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ABSTRACT

The potential release of fission and activation products from spent nuclear fuel into groundwater after container failure in the Swedish deep repository is discussed. Data from studies of fission gas release from representative Swedish BWR fuel are used to estimate the average fission gas release for the spent fuel population. Information from a variety of leaching studies on LWR and CANDU fuel are then reviewed as a basis for estimating the fraction of the inventory of key radionuclides that could be released preferentially (the Instant Release Fraction or IRF) upon failure of the fuel cladding. The uncertainties associated with these estimates are discussed.

ABSTRACT (in Swedish)

Den här rapporten diskuterar de potentiella frigörelsen av fissions- och aktiveringsprodukter från använt kärnbränsle till grundvatten efter en kapselskada i ett svenskt djupförvar. Data från studier av fissionsgasfrigörelse från representativt svenskt BWR bränsle används för att uppskatta medel-fissionsgas frigörelsen för allt bränsle. Information från en mängd lakstudier av LWR och CANDU bränsle har granskas som en grund för uppskattning av fraktionen av viktiga radionuklider som skulle kunna frigöras preferentiellt ("Instant Release Fraction" eller IRF) vid genombrott på bränslekapslingen. Osäkerheterna kopplade till dessa uppskattningar diskuteras också.

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1 INTRODUCTION

Safety assessment of the disposal of spent nuclear fuel requires information on the quantities and rates of release of the various radionuclides upon potential contact with groundwater in the repository. Spent fuel is a heterogeneous material, thus radionuclides are released into groundwater according to several different mechanisms. The majority of the radionuclide inventory is uniformly distributed throughout the UO_2 matrix and is released at the rate at which the fuel matrix dissolves. A small percentage of the inventory of a few radionuclides is located at the fuel-clad gap and at grain boundaries in the fuel. Figure 1 illustrates conceptually the distribution of various important radionuclides in spent fuel, based on many studies of fuel rod chemistry and spent fuel leaching. The radionuclides in the gap and at grain boundaries may be present as salts (e.g. CsI), metal inclusions (e.g. Tc), oxide inclusions (e.g. Zr), or gas (Kr).

The fraction of the radionuclide inventory present in the fuel-clad gap has been shown to be released very rapidly upon contact with groundwater. This fraction has been shown in many studies to be comparable to the fission gas release (FGR) to the fuel-clad gap during reactor operation (Johnson and Shoesmith 1988). The radionuclides present at grain boundaries in the fuel dissolve more slowly, but still rapidly in comparison to those released during the much slower matrix dissolution. The gap and grain boundary release fractions are frequently represented as a combined source term in performance assessment calculations. This combined source term is referred to here as the Instant Release Fraction (IRF).

Although the total number of fuel rods studied to determine the quantities of radionuclides in the gap and at grain boundaries is small, it is nonetheless possible to estimate the average IRF for all the fuel in the repository because gap and grain boundary inventories can be correlated with FGR for individual fuel rods and because an average FGR for the entire spent fuel population can be reliably estimated. In the first section of this report, the average FGR of spent fuel in the Swedish repository is estimated based on published studies of BWR fuel, the major fuel type in the repository. The gap and grain boundary leaching data for various radionuclides and for several fuel types (BWR, PWR and CANDU) are then discussed, along with their relationships to FGR. For each radionuclide, a best estimate and pessimistic value of the average IRF for the spent fuel population is then derived. The release of radionuclides from cladding is not discussed.

2 FISSION GAS RELEASE IN BWR FUELS

There are a number of sources of data for FGR measurements from BWR fuels, including Schrire et al. (1997), Grapengiesser and Schrire (1993), Barner et al. (1990, 1993), Guenther et al. (1991), and Koizumi et al. (1987). The fuels were irradiated in commercial BWR reactors at burnups from about 25 MWd/kg to 50 MWd/kg and peak Linear Power Ratings (LPR) ranging from 18 to 50 kW/m.

Barner et al. (1990, 1993) measured fission gas releases from a number of fuel types (Table 1). Fission gas release from four KWU/CE BWR rods ranged from 0.1 to 0.4% (peak LPR ~30 kW/m); FGR from eight GE BWR fuels (peak LPR 18 to 20 kW/m) ranged from 0.1 to 5.2%; and FGR from nine TVO-1 BWR fuels (peak LPR 18 to 27 kW/m) ranged from 0.3 to 17.3% (average 6.6%). In the latter TVO rods, the FGR decreased with increasing distance from control blades, thus a lower core average LPR and FGR may result. Figure 2 compares the TVO-1 data with other ABB Atom fuel rod gas release data (Barner et al. 1993). The estimated average release of the TVO/ABB data is about 2.5 %.

