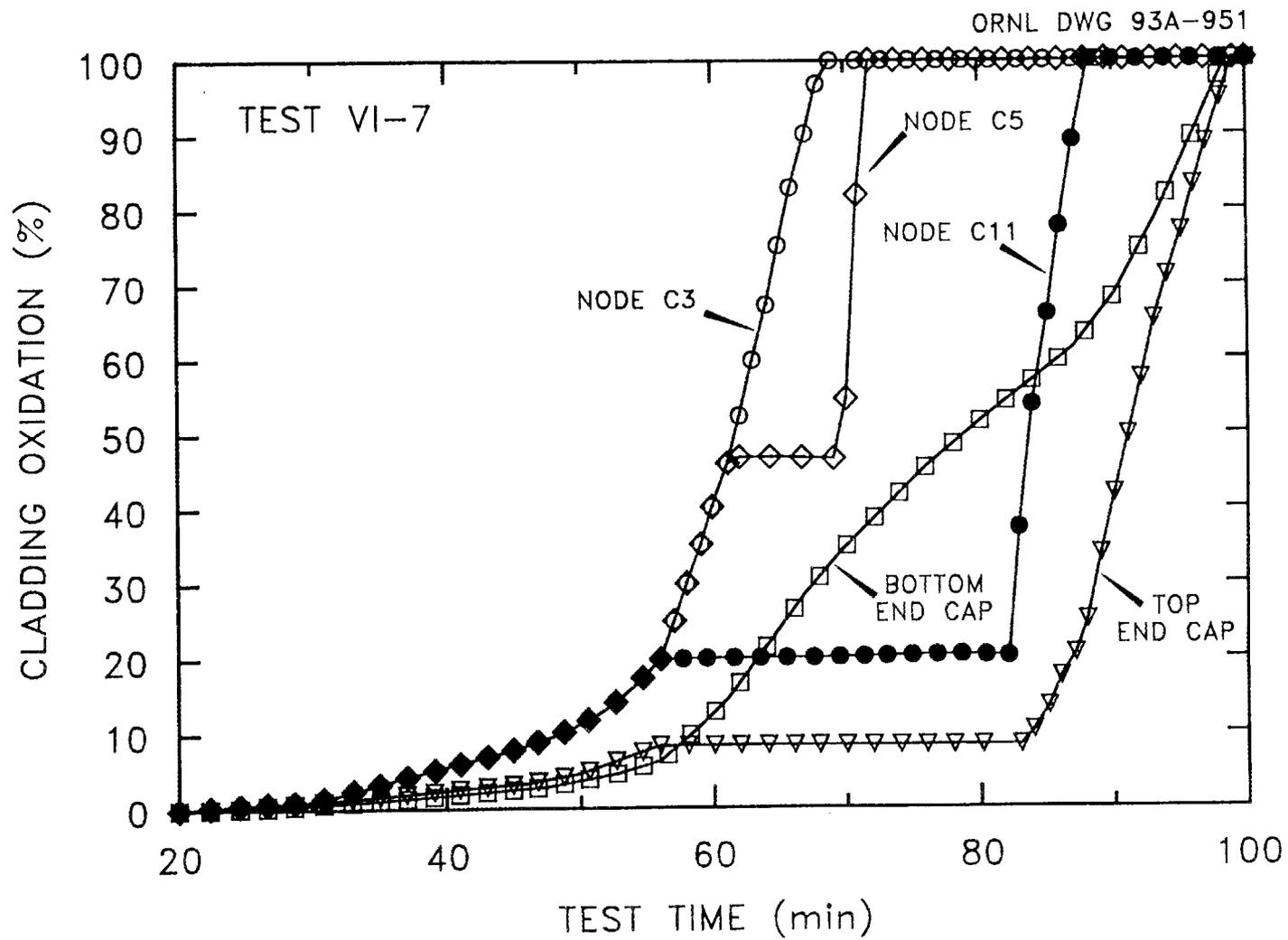


Fig. 10. Calculated extent of Zircaloy oxidation for selected locations in Test VI-7.

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