BEHAVIOUR OF SHORT-LIVED FISSION PRODUCTS WITHIN OPERATING UO_2 FUEL ELEMENTS

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Abstract

We have carried out experiments using a "sweep gas" technique to determine the behaviour of short-lived fission products within operating, intact UO_2 fuel elements. The Zircaloy-4-clad elements were 500 mm long and contained fuel of density 10.65 - 10.71 Mg/m^3. A He-2% H_2 carrier gas swept gaseous or volatile fission products out of the operating fuel element past a gamma spectrometer for measurement.

In tests at linear powers of 45 and 60 kW/m to maximum burnups of 70 MW.h/kg U, the species measured directly at the spectrometer were generally the short-lived xenons and kryptons. We did not observe iodine or bromine during normal operation. However, we have deduced the behaviour of I-133 and I-135 from the decay of Xe-133 and Xe-135 during reactor shutdowns.

Plots of R/B (released/born) against λ (decay constant) or effective λ for all isotopes observed at 45 and 60 kW/m show that a line of slope -0.5, corresponding with diffusion kinetics, is a good fit to the measured xenon and krypton data. Our inferred release of iodine fits the same line. From this we can extrapolate to an R/B for I-131 of about 5×10^{-3}. The ANS 5.4 release correlation gives calculated results in good agreement with our measurements.

1. INTRODUCTION

Considerable data are available on the behaviour of stable fission products, particularly gases, in irradiated oxide fuel. See, for example, Matzke's recent review (1). However, there is a current need for studies on the release of short-lived fission products. The equilibrium gap inventory of active species in an operating fuel element gives a measure of the activity that can be immediately released during an accident, thus providing a starting point for transient or accident analysis.

Considerable work has been carried out in the U.K. on active species swept from small, isothermally heated UO_2 samples (2). Useful data on the diffusion behaviour of krypton, xenon and iodine have been accumulated. French work on UO_2 elements at a linear power of 40 kW/m is also pertinent (3), as are recent Halden data on a swept UO_2 element at 25 - 30 kW/m (4).

In this paper we report steady state release results for short-lived fission products in two sweep gas tests on intact UO_2 elements in the NRX reactor at Chalk River Nuclear Laboratories. This text is a revised version of the draft presented at Preston, 1982 March 14-19. Experiment FIO-122 operated at a linear power of 45 kW/m; experiment FIO-124 at 60 kW/m. Both tests were terminated by sheath defects associated with fabrication. Release results from the pre-defect and defect periods of the test will be reported separately, as will release data from reactor shutdown and start-up transients.

2. EXPERIMENTAL

Table 1 outlines fuel, sheath and operating characteristics for the two tests. The sweep gas elements were irradiated in two separate trefoil assemblies, each trefoil with two UO_2 filler elements. Characteristics of the filler elements were identical with those of the swept elements. Pellets in the swept elements had six and four 1.15 mm x 1.15 mm cross-section grooves in the FIO-122 and FIO-124 tests, respectively, to permit easy gas flow.

Figure 1 is a schematic diagram of the sweep gas system. During operation, bottled He-2% H_2 is introduced into the fuel element at an operating pressure of about 3 MPa, after passing through an oxygen remover and moisture trap. The carrier gas sweeps gaseous fission products out of the fuel element past a gamma spectrometer for measurement. Volume flow rates were typically 3 mL/s. At this rate, residence time of the sweep gas in the element is about one second; transit time from element to spectrometer is about 10 minutes. Isotopic data recorded at the spectrometer were converted to concentrations and releases. After passing the spectrometer, the carrier gas and fission products pass through parallel iodine traps into delay tanks. A gamma monitor scans the stream from the delay tanks; high counts shut off the gas flow. Delay time is approximately 12 hours, sufficient to reduce contributions from short-lived kryptons and xenons to less than 10% of the administrative level.