Guenther et al. (1991) characterized a Cooper BWR fuel assembly for a standard fuel rod test material (ATM-105). The fission gas release from two elements were 1.4 and 11.2% at a peak bundle average LPR of 28 kW/m.

The data of Barner et al. (Figure 2) demonstrates that for burnups less than about 20 MWd/kg M, fission gas releases are less than 1% and that for the majority of fuels with higher burnups, fission gas releases are less than about 5%. This can be compared to BWR gas release data presented by Koizumi et al. (1987) who showed that for fuels with burnups less than 10 MWd/kg M, gas releases were less than 1%; releases ranged up to 20% for burnups ranging from 10 to ~33 MWd/kg M. Koizumi et al. (1987) demonstrated that there was a correlation between FGR and the maximum power at burnups greater than 10 MWd/kg M (Figure 3) and showed that FGR is <3% if LPR's remain below about 27 kW/m. A subsequent study with BWR fuel burnups from 30 to 39 MWh/kg M (Koizumi et al. 1991) demonstrated a similar behaviour to their previous data and they suggested that FGR depends strongly on maximum LPR and less on burnup.

Hallstadius and Grapengiesser (1991) conducted an analysis of FGR (Figure 4) from a number of 8x8 ABB Atom rods and concluded that FGR was higher from the control rod corner elements even though burnups were similar to off-corner elements. This was attributed to higher ^{239}Pu production in the corner elements resulting in an increased oxygen potential that increased the diffusion constant of Xe as well as higher element centerline temperatures. Releases from corner elements ranged from about 8% to more than 20%. The majority of analysed elements, however, had FGR less than about 5%, even at burnups as high as 50MWd/kg M. The estimated average FGR for their ABB data is about 3.5%

Schrire et al. (1997) measured FGR from over 100 10x10 SVEA fuel assemblies; the FGR increased with burnup reaching a maximum value of 5% at average rod burnups of 50 MWd/kg (Figure 5) and they observed that releases are far lower than from 8x8 fuels. Although the 10x10 fuel rods selected for this analysis preferentially favoured high power and control rod neighbourhood positions, the selection probably represents an average of rods in current reactors fairly well, given the recent trend to higher burnups (Grappengiesser, private communication). The estimated average fission gas release for their data is about 1.2%.

It appears from the above data on BWR fuels that even within a fuel assembly with similar element burnups, there will be a distribution of FGR that can range from ~0.1% to >20%. The data of Barner et al. (1993) (Figure 2) suggest that about 65% of the fuel elements examined have a FGR <1%, 80% are <5%, and about 90% are <10%. The data of Grappengiesser and Schrire (1993) also indicate that the majority of fuel elements examined have gas releases less than about 5%. More recent data (Schrire et al. 1997) on 10x10 SVEA fuels suggest that fission gas releases are <5%, even for high burnup fuels and fuels that would be representative of those from high-power locations, such as near control rods.

The modelled data of Forsberg et al. (1996) describes the distribution of FGR and burnups in a BWR at any particular time. Their calculated data indicate that 75% of the fuel in reactor has a FGR less than 5%, 95% of the fuel has a FGR less than 10% and 99.3% has a FGR less than 15%. The weighted average FGR from their data is about 4%, which is likely an over-estimate of the average FGR at discharge because their data represents an in-core instantaneous distribution of fuel assembly FGR rather than discharge values.

Based on these data, and in particular, the more recent data of Schrire et al. (1997) shown in Figure 5, a best estimate of the average FGR would be about 2%, given that most of the data referred to above are biased towards observation of high power, high burnup fuels. A pessimistic average FGR would be 4%.

3 IRF FOR KEY RADIONUCLIDES

3.1 THE IRF FOR ^{137}Cs AND ^{135}Cs

Cesium is present in UO_2 fuel as CsI and Cs uranates. Many studies have been published that illustrate the close relationship between Cs leaching and FGR for LWR and CANDU fuel (Forsyth and Werme 1992, Gray et al. 1992 and Stroes-Gascoyne 1996). The Cs leaching and FGR data for various LWR fuels, including Swedish BWR fuel, are shown in Table 2. For FGR in the range from 0 to 1%, gap releases of Cs are very similar to FGR. For the few cases of high gas release fuels that have been studied, gap releases are about ¼ of the FGR. For CANDU fuel, FGR and Cs releases are approximately equal in the range from 0.1 to 10% release, as shown in Figure 6 (Johnson and Shoesmith 1988). From the various studies, it is concluded that the average fractional release of Cs from the gap would be the same as the average FGR.

