
OECD HALDEN REACTOR PROJECT

**THE TREATMENT OF RADIOACTIVE FISSION GAS RELEASE MEASUREMENTS AND
PROVISION OF DATA FOR DEVELOPMENT AND VALIDATION OF THE ANS 5.4
MODEL**

by

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February 2001

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<p>This report reviews the experimental data from the Halden Sweep Gas Experiments available to develop and validate a revised ANS 5.4 radioactive fission gas release model. The isotope of particular interest because of its radiological importance is ¹³¹I and the report presents methods of scaling measured release of the radioactive gases, in particular ^{85m}Kr, to supplement the small number of direct measurements available.</p> <p>It is concluded that the best approach utilizes the recent analysis of White who describes the release process as one of single atom diffusion to a surface which can be regarded as having a fractal nature. A comparison of ^{85m}Kr release data scaled to that of ¹³¹I using this method shows good agreement with 'directly' measured iodine under identical irradiation conditions. Although the report has concentrated on obtaining data for ¹³¹I, the method of scaling can be used to obtain data for any other volatile species difficult to measure. This includes the other isotopes of iodine: ¹³³I and ¹³⁵I. However, caution should be used before extending the method to longer lived isotopes and to species where the diffusion coefficient may be different to that of the rare gases.</p>				
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1. INTRODUCTION

Two types of experiments have been used in the Halden reactor to investigate the release of fission gases from LWR fuel. The first employs internal pressure sensors from which the kinetics and quantity of stable gases can be measured during irradiation. The second is the use of a sweep gas to carry released fission gases from the fuel rod to a detector situated outside the reactor. With this equipment, it is possible to measure, using gamma spectroscopy, both radioactive and *stable* fission product release. In conjunction with fuel centreline thermocouples to measure fuel temperatures, these techniques have been successful in improving our understanding of the release process and the factors affecting it. The data generated have been used in many member countries to develop models and validate fuel performance codes used in reactor safety assessments.

In the sweep gas experiments, gas lines are attached to both ends of the fuel rod. This allows the introduction of a gas to pass through the free volume and carry entrained gases released from the fuel to a gamma detector situated outside but adjacent to the reactor. In this way, stable fission gas release can be inferred from measurements on the long lived isotope ^{85}Kr with a half life of ~ 10 years as well as the release of radioactive fission products, e.g. the radiologically important isotope ^{131}I from measurements of short lived krypton and xenon with half lives spanning ~ 90 secs to around 5 days. The release of ^{131}I is of greatest importance primarily due to its long half-life. Iodine release data are obtained by measuring the decay product $^{131\text{m}}\text{Xe}$. Estimates of ^{131}I inventories are also obtained by measurements on $^{85\text{m}}\text{Kr}$. Experiments to measure the release of short lived rare gases have made a significant contribution to our understanding of stable gas release, in particular in the determination of the in-pile diffusion coefficient and the processes occurring at grain boundaries.

The American Nuclear Society has recently formed a work group to review the current ANS-5.4 model with a view to improving it. Halden Project will take active part in this committee and supply experimental data. This paper will discuss measurements made with in-pile sweep gas experiments and the form and extent of the data to be used in revising the ANS model.

2. HALDEN EXPERIENCE WITH SWEEP GAS EXPERIMENTS

The experiments and method of analysis has been thoroughly reviewed by White and Turnbull in reference /1/. The following is a brief summary of the experiments and their analysis as presented previously.

2.1. The Experiments

The first Halden gas flow experiment was designated IFA-430 (Instrumented Fuel Assembly) which commenced irradiation in late 1978 and remained in the reactor for six years. The rig incorporated 4 PWR design rods, although only two were designed as gas flow rods. The principal aim of the experiment was to investigate axial gas flow as a possible limiting factor for clad ballooning during LOCA. All four rods employed 1.2 m long fuel stacks with the gas flow rods comprising 3 sub-stacks with pressure transducers fitted between to measure axial pressure gradients during steady and transient flow conditions. Although many useful fission product release data were obtained from this assembly, the information was of limited applicability

because of the long fuel stack and the range of power and burn-up along the length of the fuel column.

This experiment was soon followed by a further gas flow rig, IFA-504 designed specifically for fission product release studies. The assembly consisted of four rods with short active fuel lengths of 700 mm of different pellet design. Two rods contained solid pellets with pellets sourced from separate manufactures, one hollow rod and one fuelled with niobia doped UO₂ pellets. This assembly was loaded in December 1980 and is still operating successfully to date at a burn-up in excess of 75 MWd/kgUO₂ after 20 years continuous irradiation. Over this time the experiment has produced unique data on fuel thermal performance, hydraulic diameter as well as fission product release.

Based on the success of IFA-504, the Central Electricity Generating Board (UK), now British Energy, commissioned a similar experiment, IFA-558 to investigate the effect of internal pressure on the release of short lived fission products. The assembly carried 6 identical fuel rods 700 mm long containing hollow pellets of BNFL CONPOR fuel. The operating pressure in pairs of rods was maintained at 2, 20 and 40 bar during the entire irradiation except when being flushed periodically to measure release. After measuring short lived species by gamma counting, the gas from individual rods was trapped on liquid nitrogen cooled charcoal and stored until short lived fission products had decayed. The cold traps were then exposed to the gamma detector and the quantity of the long lived ⁸⁵Kr measured. Thus data were obtained simultaneously for both short and long lived fission gases.

The sweep gas technique has also been used on two experiments, IFA-563 and IFA-569 where the fuel was in the form of 1 mm thick disks of UO₂ sandwiched between disks of molybdenum to carry away most of the heat generated by fission. In this way the fuel operated at a near uniform temperature controlled by the fuel-clad gap and its gas composition.

Finally, sweep gas lines have been attached to re-instrumented fuel pre-irradiated to high burn-up in commercial reactors. Several loadings of this series of experiments, IFA-610, have been performed containing pre-irradiated UO₂ and MOX fuel.

2.2. Analysis of Sweep Gas Experiments

The fractional release of a radioactive fission product is measured as the ratio of the rate of release from the fuel, R in atoms/s, divided by the birth rate, also in atoms/s. For constant irradiation conditions over a period of around three half-lives, the system is at radioactive equilibrium and the total release (R/B)_{total} is the sum of two terms:

$$(R/B)_{\text{total}} = (R/B)_{\text{diffusion}} + (R/B)_{\lambda\text{-independent}} \quad (1)$$

The first term results from release by atomic diffusion and is dependent on temperature via a diffusion coefficient and a radioactive decay constant λ ; for a surface to volume ratio (S/V) and (R/B) < 0.1:

$$\left(\frac{R}{B}\right)_{\text{diffusion}} = \left(\frac{S}{V}\right) \sqrt{\frac{\alpha \cdot D}{\lambda}} \quad (2)$$

where α is a constant for each isotope representing diffusion by the precursor to the isotope under consideration. The second term is also proportional to (S/V) but is independent of half life and represents the contribution from knock-out and direct recoil. The parameters of interest in these equations are: the diffusion coefficient D which is common for all isotopes of krypton and xenon and hence the stable isotopes of these species, the surface to volume ratio (S/V), as this changes from one material to another and with irradiation. The equation for $(R/B)_{total}$ is often written in a phenomenological form:

$$\left(\frac{R}{B}\right)_{total} = A + B\sqrt{\frac{\alpha}{\lambda}} \quad (3)$$

where the coefficients A and B are referred to as the *recoil* and *slope* respectively and for a given gas flow measurement, are common for a range of species.

Referring to equation (3), the two unknowns in any experiment are (S/V) and the diffusion coefficient D, since α and λ can be defined separately and uniquely.

