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MASTER

Fission Product Release

from

Defected LWR Fuel Rods*

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FISSION PRODUCT RELEASE FROM DEFECTED LWR FUEL RODS*

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Experiments conducted at Oak Ridge National Laboratory both with fission product simulants and with irradiated commercial fuel have been utilized to develop a semi-empirical model of fission product release from defected Light Water Reactor (LWR) fuel rods. At fuel temperatures less than 1200°C, releases occur from fission products previously accumulated in the pellet-to-cladding gap region. In this temperature range, the release of species of moderate volatility is postulated to result from two processes. The first of these, which occurs during the period of fuel clad rupture, is due to the transport of the fill and fission product gases as they are vented through the cladding defect. This mode of release is virtually instantaneous. The second mechanism for release, which is time-dependent, involves the diffusional transport of the semi-volatile species to the point of clad rupture through the interconnected voids (the pellet-to-cladding gap and cracks in fuel pellets) within the fuel rod.

The burst release component in the temperature range 900 to 1200°C can be described by the mathematical relation

$$M_B = \alpha V_B (M_o/A)^a \exp(-C/T), \qquad (1)$$

in which M_B denotes the mass (in g) of fission product released during the period of cladding rupture, V_B is the volume (in cm^3) of plenum gas vented, as determined at 0°C and system pressure, M_o is the inventory (in g) of the fission product that is associated with interconnected voids, A is the internal area (in cm^2) of cladding, T represents the temperature (in K) at the point of rupture, and the parameters α, a, and C are adjustable constants.

The diffusional release component has the form

$$M_D = M_o \left[1 - \exp\left(-R_o t/M_o\right)\right], \qquad (2)$$

in which M_D is the mass (in g) of fission product released due to diffusional transport during the time period t (in h) following rupture, and R_o is the initial rate of diffusional release (in g/h). Note that Eq. (2), as written, contains no provision for radioactive decay.

The initial rate of release by diffusional transport in the temperature range 500 to 1200°C is given by the expression

$$R_o = (\delta W/P)(M_o/A)^a \exp\left(-\gamma/T\right), \qquad (3)$$

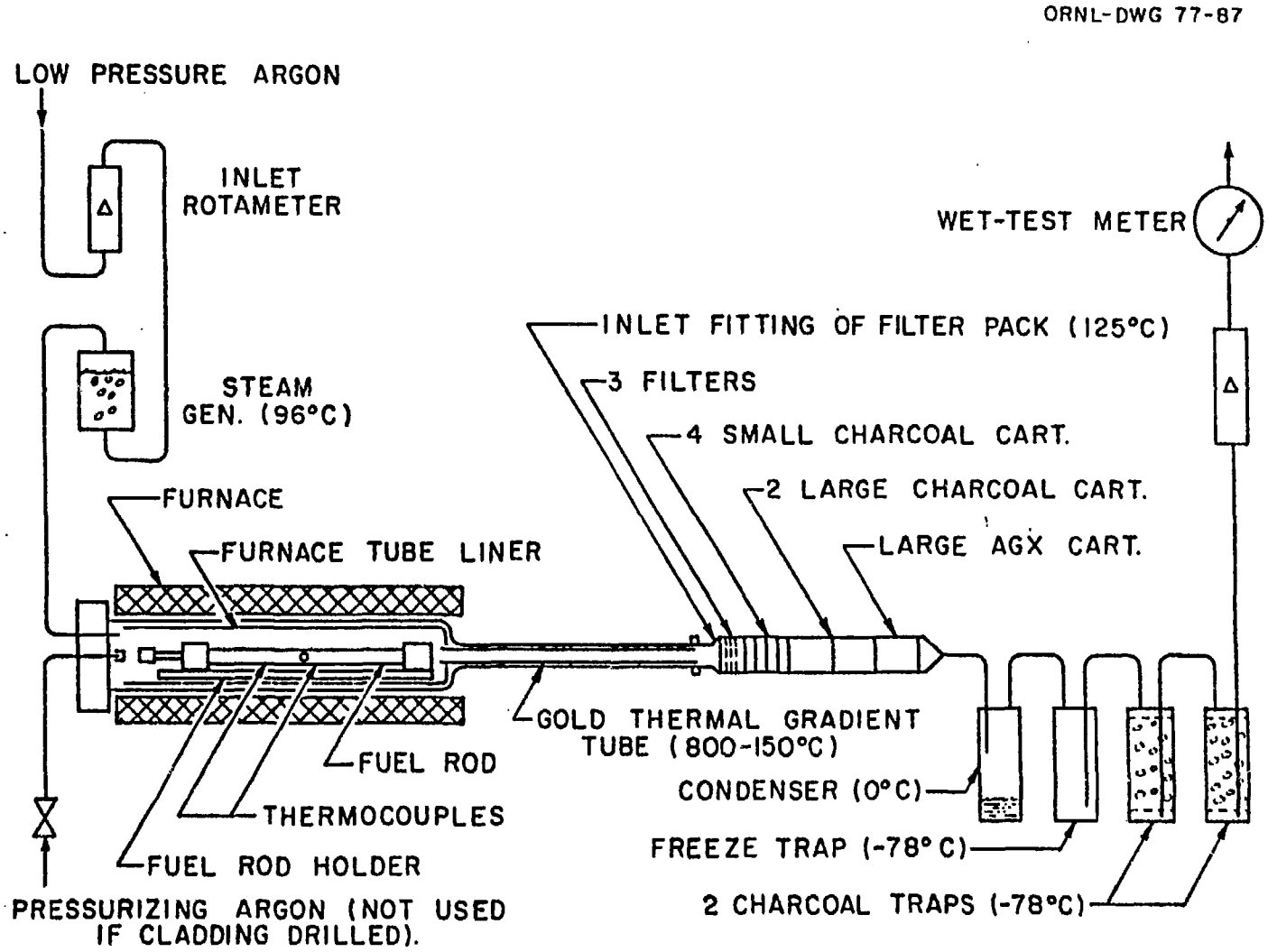
in which W is the width (in μm) of the radial gap in the defected rod, P is the system pressure (in MPa), and δ and γ are adjustable constants. (The amount of cladding expansion and the size of the rupture opening that occur in our tests result in an equivalent gap width, W, of 200 μm.)