The data on release of Cs from grain boundaries are much more limited. The study of Gray et al. (1992) determined the grain boundary inventory of Cs for four fuels covering a large range of FGR. The data are summarized in Table 2, and illustrate that although gap releases of Cs increase as FGR increases, grain boundary inventories appear to stay constant in the range of 0.5 to 1%. A similar observation has been made for CANDU fuel (Stroes-Gascoyne 1996), although the grain boundary inventories are greater than for LWR fuel.

It is concluded that a best estimate of the IRF for Cs would be 3% (2% gap and 1% grain boundary), with a pessimistic value being 6%.

3.2 THE IRF FOR ^{129}I

Iodine is present in UO_2 fuels as CsI and its fractional release during leaching has been shown in CANDU fuel leaching studies to be similar to that of FGR during reactor irradiation. Figure 6 shows the correlation between fractional release (gap release) of Xe, Cs and I for CANDU fuel, which has been studied in detail (Johnson and Shoesmith 1988). The data for LWR fuel are very limited, as seen in Table 2. In most cases the measured ^{129}I gap release is much less than the fission gas release and in one case is slightly larger. It is concluded from the available data that the gap release of ^{129}I is likely to be less than or the same as the FGR.

There are no published data available for grain boundary inventories of ^{129}I for LWR fuel. The study of Stroes-Gascoyne (1996) determined that grain boundary inventories of ^{129}I in CANDU fuel did not increase with increasing FGR. This is similar to the

observation of Gray et al.(1992) for ^{137}Cs. It is concluded that best estimate and pessimistic values of the ^{129}I IRF would be the same as those for ^{137}Cs, i.e., 3 and 6%, respectively.

3.3 THE IRF FOR ^{36}Cl

Chlorine is a ubiquitous impurity that will occur in both fuel assembly materials and the UO_2 fuel. The natural isotope of ^{35}Cl (75% natural abundance) will be neutron activated to ^{36}Cl in-reactor via $^{35}Cl(n,\gamma)^{36}Cl$. Chlorine is not generally specified as an impurity in fuel assembly or UO_2 materials; however, ASTM standards quote a limit of 25 ppm for Cl in sintered LWR UO_2 pellets (ASTM). Analysis of PWR fuel indicates that levels are below 10 ppm (Guenther et al. 1991b) and characterization studies on BWR (Guenther et al. 1991a,b) and PWR (Guenther et al. 1988) fuels have assumed a 5 or 10 ppm Cl impurity in UO_2, respectively, for calculating the ^{36}Cl inventories produced during irradiation. It is recommended that, for the purposes of calculating ^{36}Cl inventories in irradiated fuels, an initial Cl impurity level in UO_2 of 10 ppm be used.

Measurements on Cl impurity levels in CANDU Zircaloy pressure tubes have indicated concentrations typically <5 ppm (Aitchison and Davies, 1993). Since the manufacture of Zircaloy pressure tubes is similar to that for Zircaloy cladding, it is assumed that similar Cl impurity concentrations would be present in cladding materials, thus Cl would be an impurity in BWR/PWR fuel and fuel assemblies. For the purposes of calculating ^{36}Cl inventories in the Zircaloy cladding, it is suggested that values similar to those used in a recent safety assessment for CANDU fuels (Johnson et al. 1996) be adopted, i.e., 5 ppm for Zircaloy cladding..

Recent measurements of Cl impurities in CANDU UO_2 fuels have indicated average concentrations of ~2.3 ppm in unirradiated and irradiated UO_2, with concentrations typically <5 ppm (Tait et al. 1997). Measurements of the "instant release" of ^{36}Cl to aqueous solution from CANDU UO_2 fuels have indicated that there is a strong positive correlation between ^{36}Cl release and the fuel burnup, the fission gas release (FGR) and the linear power rating (LPR); this is due to the increased production of ^{36}Cl with in-reactor residence time and the migration of ^{36}Cl to the gap regions in the fuel element with increased power and fuel temperature. There appears to be little evidence for ^{36}Cl at UO_2 fuel grain-boundaries (Tait et al. 1997). Instant release measurements indicated that from 0.5 to 20% of the total ^{36}Cl inventory was released to solution over time periods from 4 to 32 days, with higher values correlating with higher fuel burnups and FGR.