2.3. The Diffusion Coefficient

By comparing the release from polycrystalline UO₂ with release from single crystal material of known (S/V), an absolute determination of diffusion coefficient was obtained for the rare gases /2,3/. This was interpreted as comprising three terms: a high temperature intrinsic diffusion coefficient, an intermediate term depending on temperature and rating and a final athermal term observed at low temperature which was assumed to be proportional to rating. However, the low levels of release at low temperatures from the small amount of material used in these experiments, (~mgs), reduced the accuracy of determining this third component. This is not the case for the sweep gas experiments performed at Halden, where the active fuel column can weigh ~ 0.5 kg. Data from IFA-430, IFA-504 and IFA-558 suggested that this third term was not athermal but had a small temperature dependence. This was explored further in the start-up ramp for IFA-563, which confirmed the earlier findings, /4/, Figure 1. Thus the final diffusion coefficient D (m²/s) in terms of the mass rating R (W/gU) and the temperature TK (K), was revised as follows:

$$D = D_1 + D_2 + D_3$$

where:

$$D_1 = 7.6 \cdot 10^{-10} \cdot e^{-35000/TK} \quad (4)$$

$$D_2 = 1.38 \cdot 10^{-16} \cdot R^{1/2} \cdot e^{-13800/TK}$$

$$D_3 = 7.67 \cdot 10^{-22} \cdot R \cdot e^{-2785/TK}$$

This diffusion coefficient has been used to analyze much of the data from the gas flow rigs with particular reference to its applicability to the lifetime of the isotopes measured. It had been noticed /3/ that employing the 3 terms in a fuel performance code to estimate stable FGR, the code systematically over predicted the release at low powers. Prior to interlinkage of grain boundary porosity, the value of (S/V) is approximately the same throughout the pellets and the resulting release of a radioactive isotope will be proportional to a volumetric average of the square root of the diffusion coefficient. If the fuel temperature is also averaged in the same way, it is possible to identify a measured (R/B) with a characteristic diffusion coefficient and temperature. In this way,

for each gas flow measurement, the diffusion coefficient can be calculated for a range of isotopes of different half lives. This was applied to a full range of isotopes including ^{131m}Xe (12 days), ^{133}Xe (5 days). A compilation of the results is shown in Figure 2, where it can be seen that these long lived species are best represented by omitting the third term D_3 . Thus with this methodology it was recommended that stable and long lived species should be modelled using $D_1 + D_2$ whilst for short lived species, the full three term diffusion coefficient should be employed. The rationale behind this difference in behaviour was ascribed to D_3 being a near surface phenomenon, thus affecting all species close to the free surface. In which case, short lived species which, because of their short lifetime originated close to the surface, were released with kinetics appropriate to this enhanced diffusivity. The longer lived species spent much of their lifetime in the bulk of the material and hence their release was rate controlled by the bulk diffusion coefficient.

2.4. Fractal Nature of (S/V)

Recently, White /5/ performed a finite difference calculation to represent diffusional release from a sphere with an enhanced diffusion coefficient close to the surface. His calculations indicated that near-surface enhanced diffusion does not provide the correct behaviour. The observations are that the release of short lived species is enhanced at low temperatures whereas his calculations showed the opposite effect. This led him to conclude that the diffusion coefficient is the same for all species and independent of distance from the free surface. In which case, he argued that in equation (2), perhaps (S/V) does not have a unique value, but is dependent on the lifetime of the species being released. To explain this, he used the analogy of measuring a coastline with rulers of different length; the longer the ruler, the shorter the coastline. This 'fractal nature' leads immediately to an effectively larger value of surface area 'S' for short lived species. The effective (S/V) is determined by how far a species diffuses in its lifetime. The fractal nature of the surface is defined by two parameters, b_0 and b_1 related to α and λ of the species under consideration by:

$$(S/V) = b_0 D^{-1/2} \left(\frac{\alpha}{\lambda} \right)^{b_1/2}$$

See equation (18) in reference /5/ and note that b_0 is a function of the diffusion coefficient otherwise (R/B) would be independent of D. White went on to show that treating the surface in this manner explained the difference in release behaviour for isotopes of different half lives and also explained the origin of the 'lambda independent' term in equation (1).

3. THE RELEASE OF RADIOACTIVE IODINE SPECIES

The release of radioactive iodine from irradiated fuel poses little problem as long as the integrity of the cladding is maintained, because the iodine contributes little to the internal pressurization of the fuel rod. The situation is radically changed in the event of clad failure, particularly if this is associated with a breach of the reactor containment as in this case, the escape of iodine can pose a radiological health hazard. The effects of radioactive iodine are generally described by the term 'Dose Equivalent Iodine' (DEI), which attempts to quantify the health hazard in terms of a weighted sum of the five radioactive iodine fission products. Approximately half of the DEI arises from ^{131}I so calculating this component of release in fault situations is of great importance.

3.1. The measurement of Radioactive Iodine

From an experimental point of view, the iodine isotopes of interest are ^{135}I , ^{133}I and ^{131}I with half lives of 6.72 hours, 20.9 hours and 8.04 days respectively. Iodine isotopes are solid except at high temperatures and are not transported through the unheated flow lines to the gamma detector. Assessment of their release therefore presents some problems and must be performed indirectly by measurement of their xenon decay products. The situation is further complicated by the fact that the decay products, ^{135}Xe , $^{135\text{m}}\text{Xe}$, ^{133}Xe and $^{131\text{m}}\text{Xe}$ are themselves released in sufficient quantities as to render at-power iodine measurement difficult. This situation is circumvented by exploiting the fact that production and release of the isotopes effectively ceases at reactor shut down. Any excess xenon detected after shut-down must have arisen from decay of iodine isotopes already released and plated out on cooler parts of the gas circuit. Therefore, at shut-down, the rods are flushed thoroughly to remove any xenon actually released as xenon. This is followed by a number of on-line flow measurements taken at intervals over the following 24 hours. In this way the release of ^{135}I and ^{133}I can be inferred from the ^{135}Xe , $^{135\text{m}}\text{Xe}$ and ^{133}Xe activity.

The determination of ^{131}I release is more complicated. 99% of its decay results in the stable ^{131}Xe isotope, the remaining 1.086% yields $^{131\text{m}}\text{Xe}$ which has a half life of 11.9 days and whose gamma energy is in close proximity to a strong gamma ray from the decay of ^{133}Xe (5.29 days half-life). The technique adopted exploits the different half-lives of the isotopes in the decay chain. If the rod is sealed, after 12-14 days, the concentration of $^{131\text{m}}\text{Xe}$ reaches a maximum and when swept out to the gamma detector, it can be distinguished from any residual amounts of ^{133}Xe and used to estimate the quantity of ^{131}I released immediately prior to shut-down. The requirement for a long shut-down for this measurement means that only a few estimates of ^{131}I release have been possible.

3.2. Iodine Diffusion Coefficient

The diffusion coefficient described in equation (4) applies strictly to the rare gases krypton and xenon. During experiments immediately preceding those from which the diffusion coefficient was derived, Turnbull and Friskney /6/ investigated the release from small samples of both rare gases and volatile species including caesium, tellurium and iodine. Here the released volatile species were trapped out on a 'cold finger' situated above the heated specimens during irradiation. This was removed at the end of each irradiation cycle and the fission products collected and measured. At a specimen temperatures of ~1400-1500 °C it was found that to a good approximation both gaseous and volatile species were released from the fuel with identical kinetics, Figure 3. This study was extended by Friskney and Turnbull /7/ to cover a wider range of temperatures, 770-1450 °C. Here, iodine diffusion coefficients derived from plate out measurements and from analysis of precursor mobility using the method of Friskney and Speight /8/ were compared to those of the rare gases. They concluded that over this temperature range, there was no discernible difference between the diffusion coefficient for iodine and that for krypton and xenon. It appeared therefore that equation (4) can be used equally for the release of iodine isotopes, and as will be seen in the next section, this conclusion has been substantiated and refined with data from Halden sweep gas experiments.

