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A Summary of ORNL Fission Product Release Tests With Recommended Release Rates and Diffusion Coefficients

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Abstract

Fission product release data from the ORNL HI test series and VI test series are summarized in this report and compared with release results from similar tests performed in France (the HEVA test series). The ORNL test results are also compared with four in-reactor tests, the SNL ST-1 and ST-2 tests, and the INEL SFD 1-3 and SFD 1-4 bundle tests. Test atmospheres range from steam to hydrogen, and the temperature range is 1675 to 2700 K.

Two improved fission product release models are proposed: The CORSOR-O model, a fractional release rate model that is similar to CORSOR and CORSOR-M, and the ORNL-Booth model, which is similar to the CORSOR-BOOTH model. Release rate coefficients and diffusion coefficients for 21 different isotopes and elements are given.

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Executive Summary

This report presents a summary of the fission product release tests performed at Oak Ridge National Laboratory (ORNL) between 1975 and 1993. Comparisons are made with several similar tests performed at Grenoble, France (HEVA), with in-reactor tests performed at Sandia National Laboratories (ST-1 and ST-2) and Idaho National Engineering Laboratories (PBF-3 and PBF-4), and with releases at Three Mile Island Unit 2.

Release results for 17 isotopes or elements are pre-sented as percentage of original fuel inventory. The results for the ORNL and other data sets are also presented as fractional release rates (release rate coefficients, min^{-1}) as used in NUREG-0772 and the CORSOR release model. A set of release rate coefficients or model called CORSOR-O ("O" for "ORNL") is presented that can be used as a stand-alone model or can be substituted for the release rate coefficients in existing CORSOR and CORSOR-M models. Some of the CORSOR-O release coefficients differ greatly from previous ones. For example, the CORSOR-O release rate for ruthenium at 2000 K is 361,000 times greater than the CORSOR-M release rate. CORSOR-O provides a simple two-level burnup dependency and atmospheric release effects not included in CORSOR-M.

The release results for the ORNL and other tests are also expressed as diffusion coefficients (cm^2/s) that can be used in any single-atom solid state diffusion equa-tion. A set of recommended diffusion equations is presented that are referred to as the ORNL-Booth Model. This name results from our choice of the Booth diffusion equations, although the recommended diffusion coefficients can be used with any valid diffusion model.

The ORNL-Booth Diffusion Model is designed so that the diffusion coefficients are compatible with the CORSOR-BOOTH Model. CORSOR-BOOTH is a diffusion-based model made available by the Nuclear Regulatory Commission. A set of scale factors is given in this report so that CORSOR-BOOTH can be easily modified to make use of the information in this report.

The ORNL-Booth Model is preferred over the somewhat simpler CORSOR-O model. This is because the diffusion equations follow experiment-derived releases much better as a function of time. The fractional release rate models (NUREG-0772, CORSOR, and CORSOR-M) overestimate releases for long time periods and large fractional releases. It is important to keep in mind that the diffusion coefficients calculated from the experimental results are the best fit for releases that occur by several different mechanisms. Releases calculated for times greater than 30 min at constant temperature will probably be overestimates.

1 Introduction

A program to study the release rate, chemical form, and transport behavior of fission products released from commercial high-burnup pressurized water reactor (PWR) and boiling water reactor (BWR) fuel was begun at Oak Ridge National Laboratory (ORNL) in 1975. As a prelude to investigations with the radioactive fission products, tests were made with nonradioactive fission product simulants to study their behavior in the apparatus proposed for the real fuel testing. The species I_2, CH_3I, Cs_2O, and CsI were studied.[1] The CH_3I decomposed rapidly in steam at 875 K (~8%/s), which demonstrated that methyl iodide could not exist in significant amounts in the reactor core or heated primary system. The importance of CsOH was discovered during these tests. Previous chemical tests and thermodynamic calculations had not included this species. A low-pressure cascade impactor was used to measure the size of particles. The mass median diameter of cesium-containing particles ranged from 0.03 to 0.4 μm, assuming a density of 3.6. Approximately 30% of the CsI was decomposed at 975 K in steam.

A short study of cesium-urania reactions was made using a Knudsen cell and mass spectrometer.[2] The partial pressure of Cs_2CO_3 was reduced by a factor of 10 when in contact with UO_2. The combination CsOH + UO_2 produced the same vapor pressure as Cs_2CO_3 + UO_2. Thermodynamic calculations showed that cesium uranates may have been formed.

A series of 11 tests were performed in which radioactively traced CsOH, CsI, and TeO_2 were coated on unirradiated UO_2 pellets to simulate fission products released to the fuel-cladding gap space.[3] In most tests, the Zircaloy-clad fuel was heated and ruptured from internal pressure. The releases of cesium and iodine were much less than expected from the vapor pressures of pure CsI and CsOH. This work provided the basis for a release model and proved that the apparatus would provide the information needed with highly irradiated fuel.

1.1 Early Tests with High-Burnup Fuel

A series of 11 tests (HBU series) was performed with fully irradiated fuel from the H. B. Robinson reactor.[4] Fission product release tests were conducted in the temperature range 775–1475 K in steam atmosphere (two in dry air). In most tests, the fuel rod segments were ballooned and ruptured using internal gas pressure. These tests showed good agreement with the simulant tests, and the combined results were used to formulate the LOCA Source Term Release Model.[5,6] This model predicts the release of cesium, iodine, and fission gas for moderate-temperature accidents, such as fuel-handling accidents, shipping accidents, and controlled loss-of-coolant accidents. The initial amounts of these fission products in the pellet-to-cladding gap space must be estimated using any good in-reactor fission gas release model. The gap inventories of cesium and iodine should be assumed to be equal to the gas release on a percentage basis. In previous sample calculations,[5,6] we had assumed that cesium and iodine gas inventories were larger than the in-reactor gas release.

A series of four scoping tests was performed with H. B. Robinson fuel in steam using direct induction coupling with the cladding to heat the fuel to the 1575–1885 K temperature range (the HT series).[7] These tests showed that at 1675 K and higher, there was significant release of cesium and iodine from the UO_2 matrix in only a few minutes. Tests at lower temperatures released only the cesium and iodine that had already accumulated in the pellet-cladding gap space. In these tests, most of the released cesium formed CsOH, which immediately reacted with the quartz (silica) tube that surrounded the fuel segment. Some of the released cesium formed CsI that condensed in the thermal gradient tubes (TGTs). From the HT and HBU tests, we concluded that as much as 99% of the iodine behaved like CsI and only ~0.5% as volatile forms of iodine.[8,9]

The BWR series consisted of four tests with fuel from the Peach Bottom-2 boiling water reactor.[10] This fuel had released ~10% of the fission gas, cesium, and iodine to the pellet-cladding gap space while in the reactor. The results of these tests confirmed the validity of the LOCA Source Term Model.[5,6] These tests also confirmed the earlier observation that the amount of volatile iodine produced is <1% of the iodine released whenever total iodine release is >1% of the total inventory. Tests BWR-4[10] and HBU-12[4] were tests in which the pellet-to-cladding gap space was purged with purified helium at temperatures up to 1475 K. These tests showed that the gap inventories of cesium and iodine were equal to the in-reactor gas release on a percentage basis.

1.2 The HI Series Tests

In 1981 the Nuclear Regulatory Commission (NRC) began sponsorship of the horizontal induction-heated (HI) test series.[11-16] The horizontal geometry was retained because of the ease of loading, unloading, and direct viewing of the fuel after the test. These tests were conducted in the temperature range 1675-2200 K, as detailed in Tables 1.1 and 1.2. The results continued to show that only small amounts of volatile forms of iodine were formed in the first second after release from the fuel. Iodine deposition profiles in the TGTs indicated the condensation of CsI as had occurred in the HT and BWR test series. Tests HI-3, 4, and 5 were run with the steam flow restricted so that unoxidized Zircaloy could melt and react with the UO_2 and fission products. The Zr-UO_2 reaction was first suggested as a possible mechanism for fission product release enhancement when the President's Commission on the TMI-2 accident met at ORNL in early June 1979. The reaction had been studied only at lower temperatures as a pellet-cladding interaction problem. The reaction was later called "liquefaction" because the melted Zircaloy penetrated the UO_2 grain boundaries, thereby weakening the pellet structure.[17] Tests HI-3, 4, and 5 showed that this reaction did not enhance the release of fission gas, cesium, or iodine. The melted cladding reacted with the UO_2, but the cladding did not run off. It appeared to be glued to the UO_2, thus restricting release pathways. Fission product chemical form was discussed in the data reports[11-16] and in additional reports.[18-22] A summary of the HI tests has been published.[23]

1.3 HS Tests

The HS tests (horizontal-simulant)[24,25] provided an opportunity to compare release rates obtained in the SASCHA tests performed at Karlsruhe, Germany, with those obtained in our fuel rod geometry. Four tests were run here using unirradiated UO_2 containing simulant fission products that were fabricated at Karlsruhe. The results showed good agreement with the SASCHA data when compared on the basis of fraction released per minute. These tests also provided an opportunity to test thoria ceramics which were needed for the 2675 K target temperature.

1.4 The VI Series Tests

In 1985 the apparatus was changed to the vertical furnace orientation for the VI (vertical induction-heated) test series. Additional improvements were made that are described in Section 2. A summary of VI tests 1-6 has been published.[26] Tests were made with atmospheres of steam, hydrogen, hydrogen followed by steam, and an air-steam mixture.[27-32] The higher test temperatures in the VI series and the use of the three sequentially operated fission product collection systems made it possible to obtain release rates for less-volatile species, such as Ba, Sr, Mo, Eu, Ru, Ce, and Pu. Test operating conditions are listed in Tables 1.1 and 1.2.

Table 1.1 Test temperatures and heatup rates

Test No.	Atmosphere	First temperature plateau			Second temperature plateau			
		Heatup rate (K/s)	Temperature (K)	Time at plateau (min)	Heatup rate (K/s)	Temperature (K)	Time at plateau (min)	Cooldown rate (K/s)
HI-1	Steam	1.2	1675	30				0.6
HI-2	Steam	1.3	2000	20				1.5
HI-3	Lim. steam	2.1	2275	20				1.4
HI-4	Lim. steam	2.3	2200	20				1.6
HI-5	Lim. steam	1.1	2025	20				1.2
HI-6	Steam	2.3	2250[a]	1				1.5
VI-1	Steam	1.0	2020	20	0.6	2300	20	0.8
VI-2	Steam	0.8	2300	60				0.5
VI-3	Steam	0.3	2000	20	0.3	2700	20	0.5
VI-4	Hydrogen	0.7	660	23	1.1	2440	20	0.6
VI-5	Hydrogen	0.25	2000	20	1.2	2720	20	1.1
VI-6	H_2/steam	0.83	2310	60				0.6
VI-7	Air/steam	0.48	2000	20	0.48	2000	20	0.7

[a] Peak temperature.

Table 1.2 Gas flow rates

Test No.	Flow to fuel segment				Flow to susceptor		Total H_2 generated[a] (L)	Pressure (MPa)
	Gas	Flow[a] (L/min)	Gas	Flow[a] (L/min)	Gas	Flow[a] (L/min)		
HI-1	Ar	0.30	H_2O	1.0	Ar	0.20	12.2	~0.099
HI-2	Ar	0.30	H_2O	1.0	Ar	0.15	16.2[b]	~0.099
HI-3	He	0.15	H_2O	0.30	He	0.15	4.9	0.100
HI-4	He	0.15	H_2O	0.32	He	0.15	13.7	0.100
HI-5	He	0.25	H_2O	0.41	He	0.15	11.0	0.099
HI-6	He	0.28	H_2O	3.0	He	0.14	11.0	0.097
VI-1	He	0.40	H_2O	1.54	He	0.40	16.4[b]	0.098
VI-2	He	0.30	H_2O	1.54	He	0.20	11.2[b]	0.101
VI-3	He	0.40	H_2O	1.60	He	0.20	11.2[b]	~0.099
VI-4	He	0.405	H_2	0.405	He + H_2	0.10	0	0.099
VI-5	He	0.40	H_2	0.40	He + H_2	0.10	0	0.099
VI-6A	He	0.40	H_2	0.40	He + H_2	0.2	0	0.099
VI-6B	He	~0.05	H_2O	~0.15	He	0.2		0.099
VI-6C_1	He	~0.05	H_2O	~0.15	He	0.2		0.099
VI-6C_2	He	0.30	H_2O	1.00	He	0.2	11.3[c]	0.099
VI-7	Air	1.0	H_2O	0.95	He	0.5	27.8	0.099

[a] Volume at 20°C and 1 bar.

[b] Cladding 100% oxidized.

[c] Total at end of test; cladding 100% oxidized.

2 Apparatus for HI and VI Series Tests

The furnaces for heating the fuel specimens in the HI and VI tests are shown in Figures 2.1 and 2.2. In both designs, a zirconia cylinder or furnace tube surrounded the fuel rod (thoria cylinder in tests VI-3 and VI-5).[33,34] A graphite cylinder outside of the furnace tube was heated by an induction coil so that heat flowed through the furnace tube to the fuel specimen. The induction coil was spaced to make up for heat loss at the ends and to preheat the gas that flowed past the fuel rod and carried released fission products into the fission product collection systems.

Each collection system consisted of a thermal gradient tube (TGT) in which the temperature dropped from ~1200 to ~400 K. The inner surface of the TGT was usually lined with a platinum insert. The insert was removed and examined for deposits at the end of the test. Platinum was chosen to decrease the chance of chemically altering the fission product species. Preoxidized stainless steel liners were used in some steam-atmosphere tests, and clean stainless steel was used in some hydrogen-atmosphere tests. The dimensions of the TGT liners and other collection system components are given in Table 2.1. Summaries of TGT results have been published.[35,36] The layout of equipment is shown in Fig. 2.3.

The filters consisted of two holders with compressed quartz wool followed by two layers of HEPA-type filters. The filters and TGTs were weighed before and after each test in order to measure the mass of the deposits.

Three cartridges containing impregnated charcoal were located immediately behind the filters. Triethylenediamine (TEDA) impregnant was used to capture all of the volatile forms of iodine (I_2, HI, CH_3I). The filters, impregnated charcoal, and lines to and from the filter assembly were heated to 400 to 420 K to prevent condensation of steam and volatilization of the charcoal impregnant.

Steam was condensed at 273 K and additional moisture removed in a freeze trap maintained at 195 K. Fission gases were collected in two large charcoal traps maintained at 195 K. Three NaI gamma detectors were used to monitor ^{134}Cs and ^{137}Cs deposited in the top of the furnace and in the TGTs, ^{134}Cs and ^{137}Cs collected on the filters, and ^{85}Kr collected in the large charcoal traps.

Beginning with Test VI-4, additional radiation monitors were used that were directed to the top and bottom of the fuel rod. They were used to monitor total radioactivity remaining in the fuel and to watch for collapse of the fuel rod.

Fuel temperatures were monitored with two-color automatic optical pyrometers.[37] A thermocouple was placed close to the fuel to measure temperatures lower than the pyrometers could detect. Beginning with the VI series tests, one or two manually operated disappearing filament optical pyrometers were used to look directly at the outside of the ceramic furnace tube through small viewing ports. Beginning with Test VI-1, a hydrogen measurement system was used to measure the rate of the steam-Zircaloy reaction.[38]

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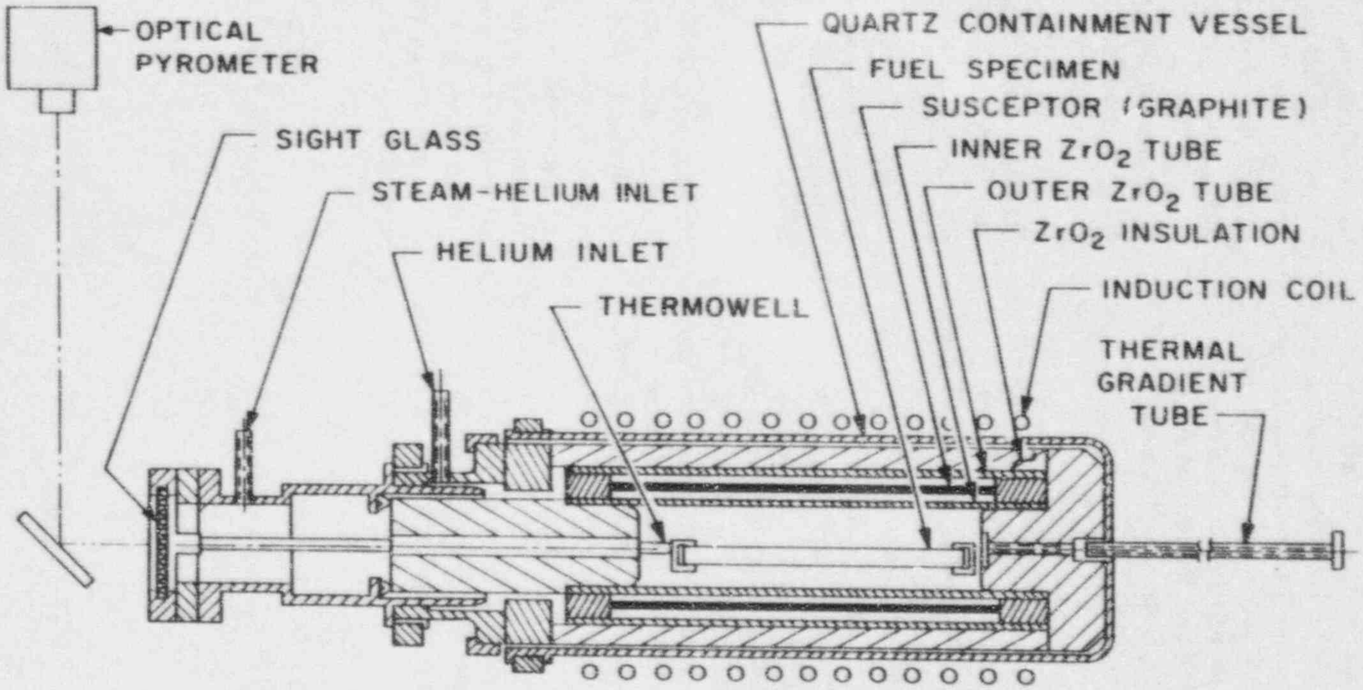

Figure 2.1 Furnace used in the HI test series

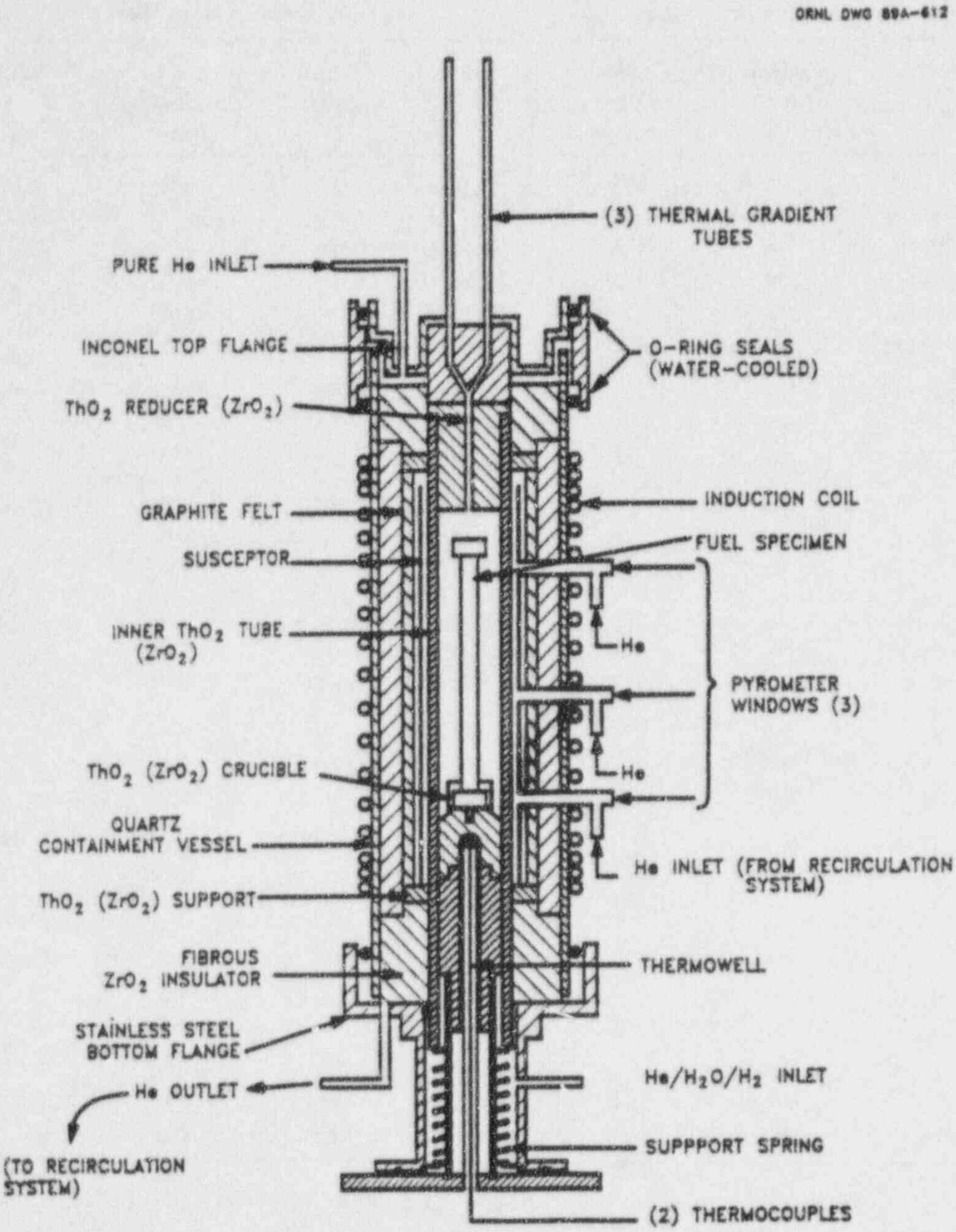

Figure 2.2 Furnace used in the VI test series

Table 2.1 Collection system dimensions

Test No.	Thermal gradient tube			Tube from TGT to filters				Filter
	Material	Length (mm)	Inside diameter (mm)	Material	Length (cm)	Inside diameter (mm)	Number of bends[a]	Diameter (mm)
HI-1	Pt	305	4.0	Teflon	7.6	3.2	2	30
HI-2	Pt + Au	356	4.0	Teflon	7.6	3.2	2	30
HI-3	Pt	356	4.0	St. steel	19	4.6	2	51
HI-4	Pt	356	4.0	St. steel	19	4.6	2	51
HI-5	Pt	356	4.0	St. steel	19	4.6	2	51
HI-6	St. steel	370	4.25	St. steel	19	4.6	2	51
VI-1A	Pt	359	7.3	St. steel	57	8.0	3	89
VI-1B	Pt	359	7.3	St. steel	73	8.0	3	89
VI-1C	Pt	359	7.3	St. steel	89	8.0	3	89
VI-2A	Pt	359	7.3	St. steel	57	8.0	3	89
VI-2B	Pt	359	7.3	St. steel	73	8.0	3	89
VI-2C	Pt	359	7.3	St. steel	89	8.0	3	89
VI-3A	304 St. steel	359	7.3	St. steel	78	8.0	3	89
VI-3B	Pt	359	7.3	St. steel	60	8.0	3	89
VI-3C	Pt	359	7.3	St. steel	47	8.0	3	89
VI-4A	Pt	359	7.3	Copper	78	8.0	2	89
VI-4B	Pt	359	7.3	Copper	60	8.0	2	89
VI-4C	Pt	359	7.3	Copper	47	8.0	2	89
VI-5A	304 St. steel	359	7.3	Copper	78	8.0	2	89
VI-5B	Pt	359	7.3	Copper	60	8.0	2	89
VI-5C	Pt	359	7.3	Copper	47	8.0	2	89
VI-6A	304 St. steel	359	7.3	Copper	78	8.0	2	89
VI-6B	304 St. steel	359	7.3	Copper	60	8.0	2	89
VI-6C	304 St. steel	359	7.3	Copper	47	8.0	2	89
VI-7A	Pt	359	7.3	Copper	78	8.0	2	89
VI-7B	Pt	359	7.3	Copper	60	8.0	2	89
VI-7C	Pt	359	7.3	Copper	47	8.0	2	89

[a] Bend radii 2.54 cm × 60° for Teflon, 5 cm × 30° for HI stainless steel, 2.54 cm × 90° for VI stainless steel, and 15 cm × 90° for copper.

