TRANSIENT FISSION PRODUCT RELEASE DURING REACTOR SHUTDOWN AND STARTUP

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ABSTRACT

Sweep gas experiments performed at CRL from 1979 to 1985 have been analysed to determine the fraction of the fission product gas inventory that is released on reactor shutdown and startup. Empirical equations were derived and applied to calculate the xenon release from companion fuel elements and from a well documented experimental fuel bundle irradiated in the NRU reactor. The measured gas release could be matched to within about a factor of two for an experimental irradiation with a burnup of 217 MWh/kgU.

1. INTRODUCTION

From mid 1979 to 1985 a total of six sweep gas experiments were done in the NRX reactor at the Chalk River Laboratories of Atomic Energy of Canada Limited. The first two experiments were designed to measure the steady state release rate of fission gases[1-3]. Later experiments measured the effects of severe transients such as loop blowdown[4-8]. During the course of the irradiations there were many reactor shutdown and startup transients and for some of these transients the fission product release was continuously monitored.

Re-examination of the records showed that two experiments, designated FIO-133 and FIO-134, had good shutdown and startup data. Two others, FIO-122 and FIO-124, had some useable data, primarily for startups. Table 1 lists the basic parameters for these four experiments.

2. EXPERIMENTAL

The sweep gas circuit and the data reduction techniques have been thoroughly described elsewhere[3]. Details of the four fuel assemblies are given in Table 1 and are briefly discussed below.

FIO-122 operated at a nominal linear power of 43 kW/m to a burnup of 86 MWh/kgU. Irradiation was halted because the swept element defected. Counting times tended to be long so little detail was obtained on the transient shutdown release. Of the total of 27 startup and shutdown cycles, fission product release was measured during six transients

and on three occasions an attempt was made to measure the dynamics of the shutdown release. Counting times were shortened, but spectrometer saturation occurred. This resulted in loss of data from the shutdown peak. Since the reactor startup rate was much slower than the shutdown rate, the peak release was also lower and there was less spectrometer saturation. As a result, startup releases were generally more reliable. Again, long counting times resulted in loss of detail of the release kinetics. Details of fuel power and burnup at each shutdown were estimated from plots of the reactor power.

FIO-124 operated at a nominal linear power of 60 kW/m to a burnup of 50 MWh/kgU. Again, irradiation was halted because the swept element defected. There were 18 reactor startup and shutdowns. Data was recorded during four shutdowns and six startups. Similar problems of spectrometer saturation during the shutdowns were experienced as for FIO-122. The fuel power and burnup at each transient was again estimated from plots of the reactor power.

FIO-133 was the first of the experiments planned for transient tests. There were 31 startup and shutdown cycles, three of which were high temperature transients. The fuel element operated at a power varying from about 52 to 62 kW/m to a burnup of 54.9 MWh/kgU. Collimator modifications minimized the problem of spectrometer saturation and counting times were generally reduced, in some cases to as short as 45 seconds. As a result, the shutdown data were generally good. However, some of the shutdowns were short, less than an hour, and there is evidence from the long lived isotopes that all of the fission gases had not been fully vented by the time the reactor started up again. As a result, the measured release on shutdown is probably low for these short transients. Fuel power and burnup at each shutdown were obtained from the original computer calculations.

FIO-134 was a high burnup experiment which operated at a linear power varying from 52 to 61 kW/m, with the average close to 54 kW/m, to a burnup of 200 MWh/kgU. At this burnup the sweep gas was changed from inert to oxidizing and irradiation continued to a burnup of 227 MWh/kgU. Data was obtained for 12 shutdowns and 10 startups. The fuel power and burnup were taken from computer printouts.

3. ANALYSIS

Figure 1 shows one of the shutdown-startup transients for FIO-133 where counting times were 45 seconds. The isotopes ^{133}Xe and ^{85m}Kr have been selected as examples to show the general behaviour. There is still evidence of some spectrometer saturation during at least the first two data points of the shutdown. Startup occurred in steps and this is reflected in the startup release rate. There is no evidence that during startup release rates were high enough to saturate the spectrometer.

The release data for each shutdown and startup for each isotope were integrated over the particular transient. The steady state inventory was calculated from the operating power immediately prior to the shutdown.

The rate of change of the number of atoms of a given isotope present in the UO_2 fuel element may be expressed by:

$$dN/dt = B - R - \lambda N \qquad (1)$$
$$= 0 \quad \text{at steady state}$$

where: N = number of atoms in the fuel,
B = number of atoms born per second, (calculated from the fuel power)
R = number of atoms released per second, (measured by the spectrometer)
λ = decay constant of the isotope (s^{-1}).