3.3. Data on Iodine release

In all, a total of 21 'direct' measurements of ^{131}I release were made from the Halden gas flow experiments and these are given in Table 1. The paucity of data is because of the difficulty in making these direct measurements. Because of this, a means of using data from the rare gases was devised in order to increase the ^{131}I database. In the early 1980s, Turnbull /9/ analysed fission gas release from a number of swept fuel experiments and based on the results of the experiments discussed above, assumed that the release of ^{131}I was essentially by single atom diffusion and hence could be calculated by simply scaling by the square root of the ratio of the half-lives of a chosen gaseous fission product and ^{131}I . Since the longest lived rare gas for which reliable data were generally found was $^{85\text{m}}\text{Kr}$ with a 4.48 hour half life, this isotope was chosen for the calculation. The release to birth ratio (R/B) for ^{131}I could therefore be written as:

$$\left(\frac{R}{B}\right)_{131} = \left(\frac{R}{B}\right)_{85\text{m}} \times \sqrt{\frac{\lambda_{85\text{m}}}{\lambda_{131}}} \quad (6)$$

This implies that (R/B) for ^{131}I is of the order of 6.56 times that for $^{85\text{m}}\text{Kr}$.

From more recent studies using the Halden experiments, it is clear that this method contains a number of conservative assumptions:

1. The scaling process neglects the fact that $^{85\text{m}}\text{Kr}$ release is enhanced by rapid diffusion of its ^{87}Br precursor. This will not be the case for iodine isotopes.
2. The scaling process ignores the presence of a recoil release component, (eq. (3)).
3. Differentiating between diffusion coefficients, release of the shorter lived $^{85\text{m}}\text{Kr}$ is best described using all three of the diffusion coefficient contributions, $D_1 + D_2 + D_3$ in eq. (4) whilst ^{131}I is best described using only $D_1 + D_2$.
4. The fractal treatment of the surface offers a better description of the release process.

Historically, some of these different methods of scaling have been used to evaluate data for the release of ^{131}I , hence it is worth considering each in turn in order to compare the values of (R/B) derived. The first assumption is relatively small and the second is easily corrected by replacing equation (6) with the following, using the nomenclature of equation (3):

$$\left(\frac{R}{B}\right)_{131} = A + B \sqrt{\frac{1}{\lambda_{131}}} \quad (7)$$

Figure 4 shows the results of comparing 'direct' measured (R/B) ^{131}I with that derived from $^{85\text{m}}\text{Kr}$ using the original scaling method, labelled 'K' and using equation (7) above, labelled 'R'. The results displayed in the figure confirm that both scaling methods are indeed pessimistic when compared with the actual measured values for ^{131}I . For the original scaling method, the predicted/measured statistics are $P/M = 4.6 \times \pm 1.74$ and for the recoil and slope scaling method $P/M = 2.71 \times \pm 1.74$. The revised method is clearly less pessimistic yet still overestimates by a factor approaching 3.

Correction for the difference in diffusion coefficient requires more computation and requires an estimate for the volume averaged temperature. In which case, with values of the coefficients A and B, derived from the rare gases, the equivalent fractional release for ^{131}I is given by:

$$\left(\frac{R}{B}\right)_{131} = A + B \times \sqrt{\frac{(D_1 + D_2)}{(D_1 + D_2 + D_3) \cdot \lambda_{131}}} \quad (8)$$

This correction has been applied to the data for IFA-504 and IFA-558 and the results identified as the points 'D' included in Figure 4. For these $P/M = 0.864 \times \div 1.72$, but if the lowest data point is removed, $P/M = 1.26 \times \div 1.62$. This method is much closer to a best estimate evaluation with predicted values occurring either side of the 'true' value.

Using the fractal approach to explain radioactive fission product release, a different scaling procedure must be used between ^{131}I and $^{85\text{m}}\text{Kr}$. Using equation (5):

$$(S/V)_{131} \left(\frac{\lambda}{\alpha}\right)_{131} = (S/V)_{85\text{m}} \left(\frac{\lambda_{85\text{m}}}{\alpha_{85\text{m}}}\right)^{b_1/2}$$

Using equation (2) for ^{131}I :

$$(R/B)_{131} = (S/V)_{131} \sqrt{D} \sqrt{\left(\frac{\alpha}{\lambda}\right)_{131}}$$

since $\alpha_{131} = 1$:

$$(R/B)_{131} = (R/B)_{85\text{m}} \left(\frac{\lambda_{85\text{m}}}{\lambda_{131} \alpha_{85\text{m}}}\right)^{(b_1+1)/2}$$

The scaling factor F is given by:

$$F = \left(\frac{\lambda_{85\text{m}}}{\lambda_{131} \alpha_{85\text{m}}}\right)^{(b_1+1)/2}$$

Using data from IFA-558 rod 3, White /5/ evaluated b_1 as ~ -0.5 , hence $F = 2.39$. He applied these to data from IFA-430, -504 and -558 as presented in Table 1 and shown in Figure 4 identified as the points labeled 'F'. In this case, $P/M = 1.36 \times \div 2.11$, again providing best estimate values of (R/B) for ^{131}I .

It is clear that the 'diffusion scaling' and 'fractal scaling' provide the best methods of obtaining reliable data for ^{131}I release. The measured values of (R/B) for $^{85\text{m}}\text{Kr}$ for all 6 rods of IFA-558 have been scaled by both the 'diffusion' and 'fractal' methods to obtain equivalent values of (R/B) for ^{131}I and the results presented along with 'direct' measurement of ^{131}I as a function of burn-up in Figures 5(a-f). It can be seen that both methods of scaling produce similar values for ^{131}I which are close to the direct values. The fractal method has clear advantages both from a physical point of view as well as an ease of application. This method alone has been used to translate data for IFA-504 as shown in Figures 6(a-d). Early in the experiment, the $^{85\text{m}}\text{Kr}$ data translate well to the directly measured values of (R/B) for ^{131}I . at face value this agreement does not persist at high

burn-up, where scaled values of (R/B) for ^{85m}Kr fall significantly below direct measurements for ^{131}I . This however is because the iodine values relate to a linear power of around 22 kW/m where the rare gas measurements could only be made at lower powers, ~11 kW/m since only at these powers was there a sufficiently large fuel-to-clad gap to allow passage of the sweep gas. There is therefore no systematic discrepancy at high burn-up with scaled krypton measurements providing adequate assessment of ^{131}I release albeit at low powers.

4. REVISION OF THE ANS 5.4 MODEL FOR CALCULATING RADIOACTIVE FISSION PRODUCT RELEASE

The current ANS 5.4 model is known to significantly over predict the quantities of ^{131}I released into the fuel-clad interspace for intact fuel rods, with values closer to 10% than the <1% observed. The American Nuclear Society has therefore set up a Working Group to revise the model in the light of data now available. Such data are available from experiments performed at AECL Chalk River, CEA Grenoble and of course Halden, as described here.

For the purposes of developing and validating the new model, Halden Project propose to make all their gas flow experimental data available. This will include direct measurements of iodine and (R/B) values derived from rare gas data corrected by methods described above. In an external paper, /10/, it was suggested that the preferred method to be employed was the 'recoil plus slope scaling' method despite it producing systematically conservative values of ^{131}I release compared to the direct measurements. With the new analysis by White it is clear that the fractal treatment of radioactive fission product release provides a better description of the release phenomenon than the proposed two and three term diffusion coefficient used earlier. Not only does it provide a better physical interpretation, but it allows a very simple translation between the measured release of an isotope, in this case ^{85m}Kr and the one required, in this case ^{131}I . Supporting this, a comparison between corrected values and direct measurement of ^{131}I under the same reactor conditions shows a good agreement. For this reason, the fractal method of correction will be applied to measured values of (R/B) for ^{85m}Kr which amount to multiplying the values by a constant factor of 2.39.

