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Fission Product Source Terms for the LWR Loss-of-Coolant Accident

R. A. Lorenz
J. L. Collins
A. P. Malinauskas

Prepared for the
U.S. Nuclear Regulatory Commission
Division of Reactor Safety Research
Office of Nuclear Regulatory Research
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FOREWORD

This document is one of a series of reports on studies of fission product release from LWR fuel. Other reports in this series are identified below:

1. Quarterly Progress Report on Reactor Safety Programs Sponsored by the Division of Reactor Safety Research for July-September 1974, ORNL/TM-4729, Vol. 1 (December 1974).
2. Quarterly Progress Report on Reactor Safety Programs Sponsored by the NRC Division of Reactor Safety Research for October-December 1974, ORNL/TM-4805, Vol. 1 (April 1975).
3. Quarterly Progress Report on Reactor Safety Programs Sponsored by the NRC Division of Reactor Safety Research for January-March 1975, ORNL/TM-4912, Vol. 1 (July 1975).
4. Quarterly Progress Report on Reactor Safety Programs Sponsored by the NRC Division of Reactor Safety Research for April-June 1975, ORNL/TM-5021 (September 1975).
5. A. P. Malinauskas, R. A. Lorenz, M. F. Osborne, J. L. Collins, and S. R. Manning, Quarterly Progress Report on Fission Product Release from LWR Fuel for the Period July-September 1975, ORNL/TM-5143 (November 1975).
6. R. A. Lorenz, J. L. Collins, and S. R. Manning, Quarterly Progress Report on Fission Product Release from LWR Fuel for the Period October-December 1975, ORNL/TM-5290 (March 1976).
7. J. L. Collins, M. F. Osborne, A. P. Malinauskas, R. A. Lorenz, and S. R. Manning, Knudsen Cell-Mass Spectrometer Studies of Cesium-Urania Interactions, ORNL/NUREG/TM-24 (June 1976).
8. R. A. Lorenz, M. F. Osborne, J. L. Collins, S. R. Manning, and A. P. Malinauskas, Behavior of Iodine, Methyl Iodide, Cesium Oxide, and Cesium Iodide in Steam and Argon, ORNL/NUREG/TM-25 (July 1976).
9. R. A. Lorenz, J. L. Collins, S. R. Manning, and A. P. Malinauskas, Quarterly Progress Report on Fission Product Release from LWR Fuel for the Period January-March 1976, ORNL/NUREG/TM-30 (July 1976).
10. R. A. Lorenz, J. L. Collins, S. R. Manning, O. L. Kirkland, and A. P. Malinauskas, Quarterly Progress Report on Fission Product Release from LWR Fuel for the Period April-June 1976, ORNL/NUREG/TM-44 (August 1976).
11. R. A. Lorenz, J. L. Collins, and O. L. Kirkland, Quarterly Progress Report on Fission Product Release from LWR Fuel for the Period July-September 1976, ORNL/NUREG/TM-73 (December 1976).

12. R. A. Lorenz, J. L. Collins, and O. L. Kirkland, Quarterly Progress Report on Fission Product Release: from LWR Fuel for the Period October-December 1976, ORNL/NUREG/TM-88 (March 1977).
13. A. P. Malinauskas et al., Quarterly Progress Report on Fission Product Behavior in LWRs for the Period January-March 1977. ORNL/NUREG/TM-122 (June 1977).
14. A. P. Malinauskas et al., Quarterly Progress Report on Fission Product Behavior in LWRs for the Period April-June 1977. ORNL/NUREG/TM-139 (September 1977).
15. A. P. Malinauskas et al., Quarterly Progress Report on Fission Product Behavior in LWRs for the Period July-September 1977. ORNL/NUREG/TM-170 (January 1978).
16. A. P. Malinauskas et al., Quarterly Progress Report on Fission Product Behavior in LWRs for the Period October-December 1977, ORNL/NUREG/TM-186 (March 1978).
17. R. A. Lorenz, J. L. Collins, and A. P. Malinauskas, Fission Product Source Terms for the LWR Loss-of-Coolant Accident: Summary Report, NUREG/CR-0091 (ORNL/NUREG/TM-206) (June 1978).
18. A. P. Malinauskas et al., Quarterly Progress Report on Fission Product Behavior in LWRs for the Period January-March 1978, NUREG/CR-0116 (ORNL/NUREG/TM-208) (June 1978).
19. A. P. Malinauskas et al., Quarterly Progress Report on Fission Product Behavior in LWRs for the Period April-June 1978, NUREG/CR-0370 (ORNL/NUREG/TM-242) (September 1978).
20. R. A. Lorenz, J. L. Collins, and S. R. Manning, Fission Product Release from Simulated LWR Fuel, NUREG/CR-0274 (ORNL/NUREG/TM-154) (October 1978).
21. A. P. Malinauskas et al., Quarterly Progress Report on Fission Product Behavior in LWRs for the Period July-September 1978, NUREG/CR-0493 (ORNL/NUREG/TM-280) (December 1978).
22. A. P. Malinauskas et al., Quarterly Progress Report on Fission Product Behavior in LWRs for the Period October-December 1978, NUREG/CR-0682 (ORNL/NUREG/TM-308) (April 1979).
23. A. P. Malinauskas et al., Quarterly Progress Report on Fission Product Behavior in LWRs for the Period January-March 1979, NUREG/CR-0917 (ORNL/NUREG/TM-332) (August 1979).
24. A. P. Malinauskas et al., Quarterly Progress Report on Fission Product Behavior in LWRs for the Period April-June 1979, NUREG/CR-1061 (ORNL/NUREG/TM-348) (October 1979).

25. R. A. Lorenz, J. L. Collins, A. P. Malinauskas, O. L. Kirkland, R. L. Towns, Fission Product Release from Highly Irradiated LWR Fuel, NUREG/CR-0722 (ORNL/NUREG/TM-287/R2) (February 1980).

FISSION PRODUCT SOURCE TERMS FOR THE LWR LOSS-OF-COOLANT ACCIDENT

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ABSTRACT

Models for cesium and iodine release from light-water reactor (LWR) fuel rods failed in steam were formulated based on experimental fission product release data from several types of failed LWR fuel rods. The models were applied to a pressurized water reactor (PWR) undergoing a hypothetical loss-of-coolant accident (LOCA) temperature transient. Calculated total iodine and cesium releases from the fuel rods were 0.053 and 0.025% of the total reactor inventories of these elements, respectively, with most of the release occurring at the time of rupture. These values are approximately two orders of magnitude less than releases used in WASH-1400, the Reactor Safety Study.

1. INTRODUCTION

The principal objectives of this study have been to determine the quantities of radiologically significant fission products released from failed light water reactor (LWR) fuel rods under accident conditions, to identify their chemical and physical forms, and to interpret the results for use as input to computer models of postulated spent fuel transportation accidents (SFTAs) and loss-of-coolant accidents (LOCAs). The purposes of this paper are to summarize the source term models developed for cesium and iodine and to demonstrate the application of these models to the analysis of cesium and iodine release during a pressurized water reactor (PWR) LOCA.