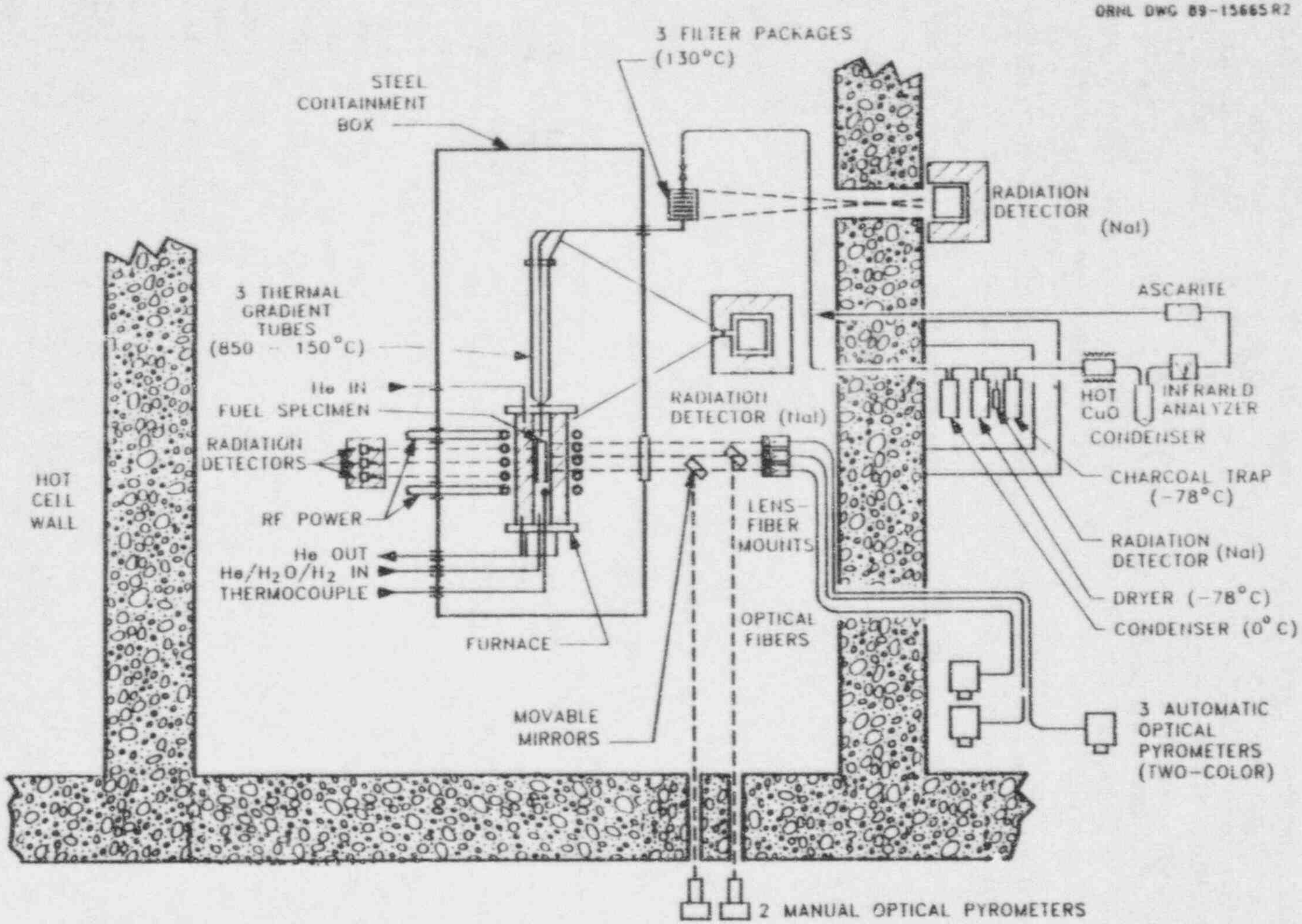

Figure 2.3 Flow diagram for the VI series tests

3 Fuel Characteristics

The characteristics of the fuel used in the ORNL tests have been reported in the individual test data reports. These characteristics are summarized in Table 3.1. Most of the fuel came from commercial PWR and BWR reactors irradiated at moderate linear heat rating typical of modern reactor design. Fission gas release while in the reactor was ≤5%. An exception was the Peach Bottom-2 fuel that had ~10% fission gas release in-reactor.

The BR3 fuel came from the Belgian research reactor BR3. Fuel from this reactor was used in several in-reactor tests because of its higher-than-average enrichment. The BR3 fuel was used in two PBF SFD tests,[39,40] four TREAT STEP tests,[41] and two SNL tests conducted in the ACRR.[42,43] BR3 fuel was used in five ORNL tests to provide a comparison with in-reactor test data. ORNL Test VI-4 and SNL Test ST-1 were run under near-identical atmosphere and temperature conditions. No significant difference in fission product release was observed between the hot cell Test VI-4 and the in-reactor Test ST-1. Also, no significant difference between BR3 fuel releases and commercial fuel releases was detected.

Fission product and actinide inventories are listed in Tables 3.2 and 3.3, respectively. These were calculated using ORIGEN-2.[44] The inventories of Tests VI-1 and HI-6 are typical of commercial PWRs and BWRs. The BR3 fuel had unusually high initial ^{235}U enrichment, espe-cially the VI-4 fuel. The production of plutonium, americium, and curium is favored by low enrichment. Some published inventories for BR3 fuel differ from these, especially for americium and curium. Inventories affect the determination of both release rate coefficients and diffusion coefficients.

Table 3.1 Fuel rod characteristics

Test No.	Reactor	Burnup (MWd/kg U)	Grain[a] diameter (μm)	Fission gas release in-reactor (%)	Fuel length (mm)	Clad[b] O.D. (mm)	UO_2 mass (g)	Zircaloy mass (g)
HI-1	H.B. Robinson	28.1	3.4	0.3	203	10.7	137.3	30.7
HI-2	H.B. Robinson	28.1	3.9	0.3	203	10.7	135.3	30.7
HI-2	H.B. Robinson	25.2	4.3	0.3	203	10.7	136.2	30.7
HI-4	Peach Bottom 2	10.1	6.6	10.2	203	14.3	254.6	51.4
HI-5	Oconee	38.3	8.9	4.1	152	10.8	107.5	25.6
HI-6	Monticello	40.3	8.0	2.0	152	12.4	126.0	44.2
VI-1	Oconee	40	9.2	0.7	152	10.8	109.2	31.2[c]
VI-2	BR3	44	12	~2	152	9.5	82.0	21.3
VI-3	BR3	44	12	~2	152	9.5	81.1	21.2
VI-4	BR3	47	12	~5	152	9.5	78.2	21.1
VI-5	BR3	42	12	~2	152	9.5	80.8	21.2
VI-6	BR3	42	12	~2	152	9.5	81.5	21.5
VI-7	Monticello	40.3	8.0	2.0	152	12.4	126.0	52.7[c]

[a] Diameter at end of test for HI tests, start of test for VI tests.

[b] All cladding Zircaloy-2 except HI-5 (Zr-4).

[c] Included two end caps on bottom end, second end cap with mounting pin.

Table 3.2 Fission product and plutonium inventories in HI and VI tests

Test No.	Amount in fuel (mg)[a]									
	Kr	Sr	Mo	Sb	Te	I[a]	Cs	Ba	Eu	Pu
HI-1	36	79	341	2.3	51	21.7	252	168	14	1038
HI-2	35	78	337	2.3	50	21.4	249	166	14	1023
HI-3	32	71	305	2.1	46	19.4	226	150	13	927
HI-4	26	59	268	2.3	39	17.3	206	116	9	1401
HI-5	39	90	364	2.9	53	22.5	287	166	18	927
HI-6	46	106	454	3.4	67	20.4	371	210	23	1123
VI-1	41	94	386	3.1	57	23.9	304	177	19	962
VI-2	39	88	327	1.6	43	17.2	244	162	12	608
VI-3	37	84	309	1.5	40	16.1	232	152	11	583
VI-4	242	97	337	1.4	41	16.1	257	159	10	491
VI-5	37	83	308	1.5	40	16.0	231	152	11	581
VI-6	37	84	310	1.5	40	16.2	233	153	11	599
VI-7	46	106	454	3.4	67	28.2	371	210	23	1123

[a] Calculated by ORIGEN-2, Reference 44. Mass of ^{129}I reduced to 0.82 times ORIGEN-2 value.

Table 3.3 Additional fuel inventory

Test No.	Amount in fuel (mg)[a]						
	Ru	Ce	La	Np	Pu	Am	Cm
HI-6	321	322	164	64	1123	63	8.48
VI-1	267	277	140	57	962	32	5.70
VI-2	195	236	120	49	608	26	0.79
VI-3	192	223	119	48	601	26	0.78
VI-4	186	243	125	41	491	16	0.19
VI-5	181	222	114	44	581	23	0.59
VI-7	321	322	164	64	1123	63	8.48

[a] Calculated with ORIGEN-2, Reference 44.

4 Release Amounts and Analysis Techniques

4.1 Release Amounts

The amounts of fission products, uranium, plutonium, zirconium, and tin (from cladding) released in the HI and VI tests are listed in Tables 4.1 and 4.2. Release amounts for other tests are given in Appendix A.

For the VI series tests, the release amounts are given for three time periods corresponding to the use of collection systems A, B, and C. Except for krypton and iodine, a significant amount of every released species deposited in the outlet end of the furnace before the effluent gas stream was divided to form the A, B, and C collection systems. A calculation was used to divide the furnace-deposited material into the A, B, and C time periods according to the amounts of each species actually found in the A, B, and C collection systems. The time of release of vapor and aerosol forms was calculated separately. Material deposited in the furnace on the upper furnace tube and the outlet end ceramic spacers were assumed to be vapor forms that had condensed at these locations. Species deposited in the thermal gradient tubes were also assumed to be vapor forms. All other deposits on the collection system components were assumed to be aerosols. Small deposits occurred on insulating material at the top of the furnace. These were assumed to be mixed vapor and aerosol forms resulting from leaks between ceramic components in the furnace. The ratio of vapor-to-aerosol at this location was assumed to be the same as that for the total collected in the three collection systems. The vapor and aerosol amounts on the furnace insulating materials were then partitioned among the A, B, and C collection trains according to the amounts of vapor and aerosol deposits in each train.

Relatively large amounts of the less-volatile released species were usually found in the upper parts of the furnace before the exit gas stream was divided into the A, B, and C collection systems. This was especially noticed for ruthenium and europium. Slightly less than half of the released Ba, Sr, Mo, U, and Pu were typically found in the furnace above the fuel rod. Twenty-five percent or more of the released antimony was also found in the upper furnace parts. As stated above, we believe that these deposits resulted primarily from the condensation of vapor species. The deposition velocity for aerosol particles is low enough that they would be carried on into the collection systems.

Users of the data given in this report should keep in mind that the amounts, rates, and diffusion coefficients given in this report are for release from fuel. The rapid deposition of many species that occurred in our test apparatus means that similar behavior can occur in real accident situations. In most cases, release from the core of a reactor will be much less than release from the fuel for the less volatile species.

Tests VI-4, ST-1, and ST-2 experienced an unusually large deposition in the upper furnace region for released species. This resulted from the very large axial temperature gradient that occurred in ST-1 and ST-2 and that was deliberately imposed in Test VI-4. In Test VI-4, 48% of the released cesium, 86% of the released antimony, and almost 97% of the released europium deposited in the cool upper furnace region. Large deposits in the common furnace region decrease the accuracy of measurement of time of release. The unusually low gas velocity in Test ST-2 further decreased the amount of released species that reached the sequentially operated collection systems.

4.2 Analysis Techniques

Our most accurate method for measuring fission product release is the radiation counting of gamma-emitting isotopes. The releases of ^{137}Cs, ^{134}Cs, ^{125}Sb, ^{103}Ru, ^{154}Eu, ^{144}Ce, and ^{85}Kr were measured by this method. Beginning with Test HI-3, most of these isotopes were measured in the fuel before and after the test in order to obtain a direct measurement of isotope release. The isotope ^{85}Kr could not be detected in the fuel; hence, it was necessary to use an ORIGEN-2[44] calculation for the original ^{85}Kr fuel inventory. The isotope ^{110m}Ag could be measured with the fresher fuel. As fuel aged, ^{110m}Ag, and ^{144}Ce could not be reliably measured.

Iodine release was measured by neutron activation of ^{129}I. The original fuel inventory for ^{129}I was obtained from ORIGEN-2 calculations that were reduced to 0.82 of the calculation. This decrease in calculated ^{129}I inventory was based on a review of 18 different comparisons between ORIGEN-2 calculations and actual measurements of ^{129}I in various fuel samples.[45] A report from the United Kingdom also suggested that the true inventory of

Table 4.1 Release during HI and VI tests

Test No.	Temperature (K)	Effective time (min)	Amount released (% of inventory)						
			^{85}Kr	^{134}Cs	^{129}I	Te (appx.)	^{125}Sb	Sn (clad) (appx.)	^{110m}Ag (appx.)
HI-1	1675	33.19	2.83	1.75	2.48	~0.25	>0.02		~0.3
HI-2	2000	20.23	51.5	50.50	65.6	~0.5	1.55		2.9
HI-3	2275	21.80	59.0	58.80	43.2	~0.3	>0.001	~1.7	>0.015
HI-4	2200	19.91	21.1	31.70	30.1	<0.4	>0.009	~1.1	>0.094
HI-5	2025	23.05	15.8	20.30	27.3		>0.33	~0.5	18.0
HI-6	2100	9.33	29.6	33.10	30.1		0.068		6.0
VI-1A	2000	24.67		28.62	23.2		0.14		14.0
VI-1B	2240	14.00		17.44	11.4		10.68		15.8
VI-1C	2300	16.15		17.17	10.6		22.34		53.7
Total	2300	27.37	46.0	63.23	45.3		33.16		83.5
VI-2A	2150	9.76		31.24	20.6		0.09	11.0	
VI-2B	2298	18.01		26.20	15.9		49.0	60.0	
VI-2C	2302	41.82		9.71	3.5		19.0	23.0	
Total				67.15	40.0		68.1	94.0	
VI-3A	2000	24.12	26.2	22.10	24.2	1.7	~0.1	6.1	
VI-3B	2530	19.53	66.2	65.74	50.8	68.8	80.6	49.3	
VI-3C	2704	22.86	7.03	12.03	4.0	28.3	18.6	21.1	
Total	2700	31.22	99.5	99.87	79.0	98.8	99.2	76.5	
VI-4A	2290	8.06	85.2	59.03	45.2		1.46	0.5	
VI-4B	2431	8.01	6.5	26.19	13.9		3.26	0.05	
VI-4C	2445	13.66	2.7	10.65	27.7		1.64	0.08	
Total	2440	24.64	94.4	95.87	86.8		6.36	0.63	
VI-5A	2005	23.23	31.4	30.85	33.5	3.2	0.18		
VI-5B	2515	7.20	57.8	59.68	34.0	25.0	9.84		
VI-5C	2720	21.38	10.8	9.43	2.7	53.3	8.04		
Total	2700	26.46	100.0	99.96	70.2	81.5	17.88		
VI-6A	2150	13.22	42.0	47.0	30.1	5.0	0		
VI-6B	2311	18.01	16.9	17.1	7.1	4.0	0.6		
VI-6C	2314	43.03	16.4	21.1	39.5	54.4	63.5		
Total			75.3	80.2	76.7	63.4	63.5		
VI-7A	2000	28.3	39	40.8	23		0		
VI-7B	2300	10.1	14	11.8	8		5.0		
VI-7C	2300	23.6	12	18.8	11		47.0		
Total	2300	30.0	69	71.4	42		52.0		

Table 4.2. Release during HI and VI tests

Test No.	Amount Released (% of Inventory)									
	Ba (appx.)	Sr (appx.)	^{154}Eu	^{106}Ru	Mo (appx.)	UO_2	Pu	^{144}Ce	Zr (clad) (appx.)	La (appx.)
HI-1	~0.008	<0.002				<0.00002			~0.006	~0.023
HI-2					~5.9				~0.002	
HI-3									~0.00008	<0.0002
HI-4	<0.04	<0.005	<0.6			<0.00004				
HI-5	~0.08		~0.021						~0.0016	
HI-6										
VI-1A					13.2	0.0001				
VI-1B					6.2	0.0076				
VI-1C					23.6	0.0013				
Total					43.0	0.0089				
VI-2A	1.89				4.6	0.00092	0.0003			
VI-2B	14.09				55.6	0.00720	0.0012			
VI-2C	2.96				25.4	0.023	0.0016			
Total	18.94				85.6	0.0311	0.0031			
VI-3A	3.2	0.7		0	7.6	0.0016				
VI-3B	16.1	1.3		0.13	53.1	0.548				
VI-3C	10.4	0.7		4.88	16.3	0.784				
Total	29.7	2.7	<0.01	5.01	77.0	1.334	0.005	<0.2		
VI-4A	7.2		18.26		0.8					
VI-4B	11.4		0.28		5.1					
VI-4C	8.2		0.09		1.0					
Total	26.8		18.63	0	6.9		0.09			
VI-5A	10.1	5.8	0.85		0.51	0.0015				
VI-5B	28.4	14.1	35.03		0.23	0.0034				
VI-5C	37.2	13.8	21.47		1.53	0.0098				
Total	75.7	33.7	57.35	0	2.26	0.0147		2.0		
VI-6A	10.5	1.57	2.4		0.46	0.0003				
VI-6B	7.9	1.14	0.1		0.24	0.0002				
VI-6C	14.0	3.11	11.2		11.91	0.0005				
Total	32.4	5.82	13.7		12.61	0.0010				
VI-7A	1.5	0.3								
VI-7B	0.8	0.2								
VI-7C	1.8	0.6								
Total	4.1	1.2	0.045	<2.8						

^{129}I was only ~80% of that calculated by ORIGEN.[46] This correction to ORIGEN-2 inventories has not been used previously in our iodine release calculations.

Components of the test apparatus received two leaches with fresh strong basic solutions for ~2 h each for the iodine analyses. Two similar leaches with nitric acid were used to leach other materials from the apparatus surfaces. The acid leaches were analyzed for U, Mo, Te, Ba, Sr, and Pu. For Tests HI-1 through VI-4, surface smears of the TGTs and filters were taken before leaching for analysis by spark source mass spectrometry (SSMS). The accuracy of the SSMS technique is reported to be +100%/−50%. Beginning with Test VI-3, most of the above elements were analyzed by the inductively coupled plasma (ICP) technique.

5 Release Rate and Diffusion Coefficient Models

There are two types of simple fission product release models used for severe accident release modeling. The first type can be described as "fractional release rate" models. In these models, the release rate coefficient, k, has the units (1/time). For low release rates, k is essentially equal to fraction released per unit time, where "fraction" refers to the inventory at the time in question, which is not necessarily the initial fission product inventory. The time unit "minute" is usually used because it is easily related to the time scale of both fission product release tests and severe accidents. The model is easily understood when the value of k, min^{-1}, is equated with the fraction released per minute. Details of this type of model are given in Section 5.1.

The second type of simple release model is the diffusion model. This model uses the classical single-atom diffusion equations and diffusion coefficient that were in use long before nuclear fission was discovered. The diffusion model was commonly used to correlate fission gas release from trace-irradiated and low-burnup fuel at moderate temperatures.[47] An ANSI standard was developed using this type model for in-reactor fission gas release, including the release of short half-lived species.[48,49] In 1985, we found that the diffusion equations created an excellent fit of fission product release at severe accident temperatures. Take Nakamura, a visiting scientist at ORNL from JAERI, quantified the results of ORNL tests. This has been called the ORNL Diffusion Release Model.[26,50] Diffusion release models are often referred to as Booth models after the AECL scientist who formulated simplified forms of diffusion equations for fission product release modeling.[51]

5.1 Release Rate Coefficients

5.1.1 Description of Fractional Release Rate Models

As explained above, the fractional release type of model uses release rate coefficients, k (min^{-1}), to express the release rates. The first models of this type were used by Albrecht et al. (1978) and later by Wichner and Lorenz in NUREG-0772.[52] A computerized version of this model was called CORSOR.[53] The most commonly used version of this type of model is CORSOR-M.[53] CORSOR-M uses a 1/T correlation to obtain values of k as a function of temperature. The release rate coefficients calculated for fission gases, cesium, and iodine by this method are essentially the same as those calculated by the NUREG-0772 curves and by CORSOR. The values of the release rate coefficient, k, for the less-volatile species in CORSOR-M were changed significantly from those in CORSOR and the NUREG-0772 report.

In addition to being easy to understand and easy to use (see below), fractional release rate coefficients are usable with any geometry. Geometries, such as large molten pools[54] or large debris beds,[55] can affect fission product release rates. Major geometry differences should be accommodated. Recommendations for fuel, structural material, and control rod material releases that may be controlled by gas flow rate (vaporization) were made in a review of the CORSOR model.

A weakness of the fractional release rate models (NUREG-0772, CORSOR, and CORSOR-M) is that they do not account for the effect of fuel morphology on fission product release. Examples of possible importance are burnup and grain size. A weakness suspected initially[52] is that the release calculated by this method overestimated release rates and releases for long heating times or large fractional releases. This was proven to be the case when ORNL Test VI-2 was performed.[28] The measured value of k for cesium release changed from 0.056 min^{-1} when 2300 K was first reached to ~0.003 min^{-1} 60 min later after 63% of the initial cesium was released.

5.1.2 Using Fractional Release Rate Models

In order to use a fractional release rate model, the first step is to determine the value of the release rate coefficient, k, for the fission product of interest. CORSOR-M uses the correlation

$$k = k_o \, e^{-\left(\frac{Q}{RT}\right)} , \tag{5.1}$$

where

k = release rate coefficient, min^{-1};

k_o = (min^{-1}) is obtained from Table 3 of Reference 53;

Q = activation energy, kcal/mol, from Table 3 of Reference 53;

R = universal gas constant, 0.001987 kcal/mol·K;

T = temperature, K.

CORSOR release rate coefficients can also be obtained from Reference 53, and the NUREG-0772 release rate coefficients are shown in Figure 4.3 of Reference 52.

5.1.2.1 "Sum of Products" Method

The next step in using fractional release rate models is to calculate the fraction of inventory released using

$$F = 1 - e^{-kt} , \qquad (5.2)$$

where

F = the fraction of original inventory released in time t;

t = time, min;

k = release rate coefficient, min^{-1}, obtained as described above.

This equation assumes that k is constant, which happens only when the temperature is constant. In all practical applications, the temperature changes with time and, therefore, the value of k changes with time. For this situation, Equation (5.2) can be used if the sum of the products $k \cdot \Delta t$ is substituted for (kt) in Equation (5.2). The time interval $\Delta t = 1$ min is satisfactory for most applications, but Δt would be the time-step value in most computer calculations.

As an example, suppose one wants to use CORSOR-M to predict the total release of cesium from UO_2 fuel heated for 100 min at 1500 K and 10 min at 2000 K. For simplicity, the heatup and cooldown periods are neglected. From Reference 52 and Equation (5.1), we find that $k = 1.01 \times 10^{-4}$ at 1500 K and $k = 0.002132$ at 2000 K. The product $k\Delta t$ at 1500 K is $(1.01 \times 10^{-4})(100) = 0.0101$, and the product $k\Delta t$ at 2000 K is $(0.02132)(10) = 0.2132$. The sum of the products is 0.2233. Substituting this "sum of products" for kt in Equation (5.2), a value of 0.200 is obtained for F. This is the total fraction of original cesium expected to be released at the end of 110 min of heating, as described above. If time steps of 1 min are used throughout, a minute-by-minute release prediction would be obtained. At the end of 1 min, the product $k\Delta t$ would be 0.000101, at 2 min the sum of the products $k\Delta t$ would be 0.000202, etc. At the end of 110 min, the sum of the products would be 0.2233. At each minute, the new "sum of products" would be used in Equation (5.2) to calculate the fraction of original inventory expected to be released by that time.

5.1.2.2 "Inventory Reduction" Method

In this method, Equation (5.2) is used without summing the "kt" products. We will rewrite it as $f = 1 - e^{-kt}$, where f = fraction of current inventory. At each time step, the value of $k\Delta t$ for that time step alone is used in Equation (5.3). The inventory must then be reduced by the amount released in the time step, since f refers to the current inventory and not the original inventory.