An indication to the extent of data available from Halden can be obtained from the following table.

IFA	Average powers (kW/m) in Burn-up range (MWd/kg)						
	1-10	10-20	20-30	30-40	40-50	50-60	>60
430	20	20-40	20				
504	20-25	20-30	15-38	15-32	25-30	20-28	15-12
558	25-35	30-40	25-40	25-30			
563C1	40-50	40-50	30-40				
563C2	45-60	50-60	35-45				
563C4	15-30						
569	100-120	90-110	60-100	80-90	70-90	80	
610						15-16	

Note that for material irradiated in the form of 1mm thick disks, the powers correspond to temperatures across the disks of:

IFA-563C1	1100-1200 °C
IFA-563C2	1150-1300 °C
IFA-563C4	800-1000 °C
IFA-569	900-1150 °C

Thus the envelope of Halden data in terms of rod power and burn-up is:

Burn-up (MWd/kg)	Rod Power (kW/m)
0-10	35
10-30	40
30-60	30
60-70	15
>70	12

With the drive to higher discharge burn-up levels in commercial reactors there is clearly a need for higher power data at high burn-up. Fortunately this will be addressed by the new Joint Programme disk irradiation experiment IFA-655 which will commence operation early this year and remain in the reactor until discharge at around 100 MWd/kg.

5. CONCLUSIONS

This report has reviewed the experimental data from the Halden Sweep Gas Experiments available to develop and validate a revised ANS 5.4 radioactive fission gas release model. The isotope of particular interest because of its radiological importance is ^{131}I . Unfortunately because this is a solid at ambient temperatures, it is not an isotope which is easily measured. Consequently, there are only a few direct measurements of ^{131}I release which have been made. There is sufficient evidence to support the assumption that the diffusion coefficient for iodine is well represented by that for the rare gases krypton and xenon. It is possible therefore to overcome this paucity of data on iodine by appropriate scaling of measured release of the radioactive gases, in particular $^{85\text{m}}\text{Kr}$. Various method of scaling have been used in the past and reviewed here.

It is concluded that the best approach utilizes the recent analysis of White /5/ who describes the release process as one of single atom diffusion to a surface which can be regarded to have a fractal nature. This essentially allows an enhancement of short-lived species compared to long-lived and stable isotopes. A comparison of $^{85\text{m}}\text{Kr}$ release data scaled to that of ^{131}I using this method shows good agreement with 'directly' measured iodine under identical irradiation conditions. Although the report has concentrated on obtaining data for ^{131}I , the method of scaling can be used to obtain data for any other volatile species difficult to measure. This includes the other isotopes of iodine: ^{133}I and ^{135}I . However, caution should be used before extending the method to longer lived isotopes and to species where the diffusion coefficient may be different to that of the rare gases. For example, it is not clear whether or not the method can be applied to radioactive isotopes of tellurium.

An overview of the Halden gas flow experiments shows that release data are available up to 70 MWd/kg, but at this burn-up, the power is rather lower than required. Fortunately this will be

remedied by loading of the new Joint Programme disk irradiation experiment IFA-655 which has a designed discharge burn-up of ~100 MWd/kg.

6. REFERENCES

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Table 1

Comparison between measured ^{131}I and release estimated from rare gas data by different methods as discussed in the text

Rod	Burn-up MWd/kgUO ₂	Measured	85mKr scaling	Recoil + slope scaling	Diffusion corrected scaling	Fractal scaling
430-2	6.5	1.2E-4	1.6E-3	1.6E-3		1.7E-3
	7.7	3.8E-4	7.2E-4	7.2E-4		3.8E-4
	12.6	1.2E-3	3.8E-3	3.8E-3		1.9E-3
	16.9	1.2E-3	1.2E-2	1.2E-2		5.8E-3
504-1	9.3	1.2E-4	4.8E-4	2.7E-4	5.4E-5	9.8E-5
	46.4	2.6E-3	1.1E-2	9.2E-3	4.87E-3	4.0E-3
504-2	2.1	2.1E-4	1.2E-3	6.7E-4	2.8E-4	3.9E-4
	9.3	1.5E-4	3.9E-4	2.6E-4	5.7E-5	9.8E-5
	49.4	9.6E-4	8.7E-3	4.0E-3	1.57E-3	1.8E-3
504-3	2.1	2.5E-4	5.9E-4	4.3E-4	1.9E-4	3.0E-4
	9.3	1.2E-4	2.2E-4	1.4E-4	3.4E-5	4.8E-5
	49.4	2.8E-3	9.4E-3	5.2E-3	4.5E-3	2.3E-3
504-4	2.1	2.5E-4	1.3E-3	7.5E-4	2.4E-4	4.6E-4
	9.3	1.2E-4	5.4E-4	5.0E-4	9.9E-5	2.2E-4
	48.4	2.6E-3	1.3E-2	8.4E-3	1.6E-3	4.0E-3
558-1	30.3	3.5E-4	1.2E-3	6.6E-4	4.8E-4	2.5E-4
	31.9	5.7E-4	1.2E-3	8.1E-4	3.1E-4	4.3E-4
	34.3	2.9E-4	1.3E-3	7.6E-4	3.4E-4	3.7E-4
558-3	29.1	4.1E-4	1.9E-3	1.1E-3	5.9E-4	4.2E-4
558-6	31.6	3.8E-4	1.5E-3	8.4E-4	3.1E-4	3.6E-4
	33.9	2.7E-4	1.5E-3	8.4E-4	3.4E-4	4.1E-4

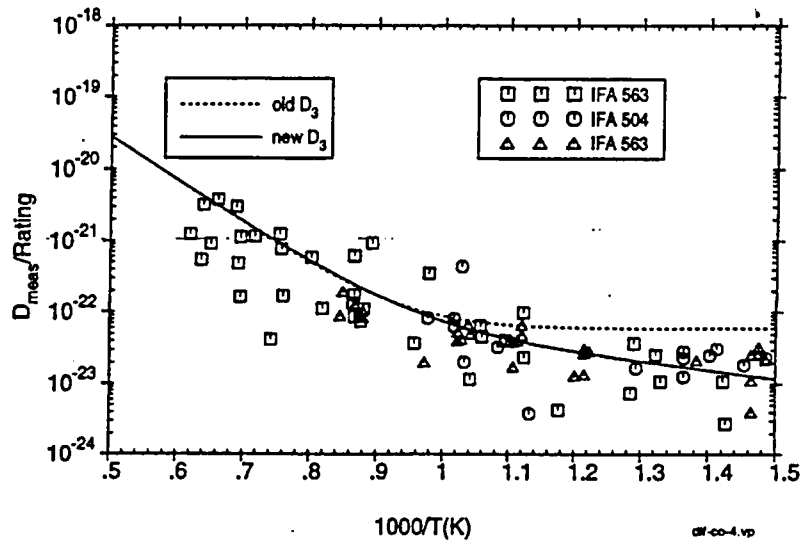


Figure 1 Revised formulation of low temperature term, D_3 , in the rare gas diffusion coefficient