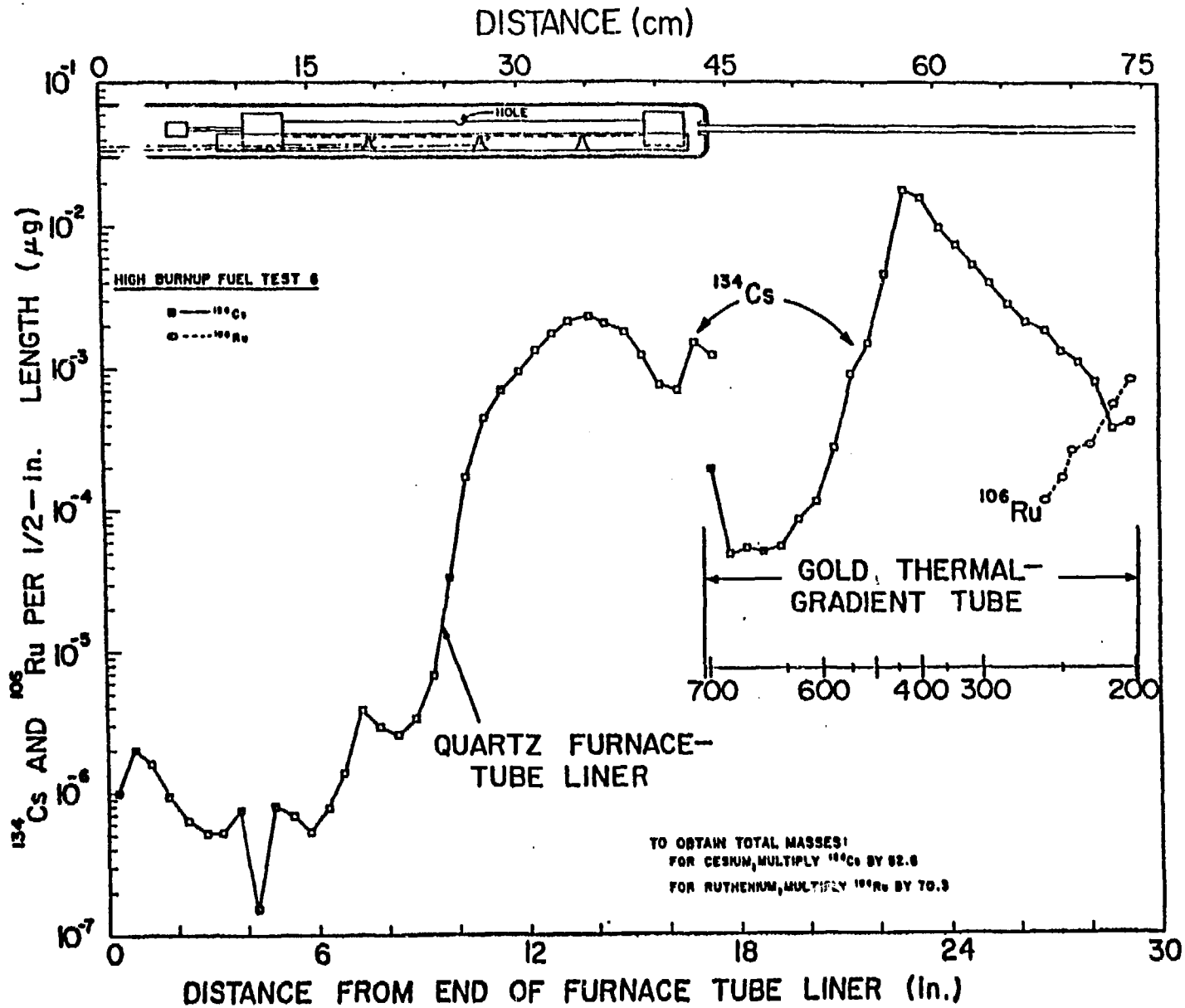
Values of the various constants which were derived from the experimentally determined release data for cesium and iodine species are presented in the following table:

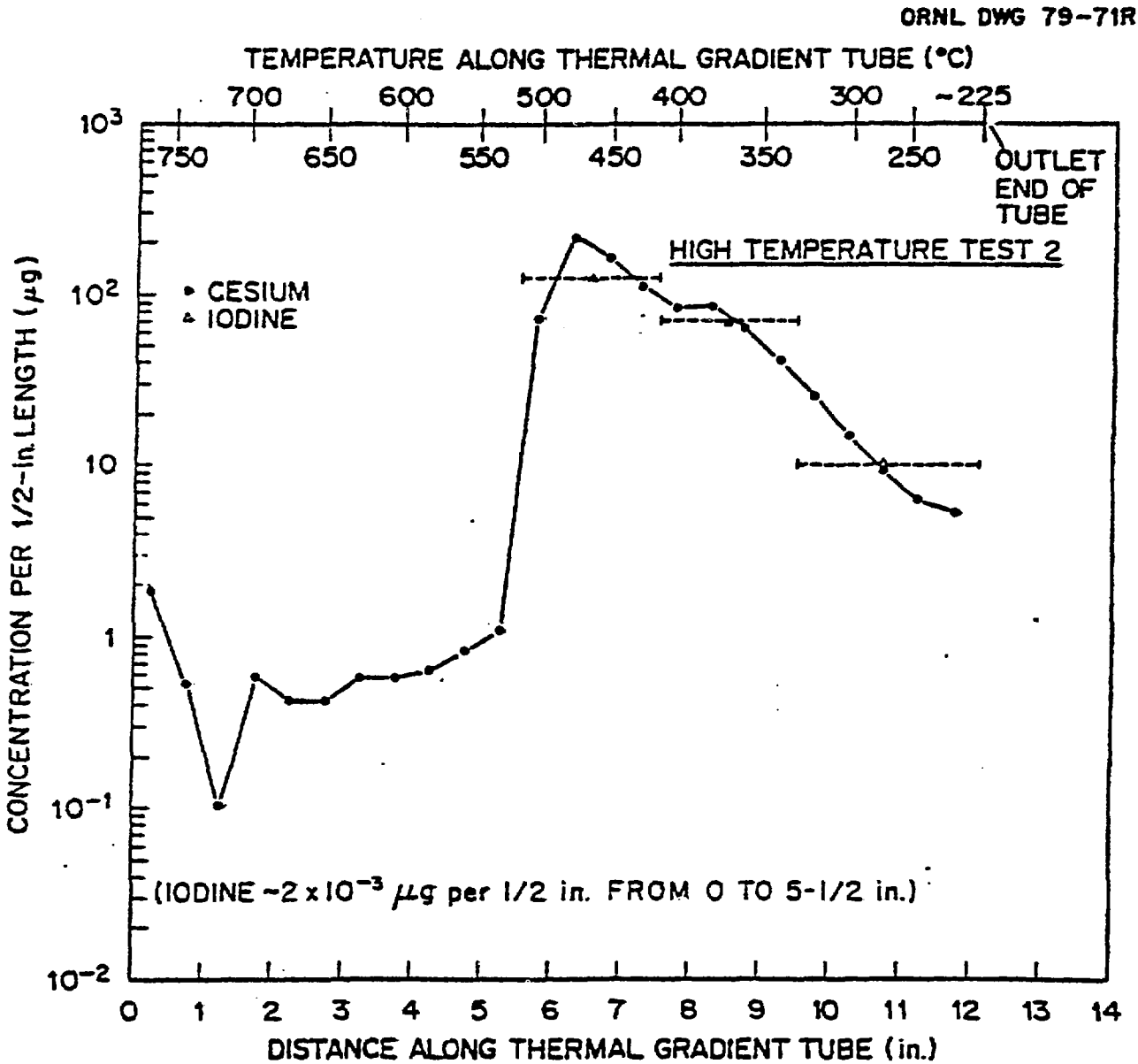
Parameter	Cesium species	Iodine species
α, $(g/cm^3)\cdot(g/cm^2)^{-a}$	3.49	0.163
a	0.8	0.8
C, K^{-1}	7.42×10^3	3.77×10^3
δ, $(g\cdot MPa/\mu m\cdot h)\cdot(g/cm^2)^{-a}$	1.90×10^3	1.22×10^2
γ, K^{-1}	1.98×10^4	1.48×10^4