As an example, we consider the heating of UO_2 at 1500 K with CORSOR-M release rate for cesium of $1.01 \times 10^{-4}\ \text{min}^{-1}$. This method is easier to follow if the inventory is traced by mass or g-atoms so that the fraction of original inventory and fraction of current inventory are not confused. Assume that the initial inventory is 100 mg Cs and that each time step is 1 min. At the end of 1 min at 1500 K (the first time step), kt (or $k\Delta t$) = 1.0×10^{-4}, $f = 1.00995 \times 10^{-4}$, and the mass of cesium released would be 1.00995×10^{-2} mg. The inventory remaining (or current inventory) becomes 99.9899 mg. The second time step would have identical values of kt (or $k\Delta t$) and f. The mass of cesium released is $(1.00995 \times 10^{-2} \times 99.9899) = 0.01009848$ mg, so that the remaining inventory (or current inventory) becomes 99.9798 mg. The procedure would be continued with each time step requiring a new calculation of the current inventory.

5.1.3 Obtaining "k" from Test Data

The value of k, the release rate coefficient, can be obtained reasonably accurately from a variation of Equation (5.1) if heatup and cooldown times were short in comparison to the time at a constant temperature:

$$k = \frac{-\ln(1 - F)}{t} , \qquad \textbf{(5.4)}$$

where k, F, and t are as defined for Equation (5.2).

In most tests, we are able to measure the releases of cesium and krypton every minute by on-line gamma counting. Release rate coefficients can be calculated from this type of data using the following equation:

$$k = -\ln[1 - (F_2 - F_1)/(1 - F_1)]/(t_2 - t_1)$$

where

F_1 = fraction of original inventory released at start of time period,

F_2 = fraction of original inventory released at end of time period,

t_1 = time corresponding to F_1,

t_2 = time corresponding to F_2.

The time interval $(t_2 - t_1)$ is usually 1 min if the gamma counting statistics are good. The fractional release data must be smoothed in order to avoid a large scatter in k values. F_1, F_2, t_1, and t_2 and the temperature all must correspond to the midpoint of the counting period when on-line gamma counting data are used. Equation (5.5) is valid for any time period as long as F_1 and F_2 are known. For example, Equation (5.5) could be used to calculate the overall release rate coefficient for a particular temperature plateau.

If the temperature changes much during a time period, Equation (5.5) is not very helpful because it does not provide the temperature that corresponds to the calculated k. An example is when our fission product collection system B is used during the temperature ramp between a low- and a high-temperature plateau. For a case like this, one must assume that the effect of temperature on k is known. Equation (5.1) with Q = 63,800 cal/mol is used at ORNL for all species.

The following is a description of the calculation procedure. A computer worksheet or spreadsheet program is used to list the input and perform the calculations.

1. Assume a value for Q in Equation (5.1). A value of Q = 63,800 cal/mol has been used at ORNL for this type of calculation. The results of this report (Section 5.2) show that a value of Q = 55,000 cal/mol would be preferable for future calculations of this type. The selected value of Q must remain constant throughout the procedure.

2. Select a starting value for k_o of Equation (5.1). This will be changed by trial and error until a satisfactory fit with test results is achieved.

3. List the measured fuel temperatures with time.

4. For each time step (time between temperature measurements), calculate the average temperature, Δt, k, the product $k\Delta t$, and the sum of the products $k\Delta t$ from the start of the test.

5. Calculate F at each time step using Equation (5.2), and substitute the sum of products $k\Delta t$ for "kt" in Equation (5.2).

6. Continue this calculation to the end of the test.

7. If the final calculated F does not agree with the measured F, try different values of k_o until satisfactory agreement is obtained. This calculation is performed with the fractional release of cesium (rather than ^{85}Kr) since our measurements for cesium are usually more accurate.

8. Using the best-fit k_o, the Q assumed in step 1, and Equation (5.1), k is calculated for the temperature most characteristic of the test. This temperature is usually the average temperature of the test plateau. If there is not a well-defined temperature plateau in the test, the temperature for the midpoint of cesium release is used. That is, half of the cesium release

occurred below this temperature and half occurred above this temperature. These values of T appear in Table 4.1.

9. It is helpful to calculate an "effective time" for each test that corresponds to the F, T, and k of steps 7 and 8. To do this, a variation of Equations (5.2) and (5.3) is used:

   $$t_{eff} = \frac{-\ln(1 - F)}{k} . \qquad (5.5)$$

   Note that both k and t_{eff} depend on the characteristic or reference temperature, T.

10. The effective time, t_{eff}, from Equation (5.5) can be used to calculate the value of k for any other element or isotope. Simply substitute t_{eff} for t in Equation (5.4) and use the total fraction released for the species of interest. Note that t_{eff} was calculated with the value of Q in step 1. A moderate change in Q usually produces only a small change in t_{eff}. At ORNL, a value for $Q = 63{,}800$ cal/mol has been used for this type of calculation. Based on the results of this summary report (Section 5.2), a value of $Q = 55{,}000$ cal/mol is preferable. The values for effective time listed in Table 4.1 were obtained using the diffusion equations described in Section 5.3.2. The two methods for calculating effective times give similar results.

11. The VI series tests used sequentially operated collection systems A, B, and C. Steps 1-7 above describe the procedure for the system A results. For the system B, the same steps are repeated, but the final k_o for system A is kept constant and the k_o for B is changed by trial and error. The sum of the products $k\Delta t$ is continued so that F at each time step is the total release from the start of the test. The calculation for system C is performed in the same way. The k_o values for A and B are fixed, and the k_o value for C is changed until a satisfactory agreement with F is obtained.

12. The effective time, t_{eff}, for systems A and C are calculated in the same method as for system A. In many cases, it is desired to know the overall A+B+C effective time. The individual t_{eff} values cannot be added because they refer to different temperatures and use different k_o values. To obtain the correct overall effective time and k value, steps 1-7 must be repeated clear to the end of the test using the same k_o all the way through. The overall t_{eff} can then be calculated using Equation (5.5).

5.2 CORSOR-O (Recommended Fractional Release Rate Model)

CORSOR-O (-O for "ORNL") consists of a set of release rate coefficients to be used with the "fractional release rate model" described in Section 5.1. CORSOR-O uses a single-temperature activation energy (55,000 cal/mol) that is used for every species. The values for K_o are different and sometimes depend on the extent of cladding oxidation or the gas atmosphere. This concept was first suggested in a review of CORSOR in 1985.[56]

The K_o value for Kr and Cs in CORSOR-O is 12,000 min^{-1}. Every other species has a release rate multiplier, C_k, so that the k_o for each fission product may be derived from data given in Table 5.1. The values of C_k were chosen to provide a good fit to the experimental results given in Section 6.

5.3 Diffusion Coefficients

5.3.1 Description of Diffusion Release Models

Two diffusion release models are featured in this report. ORNL-Booth, new with this report, replaces the ORNL Diffusion Release Model.[26,50] ORNL-Booth includes diffusion coefficients for a wide range of fission products, whereas the ORNL Diffusion Release Model was structured only for the volatile fission products. ORNL-Booth also includes diffusion coefficients for structural components (Zr, Sn in the cladding, Fe, Cr, Mn, UO_2) whose releases are obviously not controlled by the grain size of the fuel. For these species, the grain radius is assumed to be 6 μm.

Table 5.1 Relative release rates and diffusion coefficients

Element			CORSOR-O relative rate (R_K)	ORNL-Booth relative "D" (R_D)	CORSOR-BOOTH scale factor (SFACT)[a]
Initial fuel condition	Reduced fuel	Oxidized fuel or cladding			
Kr, Cs	Kr, Cs	Kr, Cs	1.0	1.0	2.0
I	I	I, Te (oc)[b]	0.8	0.64	1.6
		Sb (oc)	0.5	0.25	1.0
Ag[c]	Ag	Ag, Sn-clad (oc)	0.4	0.16	0.8
		Mo	0.25	0.0625	0.5
	Ba		0.2	0.04	0.4
	Sr, Eu	Mn[c]	0.1	0.01	0.2
Ba, Te, Sb, Sn-clad	Te, Sb, Sn-clad	Ba, Ru, Fe,[c] Cr,[c] Co,[c]	0.02	4.0×10^{-4}	0.04
Sr, Mo	Mo	Sr	0.01	1.0×10^{-4}	0.02
	Ce		0.002	4.0×10^{-6}	0.004
UO_2[c]		UO_2	0.0006	3.6×10^{-7}	0.0012
Zr-clad,[c] Ce, La	Zr-clad, La	Zr-clad, Ce, La	0.0002	4.0×10^{-8}	0.0004
Eu	UO_2	Eu	0.00006	3.6×10^{-9}	0.00012
Ru	Ru		0.00004	1.6×10^{-9}	0.00008
Pu	Pu	Pu	0.00002	4.0×10^{-10}	0.00004

[a] This is the "scale factor" (SFACT) in CORSOR-BOOTH where $D = 0.0025\, e^{(-91,100/1.99/T)}$ for the cesium high-burnup range.

[b] oc = oxidized cladding.

[c] Use 0.0006 cm for "grain radius" for all non-fission-product diffusion coefficient calculations. Use high-burnup correlation regardless of fuel burnup.

The second diffusion model is CORSOR-BOOTH. ORNL-Booth is structured so that scale factors for the two models can be exchanged easily. Additional information about CORSOR-BOOTH is given in Section 5.5.

5.3.2 Using Diffusion Release Models

The diffusion release models are almost as easy to use as the fractional release rate models. One main difference is that the diffusion coefficient cannot be directly linked with a quantitative release rate (fraction/min) as can be done with release rate coefficients.

The first step in using diffusion release models is to calculate the diffusion coefficient. All diffusion models use a 1/T correlation similar to that in Equation (5.1) for release rate coefficients:

$$D = D_o e^{-\left(\frac{Q}{RT}\right)},$$

where

D = diffusion coefficient, cm^2/s;

D_o = multiplier, cm^2/s;

Q = activation energy, cal/mol;

R = universal gas constant, 1.987, cal/mol·K;

T = temperature, K.

The values of D_o and Q for the ORNL Diffusion Release Model are given in References 26 and 50. This model is being replaced by the ORNL-Booth model described in Section 5.4.

The next step is to calculate the fraction released. Any valid mathematical expression for single-atom diffusion can be used. The Booth approximations are convenient to use.[51] This requires two equations to cover the entire release range.

For small fractional releases, when $Dt/a^2 \leq 0.1547$,

$$F = 6\sqrt{\frac{Dt}{\pi a^2}} - 3\,\frac{Dt}{a^2}, \tag{5.7}$$

where

F = fraction released;

D = diffusion coefficient from Equation (5.6), cm^2/s;

t = time, s;

a = measured initial grain radius, cm.

At ORNL, the initial grain radius, a, is used throughout the calculations even though the grain size will actually grow at high temperature. We assume that the values of D_o and Q in Equation (5.6) include the grain growth effect, just as the values of D_o and Q must include the effects of other release mechanisms.

For higher release fractions where $Dt/a^a > 0.1547$, the following equation is used.

$$F = 1 - \frac{6}{\pi^2}\exp\left[\frac{-\pi^2 Dt}{a^2}\right]. \tag{5.8}$$

Equations (5.7) and (5.8) approach each other but do not become identical. The closest point is when Dt/a^2 = 0.1547, where Equation (5.7) = 0.8673, and Equation (5.8) = 0.8679.

When the temperature changes during a test or accident sequence, the value of D changes as from Equation (5.6). This can be accounted for by summing the product $D\Delta t$ for each time step and substituting the sum of the products in Equations (5.7) and (5.8) in a manner similar to that for release rates in Section 5.1.2.1. In the literature, the symbol D′ is often used for the quantity D/a^2.

5.3.3 Obtaining "D" from Test Data

The procedure used at ORNL to calculate diffusion coefficients is very similar to the procedure for calculating "k" that is described in Section 5.1.2.3. A value of Q (or Q/R) is selected to use in Equation (5.6). A value of Q/R = 45,779 is recommended, which is the value for Q/R used in CORSOR-BOOTH. Slightly different values of Q/R are used in the FORTRAN provided with CORSOR-BOOTH and in CORSOR-BOOTH used in MELCOR. D_o is varied by trial and error until Equation (5.7) [or (5.8)] results in a value of F that is equal to the test value. The sum of products $D\Delta t$ must be substituted for Dt in these equations. This is the procedure used at ORNL to determine the best values of D for cesium for the A, B, and C time periods. The effective times, t_{eff}, for periods A, B, and C are calculated by dividing the best-fit D into the sum Dt for each time period.

The diffusion coefficients for other released species are determined by first calculating the values of Dt/a^2 at the ends of A, B, and C operation for each species using the following equations:

$$Dt / a^2 = \frac{2}{\pi} - \frac{f}{3} - 2\sqrt{\frac{1}{\pi^2} - \frac{f}{3\pi}} \text{ for } f < 0.85 \qquad (5.9)$$

or

$$Dt / a^2 = -\frac{1}{\pi^2} \ln\left[\frac{\pi^2(1-f)}{6}\right] \text{ for } f > 0.85\,, \qquad (5.10)$$

where f is the total release from the start of the test measured at the end of time periods A, B, and C.

To calculate D for time period A, divide the value for Dt/a^2 calculated from Equation (5.9) or (5.10) by the quantity t_{eff}/a^2, where t_{eff} is the effective time as calculated above for time period A. To calculate D for time period B, the value of Dt/a^2 for time period A must be subtracted from Dt/a^2 calculated from Equation (5.9) or (5.10) using the total fraction released at the end of time period B. Then divide this difference in Dt/a^2 by t_{eff}/a^2, where t_{eff} is the effective time calculated previously for time period B. To calculate D for time period C, use the same procedure but subtract Dt/a^2 for the end of time period B (f = combined releases during A+B operation) from Dt/a^2 calculated, where f = total release for the test. Divide the difference in Dt/a^2 by t_{eff}/a^2, where t_{eff} is the effective time for period C operation.

5.4 ORNL-Booth (Recommended Diffusion Release Rate Model)

The ORNL-Booth model is a refinement and expansion of the earlier ORNL Diffusion Release model.[26] The temperature (1/T) activation energy is the same as used in the CORSOR-BOOTH fission product release model (see Section 5.5). CORSOR-BOOTH used an approximate average of the 1/T activation energies used for the high- and low-burnup correlations in the old ORNL Diffusion Release Model. The new ORNL-Booth model uses the CORSOR-BOOTH 1/T slope for all fission products and other materials since it appeared to be valid within the data error band. A possible exception is the release of material by pure vaporization, such as may occur with UO_2.

ORNL-Booth shares the release rate scale factors with CORSOR-O except that it is necessary to square the "linear" CORSOR-O scale factors for ORNL-Booth in order to obtain the same relative releases. Releases calculated by the two models are not always proportional because release with time is calculated differently by the two models. The D_o for ORNL-Booth is 0.01 cm^2/s, Q is 91,100 cal/mol, and R = 1.99 cal/ mol·K. We use the same value of Q and R as used in the CORSOR-BOOTH model. Using these numbers in Equation (5.6) and adding the relative diffusion coefficient multiplier for each element, R_o from Table 5.1, the ORNL-Booth Model becomes

$$D = 0.01\ R_D\ e^{(-45,779/T)}\,.$$

5.5 CORSOR-BOOTH

The scale factors determined for CORSOR-O can be used to modify the CORSOR-BOOTH computer program. CORSOR-BOOTH is one of the optional fission product release models in the MELCOR Radionuclide (RN) package of MELCOR Version 1.8.2. It has also been given a separate limited distri-bution.[*] CORSOR-BOOTH is a diffusion release model that uses a single-temperature activation energy with scaling factors (release rate multipliers) for each fission product and UO_2.

CORSOR-BOOTH can be modified using the correlations given in this report. The CORSOR-O scale factors (column 2 of Table 5.1) can be substituted directly for the scale factors in CORSOR-BOOTH if the D_o in CORSOR-BOOTH is changed from 0.0025 to 0.01. If it is desired to keep the D_o value of 0.0025 in CORSOR-BOOTH, the scale factors in column 5 of Table 5.1 should be used. Either of these changes will make CORSOR-BOOTH give the same results as ORNL-Booth.

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[*] M. Ramamurthi and M. R. Kuhlman, "Refinement of CORSOR — An Empirical In-Vessel Fission Product Release Model," October 31, 1990 (personal communication).

It is not easy to change the scale factors in CORSOR-BOOTH because many of them are written into various computer program statements. Also, some additional statements must be added in order to include additional dependencies on atmosphere or cladding oxidation.

The CORSOR-BOOTH scale factors are linear with fractional release rate as in CORSOR-O because the scale factors are applied to a release rate that is calculated after the diffusion release computation.

6 Experimental Release Rate and Diffusion Coefficients

Experimental results are displayed graphically and discussed in this section. The release rate coefficients and diffusion coefficients calculated for the ORNL test are listed in Tables 6.1–6.3. The results from PBF SFD 1-3, PBF SFD 1-4, and TMI-2[57] are shown for reference only. Their large temperature gradients and uncertain time at temperature (especially TMI-2) make it impossible for their results to contribute to selection of the best scale factor or release correlation time. We used the method of Osetek to obtain release rates and diffusion coefficients for these three events.[58] He used the time above 2200 K and the release from the bundle region (PBF SFD) or core (TMI-2).

The release rates from these three events should be comparable to release rates calculated for full-scale reactor accidents. The comparisons between small system test results and the PBF/TMI-2 results are useful in judging whether reducing atmospheres or oxidizing atmospheres exerted more control of fission product release in the large configuration events.

6.1 Krypton and Cesium

Release rate and diffusion coefficients for ORNL tests are shown in Figures 6.1 and 6.2 (Kr) and 6.3 and 6.4 (Cs). Results for cesium release from other tests are shown in Figures 6.5 and 6.6. At temperatures above 1700 K, the krypton and cesium release rates are essentially identical. At lower temperatures, the release of krypton is faster than cesium, but this can be ignored for severe accidents. The first release of krypton is "plenum" gas released when the cladding first fails. A slower low-temperature release is from gas believed to be shallowly embedded in fuel and cladding surfaces. Typically, this release is 1 to 2% of the fuel inventory and occurs in the temperature range 500–900 K.[4]

The percent release of cesium is less than that of iodine at temperatures below 1600 K. The lower release of cesium is usually attributed to the formation of cesium uranates in this temperature range.[2,4,59]

6.2 Iodine

The release rates and diffusion coefficients for iodine are shown in Figures 6.7 and 6.8 for ORNL test data and in Figures 6.9 and 6.10 for results from other tests. There is more scatter in the iodine results than in the krypton and cesium results. This scatter undoubtedly is related to the method used to measure iodine release in the HI and VI tests. The ^{129}I analysis requires that the apparatus surfaces must be leached thoroughly and the ^{129}I separated from ^{137}Cs and other fission products in preparation for neutron activation to form and measure ^{130}I. Two basic leaches (both ammonium hydroxide with added hydrogen peroxide), usually 2 h each, were combined for the ^{129}I analysis. Early tests with this leaching procedure showed that on the order of 95% of the ^{129}I was successfully removed from the apparatus surfaces. No correction was made for the lack of complete dissolution of ^{129}I.

Another possible error in the determination of ^{129}I release is that we depend on ORIGEN-2[44] to determine the correct total inventory in the fuel. An investigation of the accuracy of ORIGEN-2 compared with experimental results showed that for 18 such comparisons, the medium result was that actual ^{129}I was 82% of that calculated by ORIGEN-2.[45] Only one comparison resulted in ORIGEN-2 being lower than what was found. Scientists from the United Kingdom believed that ORIGEN-2 was using an ^{129}I yield that was too high.[46] For this report (but not for any earlier ORNL report), we have reduced the ^{129}I initial inventory to 82% of the ORIGEN-2 value.

As illustrated in Figures 6.7 and 6.8, the iodine release rates dropped precipitously at the highest temperatures. Such behavior is not realistic and probably results from incomplete leaching of all of the apparatus surfaces, loss of iodine during the time prior to leaching, while the leach solutions were stored before analysis, or inadequacies with the analytical procedure. Long delays occurred with the leaching and analysis for ^{129}I in Tests VI-4, VI-5, and, especially, VI-3. This resulted from the nuclear reactor used for activation analysis being shut down for ~2 years and from priority schedules within our own NRC program and within the ORNL Analytical Chemistry Division. We believe that we were not completely successful in measuring all of the

Table 6.1 Release rate coefficients for HI and VI tests

Test No.	Release rate coefficient (min^{-1})													
	^{85}Kr	^{134}Cs	^{129}I	Te (appx.)	^{125}Sb	Sn (clad) (appx.)	Ba (appx.)	Sr (appx.)	^{154}Eu	Mo (appx.)	^{106}Ru	UO$_2$	^{144}Ce	^{110m}Ag (appx.)
HI-1	8.6-4	5.3-4	7.6-4	7.5-5			2.3-6	<6.0-7		4.4-7		6.0-9		9.1-5
HI-2	3.6-2	3.5-2	5.3-2	3.4-2	7.7-4					3.0-3				1.4-3
HI-3	4.1-2	4.1-2	2.6-2	1.4-4	4.6-5	8.0-4	2.7-4			2.7-4				>6.9-6
HI-4	1.2-2	1.9-2	1.8-2	2.0-4	4.5-4	5.3-4	2.2-5	<2.5-6		9.4-5		2.0-8		>4.7-5
HI-5	7.5-3	9.8-3	1.4-2		1.4-4	2.2-4	3.3-5	<3.0-4		6.0-5				8.6-3
HI-6	3.8-2	4.3-2	3.8-2		6.6-3			9.1-6						6.6-3
VI-1A		1.4-2	1.1-2		5.7-5					5.7-3		4.1-8		6.1-3
VI-1B		2.0-2	1.1-2		8.1-3					5.3-3		5.4-6		1.4-2
VI-1C		2.4-2	1.1-2		1.8-2					2.1-2		7.4-7		9.0-2
VI-1ABC	2.3-2													
VI-2A		3.9-2	2.4-2		9.2-5	1.2-2	2.0-3			4.8-3		9.4-7		
VI-2B		2.7-2	1.2-2		3.7-2	6.3-2	8.6-3			4.9-2		4.0-6		
VI-2C		6.2-3	1.4-3		1.1-2	3.7-2	8.6-4			2.4-2		5.5-6		
VI-3A	1.3-2	1.0-2	1.2-2	7.1-4	4.1-5	2.6-3	1.3-3	2.9-4		3.3-3		6.6-7		
VI-3B	1.2-1	9.5-2	5.7-2	6.2-2	8.4-2	3.8-2	9.3-3	6.7-4		4.4-2	6.7-5	2.8-4		
VI-3C	1.2-1	2.0-1	7.7-3	1.4-1	1.4-1	2.8-2	6.0-3	3.3-4		2.3-2	2.2-3	3.5-4		
VI-3ABC									3.2-6				<6.4-5	
VI-4A	2.4-1	1.1-1	7.5-2		1.8-3	6.2-4	9.3-3		2.5-2	9.8-4				
VI-4B	7.2-2	1.3-1	3.7-2		4.2-3	6.3-5	1.6-2		4.3-4	6.7-3				
VI-4C	2.9-2	9.3-2	8.3-2		1.3-3	5.9-5	7.7-3		8.1-5	7.5-4				
VI-5A	1.6-2	1.6-2	1.8-2	4.3-4	7.8-5		4.5-3	2.6-3	3.7-4	2.2-4		6.5-7		
VI-5B	2.6-1	2.8-1	9.9-2	1.4-3	1.4-2		5.3-3	2.3-2	6.1-2	3.2-4		4.7-6		
VI-5C	3.3-1	2.6-1	4.1-3	1.8-1	4.4-3		4.3-2	8.8-3	1.9-2	7.2-4		4.6-6		
VI-5ABC													7.6-4	
VI-6A	4.1-2	4.8-2	2.7-2	3.9-3	<3.3-4		8.5-3	1.2-3	1.9-3	3.5-4		2.3-7		
VI-6B	1.9-2	1.4-2	5.9-3	2.4-3	3.3-4		5.1-3	6.5-4	5.7-5	1.3-4		1.1-7		
VI6-C		1.7-2	2.3-2		2.3-2		4.4-3	7.6-4	2.8-3	3.0-3		1.2-7		
VI-6C1	3.6-3			0[a]										
VI-6C2	1.8-2			3.6-2										
VI-7A	1.7-2	1.9-2	9.2-3				5.2-4	1.1-4		2.8-5				
VI-7B	2.6-2	2.2-2	1.1-2		4.9-3		2.3-4	2.7-4		1.2-4				
VI-7C	1.8-2	2.1.2	7.2-3		2.9-2		1.8-3	2.4-4		2.2-3				
VI-7ABC									1.5-5					

[a] Assumes entire C-phase release occurred during high steam flow period.