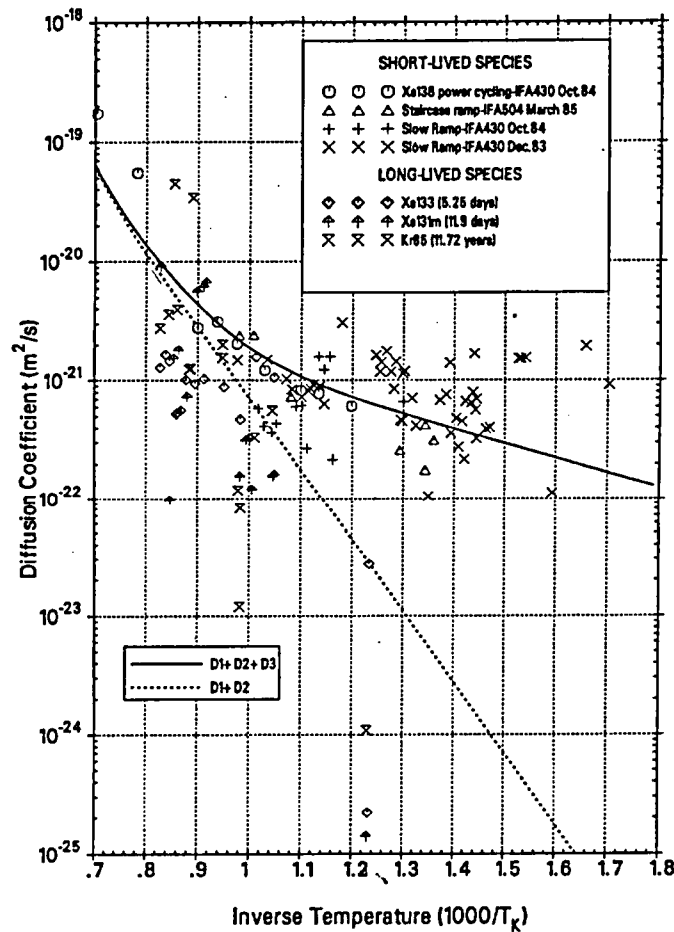


Figure 2 Plot of diffusion coefficient and its evaluation from long-lived and short-lived isotope release from Halden Gas Flow Experiments.

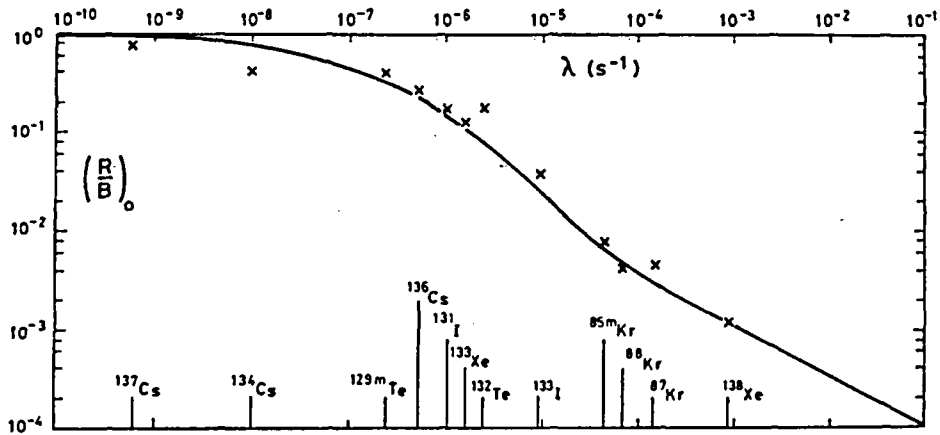


Figure 3 Fractional release (R/B) for a range of gaseous and volatile fission products versus decay constant taken from reference /6/.

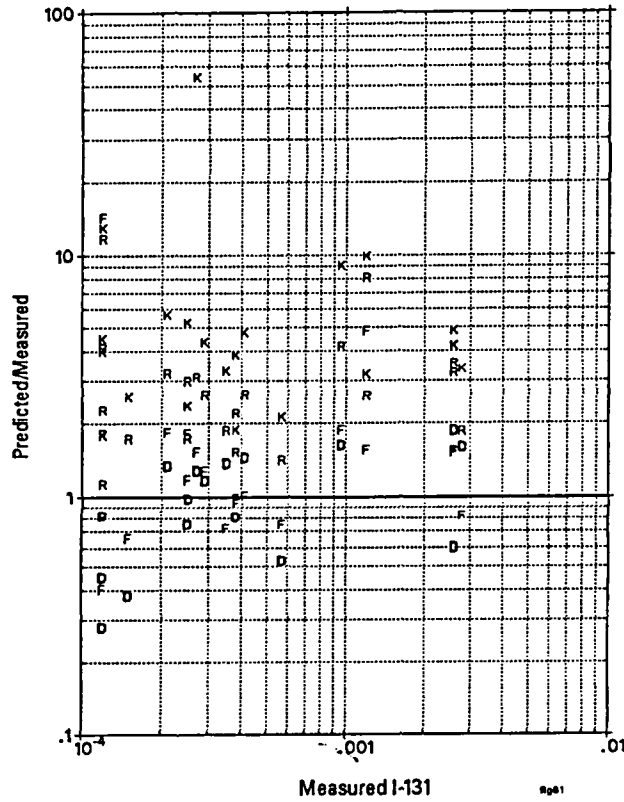


Figure 4 Values of Predicted to Measured ratio (P/M) for ¹³¹I release using various methods of derivation from measured rare gas data: K, original scaling by half life from ^{85m}Kr, R, recoil plus slope method, D diffusion coefficient corrected and F, fractal method of scaling from ^{85m}Kr. See text for explanation.

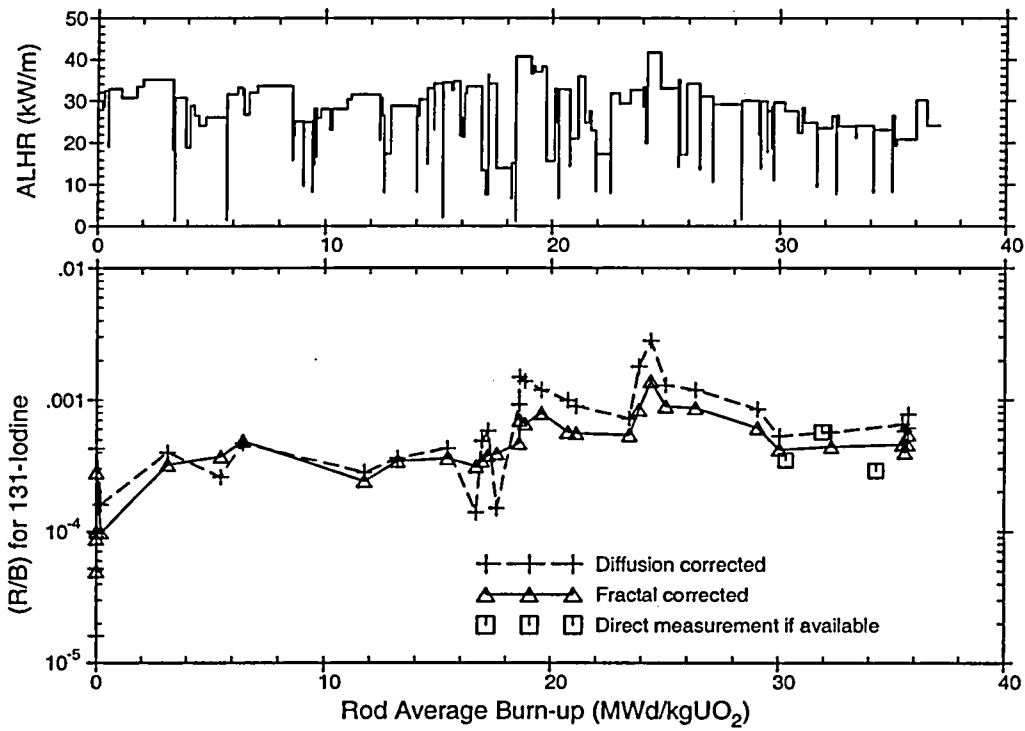


Figure 5(a) Calculated values of (R/B) for ¹³¹I from measured ^{85m}Kr; IFA-558 rod1