The models of fission product release in steam are based on three separate sets of experiments that were conducted over a temperature range of 500-1200°C. One test series employed simulants (CsI, CsOH, and TeO₂) that were applied to the surfaces of unirradiated UO₂ fuel pellets.¹

A second series used fuel capsules that were designed to boiling water reactor specifications and that had been irradiated to 1000 MWd/MT at high heat rating (560-660 W/cm),^{2,3} and the third set of experiments involved tests with commercial fuel that had been irradiated to 30,000 MWd/MT in the H. B. Robinson PWR at low heat rating (175-320 W/cm).³

A summary of the test data is given in Table 1. In half of the tests the fuel rod was pressurized with inert gas and ruptured at either 700 or 900°C; additional time was then allowed for release of fission products from the rupture opening by gas-phase diffusion. The cross-sectional area of the rupture openings ranged from 0.002-0.8 cm², with most in the range 0.02-0.04 cm². The other tests were designed to measure only diffusional release from the failed rod; the defect constituting the failure was formed in the unpressurized rod by drilling a 0.159-cm-diam hole through the cladding. Test HBU-11 was an exception. For this test the previously ruptured fuel rod segment from test HBU-7 was employed in a diffusion release test at a higher temperature.

2. GAP INVENTORIES

During the course of the experimental work, it became apparent that the concentrations of cesium and iodine in the gas vented with rupture and the mass diffusional release rates were dependent upon the amount of each species in the gap space. In the tests with simulants the amounts of fission product simulants placed in the gap space were measured directly. Experimental determination of the amounts of fission gases within the interconnected voids of the Robinson fuel indicated that 0.25% of the long-lived fission gas inventory was released to the plenum and void spaces during reactor operation.³ The amount of cesium and iodine in these spaces (the gap inventory) subsequently was found to be approximately 0.3% of the total fuel rod inventory.³ These low release values are believed to be primarily the result of knockout (i.e., the release of atoms from the pellet surface region by the action of fission fragment recoil).

Fuel irradiated at higher heat rating (and thus reaching higher fuel temperatures) would be expected to release additional quantities of

Table 1. Summary of fission product release tests

Test No.	Temperature (°C)		Diffusion time (hr)	Volume of gas vented at 1 atm (cm ³ , 0°C)	Radial gap width ^a (μm)	Initial gap inventory (μg/cm ² , cladding)		Mass released with rupture ^b (μg)		Mass released ^b by diffusion ^b (μg)	
	Rupture	Diffusion				Cs	I	Cs	I	Cs	I
Implant-1 ^c	700	700	1.0	229	127	23.1	22.0	179.5	356	0.54	6.8
Implant-2	700	700	1.5	229	127	0.47	0.45	2.45	4.9	0.036	0.41
Implant-3	900	900	2.0	172	200	87.1	8.66	290	64.6	196	96.9
Implant-4		1100	0.8	0	127	67.6	6.05			1237	145
Implant-5	700	700	2.0	348	127	96.9	8.38	378	103	3.38	5.9
Implant-6		500	20.0	0	127	141.1	12.37			<43	5.8
Implant-8	900	1100	0.96	293	200	126.7	8.38	1918	240	1393	161
Implant-10		700	5.0	0	127	97.2	9.07			4.84	12
Implant-11		1300	0.25	0	127	113.9	7.97			2320	26
Implant-12	900	900	2.0	172	200	14.6	0.69	217	23.3	47	6.9
LBU-1 ^d		700	5.0	0	26 ^c	68.9	3.40			0.046	0.17
LBU-2		900	2.0	0	27	106.7	5.27			19.3	20.6
HBU-1 ^f		700	5.0	0	20	13.1	1.20			0.123	0.93
HBU-2		900	2.0	0	20	12.7	1.17			2.82	1.76
HBU-4		500	20.0	0	20	13.1	1.20			0.017	0.105
HBU-7	900	900	0.2	96	200	12.7	1.17	130.4	11.1	0.22	0.018
HBU-8	900	900	1.0	97	200	12.7	1.17	26.6	9.1	13.0	4.44
HBU-9	900	1100	0.14	96	200	13.1	1.20	94.1	14.2	18.2	2.74
HBU-10	900	1200	0.17	98	200	11.2	1.03	223.3	11.1	56.2	2.80
HBU-11		1200	0.45	0	200	11.3	1.05			142.0	20.2

^aFor pressure-ruptured test segments, a value of 200 μm represented the combined gap width and hole size.

^bFor pressure-ruptured tests, the fraction released by diffusion was estimated using the diffusion test data as a guide.

^cImplant Test Series cladding, 0.965 cm ID; fuel length 30.48 cm; 24-cm length of pellets coated with simulated fission products.

^dLow Burnup Fuel Test Series cladding, 1.27 cm ID; fuel length, 15.24 cm; in-reactor fission gas release, 11.6% (LBU-1) and 18.9% (LBU-2).

^eThe unusually low cesium and iodine release values suggest that the hole through the cladding might have been drilled where the gap width was much less than the average value of 26 μm.

^fHigh Burnup Fuel Test Series cladding, 0.948 cm ID; fuel length; 30.48 cm; in-reactor fission gas release, 0.25%.

fission gas, cesium, and iodine to the gap space by diffusion or other mechanisms at rates dependent upon the physical and chemical characteristics of each element. Postirradiation heating tests⁴ have shown that the rates of iodine and cesium release from the irradiated fuel matrix (1000-4000 MWd/MT) at high temperatures are approximately 2.5 times greater than the rates for xenon.

In the modeling calculations, in-reactor knockout release from the fuel surfaces was assumed to be 0.25% for all isotopes of xenon, cesium, and iodine. For fuel rods with in-reactor fission gas release greater than 0.25%, additional cesium and iodine releases to the gap were assumed to be 2.5 times the amount of fission gas in excess of 0.25%. As shown in a recent review,⁵ diffusion equations predict that the fractional releases of radioactive isotopes to the gap during normal reactor operation will be less than those of stable isotopes. According to the diffusion equations, the ratio of stable (and long half-life) isotopes to short half-life isotopes that are released to the fuel rod void spaces is

$$\frac{F_s}{F_r} = \frac{4}{3} \left(\frac{t\lambda}{\pi} \right)^{0.5},$$

where

F_s = fraction of stable and long half-life isotopes released

$$(F_s < 0.2),$$

F_r = fraction of the short half-life isotope (at production-decay equilibrium) released ($F_r < 0.2$),

t = total irradiation time in sec, and

λ = decay constant of the short half-life isotope in sec^{-1} .

Although the model equations (Sect. 3) indicate equal fractional escape from the gap regardless of half-life, the overall release based on the total fuel rod inventory will accordingly be less for the isotopes with shorter half-lives.

3. THE SOURCE TERM MODEL

Sufficient results have been obtained to permit the formulation of preliminary empirical models for the release of cesium and iodine in steam. The models assume that the release is the sum of two components: burst release (carried out with escaping plenum gas when the rod ruptures) and diffusional release (diffused from the gap space after the plenum gas has vented). The following two sections describe the derivation of the burst and diffusional release components of the model.

3.1 Burst Release (Release at Time of Rupture)

When the cladding ruptures, the fill and fission gases flow mainly from the plenum through the pellet-to-clad gap space and out the rupture opening. The escaping gas tends to become saturated with fission product vapors and reaches a maximum concentration at the rupture location, which is typically at the highest axial temperature. The mass of each fission product species released is therefore equal to the concentration of the species in the gas phase at the rupture location (assuming surface-to-gas-phase mass transfer equilibrium) multiplied by the volume of gas flowing out the rupture opening. The model assumes saturation, only as an upper limit on release. However, kinetic factors that determine in part the gas-phase concentrations of the fission product species are treated empirically.

Pertinent data from the implant and high burnup burst tests are listed in Table 2. In each test the masses of cesium and iodine released during rupture were determined by subtracting the estimated diffusional release component from the total released. As shown in Table 1, the amounts released following rupture were usually less than those released at the time of rupture. The volume of gas vented was calculated from the plenum and fuel rod void of rupture and the temperature distribution. The concentrations of cesium and iodine were then calculated simply as the ratio of mass released to volume of vented gas, with the volume calculated at 0°C and external system pressure.