Two series of tests were conducted to define the limitations of the model in describing the releases of cesium and iodine species. The first of these involved an extension of the temperature range of the experiments from 1200°C to 1600°C. These tests indicate that release from high-burnup fuel proceeds by a third process over the approximate temperature range 1200 to 1450°C. This process is probably due to linkage of fission gas bubbles which are formed at grain boundaries. Beyond 1450°C, releases appear to result from diffusion of the fission products through the UO_2 matrix.

The second series of experiments involved an extension of the studies to moderate-burnup fuel that had been irradiated at a higher temperature and thus contained larger quantities of fission products associated with interconnected voids (the "gap inventory"). The results indicate that the model correctly predicts releases from fuel rods which have gap inventories up to 12% of total inventory.


ORNL-DWG 77-87
LOW PRESSURE ARGON
INLET ROTAMETER
STEAM GEN. (96°C)
FURNACE
FURNACE TUBE LINER
FUEL ROD
THERMOCOUPLES
FUEL ROD HOLDER
PRESSURIZING ARGON (NOT USED IF CLADDING DRILLED).
GOLD THERMAL GRADIENT TUBE (800-150°C)
INLET FITTING OF FILTER PACK (125°C)
3 FILTERS
4 SMALL CHARCOAL CART.
2 LARGE CHARCOAL CART.
LARGE AGX CART.
CONDENSER (0°C)
FREEZE TRAP (-78°C)
2 CHARCOAL TRAPS (-78°C)
WET-TEST METER


DISTANCE (cm)
0
15
30
45
60
75
HOLE
HIGH BURNUP FUEL TEST 6
134Cs
106Ru
GOLD, THERMAL-GRADIENT TUBE
700
600
400
300
200
QUARTZ FURNACE-TUBE LINER
TO OBTAIN TOTAL MASSES:
FOR CESIUM, MULTIPLY 134Cs BY 52.6
FOR RUTHENIUM, MULTIPLY 106Ru BY 70.3
134Cs AND 106Ru PER 1/2-in. LENGTH (μg)
10-1
10-2
10-3
10-4
10-5
10-6
10-7
0
6
12
18
24
30
DISTANCE FROM END OF FURNACE TUBE LINER (In.)