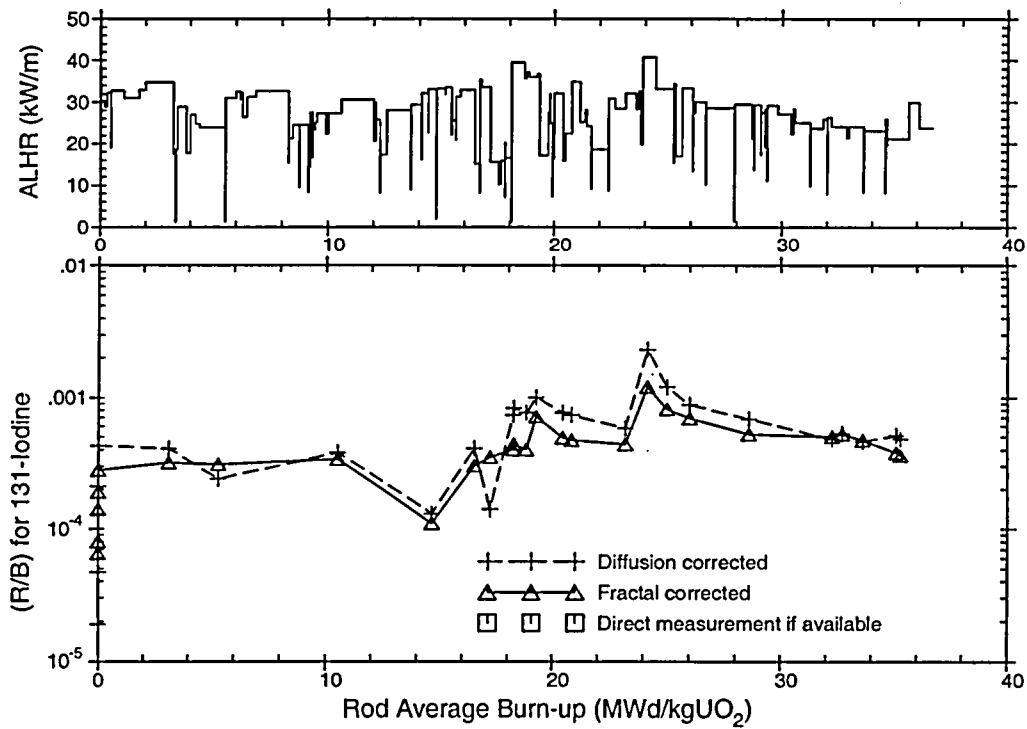


Figure 5(b) Calculated values of (R/B) for ¹³¹I from measured ^{85m}Kr; IFA-558 rod2

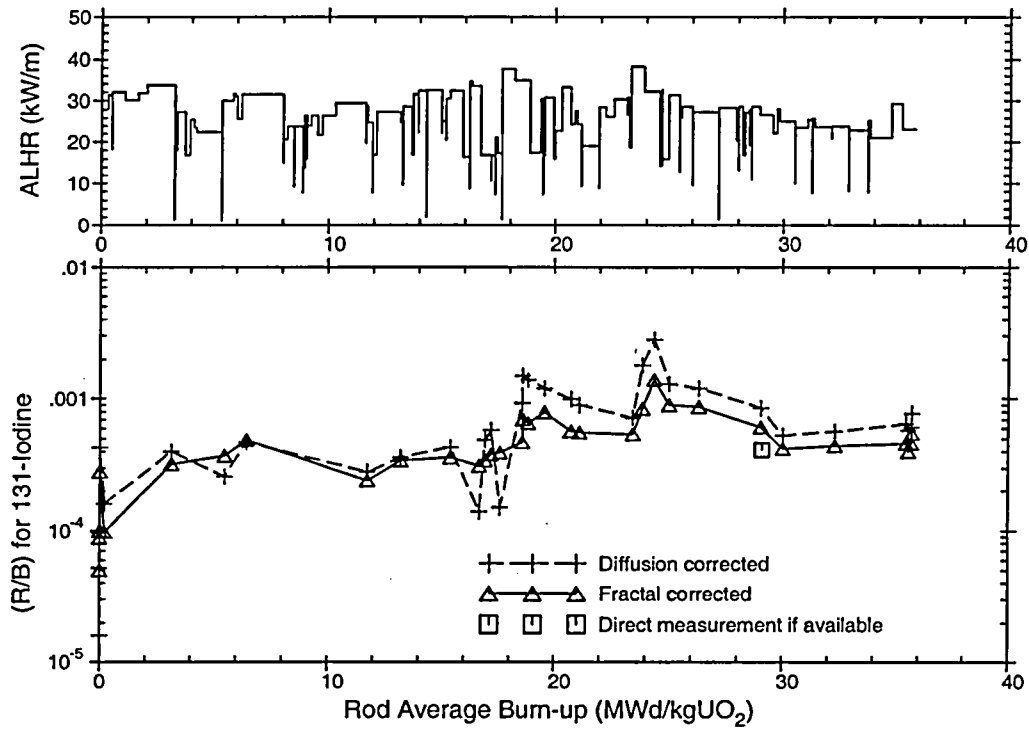


Figure 5(c) Calculated values of (R/B) for ¹³¹I from measured ^{85m}Kr; IFA-558 rod 3

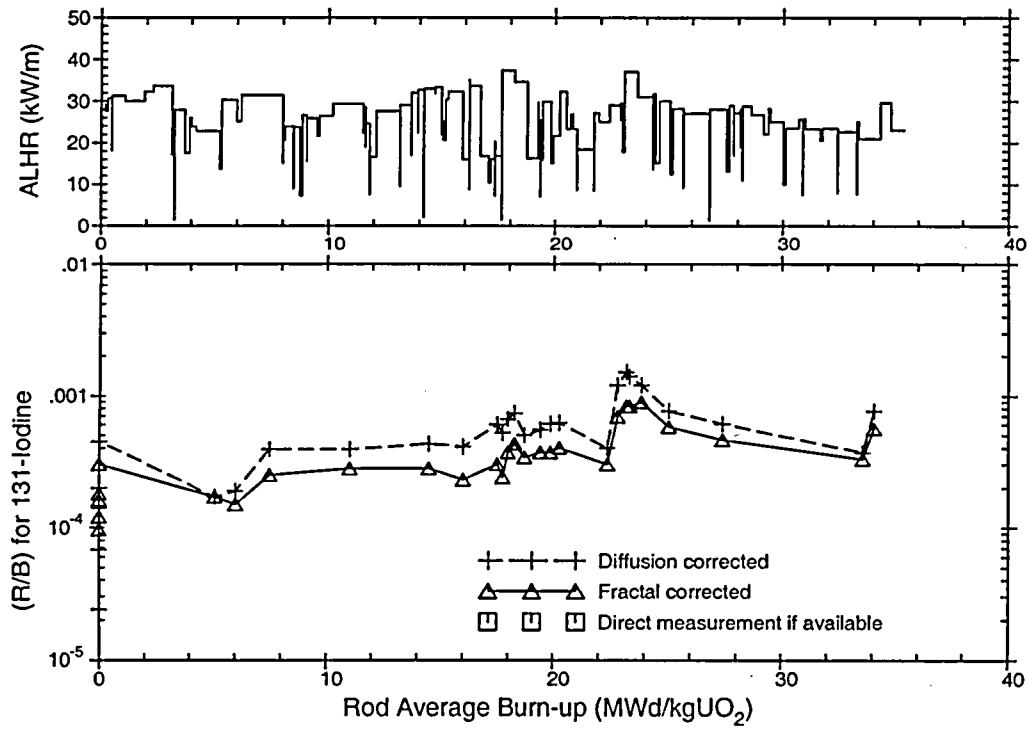


Figure 5(d) Calculated values of (R/B) for ¹³¹I from measured ^{85m}Kr; IFA-558 rod 4