Table 6.2 Diffusion coefficients from HI and VI tests

Test No.	Diffusion coefficient (cm^2/s)					
	^{85}Kr	^{134}Cs	^{129}I	Te (appx.)	^{125}Sb	Sn (clad) (appx.)
HI-1	1.03-15	3.91-16	7.9-16			
HI-2	1.03-12	9.81-13	1.9-12		6.6-16	
HI-3	1.64-12	1.63-12	7.6-13		3.1-18	7.3-15
HI-4	4.00-13	9.68-13	8.6-13		6.5-16	2.9-15
HI-5	3.41-13	5.78-13	1.1-12		1.3-16	5.8-16
HI-6	2.61-12	3.35-12	2.7-12		9.3-14	
VI-1A		1.21-12	7.7-13		2.5-17	
VI-1B		4.18-12	1.9-12		2.7-13	
VI-1C		6.72-12	2.4-12		2.3-12	
VI-1ABC	3.22-12					
VI-2A		6.51-12	2.6-12		4.4-17	7.2-13
VI-2B		1.11-11	3.5-12		9.7-12	2.5-11
VI-2C		3.29-12	4.9-13		5.7-12	2.3-11
VI-3A	1.74-12	1.20-12	1.5-12	6.3-15	<2.2-17	8.4-14
VI-3B	6.29-11	4.86-11	2.6-11	2.3-11	3.6-11	1.2-11
VI-3C	7.21-11	1.20-10	4.7-12	8.5-11	8.6-11	1.5-11
VI-4A	1.07-10	3.46-11	1.8-11		1.4-14	1.6-15
VI-4B	4.39-11	7.25-11	1.7-11		1.4-13	3.4-16
VI-4C	1.75-11	5.67-11	4.8-11		7.3-14	3.6-16
VI-5A	2.69-12	2.58-12	3.1-12	2.3-15	<7.3-17	
VI-5B	1.37-10	1.49-10	4.6-11	2.2-14	7.4-13	
VI-5C	1.99-10	1.55-10	2.3-12	9.7-11	6.2-13	
VI-6A	9.14-12	1.19-11	4.3-12	1.0-13	<1.1-15	
VI-6B	8.68-12	6.72-12	1.9-12	2.5-13	<1.1-15	
VI-6C		9.30-12	1.2-11	7.8-12	7.9-12	
VI-6C$_2$	1.86-12			0[a]	0[a]	
VI-6C$_2$	1.12-11			1.3-11	1.4-11	
VI-7A	1.60-12	1.77-12	4.9-13			
VI-7B	4.84-12	4.17-12	1.2-12		5.5-14	
VI-7C	4.07-12	5.00-12	1.1-12		3.7-12	

[a] Assumes that entire C-phase release occurred during high steam flow period.

Table 6.3 Additional diffusion coefficients from HI and VI tests

Test No.	Ba (appx.)	Sr (appx.)	^{154}Eu	Diffusion coefficient (cm^2/s)							
				Mo (appx.)	^{106}Ru	UO$_2$	Pu	^{144}Ce	^{110m}Ag (appx.)	Zr (appx.)	La (appx.)
HI-1	7.6-21	5.1-22		2.7-22		5.1-26			1.1-17	4.6-21	6.7-20
HI-2				9.8-15					2.3-15	6.1-21	
HI-3	1.1-16			1.1-16					6.9-20	9.0-24	
HI-4	1.5-18	2.0-20		2.8-17		1.3-24			7.0-18		<3.1-23
HI-5	7.2-18			2.4-17					4.5-13	3.4-21	
HI-6									9.2-14		
VI-1B				2.3-13		1.2-23			2.6-13		
VI-1B				5.1-13		1.3-19			1.9-12		
VI-1C				3.9-12		3.8-20			2.7-11		
VI-2A	1.9-14			1.2-13		4.5-21	4.8-21				
VI-2B	8.0-13			1.6-11		1.9-19	6.3-21				
VI-2C	1.5-13			1.4-11		1.1-18	9.2-21				
VI-3A	2.3-14	1.0-15		1.3-13		5.6-21					
VI-3B	1.1-12	9.5-15		1.5-11	4.5-17	8.1-16					
VI-3C	1.5-12	8.0-15		1.3-11	5.9-14	3.4-15					
VI-3ABC			<1.7-19					<6.7-17			
VI-4A	3.6-13		2.4-12	4.1-15							
VI-4B	2.2-12		7.9-14	2.3-13							
VI-4C	1.8-12		1.5-14	5.0-14							
VI-5A	2.4-13	7.8-14	1.6-15	5.9-16		5.1-21					
VI-5B	1.3-11	3.0-12	1.2-11	4.0-15		1.6-19					
VI-5C	2.2-11	2.3-12	8.2-12	1.1-14		4.7-19					
VI-5ABC								8.0-15			
VI-6A	4.7-13	9.8-15	2.4-14	8.4-16		3.6-22					
VI-6B	7.5-13	1.4-14	1.5-15	8.1-16		4.7-22					
VI-6C	1.1-12	3.4-14	2.4-13	2.1-13		9.1-22					
VI6-C$_2$				3.6-13							
VI-7A	1.8-15	8.4-17		5.3-18							
VI-7B	7.1-15	5.7-16		9.2-17							
VI-7C	1.1-13	9.9-16		2.9-14							
VI-7ABC			3.4-18								

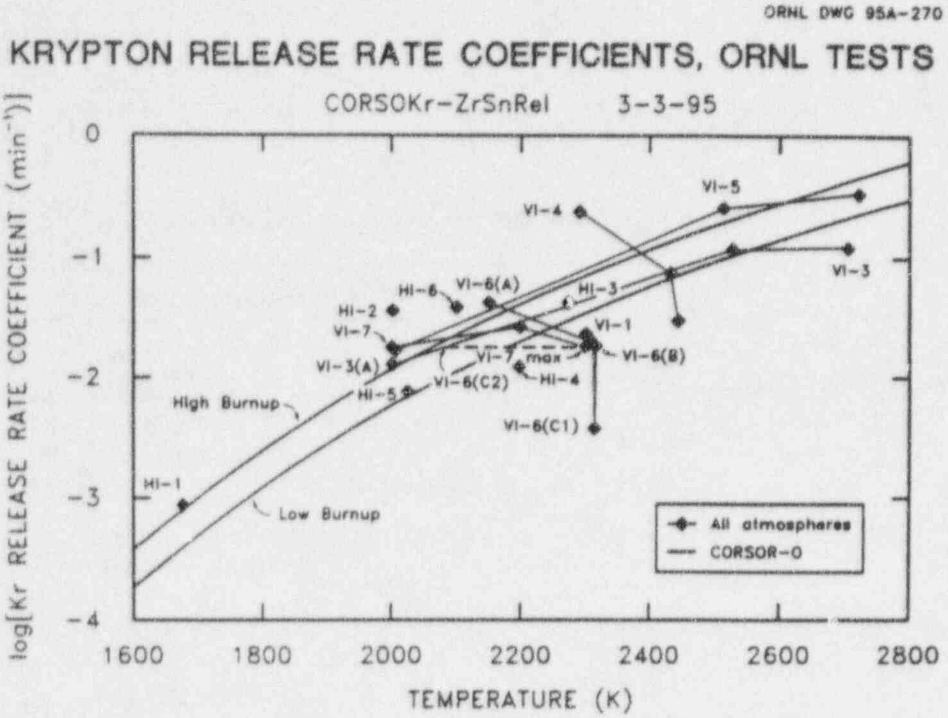

Figure 6.1 Release rate coefficients for krypton, ORNL tests

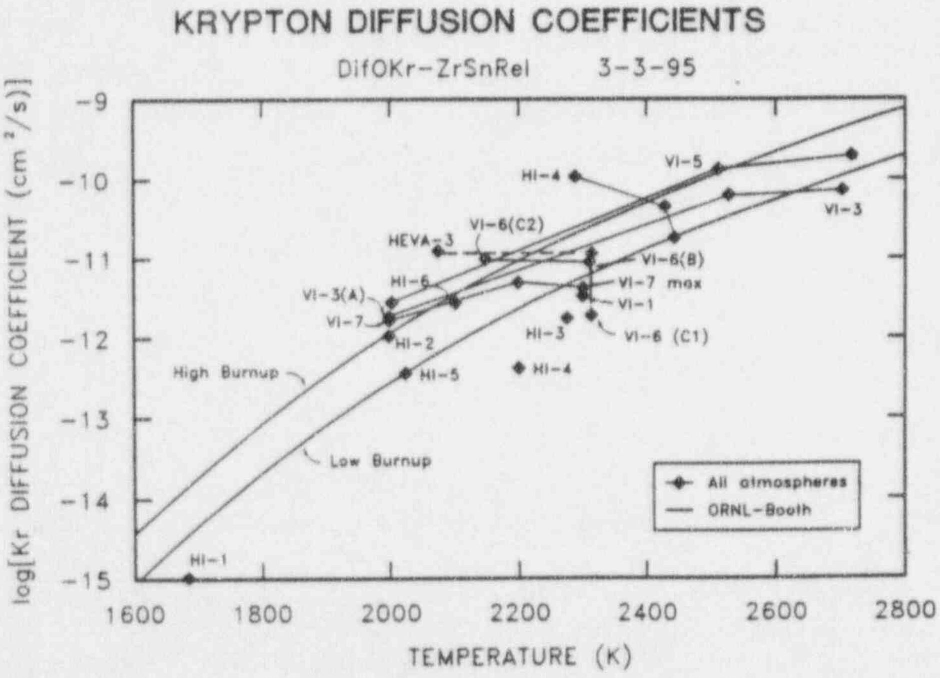

Figure 6.2 Diffusion coefficients for krypton, ORNL tests

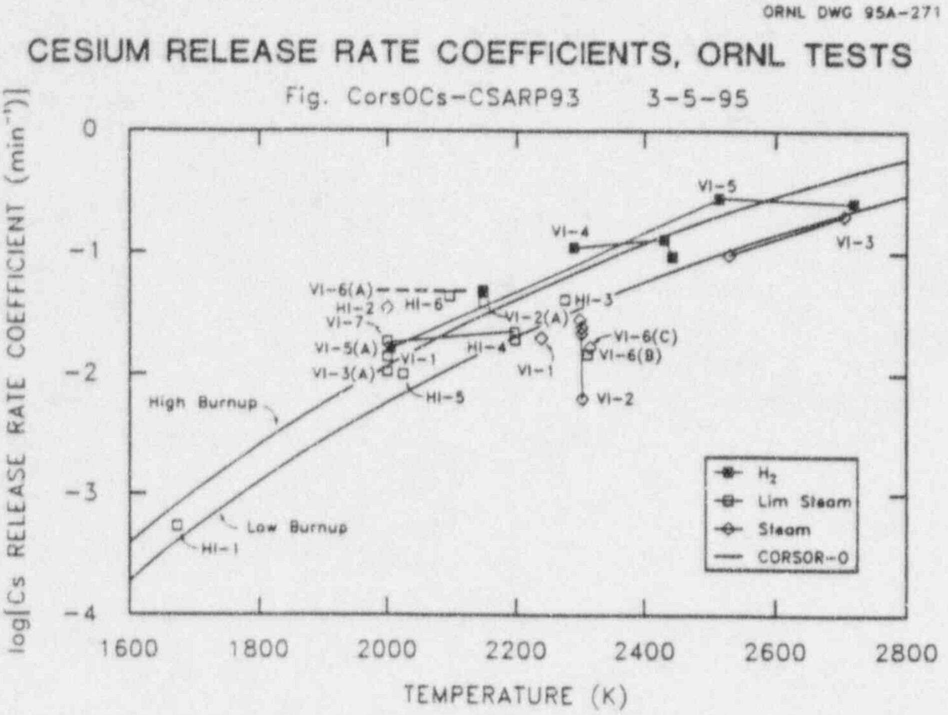

Figure 6.3 Release rate coefficients for cesium, ORNL tests

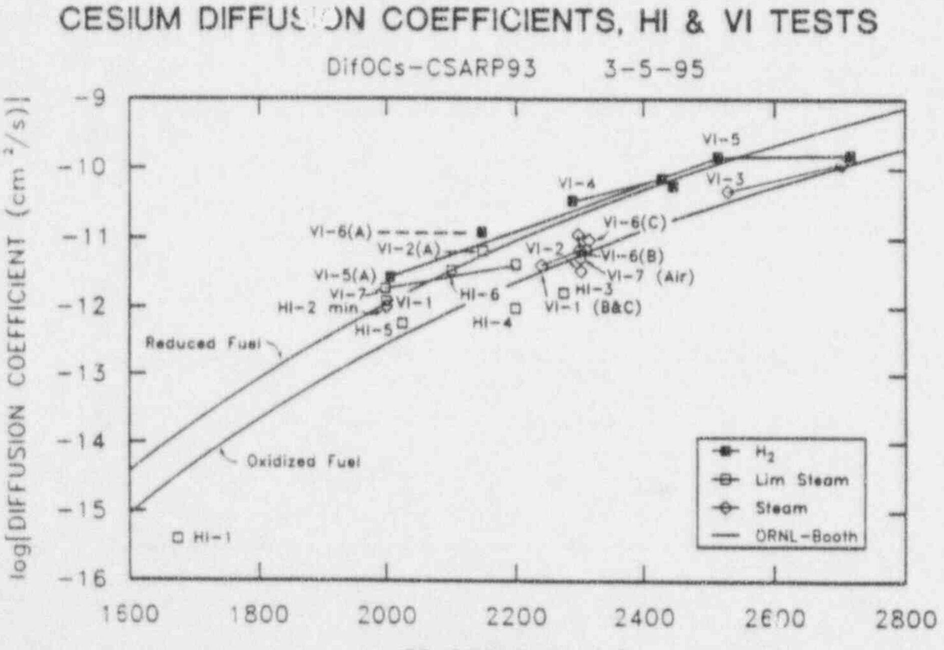

Figure 6.4 Diffusion coefficients for cesium, ORNL tests

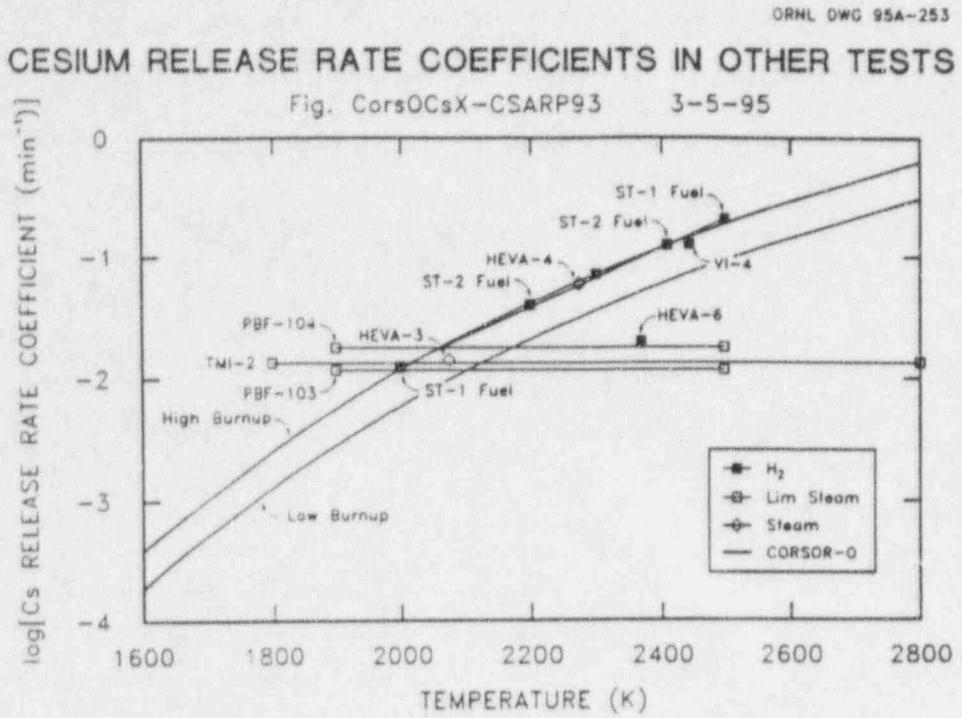

Figure 6.5 Release rate coefficients for cesium, other tests

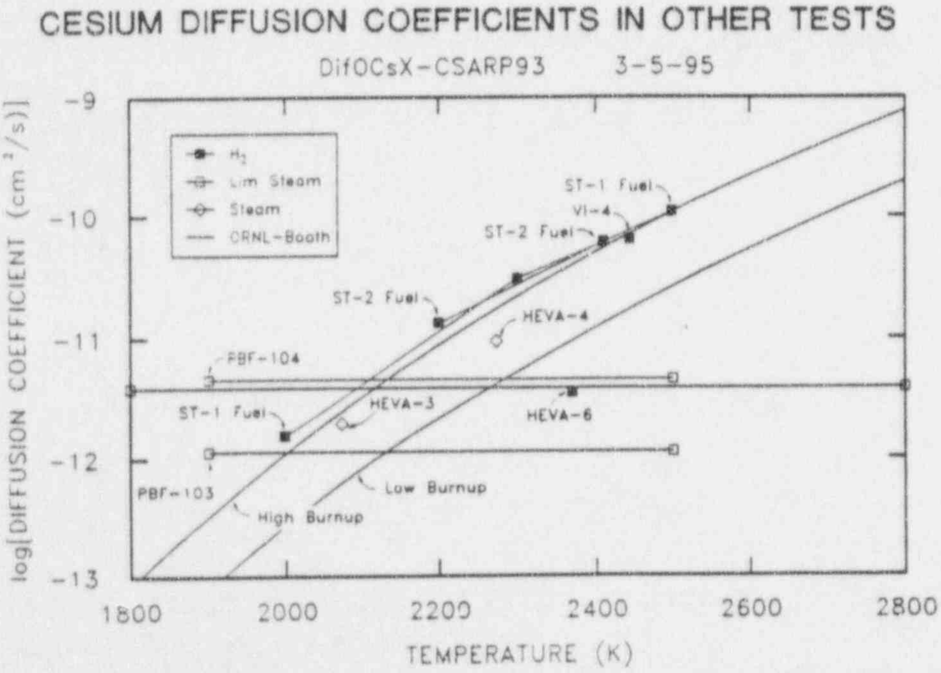

Figure 6.6 Diffusion rate coefficients for cesium, other tests

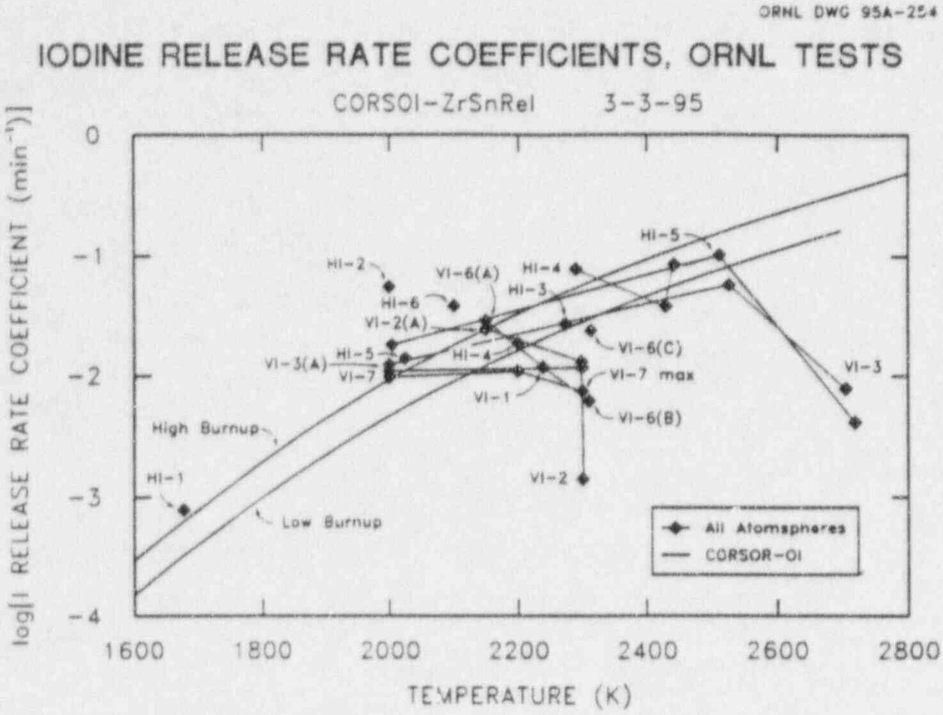

Figure 6.7 Release rate coefficients for iodine, ORNL tests

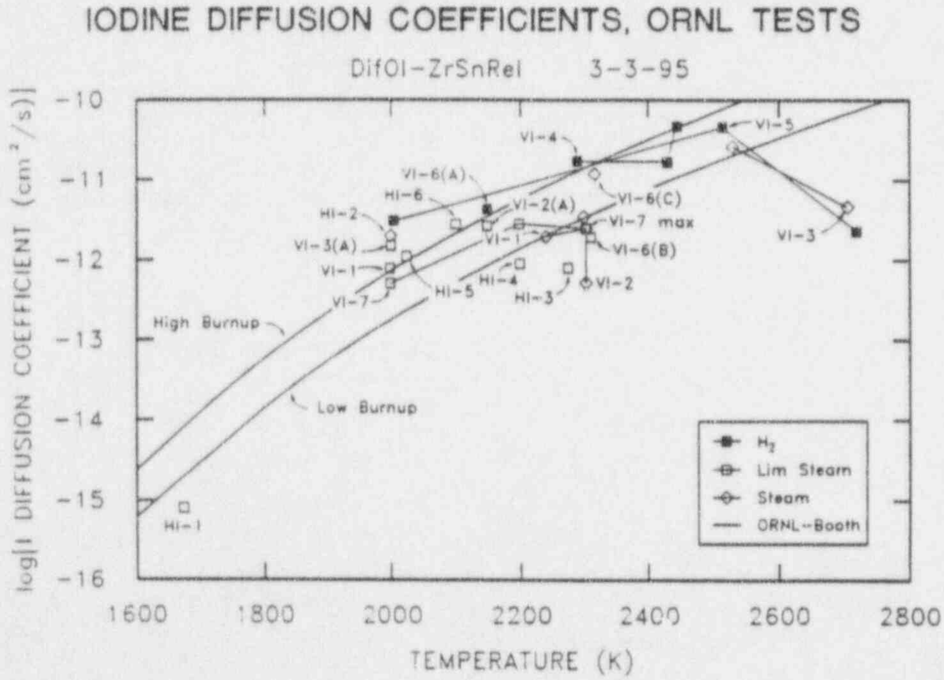

Figure 6.8 Diffusion rate coefficients for iodine, ORNL tests

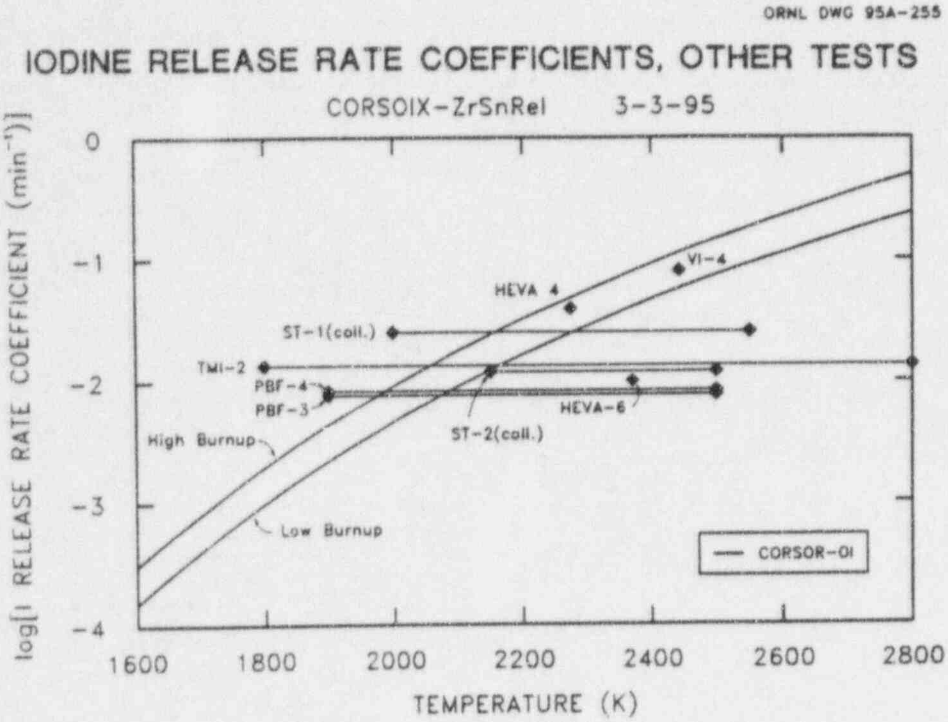

Figure 6.9 Release rate coefficients for iodine, other tests

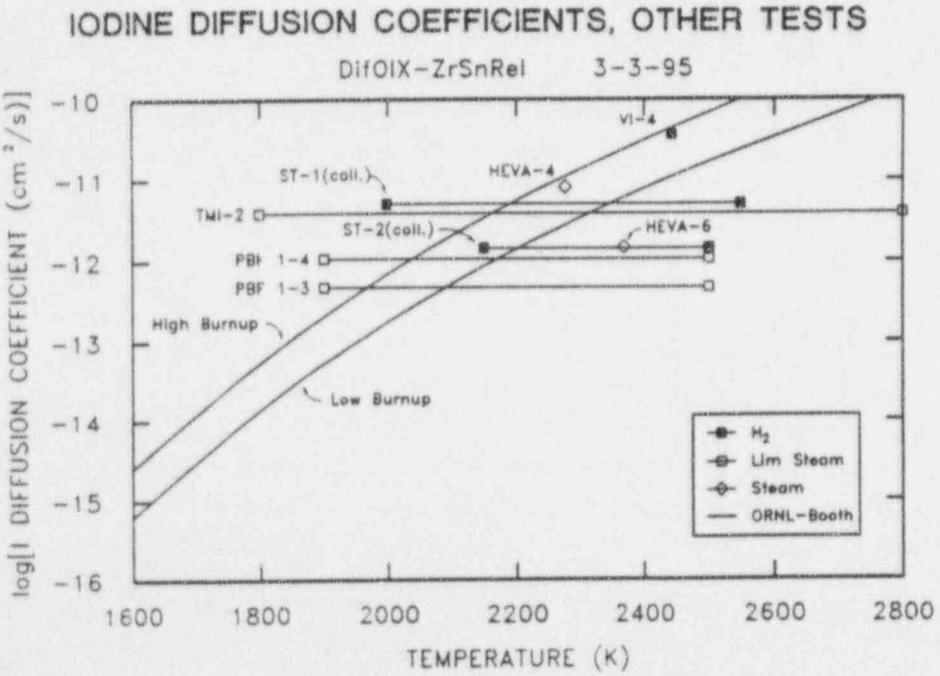

Figure 6.10 Diffusion coefficients for iodine, other tests

^{129}I that was released in Tests VI-3, 4, and 5, and that is the reason for what appear to be low release rates in these tests. The possibility does exist that iodine release rates are actually somewhat lower than those for krypton and cesium at very high temperatures. For CORSOR-O and ORNL-Booth, we have selected the C_k multiplier of 0.8 for iodine at all temperatures.