Therefore, at steady state conditions:

$$N = (B - R)/\lambda \qquad (2)$$

Let the number of atoms released during the transient be n. This will be some fraction of N and therefore will have the same dependence on λ. It is also a measured quantity since

$$n = \int_{\text{transient}} R(t)\, dt \qquad (3)$$

The transient release fraction can then be defined by

$$F = n/N = \lambda \int_{\text{transient}} R(t)\, dt \,/\, (B - R) \qquad (4)$$

where all the quantities are either known or measured.

As may be seen from Table 1, the fuel elements for the four experiments had different dimensions. The larger pellets of FIO-133 seem to have had little effect on the results, which have been expressed as release fractions. We were able to consider all experiments as one data set. Fuel stack length also varied. For convenience, all release rates were converted to atoms/s per meter of fuel length.

Release fractions were calculated for each shutdown. Figure 2 shows an example. ^{133}Xe, ^{135}Xe and ^{135m}Xe release fractions were always high. Subtraction of the iodine contribution often made the result slightly low. This correction was done by extrapolating the long term release rate of the xenon, where the rate had become that of the iodine precursor, back to time zero for the shutdown and subtracting the integral below this extrapolated curve. The large extrapolation introduces considerable potential for error. In the example shown, the correction for the iodine contribution appears to be about right for ^{133}Xe, too large for ^{135}Xe, but not enough for ^{135m}Xe. Release fractions for isotopes with half-lives shorter than ^{89}Kr (3.2 minutes) were also often apparently in error, most likely because of the large corrections that had to be made for the flight time, which was usually at least 200 seconds from the fuel to the spectrometer. This is five or more half-lives for ^{139}Xe and ^{90}Kr. As shown in the example taken for Figure 2, the isotopes with very short half-lives, ^{137}Xe and beyond, show a consistent trend of over correction for the flight time

between the fuel element and the spectrometer.

The shutdown release fraction was found to depend strongly on both fuel power and burnup. With the exception of very low burnup, less than about 10 MWh/kgU, where the fuel is still sintering, (see section 3.2) both dependencies increase exponentially.

3.1. Power Dependence of Shutdown Release

The analysis was complicated because of the strong dependence of the release on both power and burnup. Initially, the combined data from all four experiments were examined, looking at small increments of burnup. Unfortunately, so much data was lost because of spectrometer saturation in FIO-122 and FIO-124 that analysis using data from all four experiments proved impossible. Finally, six shutdowns from FIO-133 spanning a burnup range from 41.3 to 47.9 MWh/kgU and a power range from 46.7 to 62.1 kW/m were selected. Spectrometer saturation still occurred, but appeared to be restricted to one, or at most two counting cycles, certainly less than two minutes total. It was assumed that the proportion of the data lost in this manner was small compared with the overall integral.

The next problem was to select representative isotopes where there was a strong gamma ray signal and little chance of interferance from extraneous peaks. The data for the ^{85m}Kr gamma-ray peak at 151.2 keV with an abundance of 0.755 was finally selected as probably being the most reliable. ^{88}Kr at 196.3 keV and a gamma abundance of 0.26 was chosen as a check. ^{133}Xe was rejected because the data was badly scattered and the 81 keV gamma ray is subject to interference from ^{131}I. There is also a problem from the contribution of the precursor ^{133}I. ^{135}Xe was also rejected because of complications caused by the long-lived iodine precursor and neutron absorption effects.

The results are shown on Figure 3. The slope of the regression line through the ^{85m}Kr data is 7.02 and that through the ^{88}Kr data is 6.04. An average value between the ^{85m}Kr and ^{88}Kr power dependence of about 6.5 is probably reasonable. This value was used for the next stage of the analysis.

3.2. Burnup Dependence of Shutdown Release

Having established a power dependence using a limited data set, where the burnup was almost constant, we used all of the data for FIO-133 and FIO-134 to establish the burnup dependence. The power was standardized to 55 kW/m using the power dependence of 6.5. Again, most weight was given to ^{85m}Kr and ^{88}Kr. Figure 4 shows the results for ^{85m}Kr. After the initial sintering portion of the curve, up to about 10 MWh/kgU, a burnup dependence of 2.4 appears to fit the data. Some data falls below the given line because of incomplete release due to the short time between a shutdown and the following startup. The same slope was also a good representation for ^{88}Kr. The correlation curve is generally conservative, providing an upper-bound for the release fraction.

From the above analysis, we now have a dependence of the release fraction, F, (Eq. 4) on both fuel operating power and burnup. We may write a correlation as follows:

$$F = 2.2\ (10^{-19})\ P^{6.5}\ B^{2.4} \qquad (B \geq 10\ \text{MWh/kgU}) \quad (5)$$

where the power, P, is in kW/m and the burnup, B, is in MWh/kgU.

We thus have an empirical equation for the release fraction of fission products resulting from a shutdown. Since the release fraction is constant for all isotopes, regardless of decay constant, we also have a measure of the release fraction of stable isotopes, which may be considered to have infinite half-life. These are the ones which will result in buildup of gas pressure in the fuel-to-sheath gap. With Eq (5) we should be able to calculate the pressure buildup due to shutdowns for any fuel element for which the power, burnup and shutdown history are known. However, as shown in Figure 1, there is also a comparable release on reactor startup that needs to be considered.