Table 2. Release of cesium and iodine at time of rupture

	Rupture temperature (°C)	Initial gap inventory, M_0/A ($\mu\text{g}/\text{cm}^2$) (cladding)		Mass released with rupture ^a (μg)		Volume of gas vented at 1 atm (cm^3 , 0°C)	Mass concentration in vented gas ($\mu\text{g}/\text{cm}^3$, 0°C)	
		Cs	I	Cs	I		Cs	I
		Implant-1 ^b	700	23.1	22.0		179.5	356
Implant-2 ^b	700	0.47	0.45	2.45	4.9	229	0.0107	0.0214
Implant-3	900	87.1		290	64.6	172	1.686	0.356
Implant-5	700	69.9	8.38	378	103	348	1.086	0.296
Implant-8	900	126.7	8.38	1918	240	293	6.546	0.819
Implant-12	900	14.6	0.69	217	23.3	172	1.262	0.135
HBU-7	900	12.7	1.17	130.4	11.1	96	1.358	0.116
HBU-8	900	12.7	1.17	26.6	9.1	97	0.274	0.094
HBU-9	900	13.1		94.1	14.2	96	0.980	0.148
HBU-10	900	11.2	1.03	223.3	11.1	98	2.279	0.113

^aBest-estimate values.

^bCsI only implanted.

Examination of the data indicated that the concentrations of cesium and iodine in the vented gas were dependent upon the amounts of these elements existing in the gap space. This dependence is shown in Fig. 1 for cesium and in Fig. 2 for iodine. The amount in the gap space is presented as mass (μg) per cm^2 of cladding area. By this we do not mean to imply that the fission products are located on the cladding; this is only a means of compensating for different fuel rod diameters. For the Implant Test Series, the amount of cesium and iodine placed in the gap space was measured directly. For the High Burnup Test Series, we assumed that the fractions of the total amounts of cesium and iodine which are associated with the gap were the same as the fractional amounts of fission gas released to the plenum and void spaces (i.e., approximately 0.0025). This was later shown to be essentially correct, when approximately 0.3% of the cesium and iodine was observed to vaporize when purified helium was purged through the gap space of a fuel rod segment.³

The scatter of the data presented in Figs. 1 and 2 demonstrates that dependence on concentration and temperature cannot be determined precisely from the limited data available. The lines drawn in the figures represent what we believe these dependences to be.

In the temperature range of 700-900°C, burst release can be expressed as

$$M_B = \alpha V_B (M_O/A)^a e^{-(C/T)}, \quad (2)$$

where

M_B = mass (in g) of cesium or iodine (total of all isotopes) released in the burst,

V_B = volume of plenum gas (in cm^3) vented at 0°C and system pressure,

M_O = inventory (in g) of cesium or iodine (total of all isotopes) in the pellet-cladding gap,

A = internal area (in cm^2) of the cladding associated with M_O , and

T = temperature (in K) at rupture location.

The parameters α , a , and C are adjustable constants that are derived from the experimental data. The pertinent values are listed in Table 3. (Note that M_O should not be confused with the total mass of cesium or iodine in the fuel.)

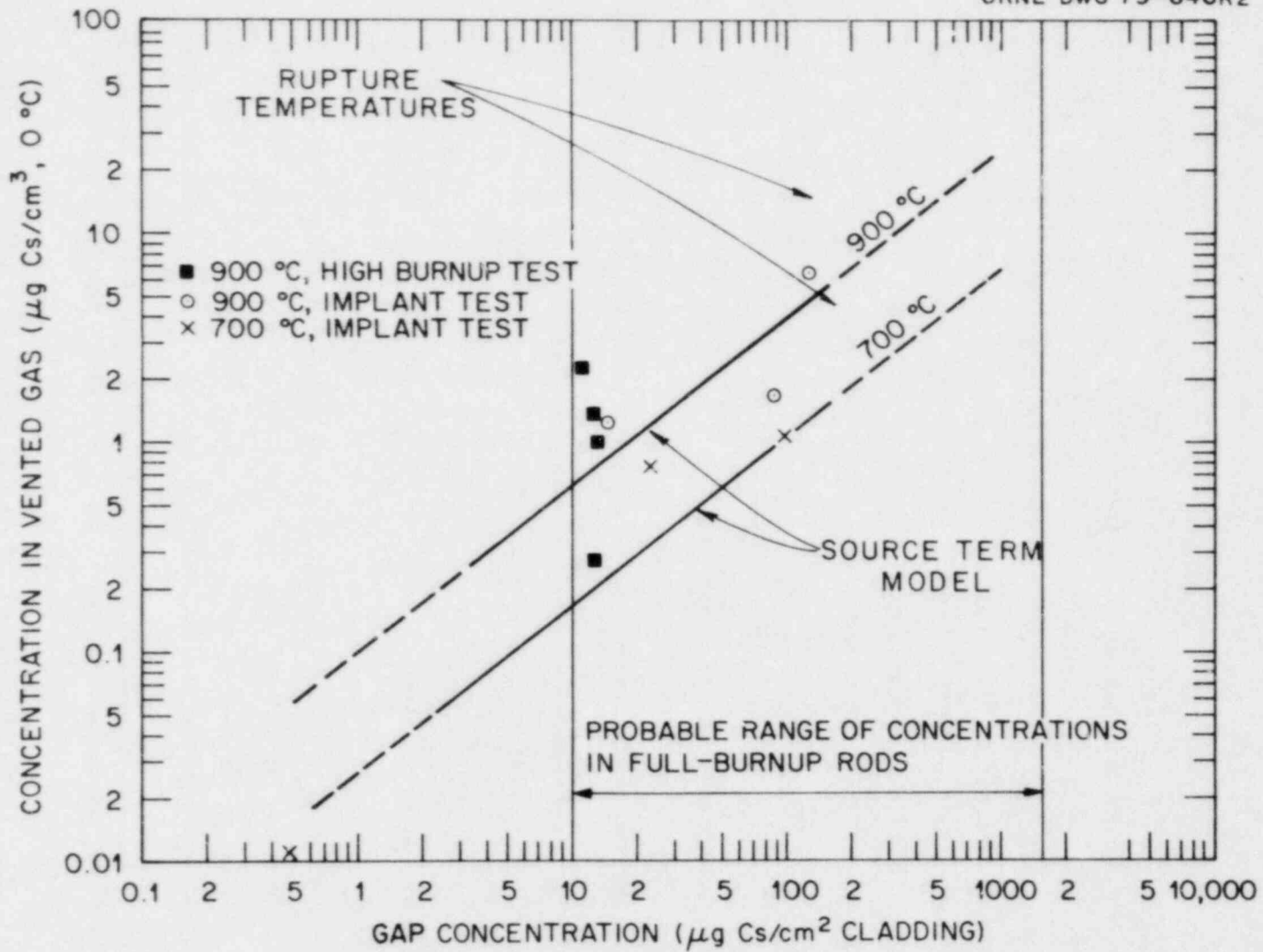


Fig. 1. Concentration of cesium in gas vented at rupture.