ORNL DWG 79-71R
TEMPERATURE ALONG THERMAL GRADIENT TUBE (°C)
700
600
500
400
300
~225
750
650
550
450
350
250
OUTLET END OF TUBE
HIGH TEMPERATURE TEST 2
• CESIUM
▲ IODINE
CONCENTRATION PER 1/2-in. LENGTH (μg)
10³
10²
10
1
10⁻¹
10⁻²
(IODINE ~2 x 10⁻³ μg per 1/2 in. FROM 0 TO 5-1/2 in.)
0
1
2
3
4
5
6
7
8
9
10
11
12
13
14
DISTANCE ALONG THERMAL GRADIENT TUBE (in.)

FISSION PRODUCT RELEASE MODEL

$$M_t = M_B + M_D$$

M_t TOTAL QUANTITY OF FISSION PRODUCT RELEASED

M_B QUANTITY OF FISSION PRODUCT RELEASED DURING RUPTURE

M_D QUANTITY OF FISSION PRODUCT RELEASED BY DIFFUSIONAL TRANSPORT

BURST RELEASE COMPONENT

$$M_B = \alpha V_B (M_o/A)^a \exp\,[-\,C/T]$$

M_B - MASS OF FISSION PRODUCT RELEASED DURING RUPTURE

V_B - VOLUME OF PLENUM GAS VENTED

M_o - INVENTORY OF FISSION PRODUCT ASSOCIATED WITH INTERCONNECTED VOIDS (THE "GAP INVENTORY")