3. RESULTS AND DISCUSSION

Table 2 shows the short-lived species directly measured at the spectrometer for FIO-122, generally the short-lived xenons and kryptons. We have not directly observed any iodine or bromine at the spectrometer during normal operation. We only see halogen transport when water/steam is inside the element, as accompanies a defect. Calculations show the limit of detectability for iodines is better than about 1% of the release to the fuel-sheath gap. Since no iodines were directly measured, we conclude that less than 1% of the gap inventory is released as a volatile, such as CH_3I. We have deduced the iodine behaviour from the decay of I-133 and I-135 to Xe-133 and Xe-135, respectively during reactor shutdowns. Figure 2 is a plot of release versus time during two reactor shutdowns, for Xe-135. The shutdown behaviour is characterized by a prompt decrease in release, followed by a decay period. The initial drop is due to xenon, and the decay is characteristic of the iodine.

Figure 3 shows results with R/B (released/born) plotted against λ (decay constant) or effective λ for xenons, kryptons and iodines observed or inferred during the test at 45 kW/m. To obtain the data, release rates were averaged over at least two periods after steady state conditions were established. R/B vs λ gives equivocal results for slope early in life of experiment, possibly because we are measuring R/inventory rather than R/B. Close to end of life, after at least three days of steady state operation, R/B vs λ gives a line of slope -0.5. Corrections have been made to R for recoil and precursor effects. Release for Xe-135 has been plotted against effective λ, which accounts for loss due to neutron capture. All isotopes with λ

greater than 8 x 10^{-4} (i.e. from Xe-138) arise from recoil. More than 85% of the contribution to Xe-135 is also from recoil. A line of slope -0.5, corresponding with diffusion kinetics, is a good fit to the measured xenon and krypton data. Our inferred iodine data fit the same line, and the resultant extrapolated R/B for I-131 is about 5 x 10^{-4}.

It is hypothesized that release of iodine into the fuel/sheath gap can provide a significant contribution to the apparent release of xenon. Those rare gases with short-lived precursors would migrate and be released from the fuel primarily as gas. Those with long-lived precursors, e.g. Xe-133, Xe-135 and Xe-135m, will be partially released as gas and partially by decay of the released iodine precursor. Since iodine is not seen at the detector, it is probably trapped in the fuel, fuel-to-sheath gap or piping where it decays to xenon. Thus release of xenons with long-lived precursors will be augmented relative to those with little precursor release.

The short-lived species directly identified at the spectrometer in the FIO-124 test were identical with those observed for FIO-122. Again, we did not observe any iodine or bromine during normal operation. Only after a defect occurred was evidence of iodines obtained by a portable spectrometer with the head near the top closure of the loop. No shutdown decay data were obtained for FIO-124, thus iodine behaviour were inferred by comparison with short-lived xenon and krypton release.

Figure 4 shows R/B versus λ plotted as for FIO-124. Corrections for recoil, precursors and neutron capture have been made. A line of slope -0.5 is again a good fit to measured xenon and krypton data, confirming that release behaviour is again diffusion-dependent, as for FIO-122. Release rates are generally 10 times higher than those measured at 45 kW/m for FIO-122. As general behaviour is similar to that for FIO-122, it is reasonable to read off R/B values for I-131, I-133 and I-135 from the graph, to allow an estimate of the inventory.

Figure 5 shows how release results for both FIO-122 and FIO-124 compare with French Contact I data at 40 kW/m (3), and Halden data (4) at 25 - 30 kW/m. Our R/B results for FIO-122 are close to those for the Halden test and recent U.K. work (5), and about a factor of 10 less than those from Contact I. From this we infer that most of our release is from the fuel surface regions, in the athermal diffusion zone. Note that the Contact I test sweeps fission products from a central fuel annulus, thus accounting for the higher observed release. Consequently one expects R/B in our tests to be independent of operating power, as long as the fuel surface temperature remains less than about 560°C, the maximum for athermal diffusion. R/B for FIO-124 is significantly higher than that for FIO-122, because of its higher fuel surface temperature, and the greater fuel volume contributing to release.