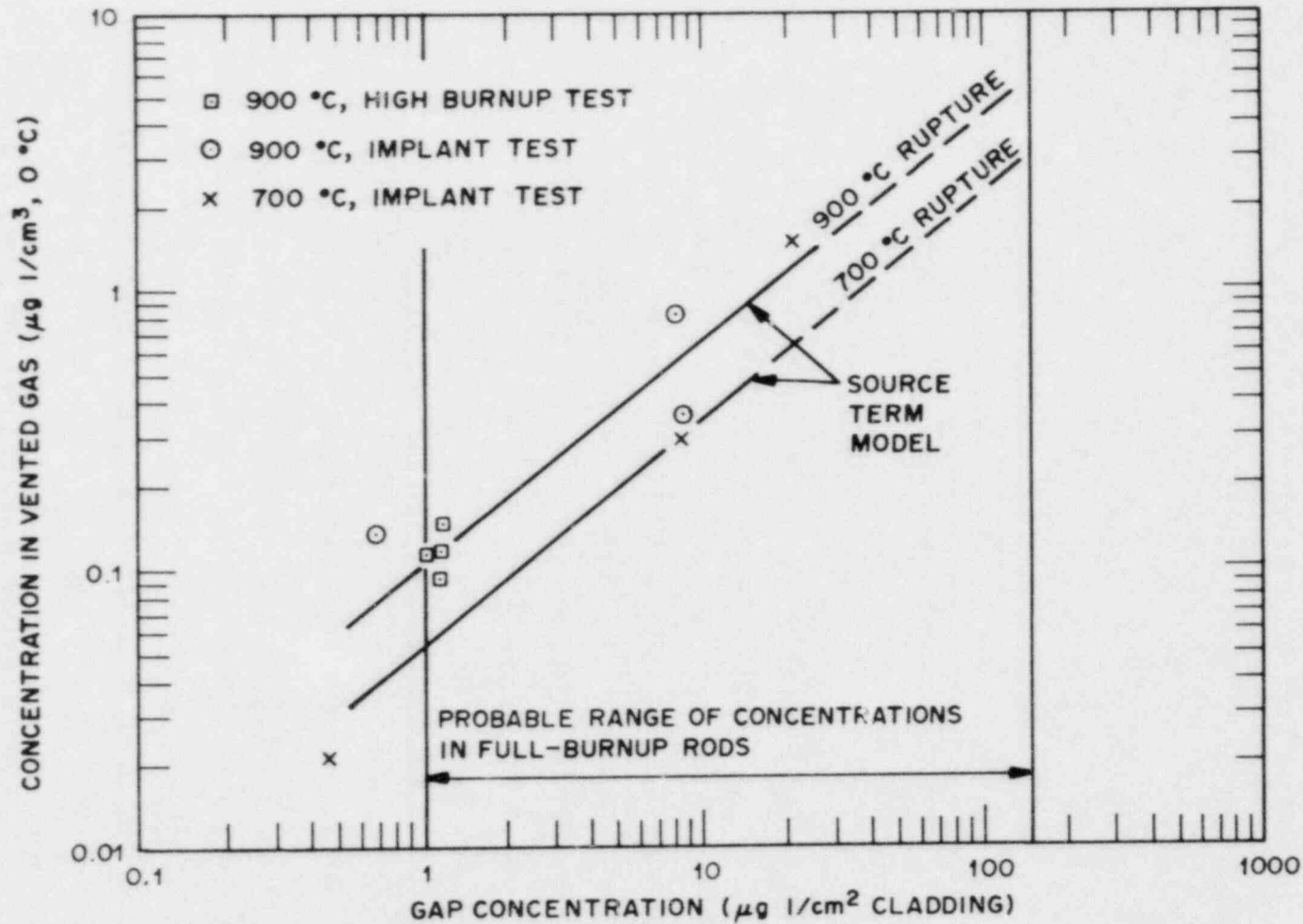


Fig. 2. Concentration of iodine in gas vented at rupture.

Table 3. Values of parameters for cesium and iodine release

Parameter	Cesium	Iodine
$\alpha, (\text{g/cm}^3) \cdot (\text{g/cm}^2)^{-a}$	3.49	0.163
a	0.8	0.8
C, K^{-1}	7.42×10^3	3.77×10^3
$\delta, (\text{g} \cdot \text{MP}_a / \mu\text{m} \cdot \text{h}) \cdot (\text{g/cm}^2)^{-a}$	1.90×10^3	1.22×10^2
γ, K^{-1}	1.98×10^4	1.48×10^4

3.2 Diffusional Release from the Gap Space

The 13 tests identified in Table 4 provided data for the diffusional release of cesium and iodine from the gap space of implant, low burnup, and high burnup fuel rods. Seven of the tests were performed with a defect made by drilling a 0.159-cm-diam hole through the unexpanded Zircaloy cladding. The cross-sectional area of these holes was 0.0198 cm^2 . In tests Implant-4 and Implant-11, the fuel rods were pressurized for rupture, but the cladding did not expand and rupture in the usual manner because of leaks at ferrule fittings. Release is believed to have occurred through crack-like openings in the cladding in a diffusional manner. Because of uncertain and atypical release conditions, the results from these tests are included only for reference.

Three tests (HBU-8, -9, and -10) were performed with fuel rods ruptured by internal pressure; the cladding had expanded and ruptured to form holes with open areas of approximately $0.02\text{-}0.04 \text{ cm}^2$. The release values measured in these three tests were a combination of burst release and diffusional release, so that only maximum and minimum values for diffusional release could be obtained. Test HBU-11 was performed with a fuel rod segment previously ruptured in test HBU-7. The test rod was reinstalled in clean apparatus for test HBU-11; thus, all of the HBU-11 release was by diffusion from the gap space.

Table 4. Diffusional release of cesium and iodine from the gap space

Test No.	Initial gap inventory, M_0/A ($\mu\text{g}/\text{cm}^2$)		Initial mass in gap, M_0 (μg)		Mass released by diffusion (μg)		Initial release rate, R_0 ($\mu\text{g}/\text{hr}$)		R_0 at $W = 200 \mu\text{m}$, $M_0/A = 12.9 \mu\text{g Cs}/\text{cm}^2$, $1.19 \mu\text{g I}/\text{cm}^2$ ($\mu\text{g}/\text{hr}$)	
	Cs	I	Cs	I	Cs	I	Cs	I	Cs	I
Implant-4 ^a	67.6	6.05	3280 ^a	293 ^a	1237	145	1941	250	812	107
Implant-6	141.1	12.37	10270	900	≤ 43	5.8	-	0.29	-	0.070
Implant-10	97.2	9.07	7070	660	4.84	12	0.97	2.4	0.304	0.74
Implant-11 ^a	113.9	7.97	5527 ^a	387 ^a	2320	26	12034	$>108^b$	3320	$>37.1^b$
LBU-1	68.3	1.47	4186 ^c	207 ^c	0.046	0.17	0.0092	0.034	0.0241	0.22
LBU-2	106.7	2.44	6486 ^c	321 ^c	19.3	20.6	9.7	10.6	13.25	44.4
HBU-1 ^d	13.1	1.20	1172	108	0.123	0.93	0.0246	0.186	0.243	1.85
HBU-2	12.7	1.17	1143	105	2.82	1.76	1.41	0.89	13.92	9.02
HBU-4	13.1	1.20	1172	108	0.017	0.105	0.00085	0.0053	0.0084	0.053
HBU-8 (max) ^e	12.7	1.17	600	55.3	≤ 39.6	≤ 13.5	≤ 41.0	≤ 15.5	≤ 41.5	≤ 15.7
HBU-9 (max) ^e	12.5	1.01	592	47.6	≤ 85.7	≤ 7.82	≤ 661	≤ 61.0	≤ 678	≤ 46.8
HBU-10 (min) ^e	8.7	0.82	411	39	≥ 161.6	≥ 3.9	≥ 1208	≥ 24.2	≥ 1655	≥ 32.6
HBU-10 (max) ^e	10.7	0.86	505	41	≤ 255.4	≤ 5.7	≤ 2093	≤ 36.1	≤ 2430	≤ 46.8
HBU-11	11.3	1.05	1012	97	142.0	20.2	340	50.3	378	55.6

^a Attempt to rupture by internal pressurization at 900°C produced only a diffusion-type leak. Above interpretation is questionable. Initial masses based on 16 cm of heated length.

