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

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1.0 INTRODUCTION

The principal objectives of the fission product release program currently in progress at Oak Ridge National Laboratory are to determine the quantity of radiologically significant fission products released from defected light water reactor (LWR) fuel rods under accident conditions, identify their chemical and physical forms, and interpret the results for use as input to computer models of postulated spent fuel transportation accidents (SFTAs) and loss-of-coolant accidents (LOCAs). The purpose of this ~~report~~ paper is to summarize the source term models which have been developed for cesium and iodine by this program, and to demonstrate the application of the source term models to the analysis of cesium and iodine release during a Pressurized Water Reactor (PWR) LOCA.

The models for fission product release in steam are based on tests conducted over the temperature range 500 to 1200°C. One series of tests, the Implant Test Series,[1] employed simulated fission products (CsI, CsOH, and TeO_2) which were coated on unirradiated UO_2 fuel pellets; a second series, the Low Burnup Fuel Test Series,[2] used fuel capsules irradiated to 1000 MWd/MT at high heat-rating (560 to 660 W/cm); and a third series of experiments, the High Burnup Test Series,[3-7] used fuel irradiated to 30,000 MWd/MT in the H. B. Robinson reactor at low heat-rating (175 to 320 W/cm).

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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 and was allowed additional time for release of fission products from the rupture opening by gas-phase diffusion. The cross-sectional area of the rupture opening ranged from 0.002 to 0.8 cm^2, with most in the 0.02 to 0.04 cm^2 range. The other tests were designed for measuring only diffusional release from the defected rod; the defect was formed in the unpressurized rod by drilling a 0.159-cm-diam hole through the cladding. Test HBU-11 was an exception in that the previously ruptured fuel rod segment from test HBU-7 was employed in a higher-temperature diffusional release test.

2.0 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 Implant Test Series, the amounts of fission product simulants placed in the gap space were measured directly. The High Burnup fuel was found to have released 0.25% of the fission gas to the plenum and void spaces during reactor operation, and we recently measured the cesium and iodine gap inventories of these fuel rods to be approximately 0.3%.[7] These low release values are believed to be primarily the result of knockout, the release of atoms from the pellet surface region by the action of fission fragment recoil.

Fuel irradiated at higher heat rating (higher temperatures) would be expected to release additional quantities of fission gas, cesium, and iodine

to the gap space by diffusion or other mechanisms at rates which could well depend upon the physical and chemical characteristics of each element. Postirradiation heating tests[8] have shown that the rates of release of iodine and cesium from irradiated fuel (1000 to 4000 MWd/MT) at high temperatures are approximately 2.5 times greater than the rates for xenon.

In our modeling calculations, we assumed that knockout release in-reactor from the surfaces of the fuel was 0.25% for all isotopes of xenon, cesium, and iodine. For fuel rods with fission gas release in-reactor greater than 0.25%, we estimated additional cesium and iodine releases to the gap to be 2.5 times the additional amount of fission gas. As shown in a recent review,[9] diffusion equations predict that the fractional releases of radioactive isotopes to the gap during normal reactor operation will be less than those for stable isotopes. Although our model equations (Sect. 3.0) indicate equal fractional escape from the gap, regardless of half-life, the overall release based on total fuel rod inventory will accordingly be less for the isotopes with shorter half-lives.

3.0 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 (that carried out with escaping plenum gas when the rod ruptures), and diffusional release (that diffusing from the gap space after the plenum gas has vented).

The mass of cesium which escapes as "burst release" was found to be equal to the volume of plenum gas vented, multiplied by the concentration

of cesium in vapor form. Cesium concentration in the vented gas has been determined to be a function of absolute temperature and the calculated inventory of cesium in the gap space. In the temperature range 700 to 900°C, cesium burst release can be expressed as

$$M_B = V_B\left[\frac{(G)^{0.8}}{7.74}\right]\left[e^{\left(6.06 - \frac{7420}{T}\right)}\right], \qquad (1)$$

where

M_B = mass of cesium released in the burst, μg Cs,

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

G = inventory of cesium (total of all isotopes) in the pellet-cladding gap per unit area of cladding, μg Cs/cm^2, and

T = temperature at rupture location, K.

In this equation the term in the first bracket is an empirical expression of the effect of the gap inventory on the concentration of cesium in the gas phase. The second bracket is a dimensional empirical expression for the concentration of cesium in the vented gas.