6.3 Tellurium, Antimony, and Tin

6.3.1 Tellurium

In 1983, a serious disagreement existed among the various researchers and users of fission product release rates concerning the true release rate for tellurium. The NUREG-0772 release rates showed lower rates for tellurium than for cesium, iodine, and the fission gases. Some experiments showed that tellurium was released at the same rate as the other "volatiles." The NRC asked ORNL to investigate tellurium behavior in more detail. The result was the finding that tellurium releases were lower in those tests in which Zircaloy cladding was used.[18] An investigation of the chemical behavior of tellurium showed that tellurium could form several chemical species with zirconium or tin (1.5% in Zircaloy) that would significantly reduce the vapor pressure of tellurium. A simple model was proposed in which the release rate of tellurium would be 1/40 that of iodine until the Zircaloy cladding was 90% oxidized and then would be changed to be equal to the iodine release rate.[18]

An early demonstration of the trapping of tellurium by Zircaloy cladding was in Implant Test 3 in which radiotraced TeO_2 coated on the dished ends of unirradiated UO_2 pellets was found to be deposited on the inside of the Zircaloy cladding.[3] The transport half-time was ~30 min at 1173 K and ~4 min at 1573 K. Te_3ZrO_5 was identified in the deposit. Separate tests with Zr and TeO_2 or Te showed that Te and ZrTe could also be formed easily.

A dramatic demonstration of the trapping ability of Zircaloy cladding occurred in Test C-9 at ORNL, a test with radiotraced tellurium placed in an unirradiated Zircaloy-clad UO_2 fuel capsule that contained a drilled hole defect.[19] Tellurium was released at a very low rate at 1975 K until the Zircaloy was essentially completely oxidized (see Figures 6.11 and 6.12 for the C-9, HI, VI, and other test results). In Test C-9, most of the released tellurium deposited in the TGT at a TGT temperature of 875 K. Equal molar quantities of Sn and Te were found in the peak, suggesting that the chemical form was SnTe. Operating conditions for Test C-9 are given in Appendix A. Another very convincing (and even more realistic) test was HEVA 4,[60] a test at Grenoble, France, in which high-burnup fuel was heated after a reirradiation so that ^{131}I and ^{132}Te could be measured continuously. The test showed that iodine and cesium began to be released immediately but the release of tellurium was delayed ~12 min until the cladding was completely oxidized. Other laboratories have also confirmed the trapping of tellurium.

The reason for reviewing the background of tellurium behavior is to show that it is well established. The HI and VI data for tellurium release are not highly precise because of our dependency on SSMS results and, for recent tests, inductively coupled plasma (ICP) analyses. The latter have not given satisfactory results for tellurium, probably because of required dilution of samples to reduce radiation exposure to the analysts. The HI and VI results are only of a qualitative nature, as seen in Figures 6.11 and 6.12. In spite of the scatter of results, it is clear that the previously established tellurium behavior prevails in recent experiments.

6.3.2 Antimony

The release of antimony in the HI and VI tests was obtained by gamma counting for ^{125}Sb. The decay rate for ^{125}Sb is low, especially for the older fuel used in Tests HI-1, 2, 3, and 4. In recent tests, we have used long counting times in order to obtain better data for ^{125}Sb. The results shown in Figures 6.13 and 6.14 demonstrate that antimony behaves much like tellurium except that the release rates are slightly different.

6.3.3 Tin (from Cladding)

Approximately 1.5% of Zircaloy is tin, an element that can be an important aerosol component. Tin was measured in the HI and VI tests by SSMS, a technique that gives only semiquantitative results. Figures 6.15 and 6.16 show tin release rates, and it is easy to see that tin from the cladding behaves in the same manner as fission products tellurium and antimony [i.e., tin release rates are much higher (~10×) after the Zircaloy cladding has been oxidized].

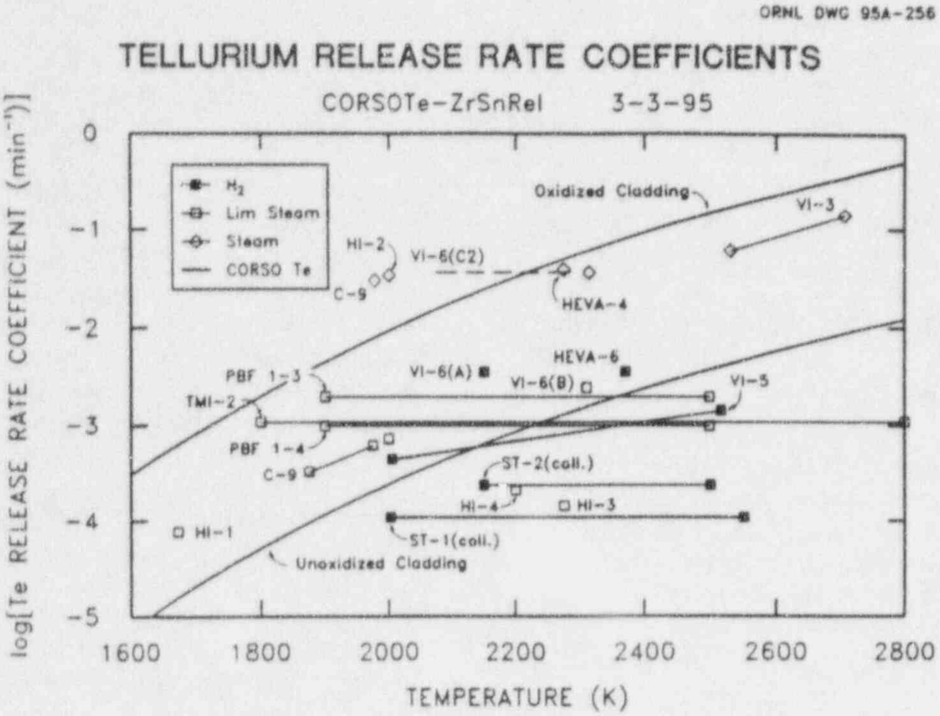

Figure 6.11 Release rate coefficients for tellurium

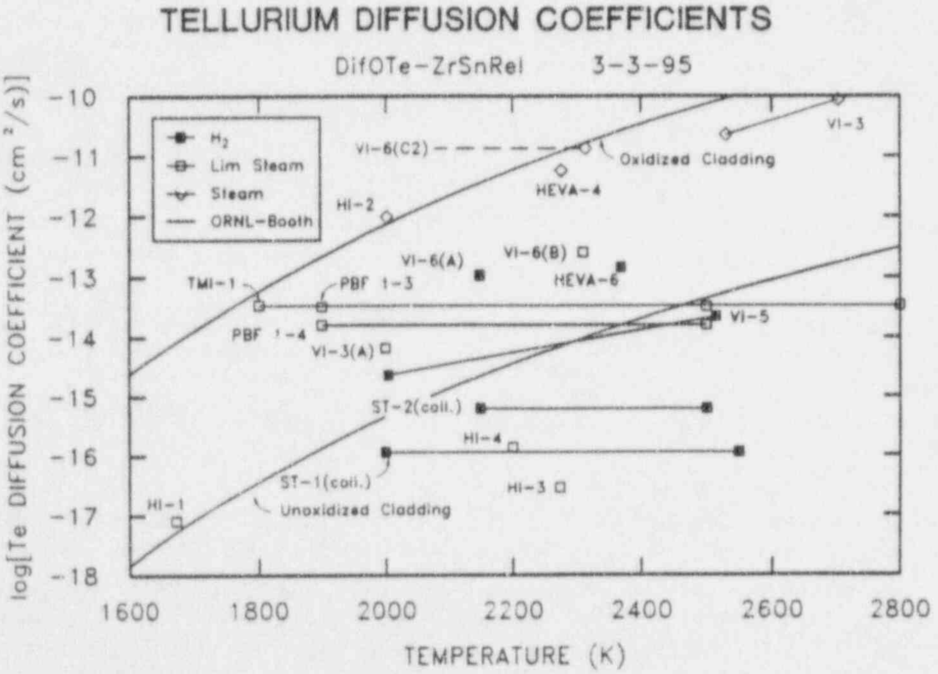

Figure 6.12 Diffusion rate coefficients for tellurium

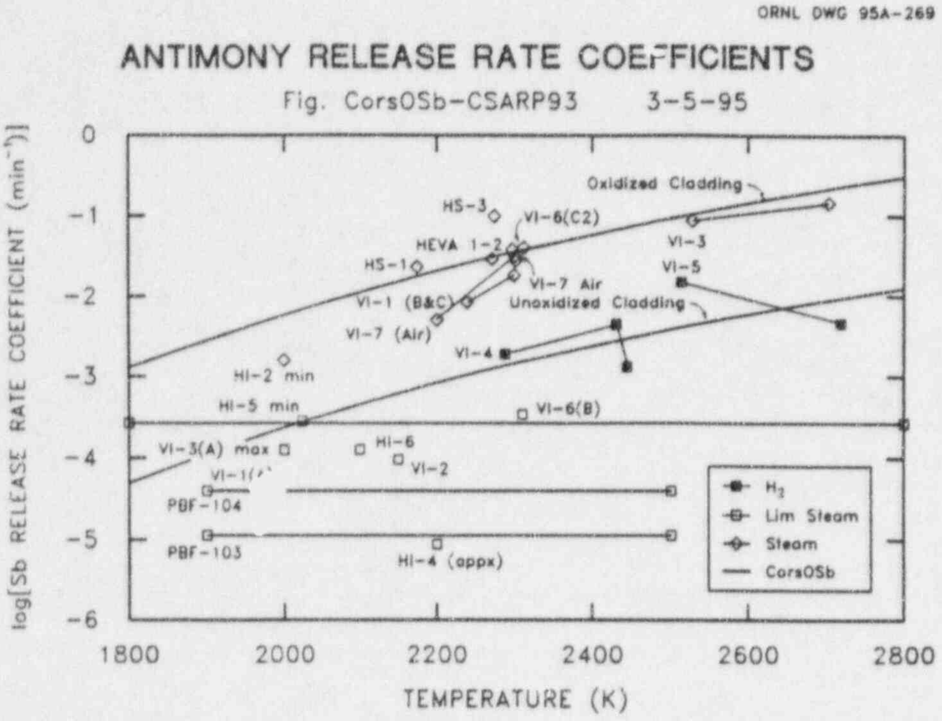

Figure 6.13 Release rate coefficients for antimony

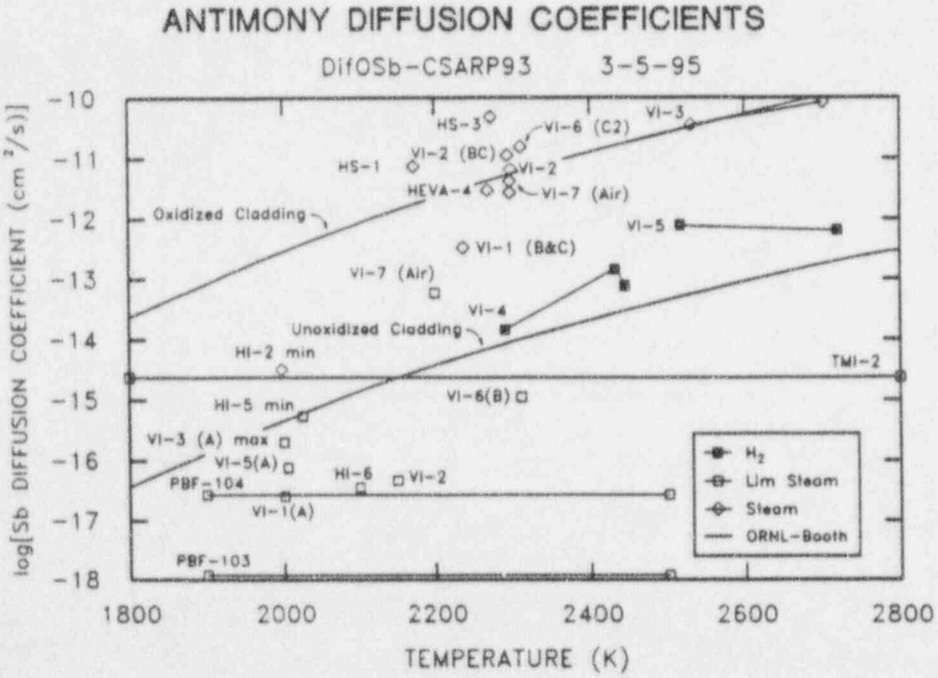

Figure 6.14 Diffusion rate coefficients for antimony

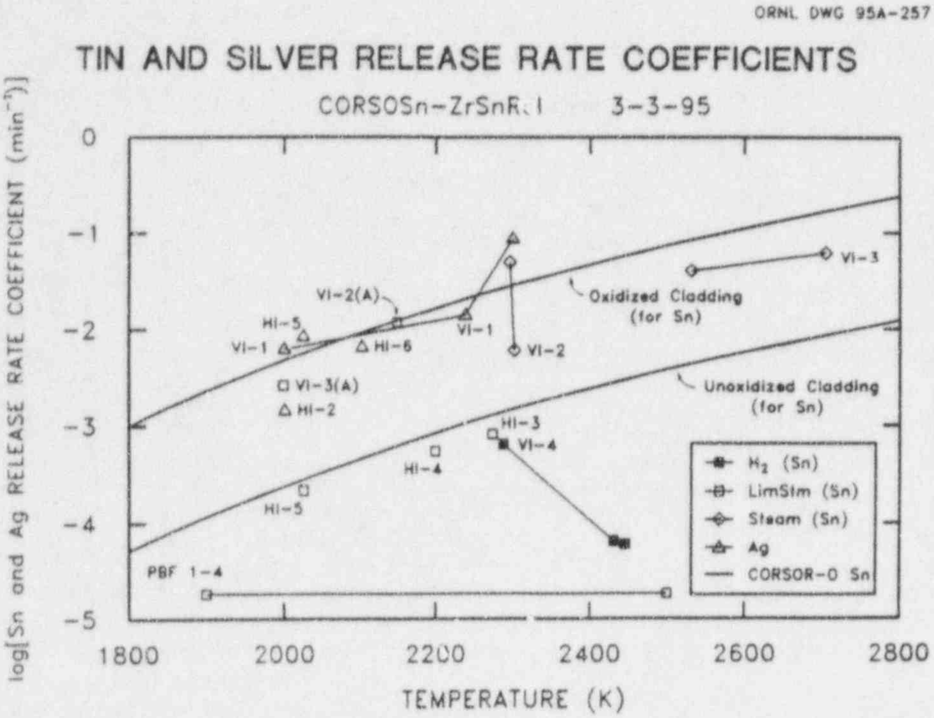

Figure 6.15 Release rate coefficients for tin (cladding)

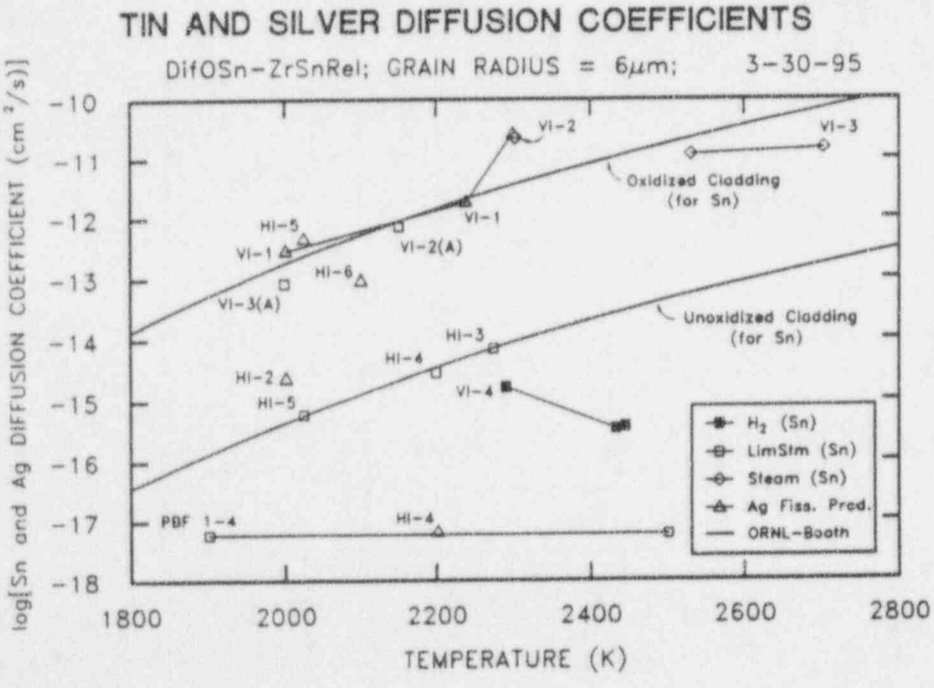

Figure 6.16 Diffusion rate coefficients for tin (cladding)

The release of tin from the cladding does not depend on the characteristics of the UO_2 fuel pellets, but calculation of diffusion coefficients requires that a "grain size" be used. We chose 6 μm for the grain radius for the release of tin for all tests regardless of the UO_2 grain size. The ORNL-Booth diffusion coefficients for all non-fission products (UO_2, Zr, Sn, Fe, Mn, Co, Ag-In, Cd) were determined using an arbitrarily chosen "grain" radius of 6 μm. This value of the grain radius must be used for these species in the diffusion equations regardless of the actual UO_2 grain size.

6.4 Barium and Strontium

The release rates for barium are shown in Figures 6.17 and 6.18 for ORNL test results and Figures 6.19 and 6.20 for results from other tests. Results for strontium are shown in Figures 6.21 and 6.22. Fission products barium and strontium behave similarly as expected from their chemical characteristics. The release of barium is slightly greater than strontium for a wide variety of experimental conditions. Their reduced atomic forms are more volatile than their oxides, whereas the oxides are the forms normally found in the UO_2 fuel. The higher volatility of the reduced forms of barium and strontium was first measured in 1962 when Zircaloy cladding was used on trace-irradiated UO_2 pellets electrically heated by a tungsten resistor rod running lengthwise through the pellets.[61] The releases of barium and strontium were several times greater than when stainless steel cladding or no cladding was used. These same tests were the first to show that the release of Te was reduced by a factor of ~40 when unoxidized Zircaloy cladding was present.

The HI and VI release rates for barium and strontium are scattered because of our dependency on SSMS and the ICP analysis of very dilute solutions. The closeness of barium and strontium release rates and the higher release in the presence of melted Zircaloy is evident. The results from Test VI-3 suggest that the percentage release of barium was 10 times that of strontium. This is counter to the normal behavior, which is that the percentage release of barium is on the order of twice that of strontium.

6.5 Europium and Cerium

6.5.1 Europium

The release of europium is dramatically increased when melted Zircaloy reacts with the fuel and some of the fission products. This is demonstrated in Figures 6.23 and 6.24. The gradual decrease in release rate exhibited by Tests VI-4, 5, and 6 can be interpreted as the result of the melted Zircaloy losing its reducing power. Fission product oxides, and especially oxygen from UO_2, will gradually satisfy the zirconium's need for oxygen.

Released europium behaves mostly as a vapor, with more than 90% of it condensing where the temperature begins to decrease. In Tests VI-4 and VI-5, we estimate that this deposition temperature was ~1800-2200 K. It is, therefore, not certain that the small fraction reaching the sequentially operated collection systems is directly proportional to the amount released from the fuel. This is especially true of Test VI-4 because of the steep axial temperature gradient imposed on the test.

Europium is measured by gamma counting ^{154}Eu. It is important to know that the initial inventory of ^{154}Eu calculated by ORIGEN-2 is almost twice what is actually present. The ST-1 and ST-2 calculations used the ORIGEN-2 inventory. Therefore, we increased the ST-1 and ST-2 amounts deposited in their filter collection systems by a factor of 1.77, the ratio of ORIGEN-2/gamma count that we measured for the VI-4 fuel sample.