^b Axial migration of iodine to the cool ends probably depleted the available inventory in an unrealistic manner.

^c Cesium and iodine gap inventories were estimated by assuming their release rate from UO_2 to be 2.5 times greater than that from xenon.

^d Gap inventories for high burnup fuel were estimated to be 0.25%, the same as for xenon. Gap inventories of cesium and iodine measured in Test HBU-12 were 0.3%.

^e Gap inventories for pressure-ruptured fuel rods calculated for postblowdown amounts. Initial masses calculated for postblowdown and 16 cm of heated length.

Although it was recognized that the rate of release should decrease as the test proceeded and the gap inventory diminished, attempts to quantify this change by monitoring iodine release in the Implant Test Series and cesium release in the Low Burnup and High Burnup Test Series were unsuccessful because of holdup on the quartz surfaces surrounding the test fuel rods.

Cesium and iodine release by diffusion from the gap space was assumed to be simple diffusion from a depleting source. Thus the rate of diffusional release can be expressed in the form

$$M_D = M_O \left[1 - e^{-(R_O t / M_O)} \right] \quad (3)$$

where

M_D = mass (in g) of cesium or iodine released by diffusion,

M_O = total calculated mass (in g) of cesium or iodine initially in the gap,

t = time (in hr) at diffusion temperature, and

R_O = initial rate (in g/hr) of release of cesium or iodine by diffusion.

In the case of the diffusional release tests, M_D was measured in each test, M_O was estimated as described earlier in the discussion of gap inventory (or measured directly for implant tests), and t was the time at temperature. With the aid of Eq. 3, R_O was then calculated. For induction-heated tests, M_O represents the gap inventory in the heated length of 16.5 cm instead of the full test rod length of 30.5 cm. (For test HBU-11, because of the slow heatup and cooldown times, an effective time at temperature of 0.45 hr was used rather than the time at maximum temperature.) The resulting values of R_O are listed in Table 4.

As indicated previously, hole sizes were of the order of 0.16 cm in diameter. For fuel rod segments with drilled-hole defects, the radial gap width of the unexpanded cladding was 20 μm . The gap width in pressure-ruptured test segments ranged from 20 μm at ends to several millimeters at the rupture location. For these segments, we estimated the effective gap width to be about 200 μm . For diffusional release, it was therefore assumed that transport along the gap, rather than through the rupture

opening, limited the rate of release. In addition, a dependence upon gap concentration, like that employed to describe the burst release component, was also assumed for diffusional release.

The experimental diffusion release data shown in Table 4 were therefore normalized by direct proportion to a radial gap width of 200 μm (the effective width for pressure-ruptured rods) and to the average gap concentration estimated to exist in the high burnup test rods using gap concentration raised to the 0.8 power. The results of this normalization are listed in the last two columns of Table 4 and are plotted in Fig. 3 for cesium and in Fig. 4 for iodine.

The resulting correlation for the initial rate of release R_0 in the temperature range of 500-1200°C is given by

$$R_0 = \delta (W/P) (M_0/A)^a e^{-(\gamma/T)}, \quad (4)$$

where

W = width (in μm) of radial gap, and

P = system pressure (in MPa).

Values of the adjustable constants δ and γ are presented in Table 3.

4. LIMITATIONS OF THE SOURCE TERM MODEL

A measure of the precision of the model was obtained by comparing the model predictions with the experimental data. This is done graphically in Fig. 5 for both cesium and iodine. The test parameters ranged as follows: V_B from 0 to 348 cm^3 (STP); M_0/A for cesium from 5×10^{-7} to 1.27×10^{-4} g/cm^2 of cladding; M_0/A for iodine from 5×10^{-7} to 2.2×10^{-5} g/cm^2 of cladding; W from 20 to 200 μm (a ruptured rod with expanded cladding was assumed to have an effective gap width of 200 μm); P was 0.101 MPa (atmospheric pressure); and the length of the test rod from 15 to 30.5 cm. The dashed lines in Fig. 5 represent a deviation of a factor of three from the model source terms. We believe this to represent good precision, considering the wide variety of types of test fuel rods and the range of five-to-six orders of magnitude in mass released. Of course, one would expect general agreement because the source term models were derived from these data.