Table 3 shows equilibrium iodine fuel-to-sheath gap inventories calculated from data for FIO-122 and FIO-124. For FIO-122, I-133 and I-135 were deduced from shutdown decay data, and I-131 inferred, assuming similar diffusion behaviour with I-133 and I-135. For FIO-124, all iodine results are inferred, assuming diffusion-dependent behaviour. For FIO-122, the I-131 inventory is 10 GBq (0.3 Ci), with total iodines 25 GBq (0.7 Ci). For FIO-124, values are about 10 times higher reflecting the higher operating power. The I-131 inventory is calculated to be about 100 GBq (3 Ci), and total iodines are 219 GBq (6 Ci). These values are significantly lower than the 10% of total fuel iodine inventory usually assumed for accident analysis (4).

4. CONCLUSIONS

1. Species swept from the element during normal operation at 45 and 60 kW/m were generally the short-lived xenons and kryptons. Iodines were not observed during normal operation.

2. R/B vs λ or effective λ for isotopes observed at 45 and 60 kW/m produced a slope of -0.5, corresponding with diffusion kinetics.

3. Our inferred release of iodine, from shutdown decay data and extrapolation, fits the same line of slope -0.5, implying effective diffusion behaviour similar to that of the short-lived xenons and kryptons.

4. The inferred equilibrium inventories of I-131 in the fuel-to-sheath gap at 45 and 60 kW/m are about 10 and 100 GBq (0.3 and 3 Ci), respectively.

5. REFERENCES

1. Hj. Matzke, "Gas Release Mechanisms in UO_2 - A Critical Review", Radiation Effects 53 (1980) 219.

2. For example, J.R. Findlay, et al., "Fission Product Release From UO_2 During Irradiation: Diffusion Data and Their Application to Reactor Fuel Pins", IAEA Specialists' Meeting on the Behaviour of Defected Zirconium Alloy Clad Ceramic Fuel in Water-Cooled Reactors", Chalk River Nuclear Laboratories, 1979 September 17-21, IAEA Report IWGFPT/6 (1980).

3. M. Bruet, M.L. Pointud, J. Dodelier and P.H. Melin, "CONTACT 1 and 2 Experiments: Behaviour of PWR Fuel Rod up to 15000 $MWd.tU^{-1}$", Specialists' Meeting on Fuel Element Performance Computer Modelling, Blackpool, U.K., 1980 March 17-21, IAEA Report IWGFPT/7 (1980).

4. A.D. Appelhans and J.A. Turnbull, "Measured Release of Radioactive Xenon, Krypton and Iodine From UO_2 During Nuclear Operation and a Comparison with Release Models", Eighth Water Reactor Safety Research Information Meeting, Gaithersburg, Maryland, 1980 October 27-31.

5. C.J. Greatly and R. Hargreaves, "The Measured Emission of Fission Product Gases From Operating UO_2 Fuel", J. Nucl. Mater. 79 (1979) 235.

6. ACKNOWLEDGEMENTS

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Table 1

Fuel, sheath and operating characteristics of tests FIO-122 and FIO-124.

Characteristic	FIO-122	FIO-124
Fuel	UO_2	UO_2
Enrichment (wt% ^{235}U in U)	5.02	4.5
Density (Mg/m^3)	10.71	10.65
Grain Size (μm)	10	10
Pellet Diameter (mm)	12.15	12.15
Fuel Stack Length (mm)	477	477
Sheath	Zircaloy-4	Zircaloy-4
Sheath Wall Thickness (mm)	0.43	0.43
Axial Clearance (mm)	2.2	2.2
Diametral Clearance (mm)	0.1	0.1
Coolant Pressure (MPa)	8.5	8.5
Coolant Flow (kg/s)	1.1	1.1
Coolant Inlet Temperature (°C)	240	240
Linear Power (kW/m)	45	60
Burnup (MW.h/kg U)	70	50

Table 2

Isotopes identified during FIO-122 and FIO-124 tests

KRYPTONS	XENONS	OTHERS
	Xe-133	
Kr-85m	Xe-135m	Rb-88
Kr-87	Xe-135	Rb-89
Kr-88	Xe-137	Rb-90
Kr-89	Xe-138	Te-132
Kr-90	Xe-139	Cs-138
		Ba-139