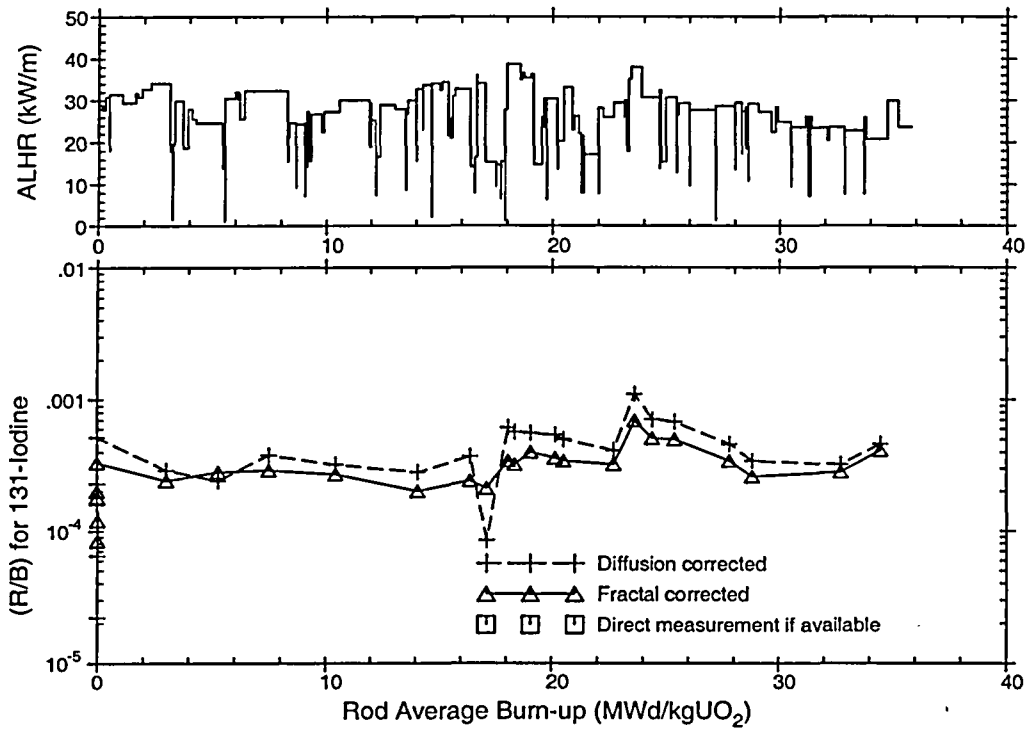


Figure 5(e) Calculated values of (R/B) for ¹³¹I from measured ^{85m}Kr; IFA-558 rod 5

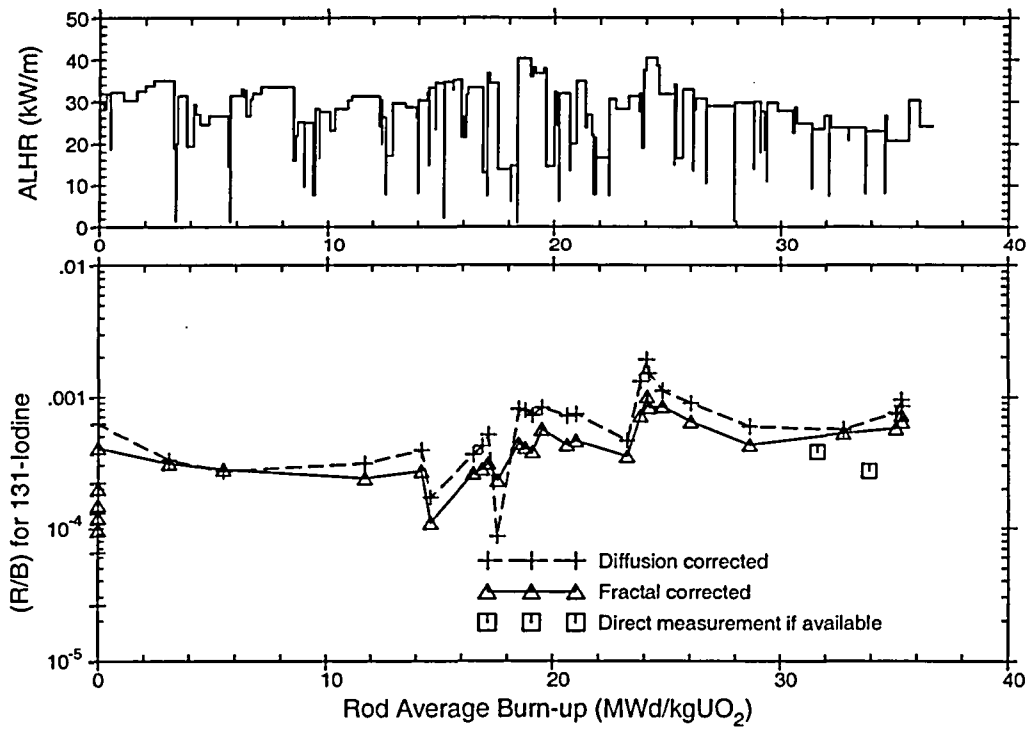


Figure 5(f) Calculated values of (R/B) for ¹³¹I from measured ^{85m}Kr; IFA-558 rod 6

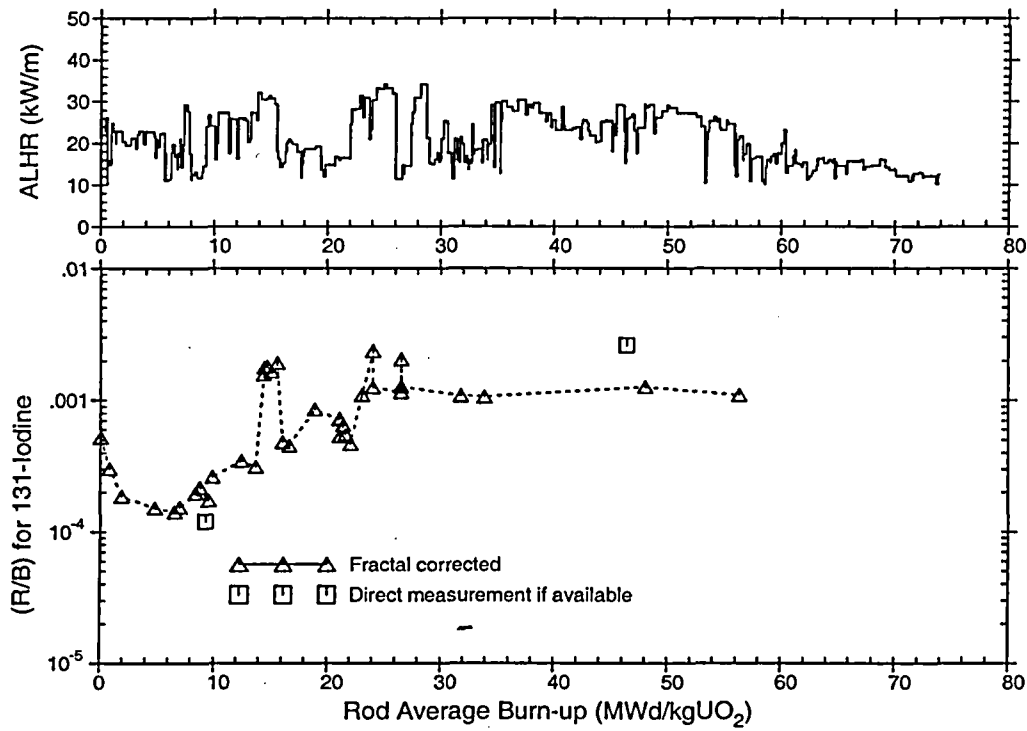


Figure 6(a) Calculated values of (R/B) for ¹³¹I from measured ^{85m}Kr; IFA-504 rod1