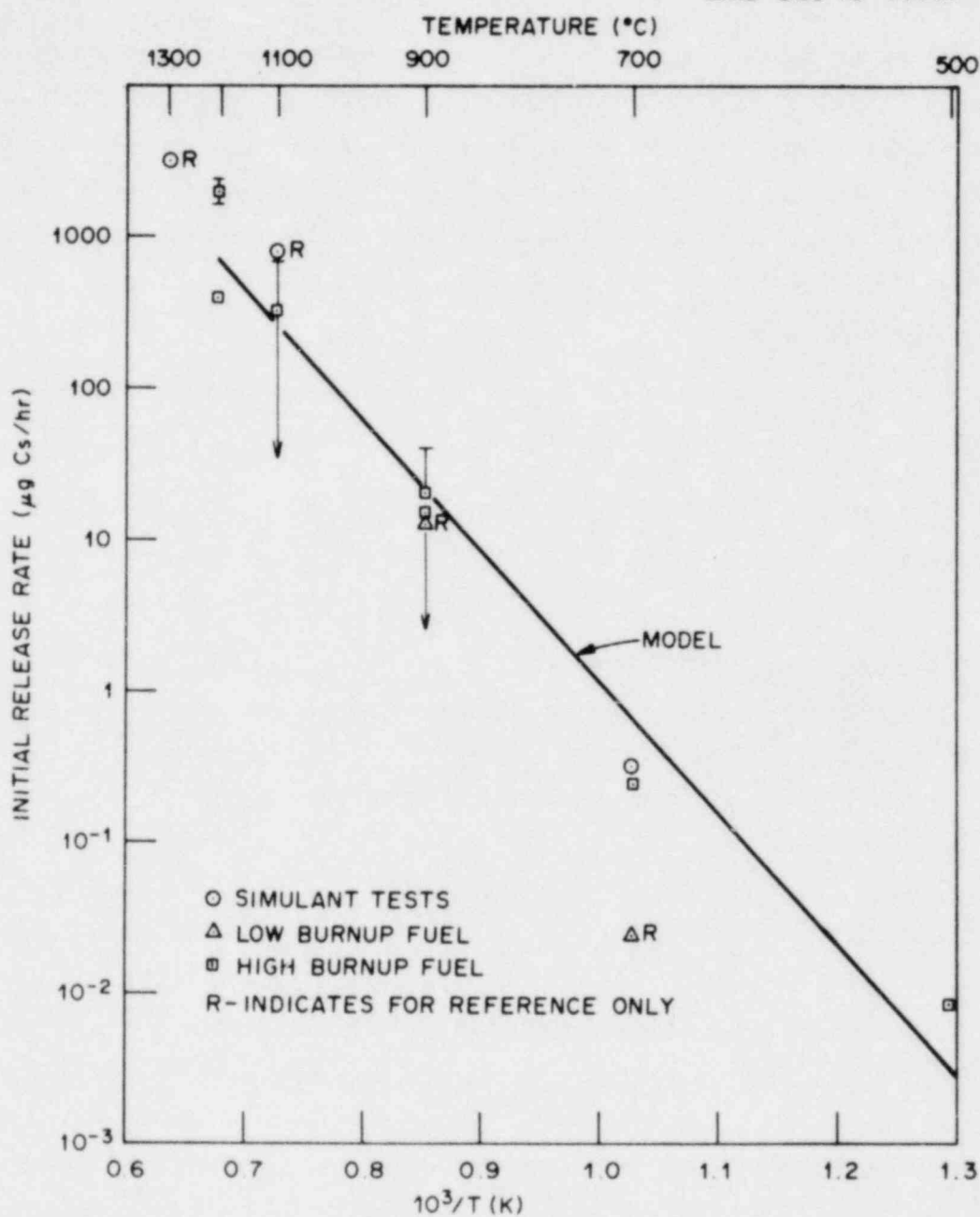


Fig. 3. Initial rate of release of cesium from a fuel rod ruptured in steam containing $12.9 \mu\text{g Cs/cm}^2$ cladding in the gap space.

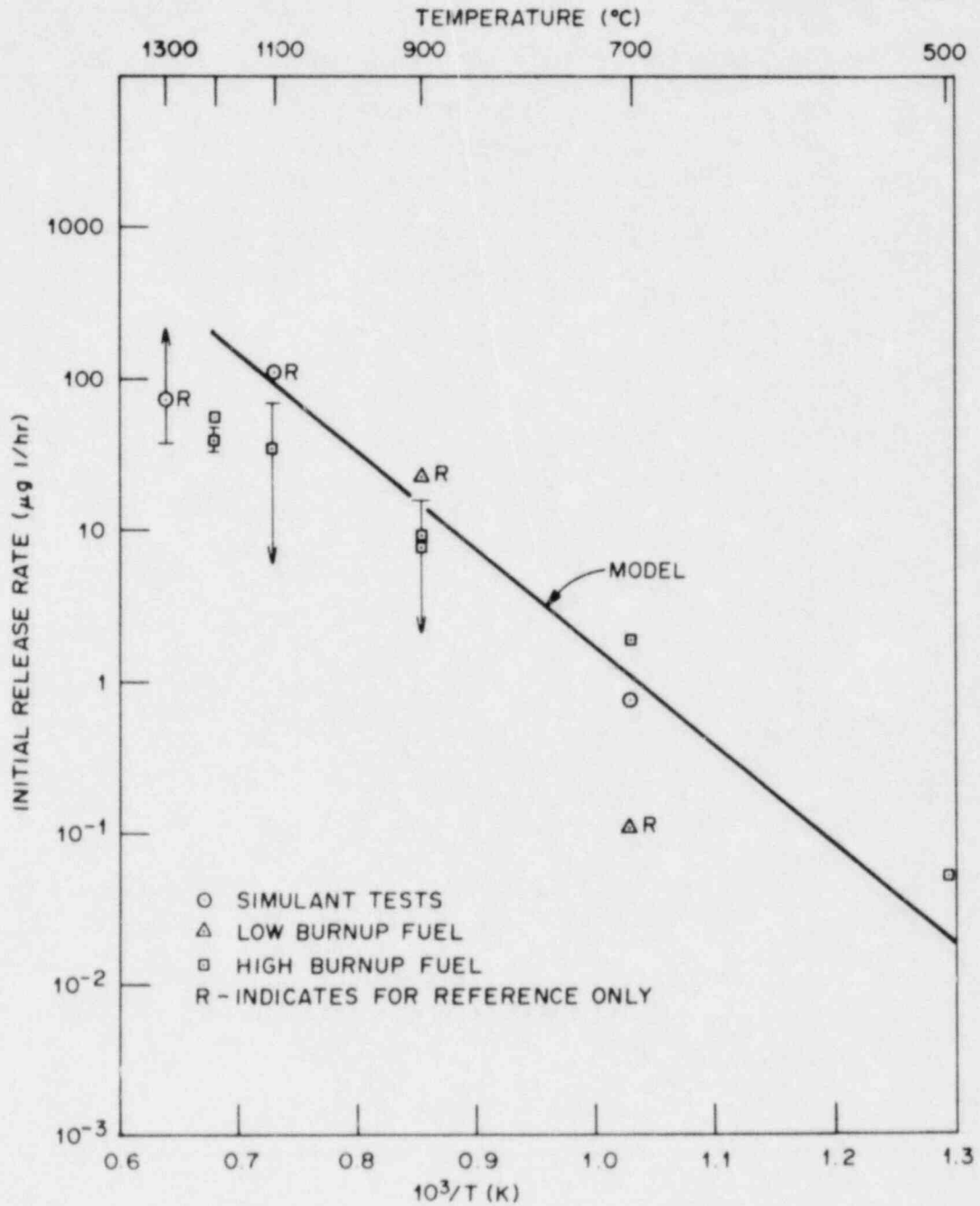


Fig. 4. Initial rate of release of iodine from a fuel rod containing $1.19 \mu\text{g I/cm}^2$ cladding in the gap space ruptured in steam.