Table 3

Fuel-to-sheath gap inventories for iodines in FIO-122 and FIO-124

TEST	ISOTOPE	INVENTORY
FIO-122	I-131	10 GBq (0.3 Ci)
	I-133	12 GBq (0.3 Ci)
	I-135	3 GBq (0.07 Ci)
	Total Iodines	25 GBq (0.7 Ci)
FIO-124**	I-131	100 GBq (3 Ci)
	I-133	100 GBq (3 Ci)
	I-135	19 GBq (0.2 Ci)
	Total Iodines	219 GBq (6 Ci)

**Assuming $\lambda^{-0.5}$ behaviour

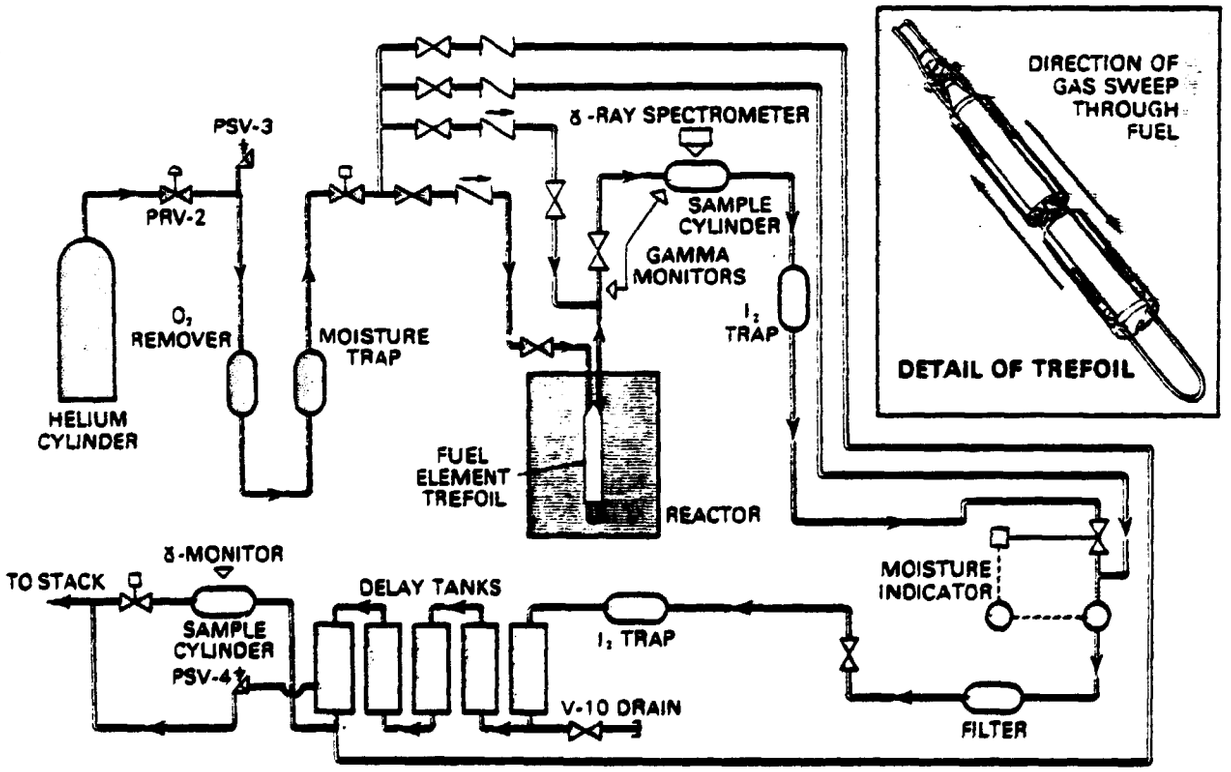

FIGURE 1 Schematic of sweep gas system.