3.3. Startup Release Analysis

Having established the release behaviour resulting from the shutdown, it is now necessary to determine the release that occurs on startup. There are two possibilities to consider. The fuel cracked on shutdown releasing stored inventory. At least a part of the startup release peak that is observed will be by diffusion from the increased surface area, until sintering heals the cracks. The second possibility is that additional cracking may occur, because of thermal stresses generated by the return to power, in which case additional stored inventory will be released as well.

The peak release for all but the very shortest lived isotopes, which required time to build up a measureable inventory, occured well before the reactor had reached full power. The actual fuel power at the time the maximum release rate was measured was obtained from operating history records for both FIO-133 and FIO-134. The measured peak release rate was compared with the steady state release rate for the same conditions, i.e. power and burnup. This required an analysis of the steady state behaviour.

3.3.1. Steady State Release Rates. Again, we are faced with the problem of analyzing something which possibly depends simultaneously on both power and burnup. We also have experiments where there were different swept areas (number and size of grooves) and fuel length (see Table 1). As before, ^{85m}Kr was chosen as the isotope upon which to place the most reliance.

The steady state release rates for all four experiments and all isotopes were plotted as a function of burnup. Figure 5 shows the results for ^{85m}Kr. The release rates have been normalized to identical fuel length, 477 mm, and identical swept areas of six grooves 1.15 mm x 1.15 mm by multiplying by the ratios of the swept areas.

Best estimate lines were drawn through the data for each isotope and the slope

calculated. Values varied from a low of 0.71 for ^{135}Xe to a high of 0.94 for both ^{87}Kr and ^{88}Kr. The value for ^{85m}Kr, i.e. the isotope which is considered to be the most reliable, was approximately 0.8. It should be noted, however, that this only applies to a burnup greater than about 10 MWh/kgU. At very low burnup, the effects of sintering of the fuel, discussed in section 3.1, also applies.

Having established a burnup dependence for the steady state release rate, the next step was to calculate a power dependence. Using a burnup dependence of 0.8, the steady state release rates were normalized to a nominal burnup of 50 MWh/kgU and plotted as a function of fuel power. The results for ^{85m}Kr are shown in Figure 6. As with other data, the scatter varied widely from one isotope to another. Best estimate lines were again drawn through the results for each isotope. The slope varied from about 3.7 to 5. Since the ^{85m}Kr is expected to be the most reliable data, the lower value is probably closer to the correct value.

3.3.2. Startup Release Fraction. The line shown on Figure 6, and similar plots for the other isotopes, were used to estimate the steady state release rate that would be expected for each isotope at the fuel power when the peak startup release rate was observed.

The long lived isotopes like ^{133}Xe generally show peak release rates hundreds or thousands of times greater than the expected steady state value for the same fuel power. However, for almost all startups, the fraction of inventory remaining at startup was large for these isotopes, i.e., usually greater than 0.95. Figure 7 shows the results for isotopes where the remaining inventory on startup was usually very much less. It shows the ratio of the observed startup peak release rate to the steady state release rate (i.e. the rate that is predicted to occur at the power at which the peak release rate was observed) versus the fraction of inventory of the isotope remaining when the reactor started up. The scatter is large, as would be expected considering all the uncertainties in picking steady state release rate values from Figures 6, and in calculating the remaining inventory of short lived isotopes when the shutdown period is probably little better known than to the nearest five to ten minutes. However, Figure 7 shows a clear trend, in spite of the large scatter. The more inventory that remains on startup, the higher the release. Therefore, there must be additional fuel cracking on startup, and hence release of stored inventory. This is consistent with the very high ratios calculated for the long-lived ^{133}Xe.

The data for ^{133}Xe was plotted separately on Figure 8 as the ratio of the peak release rate to the steady state release rate versus the fuel power at the time the peak release was measured. The figure shows no clear dependence on the fuel power. As a result, we conclude that the amount of fuel cracking that occurs on startup is an inherent characteristic and is not power dependent. This is not surprising if one considers that the fuel temperature depends on fuel power and above some temperature the UO_2 will become plastic and no longer crack due to imposed thermal stresses. Thus, we could expect that fuel will always crack on reactor startup until the appropriate power, and hence temperature, is reached. This should be true for all fuel elements.

Having established that additional cracking does occur on startup, the next problem is to try to calculate the magnitude of the release, particularly for the stable fission gas.