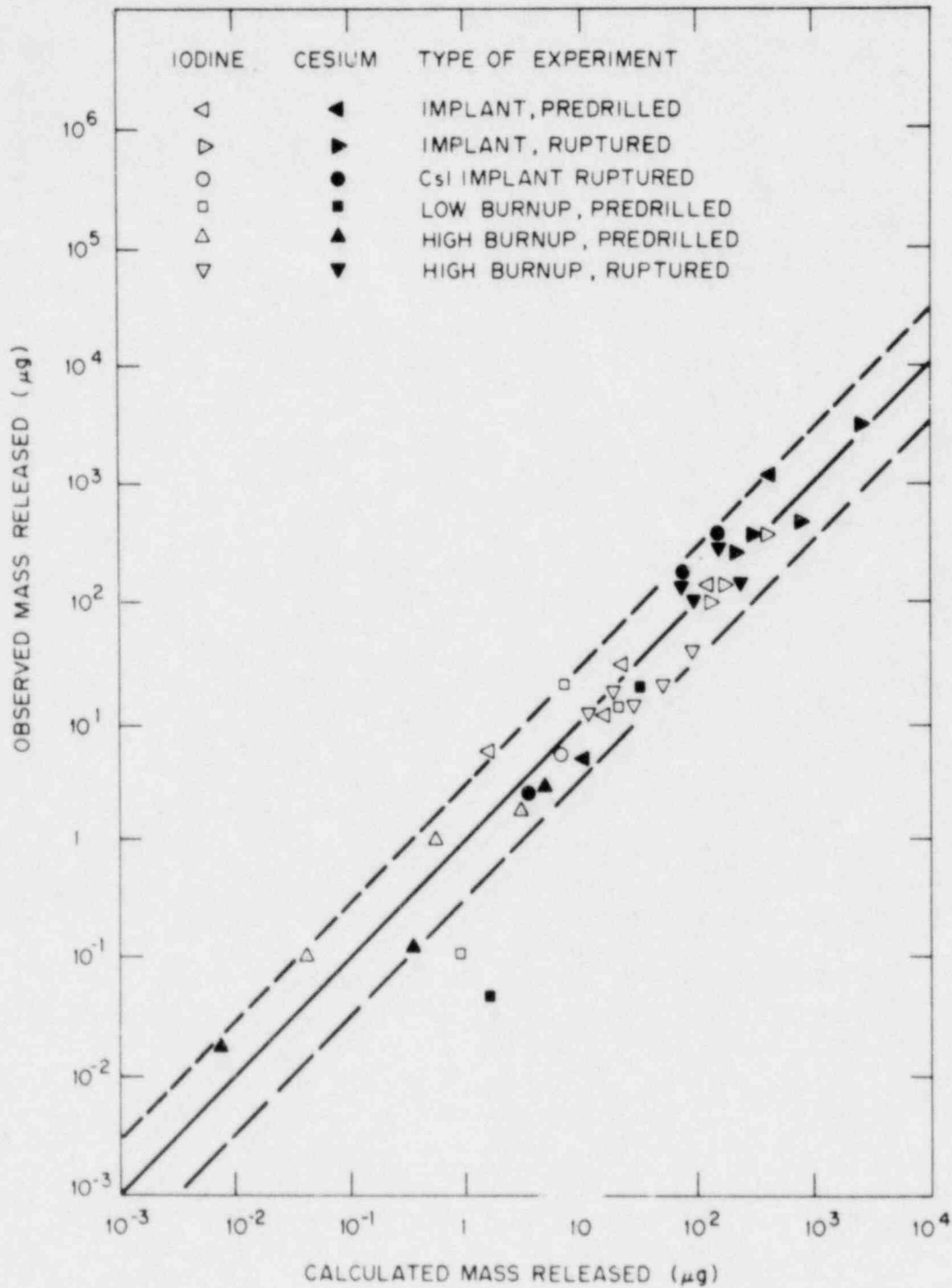


Fig. 5. Comparison of calculated and observed releases of cesium and iodine in steam.

The burst release component for both cesium and iodine is essentially a product of two terms that is integrated over the time period defining burst release. These components are the flux of plenum gas released during rupture and the effective concentration of the particular fission product in the gas phase during the rupture period. The former of the two components is virtually independent of the position at which cladding failure occurs. On the other hand, the concentration of fission product in the gas phase is temperature dependent, so the position at which cladding failure occurs will influence the amount released upon failure. However, as our experiments indicate, so little gap inventory is released over the temperature range of interest that the length of the fuel rod would have little influence on the mass of material released during depressurization of the rod. (There would be an artificial dependence on fuel rod length, on the other hand, if release were described as a fraction of the total inventory or as a fraction of the gap inventory.)

Similarly, the diffusion release component over relatively brief time periods (on the order of 10 min at 1200°C, for example) involves only the fission product material that is located within a few centimeters of the point of rupture. For most applications, therefore, the length of the fuel rod has little effect on the amount (mass) of material released.

In general we recommend applying the source term models only to situations within the range of the test parameters. It is obvious that the models must be used with full length fuel rods in order to have practical value. The source terms were written with this in mind, and we believe that they are fully applicable. In the case of burst release, the full-length rods may contain larger quantities of gas (measured in moles) than our test rods, but rupture in a LOCA will likely occur with the pressure vessel at several atmospheres of pressure, so that the actual volume of gas vented at system pressure will be within the recommended range. Because of the greater distance between plenum and rupture location, the full-length rods will vent at a slower rate (if the gap width is small), except for the quantity of gas held within the expanded cladding near the point of rupture. Very slow venting might increase

the degree of saturation of fission product vapors in the escaping gas, but we do not believe that this effect is significant when viewed in terms of the normal scatter of experimental release data.

It is probable that some of the higher-powered fuel rods in a reactor will contain gap inventories of cesium and iodine higher than those employed in the tests conducted to date (Figs. 1 and 2). Additional tests with another group of irradiated fuel rods containing higher gap inventories are planned to demonstrate the applicability of the release models to this type of fuel.⁶

5. APPLICATION OF THE MODEL TO A PWR LOCA

The models were applied to the analysis of a PWR LOCA in which the reactor characteristics were assumed to be as follows: 33,000 fuel rods, each containing 2500 g of UO_2 and operated so that equal numbers of rods have average burnups of 10,000, 20,000, and 30,000 MWd/MT. The fuel rods were divided into six groups, each containing the amount of released fission gas shown in Table 5. This distribution is based on calculations of fission gas release for a typical PWR.⁷ The amount of pressurized helium fill gas, the estimated rupture temperature, and the calculated amount of vented gas V_B (vented to the primary vessel at 0.303 MPa) are also shown. Because the fuel rods would rupture near their centers, the cesium and iodine gap inventories were based on peak burnups that are estimated to be 10% higher than the average burnups discussed above. It was further assumed that all of the fuel rods ruptured during the course of the accident and, moreover, experienced a temperature transient to 1200°C for an effective time of 10 min.

Calculations using the models are summarized in Tables 6 and 7. Total reactor inventories are 1.208×10^5 g of cesium and 1.112×10^4 g of iodine; the releases of cesium and iodine are equivalent to 0.025% and 0.053%, respectively, of these totals. The fission gas initially in the plenum and void spaces, 1.27% of the total inventory, will also be released. However, based on our tests with high burnup fuel, we would expect that during the heatup to 1200°C additional fission gas release

Table 5. Assumed characteristics of fuel rods in example PWR

Number of rods ^a	Fission gas initially in plenum at STP		Helium fill gas at STP (cm ³)	Rupture temperature (°C)	Amount of vented gas at 0°C and 0.303 MPa ^b
	%	cm ³			
130	8	163.3	750	830 ^c	291.4 ^c
130	8	108.9	750		
130	8	54.4	750		
920	4	81.6	750	855 ^c	264.2 ^c
920	4	54.4	750		
920	4	27.2	750		
1990	2	40.82	750	860 ^c	250.6 ^c
1990	2	27.21	750		
1990	2	13.61	750		
3450	1	20.41	750	860 ^c	243.8 ^c
3450	1	13.61	750		
3450	1	6.80	750		
2640	0.5	10.20	750	860 ^c	240.4 ^c
2640	0.5	6.80	750		
2640	0.5	3.40	750		
1870	0.25	5.10	750	860 ^c	238.7 ^c
1870	0.25	3.40	750		
1870	0.25	1.70	750		

^aThe three groups of equal rod numbers correspond to burnups of 30,000, 20,000 and 10,000 MWd/MT; fuel length, 365.8 cm; cladding internal area adjacent to fuel, 1069.3 cm².

^bAssumes that 13 cm³ (at 0°C and 0.303 MPa) remains in fuel rod.

^cFor simplicity, a single rupture temperature and amount of vented gas was used for each group of rods.