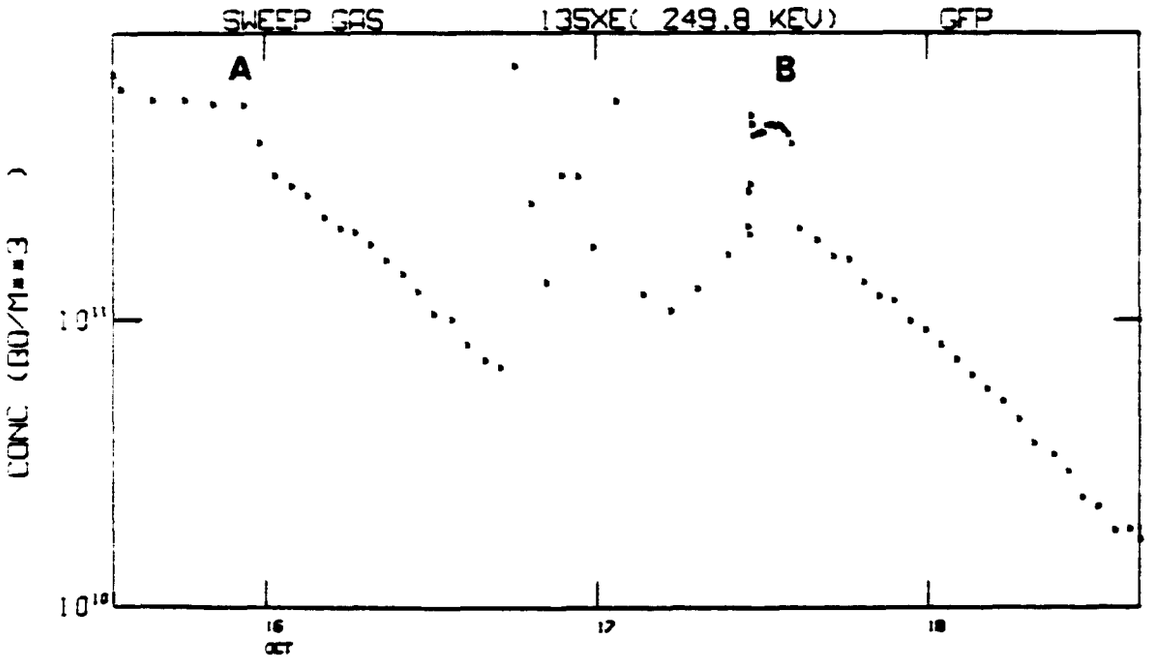

FIGURE 2 Concentration versus time for Xe-135 during shutdowns at A and B.