The only instant release data for ^{36}Cl from UO_2 fuels is that from CANDU fuels (Tait et al. 1997). Given the strong correlation between ^{36}Cl release and FGR, a best estimate for the ^{36}Cl IRF would be 3 times the FGR. This would slightly underestimate release at low FGR (<0.5%) but would be conservative for higher FGR's.

The FGR data from BWR fuels suggest an average FGR of 2% which would give a mean ^{36}Cl IRF of 6%. A pessimistic estimate of the IRF would be 12%, i.e., three times the pessimistic estimate of average FGR.

3.4 THE IRF FOR ^{99}Tc

Technetium is present in spent fuel in metallic form, typically in alloy inclusions (Kleykamp 1985, Johnson and Werme 1994). Although a significant fraction of the inventory of ^{99}Tc may be present in metallic inclusions, the fraction at grain boundaries is likely to be small, because Tc diffusion coefficients in UO_2 are lower than those of Xe, Cs and I (Prussin et al. 1988). The results of leaching studies summarized in Table 2 show that ^{99}Tc gap releases are less than ~0.1%, and that releases from grain boundaries are similarly small. This may well be partly because of the resistance of the alloy inclusions to dissolution in the groundwater. The highest releases observed are in a study involving leaching of CANDU fuel that was oxidized in air to U_3O_8 powder (Stroes-Gascoyne and Sellinger 1986). Releases were as much as 4.5%; however, high temperature air oxidation represents extreme conditions not relevant to repository conditions.

It is concluded that a best estimate of the IRF for Tc would be 0.2 %, with a pessimistic estimate being 1%.

3.5 THE IRF FOR ^{90}Sr

Strontium has been detected at grain boundaries in spent LWR fuel (Jeffery1967), although it is mainly dissolved in the fuel matrix (Kleykamp 1985). Gap and grain boundary leaching data for ^{90}Sr for BWR and PWR fuel are shown in Table 2. Gap releases are generally a small fraction of FGR , in the range of 0.01 to 0.1%. Grain-boundary inventories in LWR fuel have been measured only in the study of Gray et al. (1992). In a some cases, the grain boundary inventory exceeds the gap inventory. Gap and grain-boundary releases of ^{90}Sr from samples of CANDU fuel have been determined by Stroes-Gascoyne (1996). These results suggest much higher releases (1 to 4%); however, CANDU fuels have appreciably higher linear power ratings, thus these results are unlikely to be relevant to the case of LWR fuel. It is concluded from the limited data in Table 2 that a best estimate IRF for LWR fuel for Sr would be 0.25%, with a pessimistic estimate being 1%.

3.6 THE IRF FOR ^{14}C

The activation of ^{14}N and ^{17}O impurities in UO_2 fuel and Zircaloy cladding will give rise to ^{14}C through the reactions $^{14}N(n,p)^{14}C$ and $^{17}O(n,\alpha)^{14}C$ with the first reaction being by far the most important due to the high neutron capture cross-section. Both ^{14}N and ^{17}O are naturally abundant isotopes (~99.6% and ~0.04% respectively) that are inevitably present in UO_2 fuel and Zircaloy. Nitrogen is likely a contaminant introduced in the

fuel manufacturing process. A third minor production path is $^{13}C(n,\gamma)^{14}C$; however, this pathway is insignificant by comparison, due to the low capture cross section and low natural abundance of ^{13}C.

The release of ^{14}C from UO_2 fuel in a disposal vault will depend on both its physical distribution and its chemical form in the fuel. It has been suggested that ^{14}C resides in the fuel in the form of carbides, oxycarbides or elemental C (Van Konynenburg et al. 1987). The release of ^{14}C from LWR fuels has been studied for time periods of several hundred days at 25 and 85°C (Wilson and Shaw 1987; Wilson, 1990a, 1990b) and releases ranged from 0.035% to 3.5% of the total measured ^{14}C inventory. Leaching tests conducted at 200°C (Neal et al. 1988) indicated ^{14}C inventory releases varying from 2.4 to 7.3%, although this may include some release from matrix dissolution as well. Measurements on the instant release fraction of ^{14}C from CANDU fuels (Stroes-Gascoyne et al. 1994) indicated IRF's ranging from 0.06% to 5.04% with an average of 2.7% ± 1.6% based on measurements of the total ^{14}C inventory.