A - INTERNAL AREA OF THE CLADDING

T - TEMPERATURE AT BURST LOCATION

α,a,C - ADJUSTABLE CONSTANTS

DIFFUSIONAL RELEASE COMPONENT

$$M_D = M_o \left[1 - \exp(-R_o t/M_o)\right]$$

t - TIME DURING WHICH DIFFUSIONAL RELEASE OCCURS

INITIAL RELEASE RATE

$$R_o = (\delta W/P)(M_o/A)^a \exp\,[-\gamma/T]$$

W - WIDTH OF THE RADIAL GAP ALONG WHICH DIFFUSION OCCURS

P - SYSTEM PRESSURE

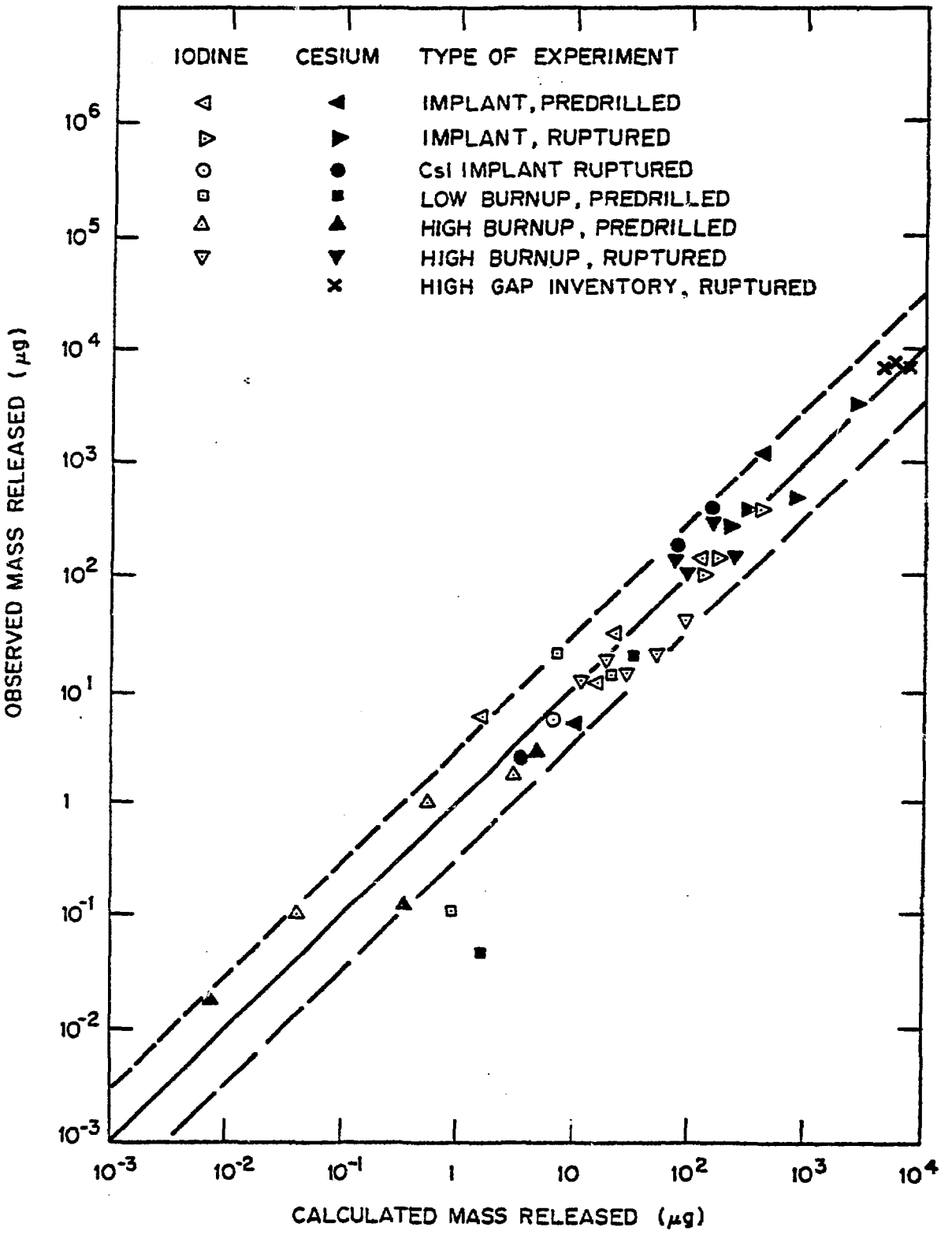
δ, γ - ADJUSTABLE CONSTANTS

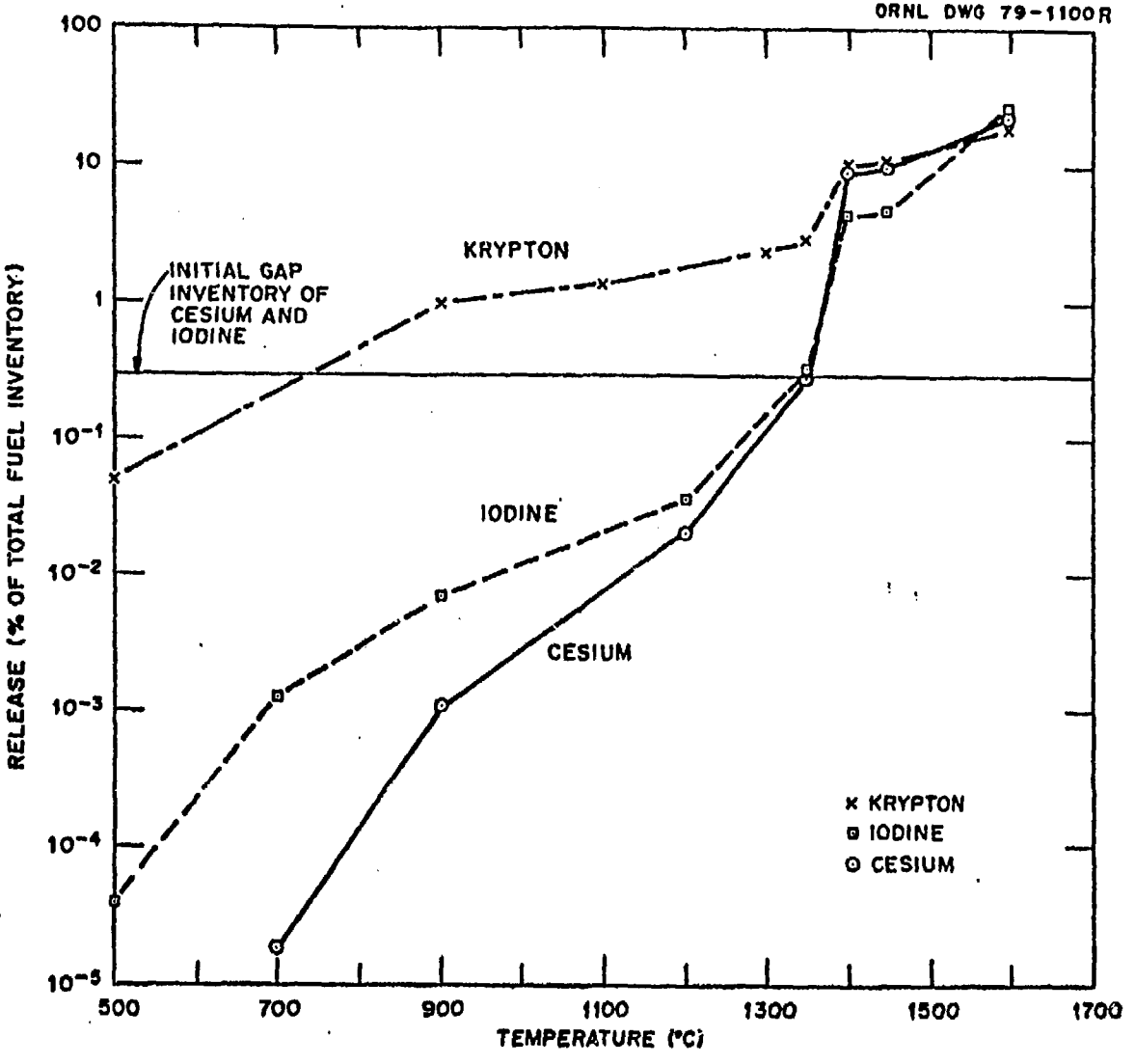
COMPARISON OF BURST AND DIFFUSIONAL RELEASE COMPONENTS