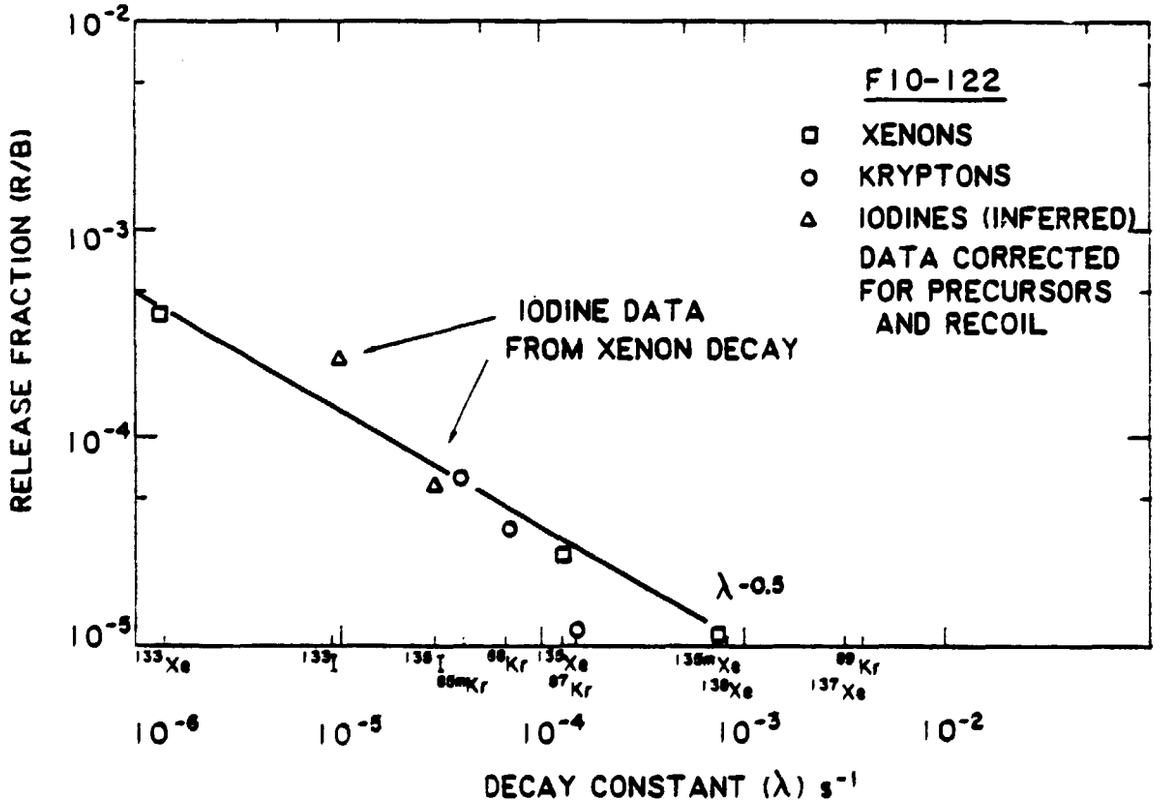

FIGURE 3 R/B versus λ or effective λ for isotopes observed or inferred in F10-122 at a linear power of 45 kW/m.