3.3.3. Stable Fission Gas Release on Startup. It was assumed that the stable fission gas release on startup would be most closely approximated by that of ^{133}Xe. Those startups were selected where the most detail, shortest counting times, was obtained so that there was a reliable value for the integrated release. Six startups were selected from each of FIO-133 and FIO-134. This integrated peak was corrected by dividing by the fraction of the inventory remaining at the time of reactor startup. A release fraction for startup was then calculated by dividing the corrected startup release by the pre-shutdown inventory. The results are shown on Figure 9 superimposed on the shutdown release fraction curve from Figure 4.

There are two major anomalies. The first data point for FIO-133 is low, but this may be because of the low burnup of 27 MWh/kgU. The datum for FIO-134 at 123 MWh/kgU, which is high by an order of magnitude, cannot be explained. The remaining data scatter about a line that would suggest that the startup release fraction is independent of burnup, with a value of about 8×10^{-4}.

4. MODEL APPLICATIONS

We now have values for the release behaviour both on shutdown and startup. On shutdown, the release fraction is independent of the decay constant as expected. It is dependent on the fuel operating power and on the burnup, and may be expressed by Eq. (5):

$$F_{SD} = 2.2(10^{-19})\ P^{6.5}\ B^{2.4} \qquad (5)$$

The release fraction on startup is independent of either power or burnup and may be expressed by the constant value obtained from Figure 9:

$$F_{SU} = 8 \times 10^{-4} \qquad (6)$$

It has been shown from both the sweep gas tests[5,6] and in-reactor tests where a pressure transducer continuously measured the internal fuel element gas pressure, reported by Notley et al[9] that the gas release during steady state operation is negligible compared with the release during power transients. The above two expressions, therefore, should give us a way of calculating the stable gas release expected from a fuel element based on its known power history. This was done for FIO-122 and FIO-124, where post irradiation gas puncture measurements were done on companion fuel elements. In addition, gas puncture results were available from several of both outer and inner elements of a fuel bundle SCA, which was irradiated in NRU in 1966 and 1967. It was irradiated to a burnup of 147 MWh/kgU on the inner elements and 217 MWh/kgU on the outer elements. Power and burnup data and the startup /shutdown history were obtained from the reactor operating logs. Gas puncture data has been previously reported[10].

The results are summarized on Table 2. Since we are calculating the fraction of the total amount of stable gas that is released, the most important shutdowns are those which occur towards the end of the irradiation. There were a total of 27 shutdowns during the

irradiation of FIO-122, 18 for FIO-124 and 117 for bundle SCA. The results for FIO-122 are within the measured range, for FIO-124 they are slightly low, and for fuel bundle SCA the calculated release for both the inner and outer elements are too high.

The history for bundle SCA showed that there were many shutdowns, particularly towards the end of the irradiation, where the time at power between startup and the next shutdown was short (in some cases less than one hour). There were two shutdowns for FIO-133 at a burnup of 7.6 MWh/kgU with only three hours between reactor startup and the second shutdown. The number of atoms released on the second shutdown were nearly two orders of magnitude lower than would be expected compared with shutdowns occurring after about 100 hours or more at power.

Unpublished photomicrographs from FIO-122 showed that cracks created in the UO_2 will resinter during subsequent irradiation. We postulate that if the fuel has only been at power for a short time before there is another reactor shutdown, the cracks produced in the UO_2 during the previous shutdown/startup cycle will not have had time to heal and the thermal stresses resulting from the new shutdown will tend to make cracks follow the paths of the previous, unhealed cracks. There would thus be little new inventory released since it had already been released by the previous shutdown and startup. The question then is how much time at power is required to complete resintering?

Plots of release rate as a function of time were available for FIO-133 and FIO-134. Examination of these plots showed that ^{135}Xe was probably the best isotope to analyze for determining the sintering kinetics. The signal is strong and unambiguous. Five startups from FIO-133 and six from FIO-134 were examined and the times to return to equilibrium were estimated directly from the plots. The times obtained for each experiment and the operating fuel power at each of these startups were averaged. Plots were not available for the other two experiments so times were estimated from the computer printout of the release rates. Only two startups from FIO-124 were suitable (i.e., the spectrometer was on for a long enough time after startup to unambiguously determine when equilibrium had been reached), and one from FIO-122. The results are shown on Figure 10. In spite of large scatter between individual measurements, the averages, as plotted, show remarkable consistency. These are probably overestimates of the time required to resinter because the reactor power is usually raised in steps so that full power is reached some time after the nominal time of the startup.

The next question is how much do the cracks have to resinter before the next shutdown will result in new cracks following a new path, and again release inventory stored in the grain boundary bubbles. We know that about 3 hours at a nominal power of 52 kW/m is insufficient since, as discussed above, the release was nearly two orders of magnitude lower than expected. Unfortunately we have no other useful data. The time required for complete resintering is almost certainly an over estimate. From Figure 10, the 3-hour data point is about 1/8 of the time required for complete resintering at 52 kW/m. Increasing the fraction of resintering required before cracks follow a new path made the predictions for both FIO-122 and FIO-124 worse, but improved them for the fuel bundle SCA. Three quarters of the time required for complete resintering was taken as a reasonable compromise. Thus, it is

assumed in the present calculation that no additional stored gas is released on shutdown or startup if the time between the previous startup and the given shutdown is less than 3/4 of that estimated to produce complete resintering. The results are also summarized on Table 2.