6.5.2 Cerium

In the HI and VI tests, cerium is detected by gamma counting ^{144}Ce, which has a half-life of 284 d. The count rate for ^{144}Ce is low in old fuel, and the release rate is very low. The only detected release of cerium was in Test VI-5 in which it was found in small fractions at two locations. Companion steam Test VI-3 was examined for the detection limit, and that value, along with the VI-5 release rate, is shown in Figures 6.25 and 6.26. With these few data, we have selected cerium C_k values for CORSOR-O. The oxidizing release rate is backed up by data from older tests with reirradiated or trace-irradiated

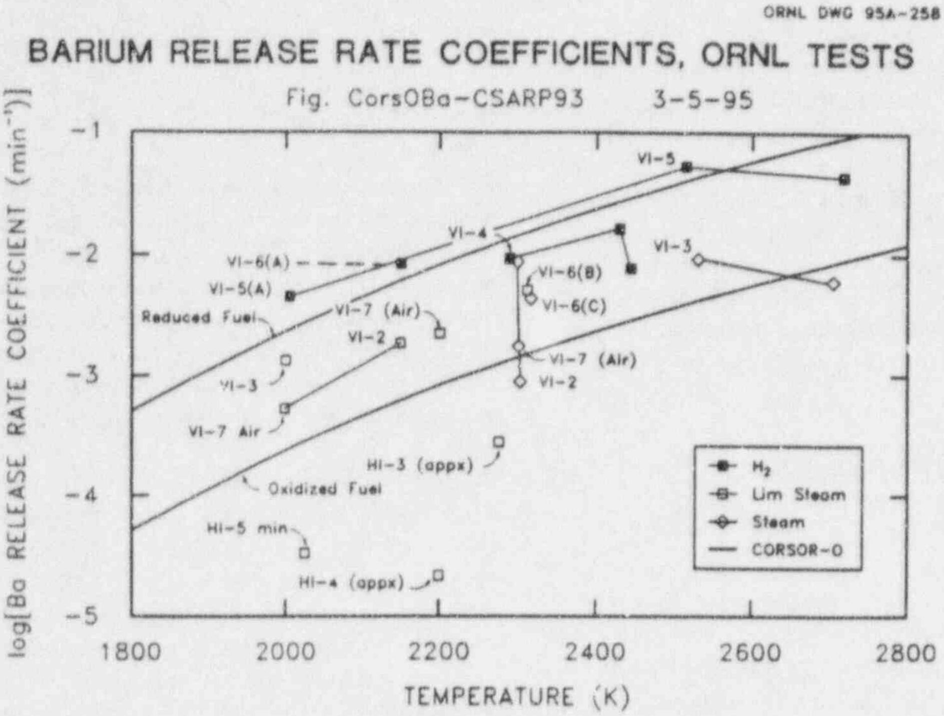

Figure 6.17 Release rate coefficients for barium, ORNL tests

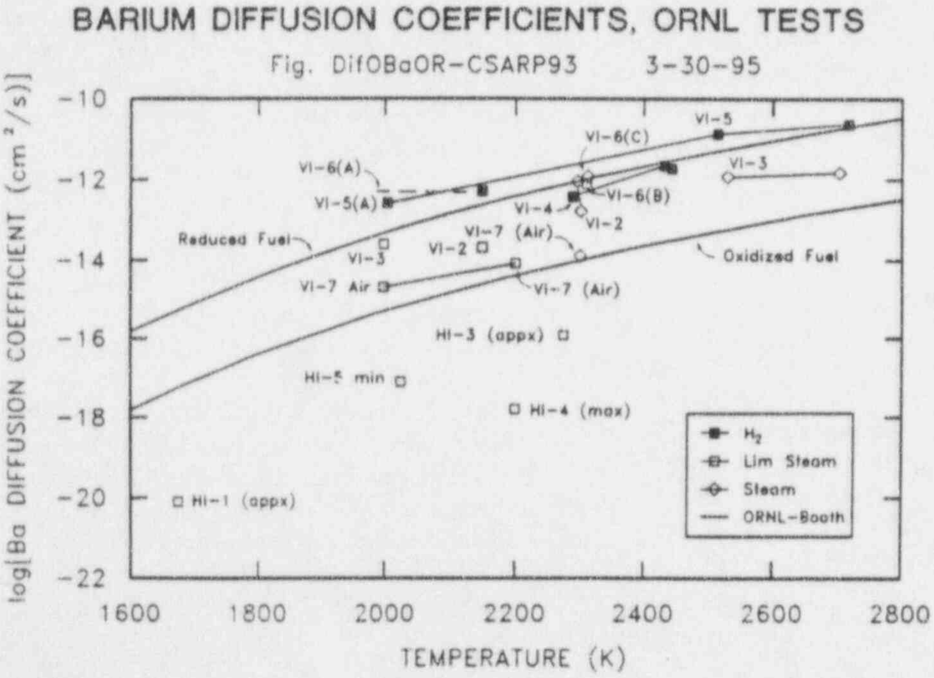

Figure 6.18 Diffusion rate coefficients for barium, ORNL tests

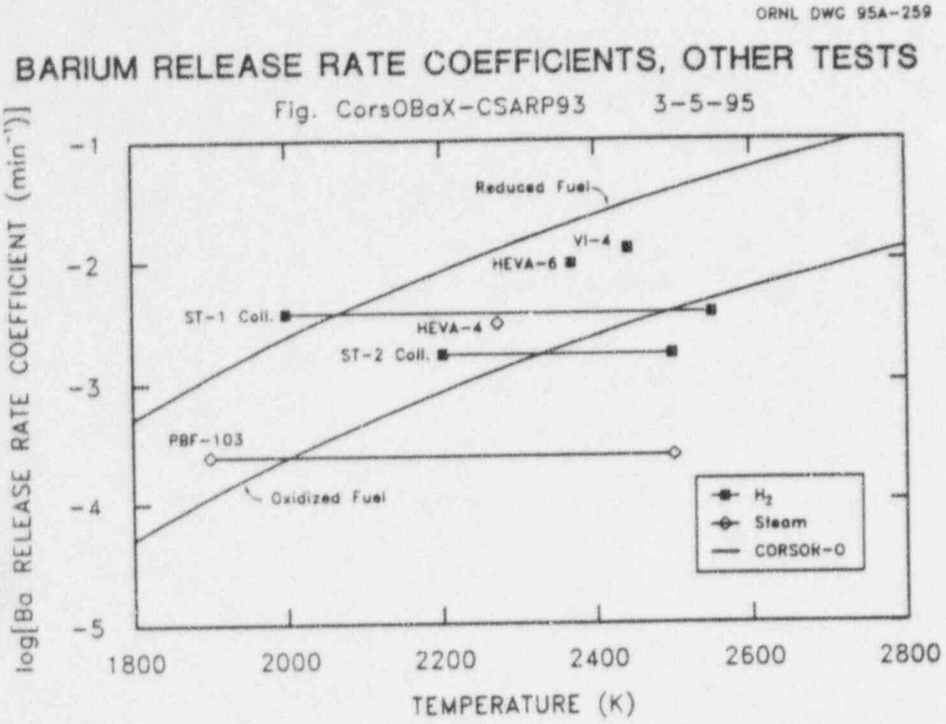

Figure 6.19 Release rate coefficients for barium, other tests

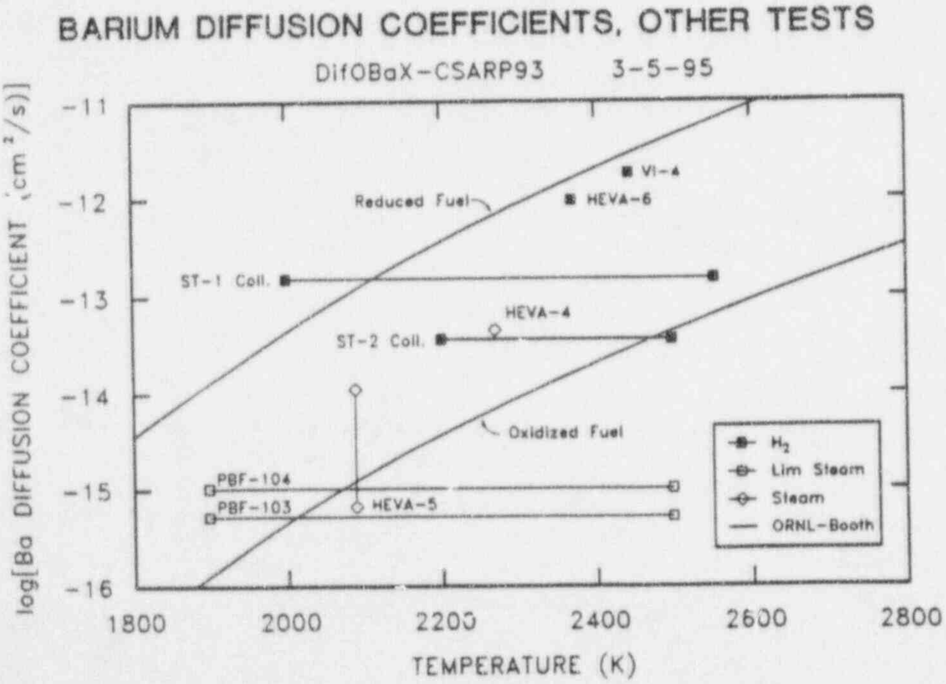

Figure 6.20 Diffusion coefficients for barium, other tests

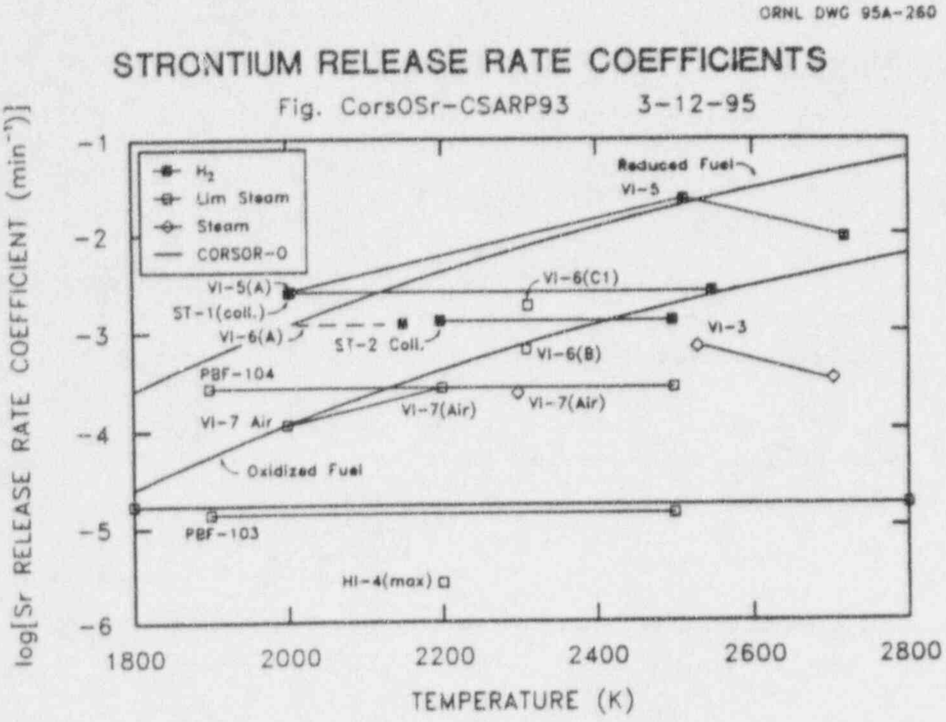

Figure 6.21 Release rate coefficients for strontium

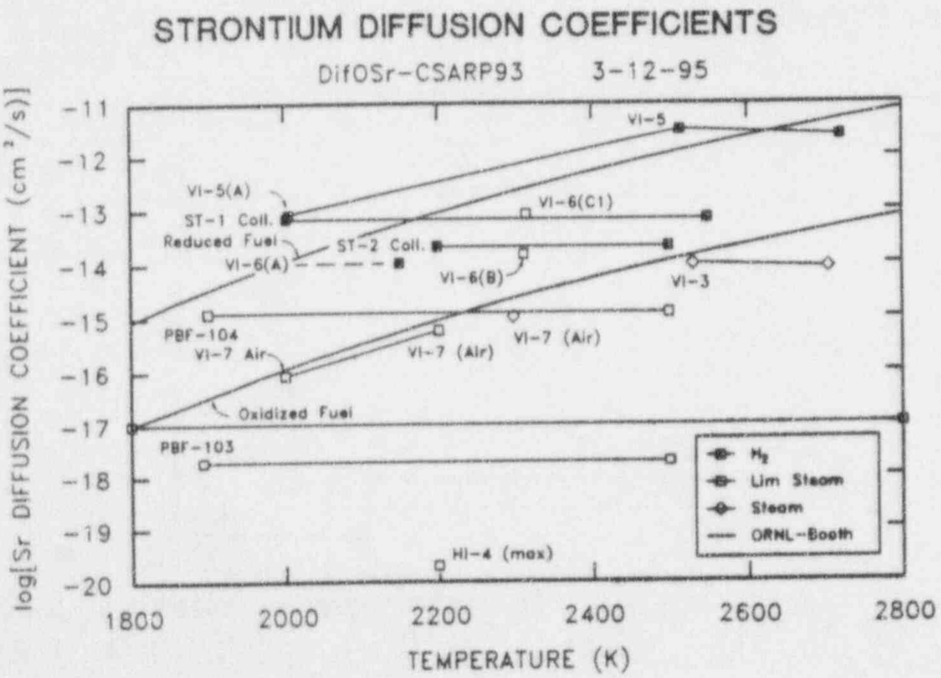

Figure 6.22 Diffusion rate coefficients for strontium

ORNL DWG 95A-261

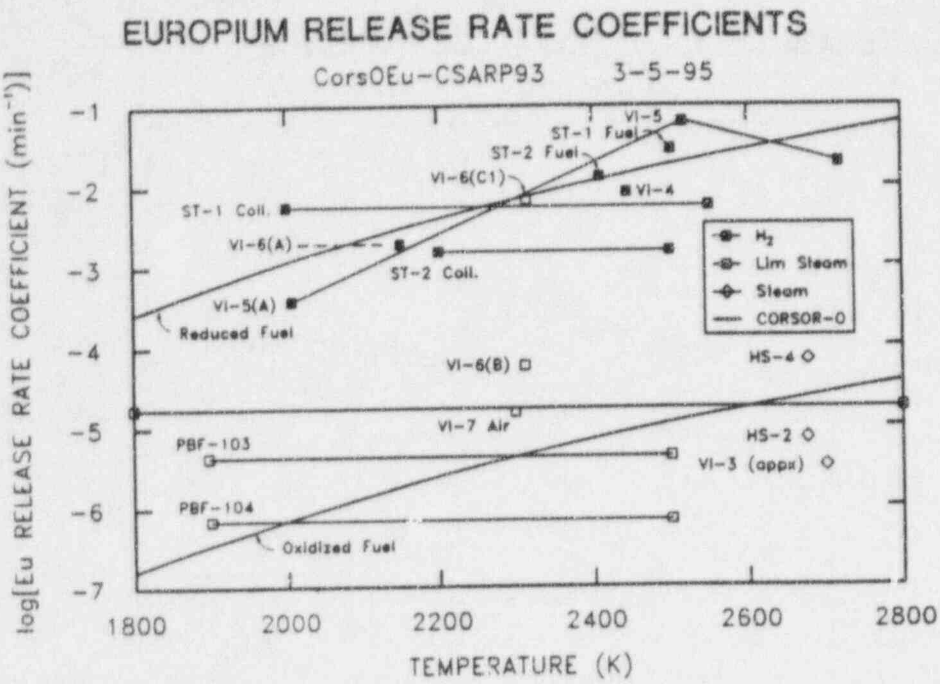

Figure 6.23 Release rate coefficients for europium

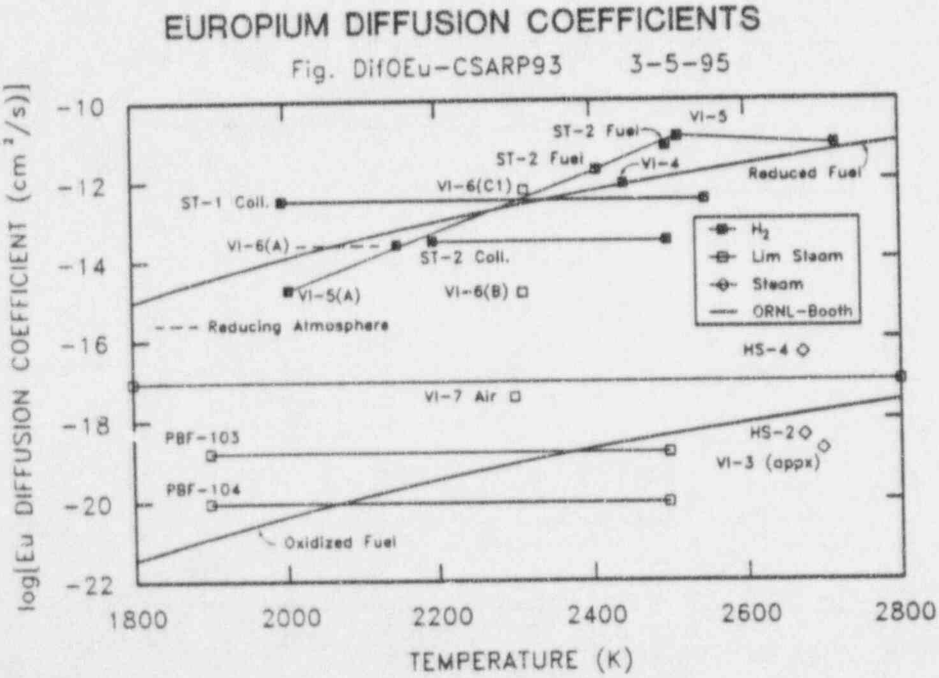

Figure 6.24 Diffusion rate coefficients for europium

ORNL DWG 95A-262

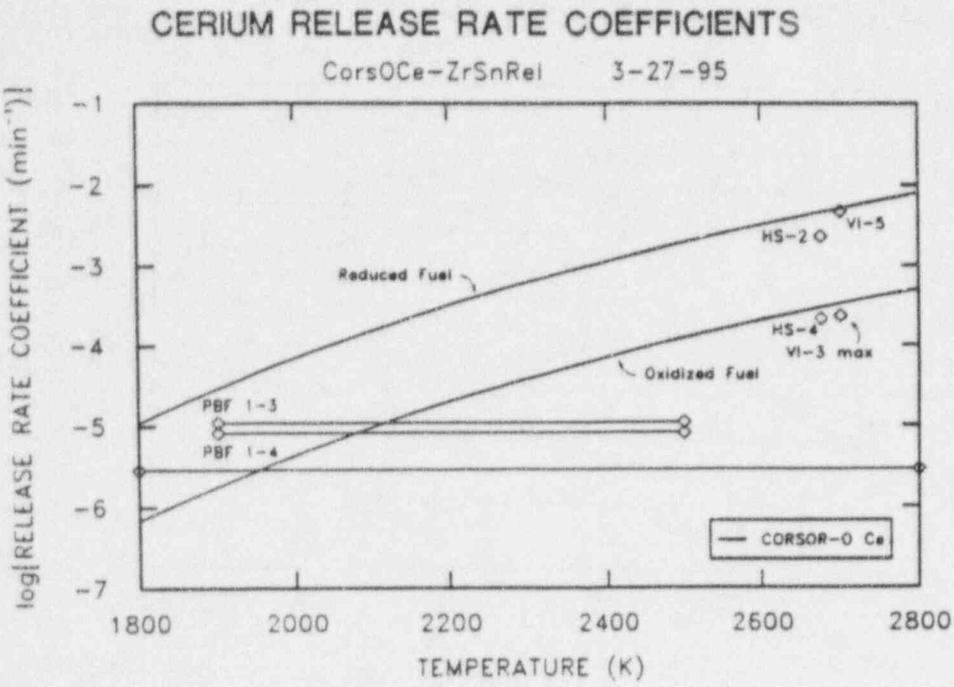

Figure 6.25 Release rate coefficients for cerium

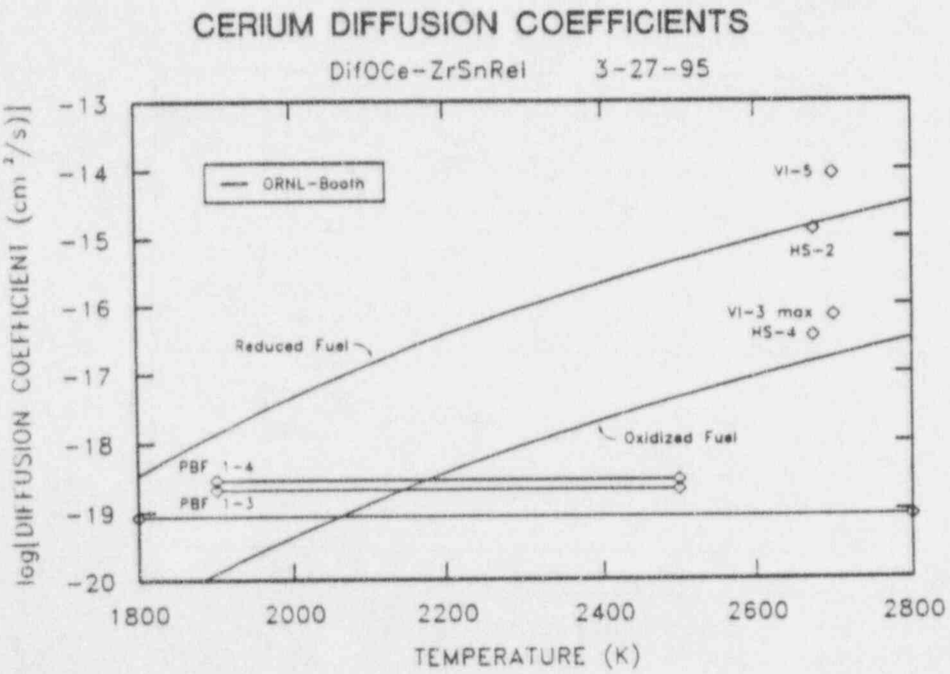

Figure 6.26 Diffusion rate coefficients for cerium

fuel with which a cerium gamma was easily measured.[54] The rate for reducing conditions is speculative.

6.6 Molybdenum and Ruthenium

6.6.1 Molybdenum

Figures 6.27 and 6.28 show that oxidizing atmospheres cause higher release rates for molybdenum. The data for the HI and VI tests were obtained by SSMS or ICP analyses. The HEVA data were obtained by direct gamma counting for ^{99}Mo since the fuel was reirradiated for the HEVA tests.[60]

6.6.2 Ruthenium

Ruthenium forms a volatile oxide in oxidizing atmospheres. In steam-helium tests, release rates for ruthenium are much lower than for molybdenum and have been detected only in ORNL Tests VI-3 and two simulant tests, HS-2 and HS-4. Figures 6.29 and 6.30 show the measured release rates. Ruthenium release was also measured in PBF 1-3 and 1-4 and at TMI-2.

The effect of steam on the release rate of ruthenium occurs slowly with clad fuel. The difference can be seen between the CORSOR-O steam curve (upper curve) and the rate of release measured from pieces of unclad UO_2 heated in steam.[62] The rate of release of ruthenium from fuel with cladding undoubtedly depends on the extent of the defects in the cladding and the length of time exposed to the steam.

A further release rate increase is observed when the fuel rod segments or unclad pieces are exposed to air.[62] Figures 6.29 and 6.30 show the ruthenium release rates for irradiated H. B. Robinson reactor fuel segments heated in air at 775 and 975 K in Tests HBU-5 and HBU-6. The other tests in the HBU series were run with steam atmospheres, and no ruthenium release was detected at test temperatures up to 1475 K. These higher-temperature tests were heated for only 20 min. In Tests HBU-5 and HBU-6, a 1.6-mm-diam hole was drilled in each fuel segment before the test. The cladding defects did not increase during the test so that all of the fuel oxidation and fission product release occurred near these holes. The release rates for these tests in Figures 6.29 and 6.30 have been extended by factors of 10 and 100 to indicate that release rates from the amount of fuel actually affected by the air were much greater than when the total fuel inventory was used as the initial inventory.

The ORNL fission product release program did not include releases or effects of control rod materials and structural materials. Using the information in Reference 56 as a guideline, we have determined that the following release rates and diffusion coefficients can be used: cadmium, same as cesium; indium, same as antimony; and silver, same as tin.

For the diffusion coefficients, a grain radius of 6 μm should be used regardless of the actual fuel grain size. The release of control rod materials is undoubtedly mainly a vaporization process, so some allowance should be used for situations in which the gas flow rate is unusually high or low. Some suggestions for adopting simple models to vaporization situations are given in Reference 56.

Some older accident computer codes did not allow melted material to flow toward cooler regions at the bottom of the core where they could solidify. Some adjustment of "k" and "D" values should be made for cases like this in order to prevent unrealistically high releases of molten materials.

The effect of air on unclad pieces of UO_2 fuel was measured at UKAEA and AECL.[62,63] The release rates from pieces of fuel are much higher because of the more rapid fuel oxidation. In a special test described in Reference 4, RuO_2 was heated in O_2 at 775 and 975 K. Ruthenium was transported, probably as RuO_4, and condensed at lower temperatures, probably as RuO_2.

6.7 UO_2 and PuO_2

6.7.1 UO_2

The UO_2 release occurs almost certainly by vaporization and is, therefore, dependent on the gas flow rate and the amount of UO_2. Older tests, which used milligram amounts of fuel on U-shaped tungsten filaments, vaporized UO_2 so rapidly at high temperature that the release rate of volatile fission products was only the order of 10 times that of the low-volatility fission products. The less-volatile fission products were released by the congruent vaporization of the UO_2.