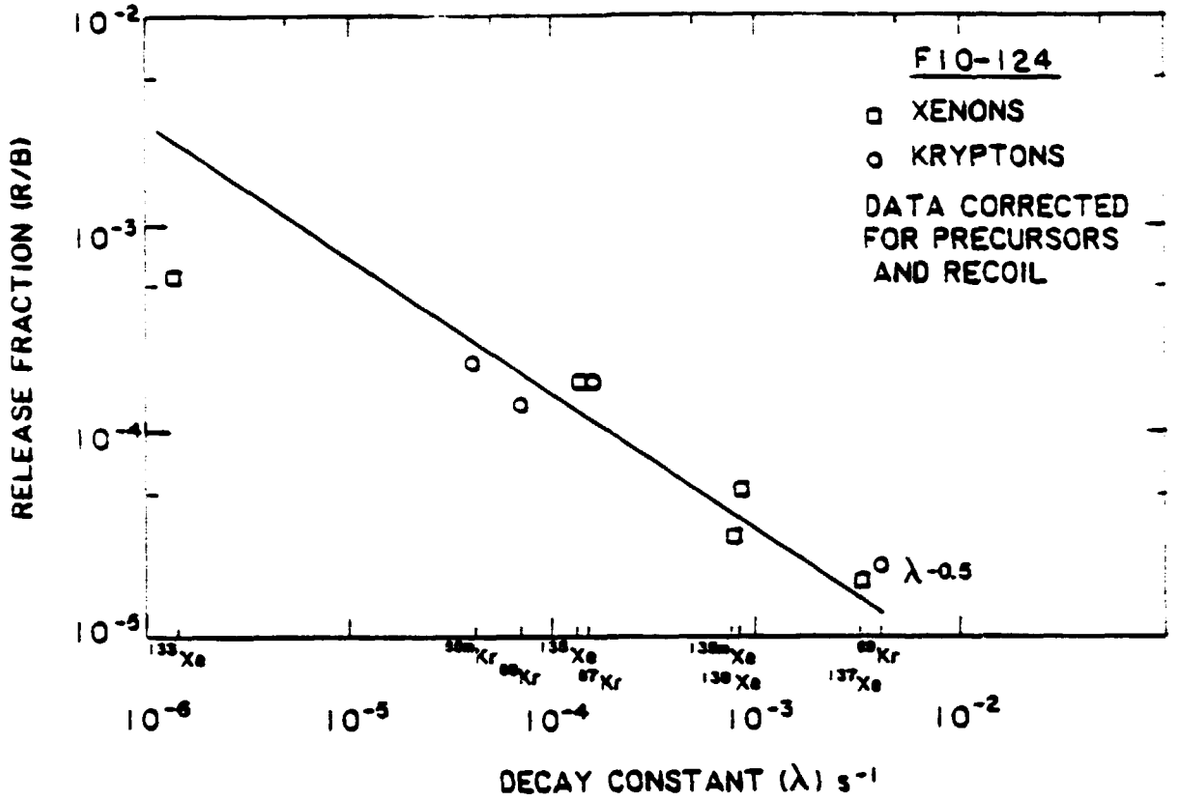

FIGURE 4 R/B versus λ or effective λ for isotopes observed in FIO-124 at a linear power of 60 kW/m.

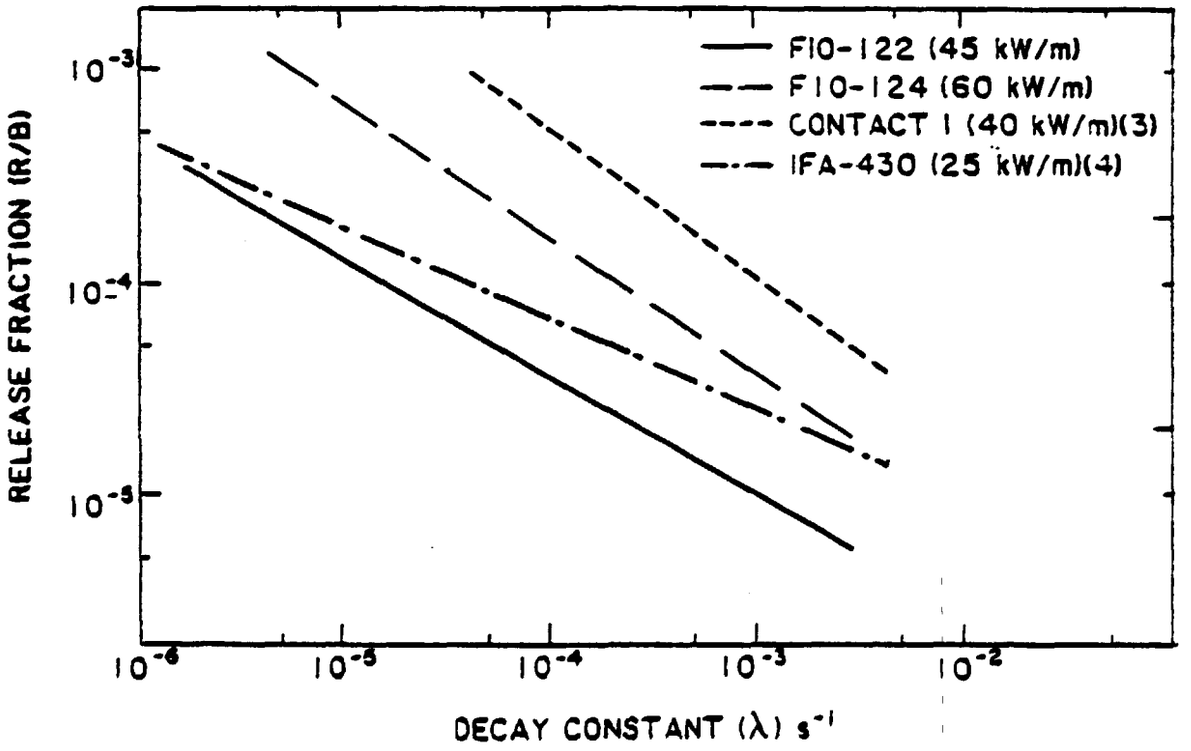

FIGURE 5 Results for FIO-122 and FIO-124 plotted with French CONTACT I data (3) at 40 kW/m and Halden IFA-430 data (4) at 25-28 kW/m.