5. CONCLUSIONS

1. A correlation has been developed to predict the fraction of the total fission gas inventory released during reactor shutdown and startup. This work is based on analysis of release rates that were measured for short-lived isotopes in sweep gas experiments at the Chalk River Laboratories (linear powers from 43 to 60 kW/m, up to a burnup of 200 MWh/kgU).

2. The fission gas released from fuel on reactor shutdown can be estimated based on fuel power P (in kw/m) and burnup B (in MWh/kgU). The empirical equation derived for the fraction of the steady-state inventory released on shutdown is:

$$F_{SD} = 2.2\ (10^{-19})\ P^{6.5}\ B^{2.4}$$

The shutdown gas release fraction depends on the extent to which the fuel cracks from the thermal shock. The fuel cracks further on startup and the amount of startup cracking is relatively independent of the fuel power and burnup. A constant value for the fraction of the total inventory released on startup is deduced to be:

$$F_{SU} = 8 \times 10^{-4}$$

3. If the operating time between two consecutive startups is insufficient to resinter the cracks, the fractional gas release is orders of magnitude less than that calculated above. About 3/4 of the time estimated to completely resinter may be required before the next shutdown will produce cracks which follow a new path through the fuel.

4. The gas release calculated over the lifetime of the fuel agrees within a factor of ≈ 2 with gas puncture measurements provided several assumptions are made. The main assumption was that no further gas was released if the time between reactor startup and shutdown was less than 3/4 of that estimated to produce complete resintering of the cracks in the UO_2.

6. REFERENCES

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TABLE 1
EXPERIMENTAL DETAILS

Characteristic	CRL Experiment designation			
	FIO-122	FIO-124	FIO-133	FIO-134
Fuel	UO2	UO2	UO2	UO2
Enrichment	5.02	4.5	1.38	5.98
Density (Mg/m3)	10.71	10.65	10.65	10.64
Grain size (μm)	7	7	7.6	10
Pellet diameter (mm)	12.16	12.16	18.09	11.63
Fuel stack length (mm)	477	477	378	477
Number of swept grooves	6	4	3	3
Groove dimensions (mm)	1.15 x 1.15	1.15 x 1.15	1.3 x 1.0	1.3 x 1.0
Sheath	Zircaloy-4	Zircaloy-4	304L SS	304L SS
Outside diameter (mm)	13.11	13.11	19.85	13.06
Wall thickness (mm)	0.43	0.43	0.82	0.635
Coolant	Pressurized water	Pressurized water	Pressurized water, fog.	Pressurized water
Pressure (MPa)	8.5	8.5	8.5	8.5
Flow (kg/s)	1.1	1.1	0.24	0.24
Inlet temperature (°C)	240	240	260-275	260
Linear power range (kW/m)	39 - 45	58 - 63	52 - 62	52 - 61
Final burnup (MWh/kgU)	86	50	54.9	227
Total shutdowns	27	18	31	34
Shutdowns with data	3	4	16	12
Startups with data	6	6	15	10

TABLE 2
SUMMARY OF STABLE GAS RELEASE

Experiment	Percent Xenon released			3/4 of the time to complete sintering (h)
	Measured %	Calculated %		
		All Shutdowns and Startups	Allowing 3/4 of the time to complete sintering	
FIO-122	1.3 - 2	1.53	0.78	46
FIO-124	2.2 - 2.5	1.69	1.02	15
Bundle SCA				
Outer elements	15.15 - 20.15	46.2	24.6	22
Inner elements	0.04 - 0.62	6.8	2.37	65

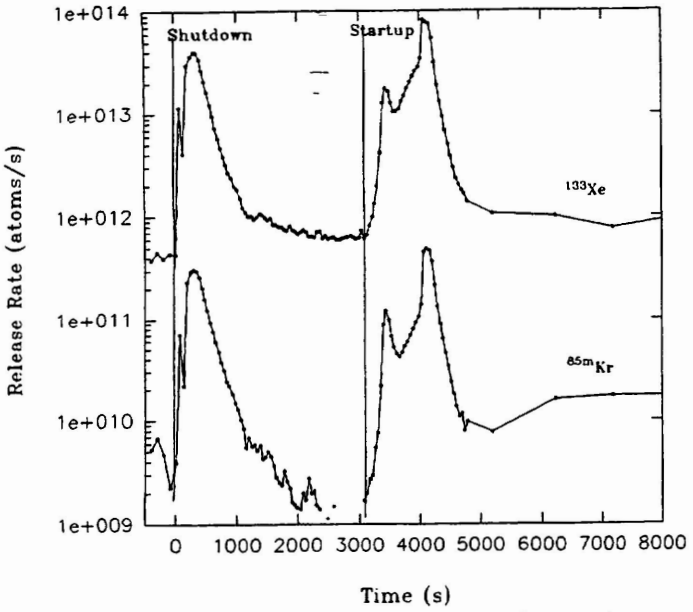

Figure 1. Shutdown and startup release rate versus time for November 16, 1981. The previouss steady state fuel power was 62.1 kW/m. and the burnup was 41.7 MWh/kgU. 19 hours of operation had occurred since the last startup.