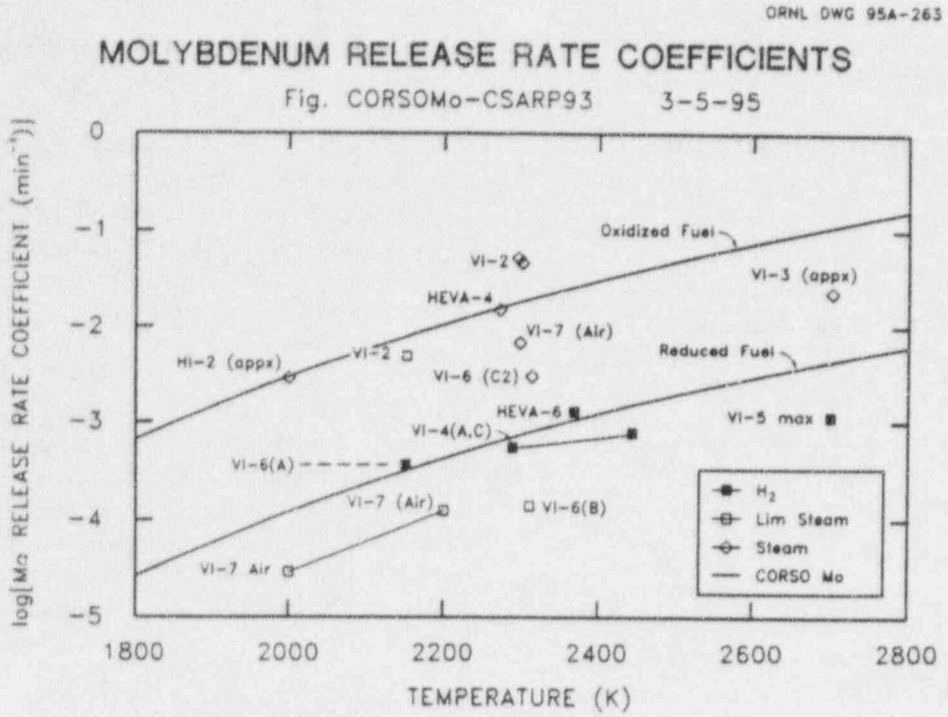

Figure 6.27 Release rate coefficients for molybdenum

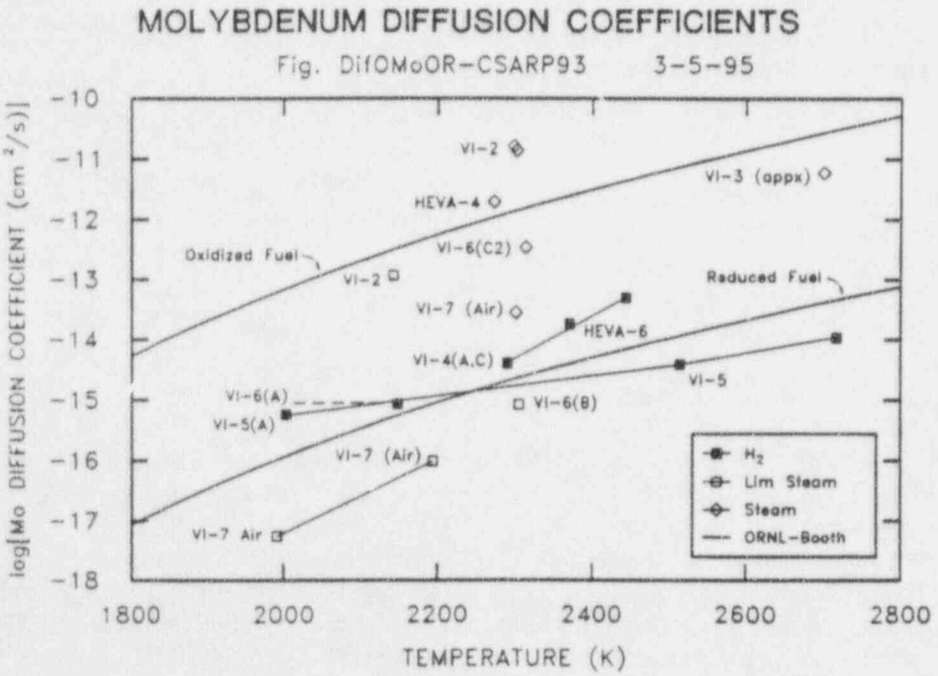

Figure 6.28 Diffusion rate coefficients for molybdenum

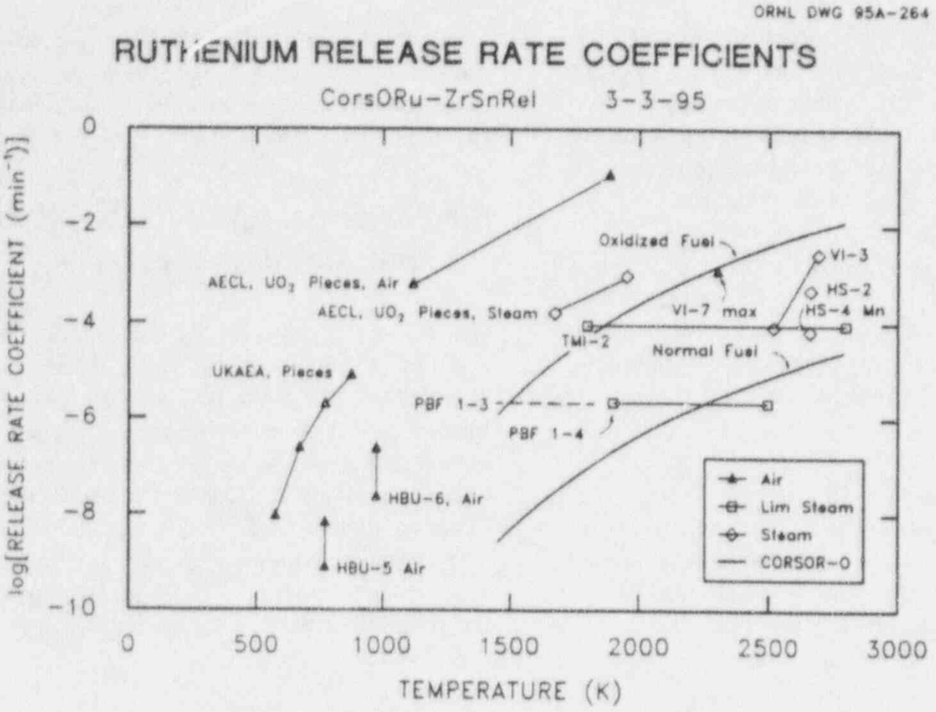

Figure 6.29 Release rate coefficients for ruthenium

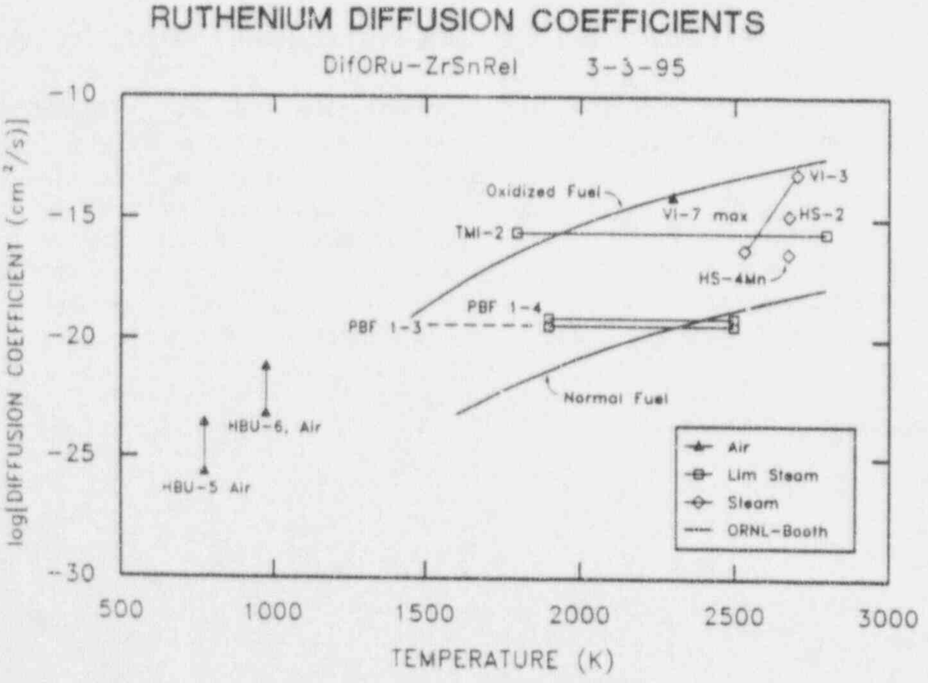

Figure 6.30 Diffusion rate coefficients for ruthenium

it is, therefore, difficult (really impossible) to specify a release rate or diffusion coefficient that applies to all test or accident conditions. Fortunately, most experiments used gas flow rates that are in the accident range for the amount of fuel used. We have, therefore, presented the test results directly without any flow adjustment.

The data shown in Figures 6.31 and 6.32 suggest that there is an atmospheric difference for UO_2 release (vaporization). Alexander found that zirconium represses the U-species partial pressures,* which results in lower partial pressures above 2000 K for the ZrO_2-UO_2 mixture.

In calculating the diffusion coefficients for UO_2 "release," we used a grain radius of 6 μm regardless of the measured grain size. This was done because we believe that UO_2 release is primarily a vaporization process that is not dependent on grain size. Use the high burnup correlations regardless of actual fuel burnup.

6.7.2 Plutonium

Very little data are available for plutonium release. We have selected a plutonium C_k value for CORSOR-O slightly below the value for UO_2, as shown in Figures 6.33 and 6.34. The plutonium release for Test VI-4 is somewhat uncertain because the uranium releases for that test did not appear to be realistic. Use the high burnup correlations and a grain radius of 6 μm.

Alexander heated a small piece of irradiated BR3 fuel in an evacuated Knudsen cell-mass spectrometer combination.[64,65] The rate of "volatilization" of plutonium (atoms/s) was slightly greater than that of cerium. Since the inventory of plutonium in BR3 fuel is ~2.5 times that of cerium, the fractional release rate of cerium was slightly greater than that of plutonium. The high rate of vaporization of UO_2 might have influenced these results. The BR3 fuel was more reducing than other fuels tested by Alexander. The predominant chemical forms released were PuO and CeO.

6.8 Structural Materials: Zirconium and Stainless Steel

Release rate coefficients and diffusion coefficients for the structural materials are shown in Figures 6.35 and 6.36. In order to calculate diffusion coefficients for these materials, it is necessary to assume a grain size. We have selected a grain radius of 6 μm, the same as for the UO_2 grain radius in many of the ORNL tests.[66] The "release" of structural materials is undoubtedly primarily a vaporization process. We have not made any adjustment for airflow. The high burnup correlation should be used for structural materials regardless of the fuel burnup.

The release rates for fission product zirconium were measured in older tests. They are similar to those of Ce and UO_2.[54] The release rate coefficients and diffusion coefficients for Zr-clad can be used, but burnup and grain size correlations should be used for fission product zirconium.

6.9 Control Rod Materials: Ag, In, Cd

Silver alloy control rods can become a significant source of aerosol matter in a severe accident. Such aerosols can participate in the transport of fission products by reaction, sorption, or agglomeration. Silver stabilizes iodine[67] by reaction, and cadmium stabilizes iodine[68] by sorption and reaction at containment temperatures. The high burnup correlations should be used for control rod materials regardless of fuel burnup. A grain radius of 6 μm should be used.

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*C. A. Alexander, U.S. NRC Partner's Meeting, Silver Spring, MD, Oct. 12-23, 1987, personal communication.

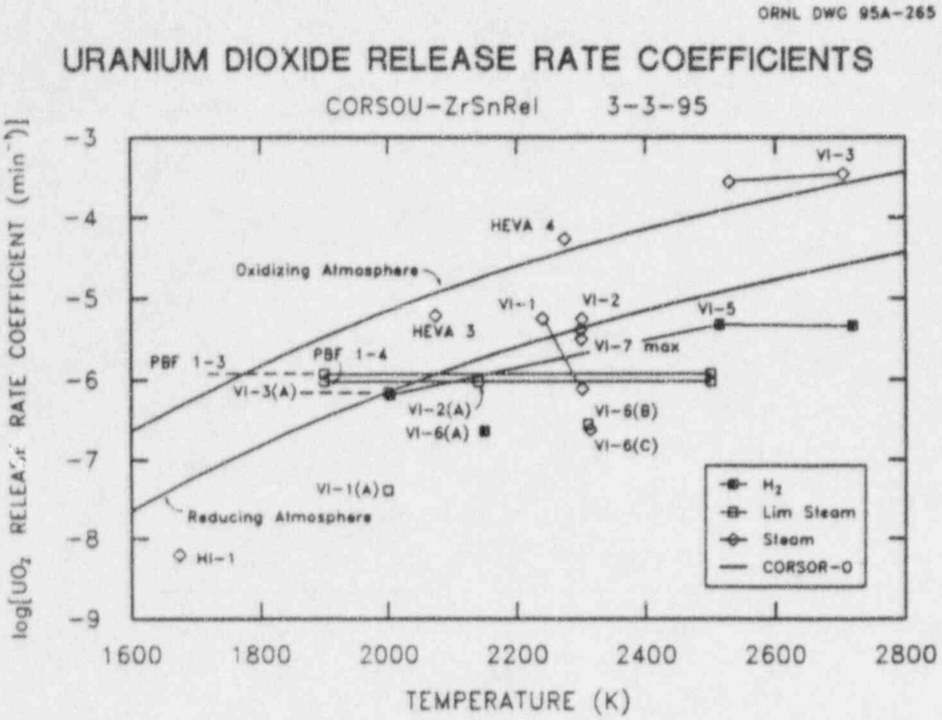

Figure 6.31 Release rate coefficients for uranium

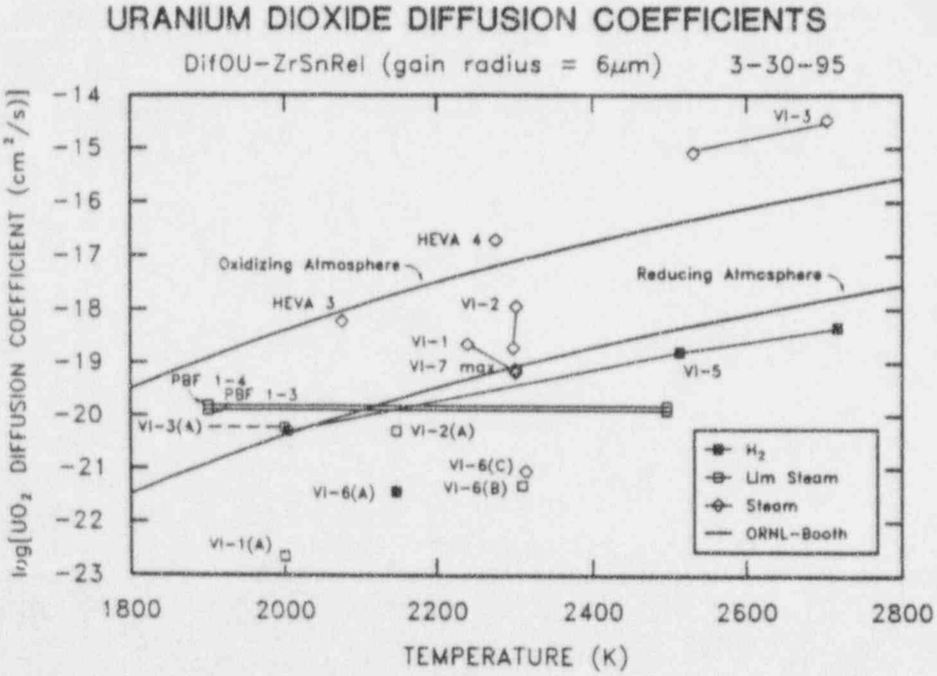

Figure 6.32 Diffusion rate coefficients for uranium

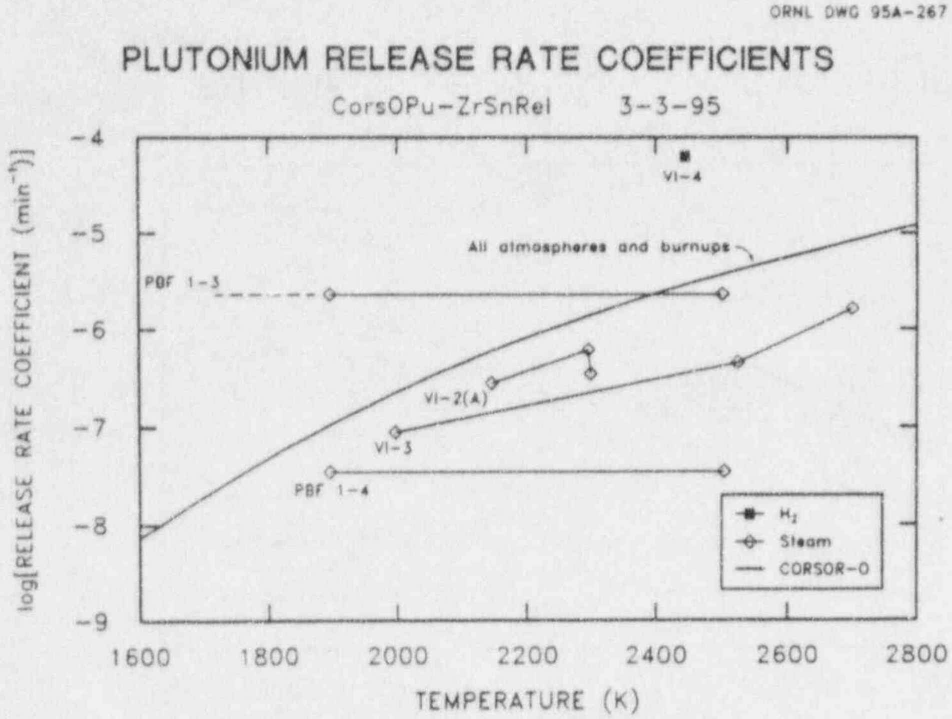

Figure 6.33 Release rate coefficients for plutonium

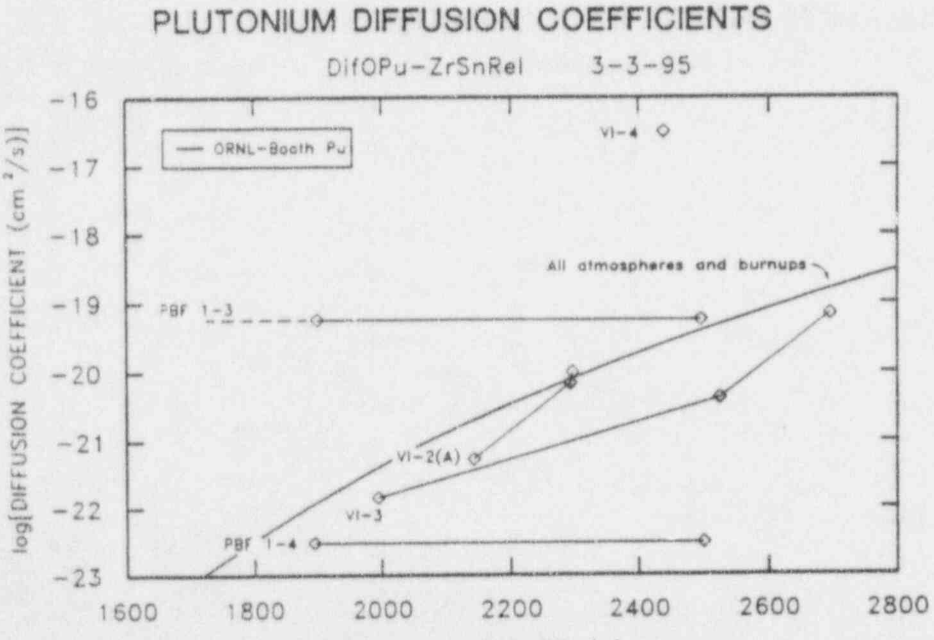

Figure 6.34 Diffusion rate coefficients for plutonium

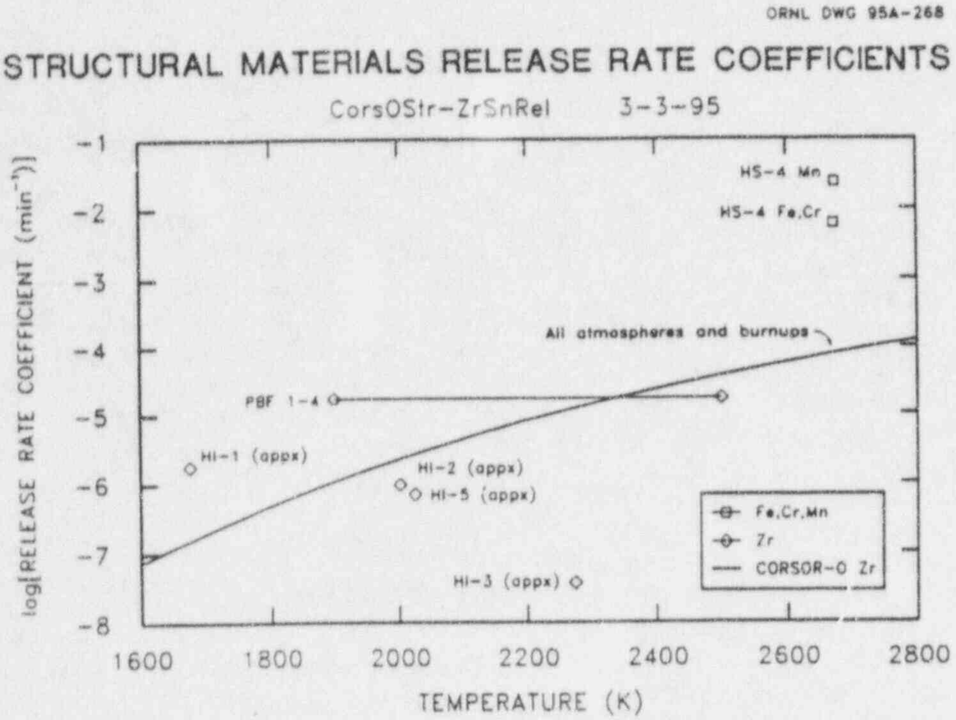

Figure 6.35 Release rate coefficients for structural materials

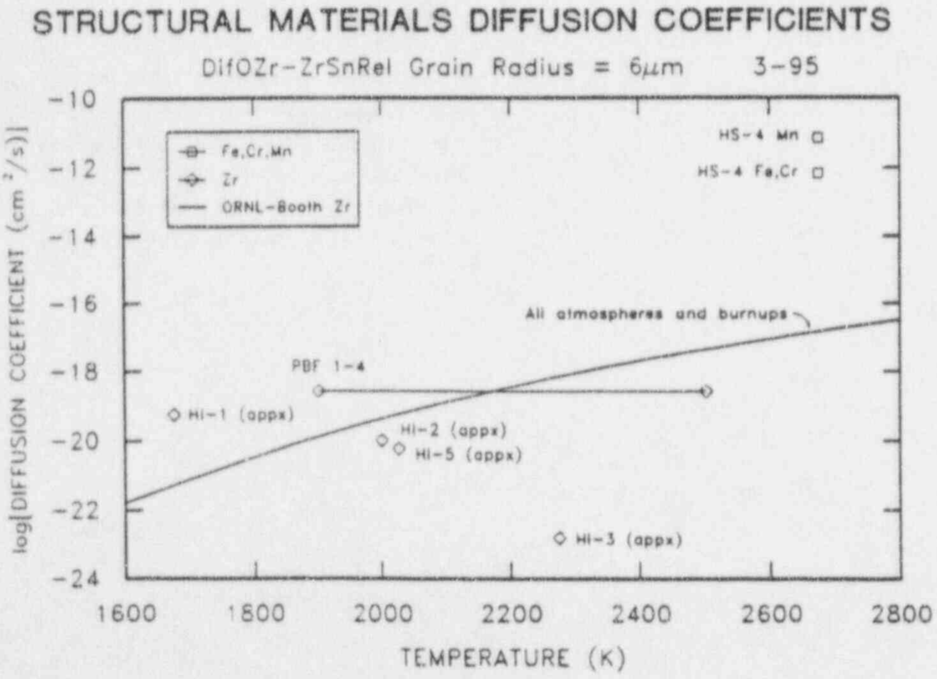

Figure 6.36 Diffusion rate coefficients for structural materials

7 A Comparison of Release Models

Figure 7.1 compares the time release behavior of the two fractional release rate models (CORSOR-O and CORSOR-M) and the two diffusion release models (ORNL-Booth and CORSOR-BOOTH). The comparison is for cesium release from high-burnup fuel that is instantaneously heated to 2000 K and held at 2000 K for 100 min. The diffusion models increase in total release faster initially but decrease more with longer times. This is characteristic of real fission product release and is the reason that the diffusion models provide a better fit with fission product release data.

The diffusion models include the effect of grain size. It is not clear from our test results how much this improves the fit of the data. This is because much of the fission product release occurs by mechanisms that are not controlled by atomic diffusion within the grains, and the diffusion coefficients that we calculate from test data are an empirical fit that includes all release mechanisms. The initial grain size is used. Any grain growth effects are accommodated by the empirical fit of the data. If the grain size is unknown, we recommend using a 6-μm radius.

Comparisons of release rates between CORSOR-O and CORSOR-M and between ORNL-Booth and CORSOR-BOOTH are shown in Table 7.1 for temperatures of 2000 and 2700 K (CORSOR-O/ CORSOR-M). The diffusion model differences are the same at all temperatures because both models use the same value of Q.