Table 6. Release of cesium from example PWR LOCA

Gap inventory (%)		Number of rods ^a	Cesium inventory, ^b M/A (10 ⁻⁶ g Cs/cm ²)	Cesium released by burst, each rod (10 ⁻⁶ g Cs)	Total burst release, all rods (g Cs)	R _o each rod ^c (10 ⁻⁶ g Cs/hr)	M _o [1 - e ^{-(R_ot/M_o)}] each rod (10 ⁻⁶ g Cs/rod)	Cesium released by diffusion, all rods (g Cs)	Total burst plus diffusion release (g Cs)
Xe	Cs								
8	19.63	130	1088	5198	0.676	7798	1299	0.169	0.845
		130	725	3759	0.489	5638	940	0.122	0.611
		130	363	2160	0.280	3238	540	0.070	0.350
					1.445			0.361	1.806
4	9.63	920	534	3095	2.847	4411	735.1	0.676	3.523
		920	356	2239	2.060	3188	531.3	0.489	2.549
		920	178	1284	1.181	1832	305.2	0.281	1.462
					6.088			1.446	7.534
2	4.63	1990	256.4	1681	3.345	2453	408.8	0.813	4.158
		1990	170.9	1216	2.419	1773	295.5	0.588	3.007
		1990	85.5	698	1.389	1018	169.7	0.338	1.727
					7.153			1.739	8.892
1	2.13	3450	117.8	878	3.029	1316	219.3	0.757	3.786
		3450	78.55	635	2.191	952	158.7	0.547	2.738
		3450	39.27	364	1.256	55	91.1	0.314	1.570
					6.476			1.618	8.094
0.5	0.88	2640	48.52	426	1.125	648	108.0	0.285	1.410
		2640	32.34	308	0.813	468	78.0	0.206	1.019
		2640	16.17	177	0.467	269	44.9	0.118	0.585
					2.405			0.609	3.014
0.25	0.25	1870	13.86	155	0.290	238	39.6	0.074	0.364
		1870	9.24	112	0.209	172	28.6	0.053	0.262
		1870	4.62	64	0.120	99	16.5	0.031	0.151
					0.619			0.158	0.777
Core total					24.186			5.931	30.117

^aThe three groups for each fission gas release percentage correspond to burnups of 30,000, 20,000, and 10,000 MWd/MT.

^bFor each rod irradiated to 30,000 MWd/MT av burnup, total mass of cesium = 5.491 g, cladding area = 1089.3 cm³, and peak-to-av burnup = 1.1.

^cFor the case where T = 1473.2, P = 0.303 MPa, and W = 200 μm.

Table 7. Release of iodine from example PWR

Gap inventory (%)		Number of rods ^a	Iodine inventory, M_0/λ (10^{-6} g I/cm ²)	Iodine released with burst, each rod (10^{-6} g I)	Total burst release, all rods (g I)	R_0 each rod ^c (10^{-6} g I/hr)	$M_0 [1 - e^{-(R_0 t/M_0)}]$ each rod (10^{-6} g Cs/rod)	Iodine released by diffusion, all rods (g I)	Total burst plus diffusion release (g I)
Xe	I								
8	19.63	130	100.2	985.6	0.1281	2210.8	368.3	0.0479	0.1760
		130	66.8	712.7	0.0927	1598.6	266.3	0.0346	0.1273
		130	33.4	409.4	0.0532	918.2	153.0	0.0199	0.0731
					0.2740			0.1024	0.3764
4	9.63	920	49.13	545.0	0.5014	1250.5	208.4	0.1917	0.6931
		920	32.75	393.8	0.3623	903.6	150.6	0.1385	0.5008
		920	16.38	226.3	0.2082	519.2	86.5	0.0796	0.2878
					1.0719			0.4098	1.4817
2	4.63	1990	23.61	292.0	0.5811	695.7	115.95	0.2307	0.8118
		1990	15.74	211.1	0.4201	502.9	83.82	0.1668	0.5869
		1990	7.869	121.2	0.2412	288.8	48.13	0.0958	0.3770
					1.2424			0.4933	1.7357
1	2.13	3450	10.85	152.5	0.5261	373.5	62.25	0.2148	0.7409
		3450	7.231	110.2	0.3802	269.9	44.99	0.1552	0.5354
		3450	3.615	63.3	0.2184	155.0	25.84	0.0891	0.3075
					1.1247			0.4591	1.5838
0.5	0.88	2640	4.466	73.92	0.1951	183.6	30.60	0.0808	0.2759
		2640	2.977	53.43	0.1411	132.7	22.12	0.0584	0.1995
		2640	1.489	30.71	0.0811	76.3	12.71	0.0336	0.1147
					0.4173			0.1728	0.5901
0.25	0.25	1870	1.276	26.95	0.0504	67.40	11.23	0.0210	0.0714
		1870	0.8507	19.48	0.0364	48.72	8.12	0.0160	0.0524
		1870	0.4253	11.19	0.0209	27.99	4.67	0.0087	0.0296
					0.1077			0.0457	0.1534
Core total					4.2380			1.6831	5.9211

^aThe three groups for each fission gas release percentage correspond to burnups of 30,000, 20,000, and 10,000 MWd/MT.

^bFor each rod irradiated to 30,000 MWd/MT av burnup, total mass of iodine = 0.5055 g, cladding area = 1089.3 cm², and peak-to av burnup = 1.1.

^cFor the case where T = 1473.2K, P = 0.303 MPa, and W = 200 μ m.

would be equal to approximately 1.5% of the total inventory. The total fission gas release would therefore be approximately 2.77%.

6. COMPARISON WITH THE WASH-1400 LOCA

It is interesting to compare these release values with those used in WASH-1400.⁸ Values for gap inventory and gap escape fraction (the fraction of gap inventory that escapes following rupture) are listed in Table 8. These best-estimate predictions of total cesium and iodine release differ by factors of 200 and 60 respectively. These large differences are due to two factors: a lower estimate for initial fission gas inventory than used in WASH-1400 and lower estimates of the gap escape fractions. The lower estimate for initial fission gas inventory in the example PWR⁶ is believed to be partly a result of the attainment and recognition of lower peak-to-average power ratios throughout the modern reactor. On the other hand, the high gap escape fractions used in WASH-1400 were based upon estimated volatilities, because the available experimental data were both sparse and inconsistent.

Table 8. Comparison of WASH-1400 and Model calculations

Element	Gap inventory ^a (% of total inventory)		Gap escape fraction (% of gap inventory)		Total release (% of total inventory)	
	WASH-1400	Model	WASH-1400	Model	WASH-1400	Model
Xe and Kr	8	1.27	100	100 ^b	8	1.27 ^b
Cs	15	2.79	33	0.89	5	0.025
I	10	2.79	33	1.91	3.3	0.053

^aCalculated for stable and long half-life isotopes. The gap inventory and total release of ¹³¹I and ¹³³Xe would be 3-9 times lower.

^bAn additional amount of fission gas, approximately 1.5% of the total inventory, would be released during heatup.

As outlined in Sect. 2, diffusion equations⁵ predict that the fractional release of radioactive isotopes from the fuel to the plenum and void spaces will be less than for stable isotopes. For the sample PWR discussed in this report, we would estimate the release of ^{131}I and ^{133}Xe to the gap and plenum to be from 3 to 9 times less than that for stable isotopes of the same elements.

7. CONCLUSIONS

The model is believed to satisfactorily predict "best estimate" cesium and iodine release from full-length fuel rods in steam, provided the range of temperatures, times, and gap inventories employed in the tests are not exceeded. Some fuel rods in operating reactors will likely have gap inventories of cesium and iodine higher than those used in our tests (approximately 20% of the rods in our sample PWR, for example).

The method employed for the estimation of gap inventories is only tentative and has not been verified by testing of irradiated fuel rods. Furthermore, in correlating our test data and for the sample PWR LOCA, it was assumed that pressure-ruptured rods with expanded cladding continue to release cesium and iodine by a diffusional process that is characteristic of transport through an effective gap width of 200 μm . If greater expansion is expected (along sufficient axial length of a fuel rod to supply the calculated quantities escaped) with a correspondingly large rupture opening, it would be appropriate to use a gap width larger than 200 μm for the diffusional release predictions.^{9,10} Moreover, if pressure fluctuations during the diffusional release period are large enough to pump out additional fission product vapors, this effect must likewise be considered in the diffusional release calculations. Also, the Source Term Model does not include the amounts of cesium and iodine ejected as fuel dust at the time of rupture. This should amount to approximately 0.003% of the fuel and its associated fission products for a full-length rod.³

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