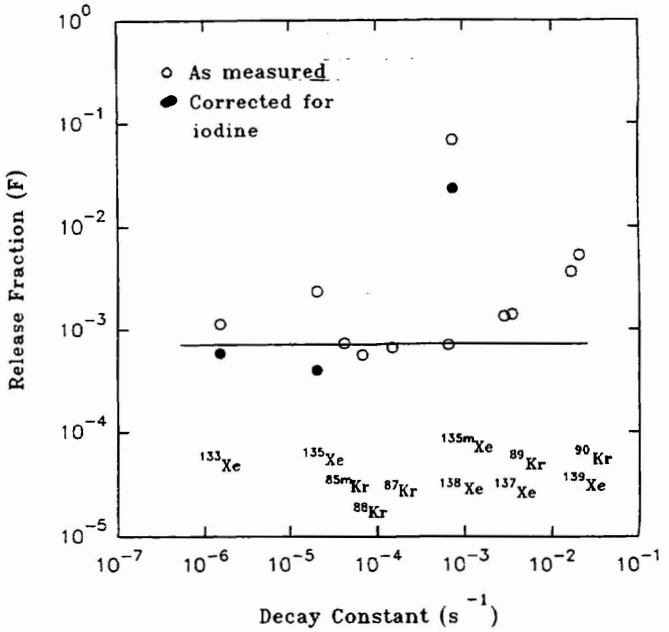

Figure 2. Shutdown release fractions for FIO-134 on October 24, 1982. 121.4 hours had occurred from the last startup. The previous steady state power was 54.9 kW/m and the burnup was 99.4 MWh/kgU.

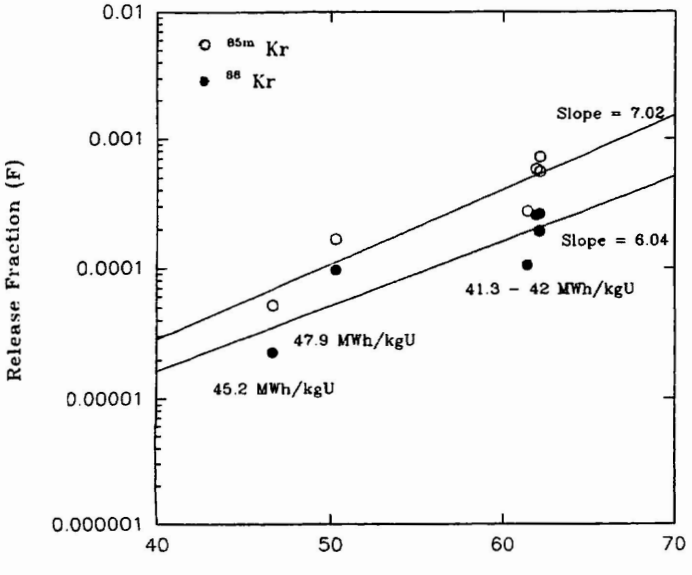

Figure 3. Release fraction versus fuel power at approximately constant burnup for two krypton isotopes. Data from FIO-133.

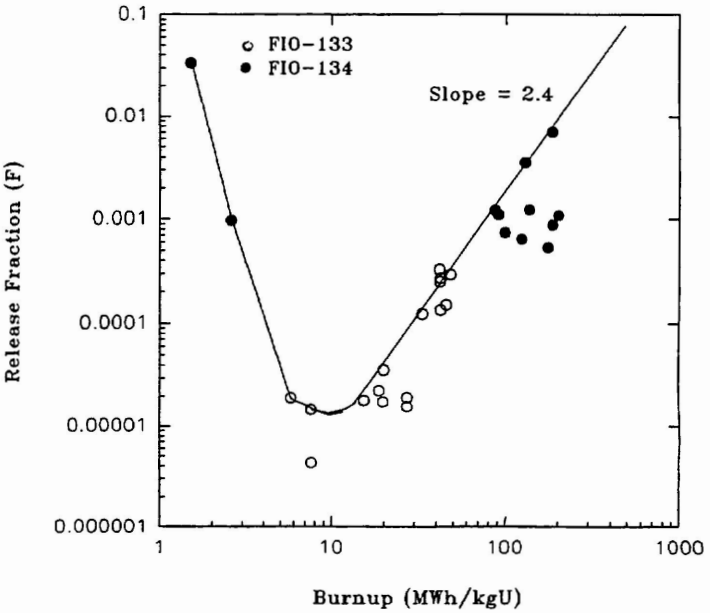

Figure 4. Burnup dependence of release fraction for ^{85m}Kr corrected to 55 kW/m fuel power.