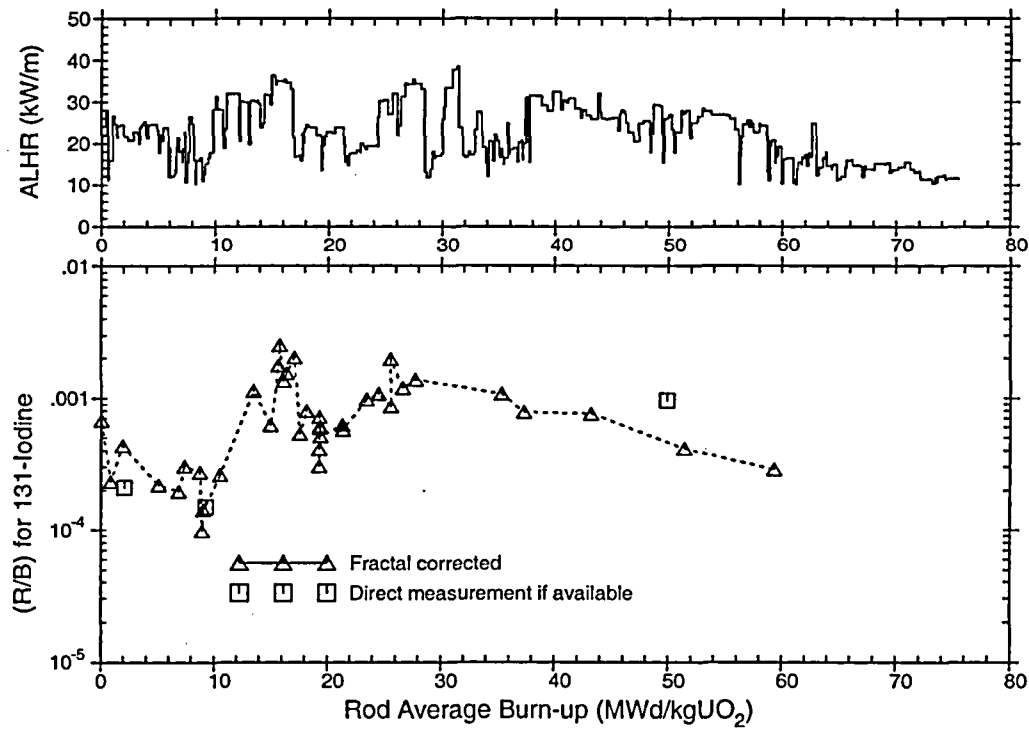


Figure 6(b) Calculated values of (R/B) for ¹³¹I from measured ^{85m}Kr; IFA-504 rod2

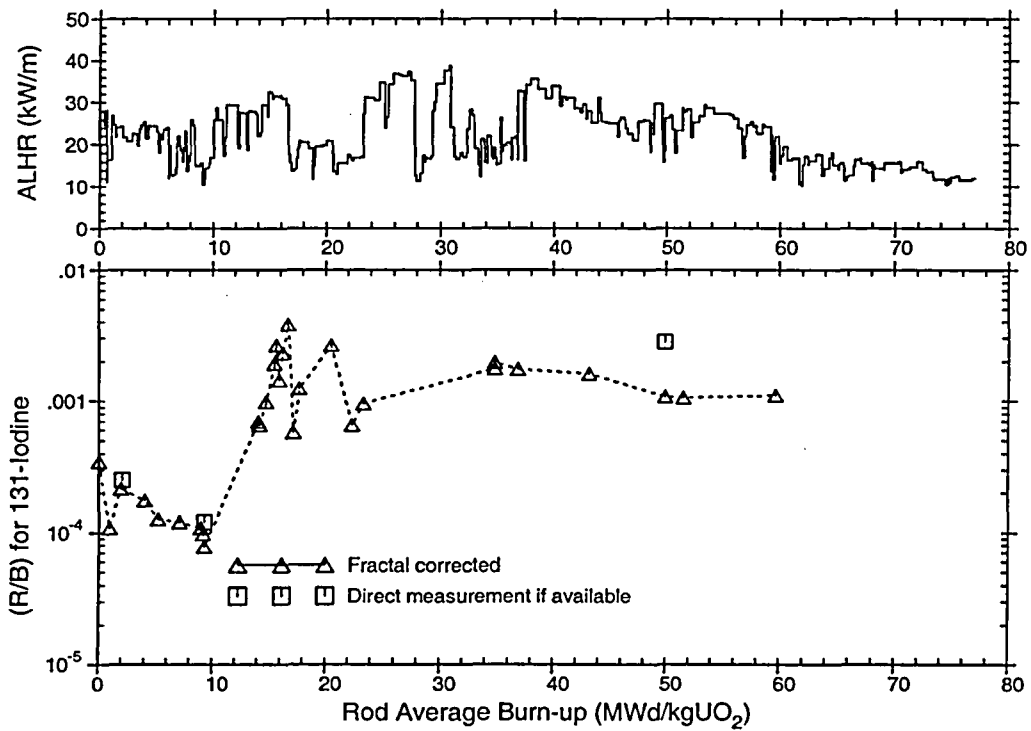


Figure 6(c) Calculated values of (R/B) for ¹³¹I from measured ^{85m}Kr; IFA-504 rod 3

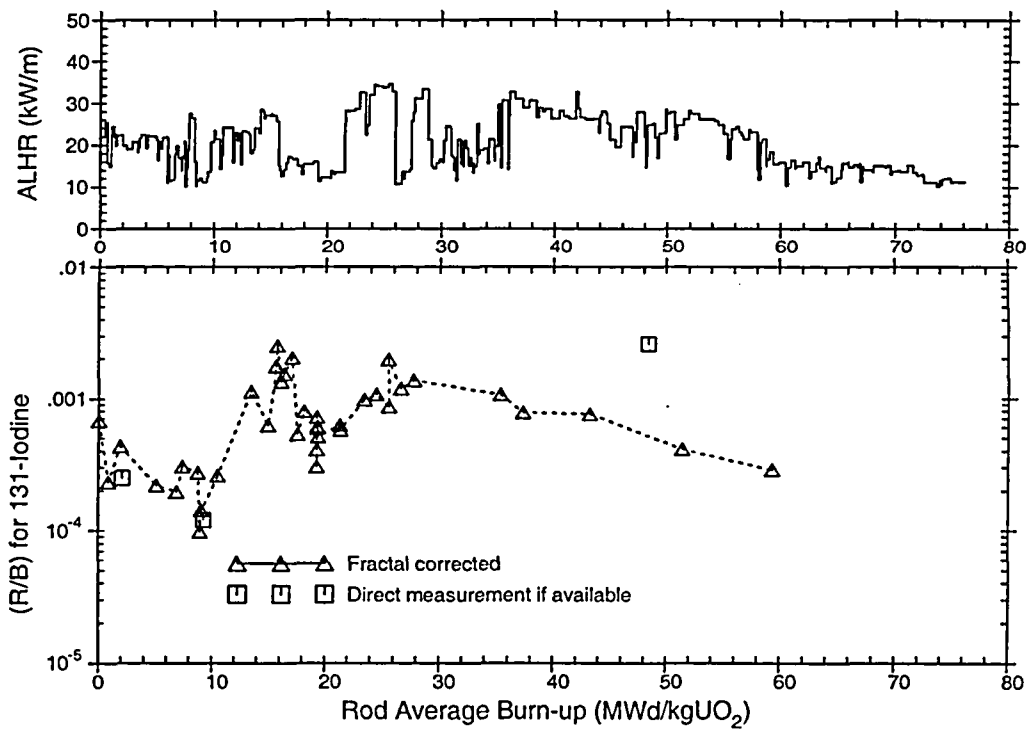


Figure 6(d) Calculated values of (R/B) for 131-I from measured 85m-Kr; IFA-504 rod 4