7.1 CORSOR-O and CORSOR-M

The rate of CORSOR-O release to the CORSOR-M release rate for each element at two temperatures is given in Table 7.1. It is obvious that there are important differences. For the less-volatile species, the release differences at 2000 K are not very significant because the release rates are very low. Although the ruthenium differences are the largest, the differences for strontium and barium are probably of more significance in source term calculations.

7.2 ORNL-Booth and CORSOR-BOOTH

The ratio of amounts released for small fractional releases is given in Table 7.1. These ratios are much closer to 1.0 than for the CORSOR models, but there are also some significant differences.

7.3 ORNL-Booth and CORSOR-O

The main difference between ORNL-Booth and CORSOR-O is the release-with-time behavior. A sample comparison is shown in Figure 7.1. For this comparison, we assume that a section of high-burnup fuel with grain radius = 6 μm is heated instantly to 2000 K for 100 min. The diffusion equation (ORNL-Booth) allows a rapid initial release followed by a rapidly decreasing release rate. This is characteristic of experimentally measured release behavior.

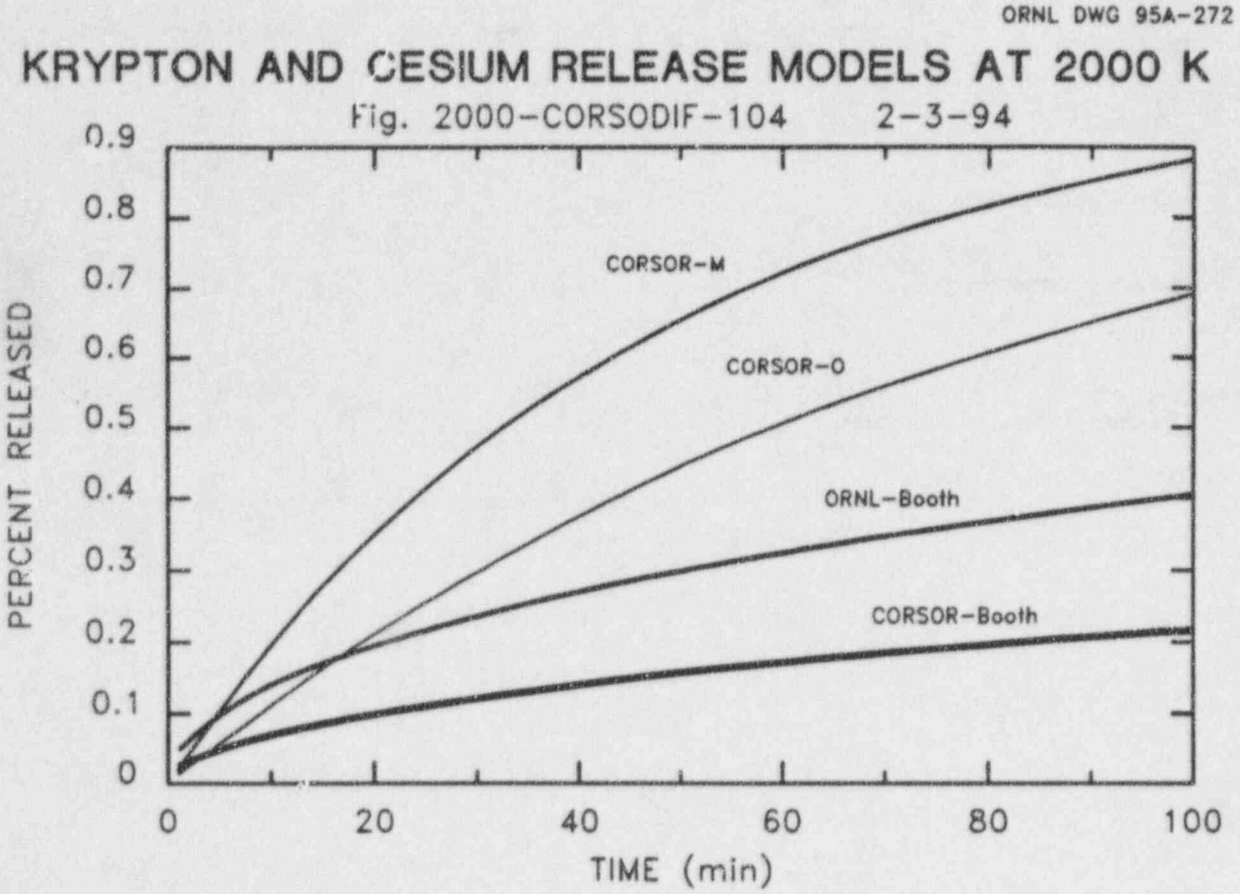

Figure 7.1 Comparison of ORNL-Booth and CORSOR-O cesium release

Table 7.1 Release rate comparison

Element	Ratio (CORSOR-O/CORSOR-M)	
	2000 K	2700 K
Kr, Cs	0.55	0.31
Te	0.44	0.25
Ba (reducing atmosphere)	708	37
Ba (oxidizing atmosphere)	71	3.7
Sr (reducing atmosphere)	16,300	285
Sr (oxidizing atmosphere)	1,630	28
Ru (oxidizing atmosphere)	361,000	612
Ru (reducing atmosphere)	36,100	61
UO_2	349	1.1
Sn (cladding, oxidizing atmosphere)	32	11.5
Sn (cladding, reducing atmosphere)	2.2	0.8
Zr (cladding)	24,100	97

8 Fission Product Transport and Chemical Form

8.1 Transport Behavior (Vapor or Aerosol)

The physical form of released fission products affects their transport behavior. At the high temperatures of severe accidents, most fission products are released as single atoms. They may remain as atoms, combine with other fission products (e.g., CsI), combine with vaporized structural or control rod materials (e.g., SnTe), nucleate to form pure CsI aerosols, or become attached to other solid (or liquid) particles. Fission products that remain in the vapor form are very mobile and will condense on fixed or aerosol surfaces at the first opportunity. Vapor species will also quickly locate any fixed or aerosol surface and immediately react if the surface material is reactive with the species.

Examples of fission products that condense in our tests and their condensation temperatures are CsI (~800 K), Eu (~2200 K), and Sb (~1200 K). The latter two occur in hydrogen atmosphere tests; their chemical forms are uncertain.

The atmosphere has a strong effect on the amount of released cesium that is transported in the aerosol form. With steam atmospheres, almost 70% of the released cesium is associated with aerosols. The remainder is in vapor forms that condense or react with fixed surfaces in the upper furnace region or in the TGTs. In hydrogen atmosphere tests, only ~30% of the released cesium is associated with aerosols. The aerosol/vapor relationship is shown in Figure 8.1 as a function of atmosphere and fuel temperature. The total mass of aerosols and deposited vapor species are greater in steam atmosphere tests, as shown in Table 8.1. One of the reasons for the larger mass of the deposits in steam atmosphere tests is that the oxides and hydrates are formed.

8.2 Chemical Form

The chemical form of fission products cesium, iodine, and tellurium in our tests has been summarized in journal articles.[19-22] The most important fission product chemical form is that of iodine. Iodine is biologically an important species, and its behavior is very sensitive to its environs.

In our tests, the fission products are collected within 1 s of release from the fuel. This is by design in order to learn fission product behavior and chemical form as close to the time of release as possible. The amount of volatile iodine formed (I_2, HI, or CH_3I) is very important since these forms can persist in the gas phase longer than less-volatile forms.[67,68] We measure the amount of "volatile iodine" by collection on charcoal that is impregnated with TEDA.

We have found in our tests that for releases above the trace level (releases more than ~1% of total inventory), the amount of volatile iodine is <1% of the amount released. This has been true for tests in which the furnace tube has been silica, zirconia, or thoria; for TGT liners made of platinum, oxidized stainless steel, or clean stainless steel; and for test atmospheres of hydrogen, restricted steam flow, or high steam flow. Exceptions were Tests VI-5 and VI-6, in which ~3% of the released iodine was collected by the charcoal. With both of these tests, there was a waiting period of 5 or 6 months during which the charcoal cartridges remained sealed up with the filter papers in the original filter/charcoal package. We believe that radiation decomposition over this time period may have decomposed some of the iodine species associated with particulates and allowed volatile forms of iodine to migrate slowly to the charcoal.

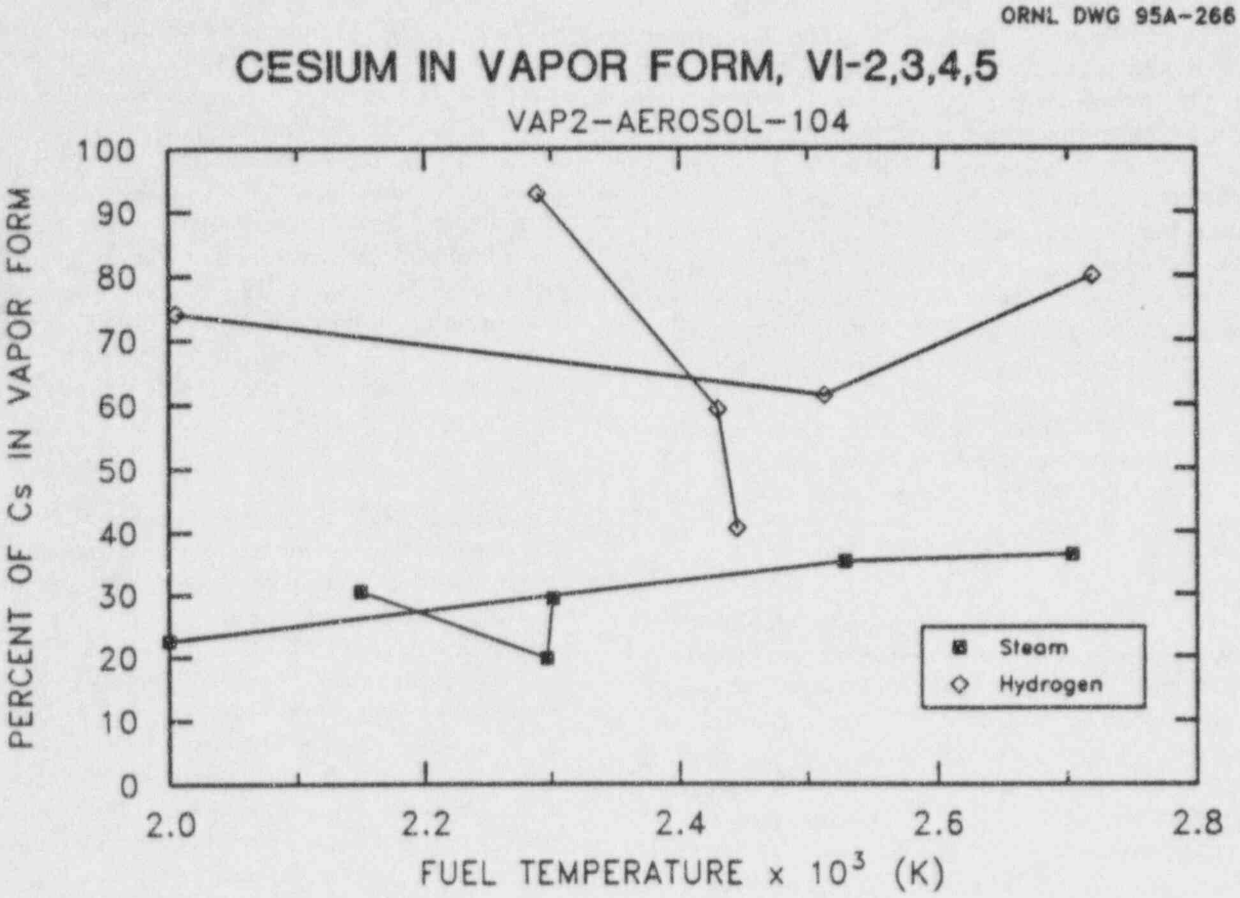

Figure 8.1 Effect of atmosphere on amount of released cesium in aerosol and vapor form

Table 8.1 Comparison of total deposits in steam and hydrogen atmospheres

	Weight of deposits (mg)	
	VI-3 (steam)	VI-5 (hydrogen)
TGT-A	61	104
TGT-B	380	50
TGT-C	241	30
Filter A	308	47
Filter B	1094	273
Filter C	1084	177

9 The Zr-UO_2 "Liquefaction" Reaction

The Zr-UO_2 reaction was first suggested as a possible mechanism for enhancement of fission product release when the TMI-2 President's Commission visited ORNL in early June 1979. Fission product release in TMI-2 was greater than could be explained by the temperature scenarios proffered by most of the thermal-hydraulic specialists. Until this time, the Zr-UO_2 reaction had been investigated only at moderate temperatures as a physical fuel-cladding interaction problem. Fission product release with Zircaloy cladding at higher temperatures had not been investigated.

Tests HI-3, HI-4, and HI-5 were run with low-steam-flow rates so that some of the Zircaloy cladding would remain unoxidized, melt, and react with UO_2 and fission products. The release of fission products was actually lower than in high-steam-flow Test HI-2. The melted cladding in HI-3, 4, and 5 remained "glued" to the fuel and appeared to act as a barrier to release.[13-15,66]

This was contrary to what many people expected. The Zr-UO_2 reaction was called "liquefaction" by some other experimenters and modelers who believed that the liquid phase present during the reaction would enhance fission product release. Tests VI-4, 5, and 6(A) were performed in hydrogen atmosphere in the vertical orientation. A small enhancement of Kr and Cs releases is suggested by the data, but the collapse of the VI-4 fuel may have affected the release rates.

10 Fuel Melt Progression

Fuel and cladding behavior under accident conditions has been found to be strongly dependent on the atmosphere.

10.1 Steam Atmosphere

Three tests (VI-1, VI-2, and VI-3) were run in the vertical apparatus with high steam flow. Test VI-6 started with hydrogen gas and ended with high steam flow. The fuel segments remained standing, although cracks in the cladding were discovered during posttest examinations of the fuel from Tests VI-1, 2, and 3. Test HI-2 was the only test run in the horizontal apparatus in which the cladding was completely oxidized. A large longitudinal crack was formed in the cladding. There is a linear growth of ~10% when the Zircaloy is oxidized. From these tests, we can see that steam atmosphere oxidation results in a fuel rod segment that is strong enough to remain standing (15-cm length) yet contains cracks in the cladding. An X-ray of the VI-3 fuel segment after the test showed that it was bowed, which demonstrates some high-temperature ductility.

Hydrogen was present during the early stages of all steam tests as a result of the H_2O-Zr reaction. The rate of hydrogen production was measured beginning with Test VI-1. The results were published in References 27-29 and Reference 38.

10.2 Restricted Steam Flow

Tests HI-3, HI-4, and HI-5 were all run with the steam flow rate low enough to prevent complete oxidation of the cladding and to permit melting of the incompletely oxidized portion of the cladding. Posttest examination revealed that some of the melted cladding had reacted with the UO_2. In all three tests (2025 to 2275 K), the runoff of cladding was low.[13-15,66] Partially oxidized cladding remained on the UO_2 pellets. Numerous bubbles were formed in the cladding.

The HEVA-6 test was performed with a short fuel section in vertical orientation.[69] The cladding was 67% oxidized at moderate temperature. The high-temperature portion of the test was then run at 2375 K in hydrogen atmosphere. The partially oxidized cladding did not run off. It appeared to be glued to the UO_2 and contained bubbles in the cladding as with the three ORNL horizontal tests. The release of cesium and iodine was lower than in high steam flow tests, suggesting that the partially oxidized and melted cladding served as a barrier to release.

10.3 Hydrogen Atmosphere

Tests VI-4 (2440 K) and VI-5 (2700 K) were run with hydrogen atmospheres. The VI-4 fuel specimen collapsed at ~2150 K, shortly after melting of the cladding. The VI-5 fuel specimen collapsed at a much higher temperature, or possibly during cooldown. The VI-6 fuel specimen was heated in flowing hydrogen/ helium to 2300 K, at which point gas flow was then changed to helium with very low steam, and later to high steam flow. It did not collapse.

The SNL ST-1 and ST-2 4-rod fuel bundles expanded into a foamlike structure while heated by internal fission and gamma heating in the ACRR. The collapsed VI-4 fuel underwent the Zr-UO_2 reaction, but only a small amount showed bubble formation similar to the ST-1 and ST-2 fuel. Regardless of the details of the mechanism, it is clear that the hydrogen atmosphere weakens the fuel column more than a steam or restricted steam flow atmosphere.

10.4 Air Atmosphere

Test VI-7 was run in an air-steam-helium mixture. All of the cladding, except for the upper 2 cm, was completely oxidized while the fuel was at 2000 K. At the time of this writing, only the krypton and cesium release quantities are available. Their release rates are similar to those in steam and hydrogen atmospheres. Gamma scans of the fuel rod taken after the test show that significantly more cesium was released from the bottom third of the fuel (gas inlet end) than from the remainder of the fuel.

Tests HBU-5 and HBU-6 were run in dry air at 975 and 775 K. Fission product simulant test Implant-7 and Implant-9[3] were also run in dry air at the same temperatures. In all four tests, there was expansion of the fuel and cladding caused by the air oxidation of the UO_2. The released iodine in the simulant tests was collected mainly on the first of a series of silver-plated screens. The iodine released in the HBU tests was collected mainly on the first charcoal cartridge (there were no silver screens in the HBU apparatus). This behavior indicates the presence of a volatile chemical form of iodine, probably I_2. These tests were run for 5 and 20 h,and similar tests were run in steam. Similar long-term tests in steam also produced mainly a volatile form of released iodine (Implant-10 and HBU-1).[4] Low release concentrations and long exposure times both promoted high proportions of released iodine in volatile forms.

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Appendix A. Results from Other Tests

The results from ORNL HI and VI tests can be compared with the results from several other test series. The temperature and heatup rates for these tests are given in Table A.1. The operating conditions for these tests are given in Table A.2, and the release amounts are given in Tables A.3 and A.4.

A comparison of operating conditions for in-reactor and hot-cell tests is given in Table A.5. There has been a long-time interest in whether the fuel pellet centerline-to-surface temperature difference created by radiation decay heat would influence the fission product release rate.

There has also been an interest in the possible effect of irradiation intensity on fission product release rate, chemical form, or transport behavior. The tests in Table A.5 are listed in order of interval heat generation, which is approximately proportional to the pellet radial temperature gradient. The pellet centerline-to-surface temperature difference in the TMI-2 accident was 10 K or less. No effect of temperature gradient or radiation intensity was reported for any of the tests listed in Table A.5. The ANL DEH tests reported that many small cracks were formed in the UO_2 in their tests that enhanced the release of fission gas.

Table A.1 Temperature and heatup rates in other tests

		First temperature plateau			Second temperature plateau			
Test No.	Atmosphere	Heatup rate (°/s)	Temperature (K)	Time at plateau (min)	Heatup rate (°/s)	Temperature (K)	Time at plateau (min)	Cooldown rate (°/s)
HS-1	Steam	1.1	1875	15	1.9	2175	30	1.7
HS-2	Steam	1.6	2275	10	1.3	2675	10	1.6
HS-3	Steam	1.1	2275	1		2275	20	1.3
HS-4	Steam	1.3	2275	5	1.9	2675	15	1.5
HT-1	Steam	6.0	1575	10				8
HT-2	Steam	15.0	1720	7				12
HT-3	Steam	7.4	1885	3				18
HT-4	Steam	13.0	1675	0.3				7
HBU-12	He	~0.3	1375	260	~0.3	1475	50	
HEVA-3	Steam	1.0	2075	30				
HEVA-4	Steam	1.0	2275	7				2.4
ST-1	H_2	1.0	1600	20	1.2	2490	~19	0.6
ST-2	H_2	1.0	1550	25	1.0	2410	20	0.5

Table A.2 Operating conditions for other fission product release tests

Test No.	Gas flow through system						
	Gas	Flow (L/min)	Gas	Flow (L/min)	Pressure (MPa)	Burnup (MWd/kg)	UO_2 (g)
HS-1	He	0.50	H_2O	1.45	~0.1	40 simulated	90
HS-2	He	0.49	H_2O	1.34	~0.1	40 simulated	90
HS-3	He	0.50	H_2O	1.35	~0.1	40 simulated	90
HS-4	He	0.51	H_2O	1.30	~0.1	40 simulated	90
HT-1	He	0.92	H_2O	0.76	0.1	31.4	210
HT-2	He	0.92	H_2O	0.99	0.1	30.6	210
HT-3	He	0.92	H_2O	1.08	0.1	31.4	210
HT-4	He	0.84	H_2O	0.98	0.1	31.1	210
HBU-12	He	1.80			0.12	31.4	199
HEVA-3	H_2	0.36	H_2O	2.97	~0.1	27.7	~40
HEVA-4	H_2	0.36	H_2O	2.40	~0.1	36.7	~40
ST-1	Ar	1.8—4.4	H_2	0.9—2.2	0.11—0.22	47.0	307
ST-2	Ar	3.16	H_2	0.081	1.8 avg.	47.0	307

Table A.3 Release during various fission product release tests

Test No.	Temperature (K)	Effective time (min)	Amount released (% of inventory)								
			^{85}Kr	^{134}Cs	^{129}I	Te	Sb	Sn	Ba	Sr	^{154}Eu
HS-1[a]	2175	35		100		92.9	51.7	100			
HS-2	2675	15									0.01
HS-3[a]	2275	25		100		97.4	89.8	92.3			
HS-4	2675	20									0.13
HT-1	1575	12	1.07	0.11	0.20						
HT-2	1720	9	5.0	4.82	2.87						
HT-3	1885	5	8.25	10.95	15.4						
HT-4	1675	2	2.80	3.05	2.13						0.05
HBU-12	1375	500		0.45	0.55		20.1				
HEVA-3	2075	33		37.9					1.3		
HEVA-4	2275	10		44.2	40.0	40	25		3.5		
HEVA-6	2375	33		48.2	28.0	10			27		
ST-1R[b]	~2500[c]	22		98.9							46
ST-1R[b]	~2300[c]	22		79.5							
ST-1R[b]	~2100	22		23.8							
ST-1R[b]	2000–2500	22		71							20
ST-1C[d]	2000–2500	22		56	38	<0.2			8	5	11.28
ST-2R[b]	2410[c]	24		95							27
ST-2R[b]	2200	24		62							
ST-2R[b]	2100–2410	24		82							15
ST-2C[d]	2100–2410	24		30	23	0.5			4	3	3.54

[a] All of the isotopes and elements shown were contained in unsintered unirradiated UO_2 pellets.

[b] Release from fuel. Measured by gamma counting before and after test.

[c] Temperature may have been ~150° higher.

[d] Amount collected in fission product collection systems.

Table A.4 Additional release results

Test No.	Temperature (K)	Effective time (min)	Amount released (% of inventory)							
			Mo	Ru	^{51}Cr	^{54}Mn	^{59}Fe	^{144}Ce	U	^{110m}Ag
HS-1[a]	2175	35								92.1
HS-2	2675	15		0.58						
HS-3[a]	2275	25								98.0
HS-4	2675	20		0.13	20.5[b]	56.9[b]	20.0[b]			100[c]
HEVA-3	2075	33						<3.2	~0.02	
HEVA-4	2275	10	~21						0.07	

[a] All of the isotopes and elements shown were contained in unsintered unirradiated UO_2 pellets.
[b] From irradiated stainless steel.
[c] From Ag-In-Cd-Sn control rod alloy.

Table A.5 Comparison of in-reactor and hot cell tests

Test	Internal heat (W/g)	Fuel burnup (MWd/kg U)	Peak temp. (K)	Peak pres. (MPa)
ORNL HBU, BWR, HT, HI, VI	0.001	10–47	2700	0.1
TMI-2 accident	0.5	3	~3200	~15.0
PNL FLHT	~1.0	30	~2400	1.4
INEL PBF SFD 1-4	2.0	40–47	~3000	7.4
SNL ST-1	2.2–4.6	47	~2500	0.2
SNL ST-2	~3	47	~2500	2.0
ANL TREAT STEP	~3	40–47		0.3
ANL DEH (electric heat)	~12	30	~3200	0.1
ORNL fuel rod bundle failure	~12	2.8	1590	0.1
INEL PBF RIA-ST	120	<0.5	3500	~15.0
ORNL ORR UO_2 melting	120	0.3–21	~3400	0.1
ORNL Carroll "sweep gas"	270	18.0	1975	0.1
ORNL TREAT UO_2 melting	6000	0.02	~3500	1.2–15.2

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Fission product release data from the ORNL HI test series and VI test series are summarized in this report and compared with release results from similar tests performed in France (the HEVA test series). The ORNL test results are also compared with four in-reactor tests, the SNL ST-1 and ST-2 tests, and the INEL SFD 1-3 and SFD 1-4 bundle tests. Test atmospheres range from steam to hydrogen, and the temperature range is 1675 to 2700 K.

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