T (°C)	M_B/M_D (1HR)	
	CESIUM	IODINE
700	220	41
800	68	14
900	25	5.9
1000	6.8*	2.2*
1100	2.2*	0.9*
1200	0.8*	0.5*

*Assumes rod rupture at 900°C

ORNL DWG 79-1321R





RELEASE FROM RUPTURED ROD BY DIFFUSION IN STEAM IN 10 min.

DISTRIBUTION OF CESIUM AND IODINE IN IRRADIATED FUEL TESTS

TYPE OF TEST	TEST TEMPERATURE (°C)	TEST TIME (min)	FURNACE TUBE		THERMAL GRADIENT TUBE AND FILTER PAPERS		CHARCOAL	
			CESIUM (µg)	IODINE (µg)	CESIUM (µg)	IODINE (µg)	CESIUM (µg)	IODINE (µg)
GAP PURGE, He	700-1200	417	100	0.1	1900	190	0.0	0.3
BURST, STEAM	900	1	110	4.0	17	7.0	0.0	0.4
DIFFUSIONAL, STEAM	1200	27	40	0.1	110	19.0	0.0	0.9
DIFFUSIONAL, STEAM	1445	7	21,000	34.0	1200	960.0	0.0	0.6
DIFFUSIONAL, DRY AIR	700	300	2	0.03	5	0.5	0.0	18.4

SUMMARY OF CHEMICAL BEHAVIOR

1. IN THE GAP-PURGE TESTS WITH PURIFIED HELIUM, IODINE APPEARED TO BE DEPOSITED IN THE APPARATUS AS CsI.
2. IN TESTS CONDUCTED IN STEAM, CsOH AND CsI APPEARED TO BE THE MAJOR CHEMICAL SPECIES OF CESIUM AND IODINE RELEASED.
3. IN TESTS CONDUCTED IN DRY AIR, THE INDICATED MAJOR CHEMICAL SPECIES OF CESIUM AND IODINE RELEASED WERE Cs_2O AND I_2.
4. ANTIMONY APPEARED TO BE RELEASED IN THE STEAM TESTS AS ELEMENTAL ANTIMONY. NO ANTIMONY WAS RELEASED IN THE DRY AIR TESTS.
5. IN DRY AIR TESTS, SIGNIFICANT AMOUNTS OF VERY VOLATILE RUTHENIUM WERE RELEASED, PROBABLY AS RuO_4. ONLY TRACE AMOUNTS WERE RELEASED IN STEAM.