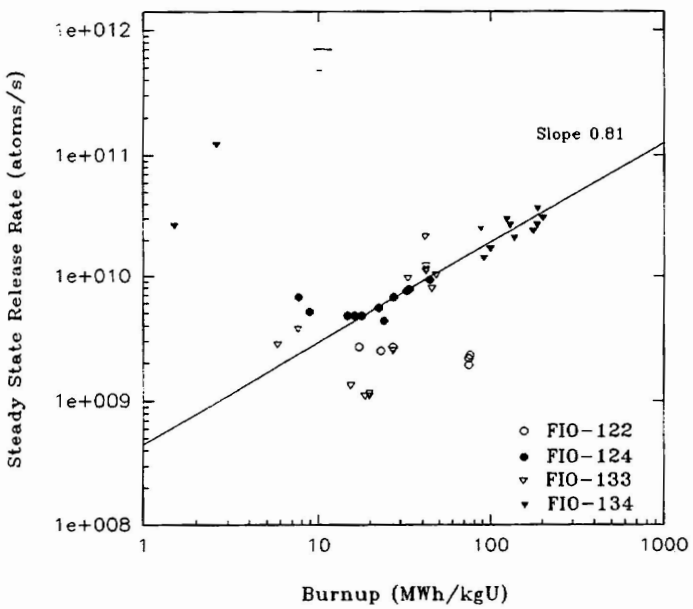

Figure 5. Steady state release rate normalized to 477 mm fuel length and six swept grooves (1.15 mm x 1.15 mm) for the isotope ^{85m}Kr.

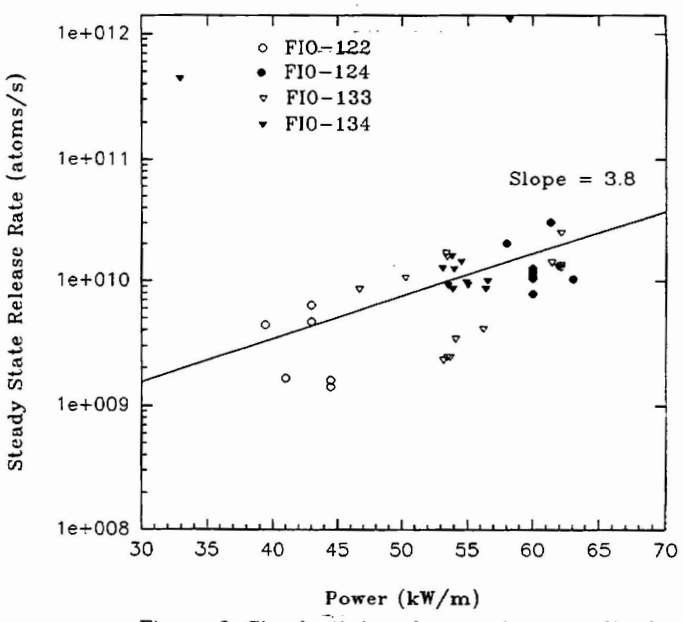

Figure 6. Steady state release rate normalized to 50 MWh/kgU burnup for the isotope ^{85m} Kr.

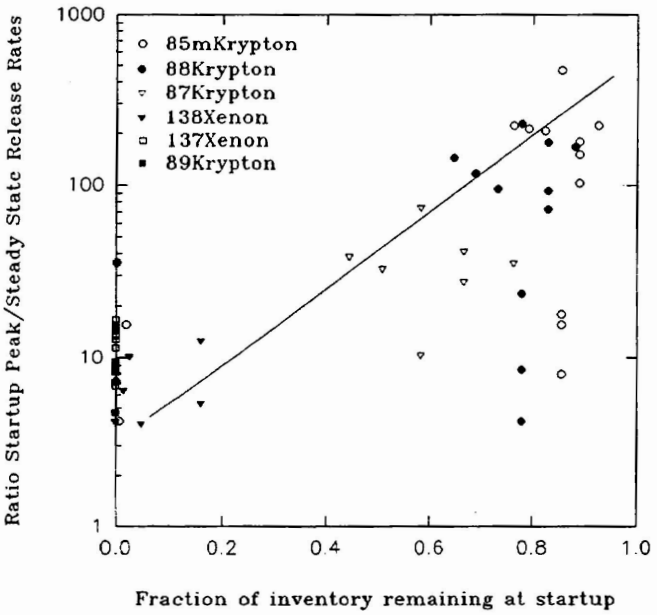

Figure 7. Ratio of startup peak release rates to the steady state release rate at the same power as the peak, for short-lived isotopes with short-lived precursors. Data from FIO-133 and FIO-134.