A simple depletion equation is used to model cesium release by diffusion in steam over the temperature range 500 to 1200°C,

$$M_D = M_o\left[1 - e^{-\frac{R_o t}{M_o}}\right], \qquad (2)$$

where

M_D = mass of cesium released by diffusion, μg Cs,

M_o = total calculated mass of cesium initially in the gap, μg Cs,

t = time at diffusion temperature, hr, and

R_o = initial rate of release of cesium by diffusion, μg Cs/hr.

The initial rate of release by diffusion, R_o, in turn, is given by

$$R_o = \left[\frac{W}{200}\right]\left[\frac{0.101}{P}\right]\left[\frac{G^{0.8}}{7.74}\right] e^{\left(19.96 - \frac{19{,}810}{T}\right)} , \qquad (2)$$

where

G = gap inventory of cesium following burst release, μg Cs/cm^2 of cladding,

W = width of radial gap, μm, and

P = system pressure, MPa.

In this equation the first two brackets account for the effects of cross-sectional area and absolute pressure on gas-phase diffusion. The third bracket is an empirical expression for the effect of gap inventory on the gas-phase concentration of the diffusing species. The fourth bracket is a dimensional empirical expression for the rate of escape by diffusion. The equation is simplistic in that it does not account for the increase in length of diffusion path as release progresses.

The model for release of iodine follows the same pattern as for cesium. The iodine burst release in the temperature range 700 to 900°C is expressed as

$$M_B = V_B \left[\frac{G^{0.8}}{1.15}\right] e^{\left(1.09 - \frac{3770}{T}\right)} . \qquad (4)$$

The release of iodine by diffusion in the temperature range 500 to 1200°C is given by Eq. (2), but R_o for iodine is given by

$$R_o = \left[\frac{W}{200}\right]\left[\frac{0.101}{P}\right]\left[\frac{G^{0.8}}{1.15}\right] e^{\left(15.3 - \frac{14{,}800}{T}\right)} . \qquad (5)$$

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. 1 for both cesium and iodine. The test parameters ranged as follows: V_B from 0 to 348 cm^3 (STP), G (cesium) from 0.5 to 127 μg of Cs per cm^2 of cladding, G (iodine) from 0.5 to 22 μg of I per 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 ranged from 15 to 30.5 cm.

4.0 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 as follows: 33,000 fuel rods, each containing 2500 g of UO_2 and operated so that equal numbers of rods had 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 2. This distribution is based on calculations of fission gas released for a typical PWR.[10] 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. Since the fuel rods would rupture near their centers, the cesium and iodine gap inventories were based on peak burnups estimated to be 10% higher than the above average burnups. It was further assumed that all fuel rods ruptured and experienced a temperature transient to 1200°C for an effective time of 10 min.

Calculations using the models are summarized in Table 3. 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. Based on our tests with high-burnup fuel, we would expect an additional fission gas release during the heatup to 1200°C equal to approximately 1.5% of the total inventory. The total fission gas release would therefore be approximately 2.77%.

5.0 COMPARISON WITH THE WASH-1400 LOCA

It is interesting to compare these release values with those used in WASH-1400.[11] Values for gap inventory and gap escape fraction (the fraction of gap inventory which escapes following rupture) are listed in Table 4. 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 PWR[10] 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, since the available experimental data were both sparse and inconsistent.

Diffusion equations[9] 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 small fractional releases, typical of our example PWR (but not typical of the WASH-1400 PWR), the reduction is inversely proportional to the square root of the decay constant for isotopes reaching production-decay equilibrium. The magnitude of the reduction also depends upon the irradiation time, but, as indicated earlier, release by knockout is essentially independent of half-life. For the example PWR discussed in this paper, 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 for stable isotopes of the same element.

6.0 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 is 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 example 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. In correlating our test data and for the example PWR LOCA, we assumed that pressure-ruptured rods with expanded cladding release cesium and iodine with 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 larger gap width than 200 μm for diffusional release predictions. If pressure fluctuations during the diffusional release period are large enough to pump out additional fission product vapors, this effect must be added to the calculated diffusional release.