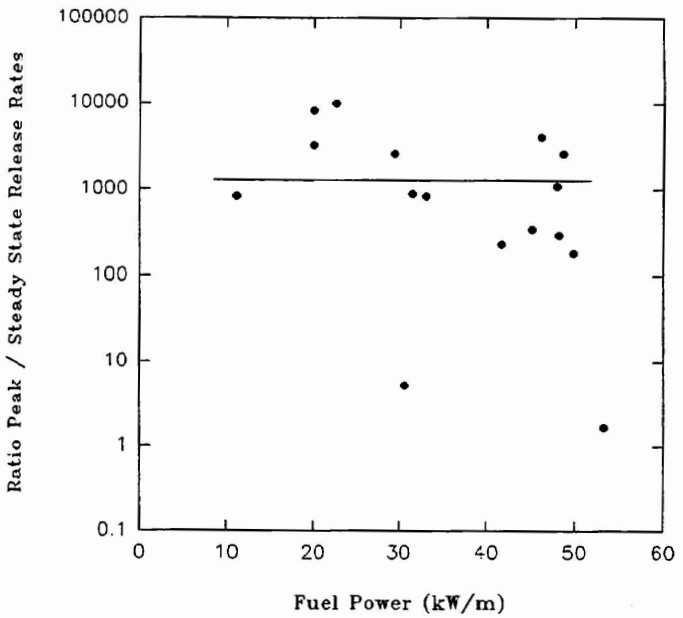

Figure 8. Ratios of the peak to the steady state state release rates at the power when the peak release was observed. Data for ^{133} Xe from FIO-133 and FIO-134.

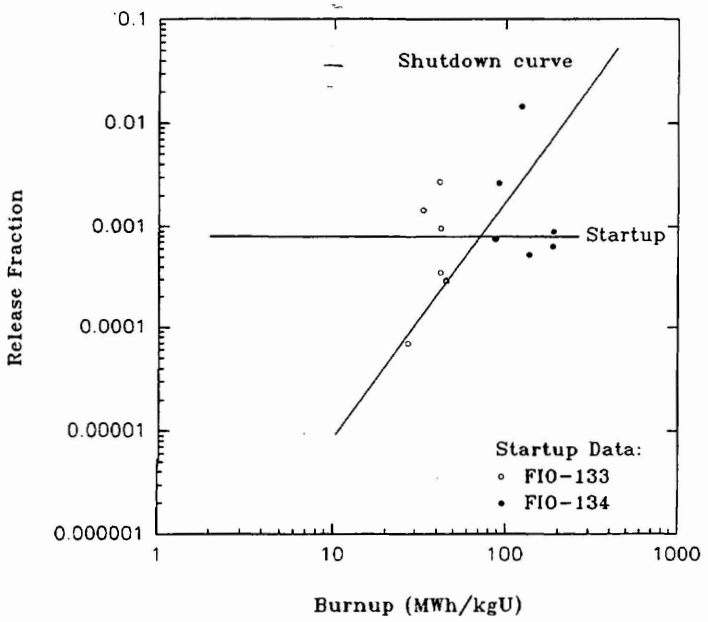

Figure 9. Startup release fraction versus burnup for ^{133}Xe. The shutdown curve from Figure 4 is included for comparison.

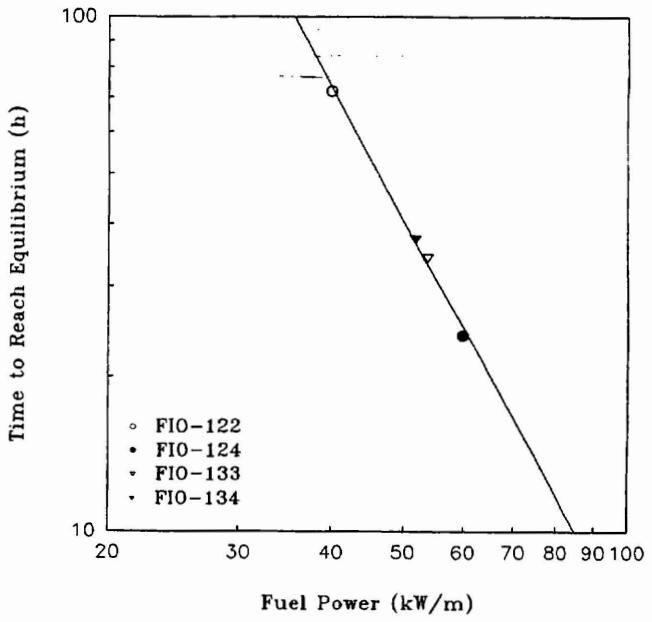

Figure 10. Estimated time required to re-sinter fuel cracks after reactor startup as a function of the fuel operating power, based on ^{135}Xe data.