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Table 1. Summary of fission product release tests

Test No.	Temperature: Rupture (°C)	Temperature: Diffusion (°C)	Diffusion time (hr)	Volume of gas vented (cm^3, 0°C, 1 atm)	Radial gap width (μm)	Initial gap inventory: (μg Cs/cm^2 cladding)	Initial gap inventory: (μg I/cm^2 cladding)	Mass released with rupture[a]: (μg Cs)	Mass released with rupture[a]: (μg I)	Mass released by diffusion[a]: (μg Cs)	Mass released by diffusion[a]: (μg I)
Implant-1[b]	700	700	1.0	229	127	23.1	22.0	179.6	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	291.7	96.9
Implant-4	-	1100	0.8	0	127	67.6	6.05	-	-	1240	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	1329	161
Implant-10	-	700	5.0	0	127	97.2	9.07	-	-	5.0	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	60.2	6.9
LBU-1[c]	-	700	5.0	0	26[d]	68.8	1.47	-	-	0.046	0.11
LBU-2	-	900	2.0	0	27	106.7	2.44	-	-	19.4	20.0
HBU-1[e]	-	700	5	0	20	13.1	1.20	-	-	0.123	0.93
HBU-2	-	900	2	0	20	12.7	1.17	-	-	2.82	1.76
HBU-4	-	500	20	0	20	13.1	1.20	-	-	0.017	0.105
HBU-7	900	900	0.02	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	0.17	0	200	11.3	1.05	-	-	142.0	20.2

[a]For pressure-ruptured tests, the fraction released by diffusion was estimated using the diffusion test data as a guide.

[b]Implant Test Series cladding 0.965 cm inside diam, 24 cm length of pellets coated with simulated fission products.

[c]Low Burnup Fuel Test Series cladding 1.27 cm inside diam, fuel length 15.24 cm, in-reactor fission gas release 11.6% (LBU-1) and 18.9% (LBU-2).

[d]The unusually low cesium and iodine release values suggest that the hole drilled through the cladding might have been at a location where the gap width was much less than the average value of 26 μm.

[e]High Burnup Fuel Test Series cladding 0.948 cm inside diam, fuel length 30.48 cm, in-reactor fission gas release 0.25%.

Table 2. Assumed Characteristics of Fuel Rods in Example PWR

Number of Rods	Fission Gas Initially in Plenum (%)	Fission Gas Initially in Plenum (cm^3, STP)[a]	Helium Fill Gas (cm^3, STP)	Rupture Temp. (°C)	Amount of Vented Gas (cm^3, 0°C, 0.303 MPa)
390	8	163.3	750	830	391
2,760	4	81.6	750	855	264
5,970	2	40.6	750	860	251
10,350	1	20.4	750	860	244
7,920	0.5	10.2	750	860	240
5,610	0.25	5.1	750	860	239

[a]Volumes listed correspond to rod average burnups of 30,000 MWd/MT. For simplicity, a single rupture temperature and amount of vented gas was used for each group of rods.

Table 3. Summary of Calculated Fission Product Release from Example PWR

Number of Rods in Group[a]	Fission Gas in Plenum (% of Total in Rod)	Cesium and Iodine in Gap Space (% of Total in Rod)	Total Released from Rod Group (g Cs)	Total Released from Rod Group (g I)
390	8	19.63	1.80	0.38
2,760	4	9.63	7.52	1.48
5,970	2	4.63	8.90	1.73
10,350	1	2.13	8.10	1.59
7,920	0.5	0.88	3.01	0.59
5,610	0.25	0.25	0.78	0.15
Total 33,000	-	-	30.11[b]	5.92[c]

[a]An equal number of rods in each group has inventories corresponding to average burnups of 10,000, 20,000, and 30,000 MWd/MT.

[b]80% released at time of rupture; 20% released by diffusion.

[c]72% released at time of rupture; 28% released by diffusion.

Table 4. Comparison of WASH-1400 and Model Calculations

Element	Gap Inventory[a] (% of Total Inventory) WASH-1400	Gap Inventory[a] (% of Total Inventory) Model	Gap Escape Fraction (% of Gap Inventory) WASH-1400	Gap Escape Fraction (% of Gap Inventory) Model	Total Release (% of Total Inventory) WASH-1400	Total Release (% of Total Inventory) 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

[a]Calculated for stable and long half-life isotopes. The gap inventory and total release of ^{131}I and ^{133}Xe would be 3 to 9 times lower.

[b]An additional amount of fission gas, approximately 1.5% of the total inventory, would be released during heatup.

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