Leaching data from CANDU fuels show no correlation with fuel power or burnup (Stroes-Gascoyne et al. 1994). This lack of correlation is somewhat expected, since N is the predominant source for ^{14}C and N impurity levels in fuel are not accurately known and are likely variable. There was, however, a consistent pattern of higher ^{14}C release with fuel power for a given fuel bundle. Measurements of total ^{14}C in CANDU fuels suggested that initial N impurity levels were about 10 ppm.

Although the ^{14}C leaching data on LWR fuels are scarce, the available data suggest that ^{14}C IRF's are <10%. The IRF's for ^{14}C in CANDU fuels were independent of FGR and typically ranged from 1 to 5 %. Since the release of ^{14}C does not seem to be dependent upon fuel power or fission gas release, it is recommended that a best estimate IRF of 5% of total ^{14}C inventory be adopted. A pessimistic estimate of the IRF for ^{14}C would be 10%, given that tests under extreme conditions (Neal et al. 1988) and on higher power CANDU fuels (Stroes-Gascoyne et al. 1994) give values much less than this.

Van Konynenburg et al. (1987) have reported a rapid release of <1% of the total ^{14}C inventory from air oxidation and water corrosion studies on Zircaloy cladding. The above IRF would thus conservatively account for uncertainties in estimating the release from oxide films on the cladding.

3.7 IRF VALUES FOR OTHER RADIONUCLIDES

A number of other radionuclides (see Table 3) need to be considered in the safety assessment with respect to their potential to segregate from UO_2 during reactor irradiation and be released preferentially in groundwater. There is little published data describing the quantities of these radionuclides released on contact with water, thus estimates of their IRF values have been derived largely from a knowledge of their solid-state chemistry during reactor irradiation. The uncertainties associated with this are discussed in Appendix A. In some cases, such as lanthanide fission products and actinide activation products, there is also direct evidence from leaching studies that the IRF values are negligible.

The radionuclides of significance in the SKB safety assessment are summarized in Table 3, along with their estimated IRF values

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5 TABLES AND FIGURES

Table 1. Fission Gas Release Data from BWR Fuels

Fuel	Burnup (MWd/kg M)	Fission Gas Release (%)
KWU/CE[a]	28.3	0.3
KWU/CE	27.9	0.1
KWU/CE	34.1	0.4
KWU/CE	33.8	0.3
GE[a]	28.9	0.1
GE	31.4	5.2
GE	24.0	0.1
GE	32.2	5.2
GE	33.5	0.2
GE	30.3	2.6
GE	32.1	<0.2
TVO-1[a]	44.6	11.2
TVO-1	45.9	12.7
TVO-1	44.9	3.4
TVO-1	43.7	1.0
TVO-1	46.6	17.3
TVO-1	47.8	1.0
TVO-1	48.5	0.3
TVO-1	45.5	6.6
TVO-1	45.5	6.2
ADD2966[b]	33.9	7.8
ADD2974	31.0	0.6

a) Barner et al. 1993
b) Guenther et al. 1991

Table 2. Gap and Grain Boundary (GB) Leaching Data for BWR and PWR Fuels

Fuel I.D.	Burnup (MWd/kg M)	Fission Gas Release (%)	Cs Gap (%)	Cs GB (%)	Sr Gap (%)	Sr GB (%)	Tc Gap (%)	Tc GB (%)	I Gap (%)	C Gap (%)
BWR (Oskarsham)[a]	42	0.7	~1							
BWR (Ringhals)[a]	20-49	1.1	0.4-0.8		0.07		0.1 to 0.7			
PWR (Ringhals)[a]	43	1.05	~1							
ATM-103[b] (PWR)	30	0.25	0.2	0.48	0.01	0.11				
ATM-106[b] (BWR)	43	7.4	2(avg)	0.5	0.11	0.03	0.13			
ATM-106[b] (BWR)	46	11.0	2.5	1.0	0.02	0.13	0.01	0.01		
ATM-106[b] (BWR)	50	18.0	4.0	1.0	0.1	0.07	0.05	0.12		
PWR-HBR[c]	31	0.2	0.8		0.024		0.03		0.008	0.01
PWR-TP[c]	27	0.3	0.32		0.012		0.04		0.0023	
PWR-HBR[d]	31	0.2							0.284	0.33
PWR-TP[d]	27	0.3	0.4				<0.01		0.076	3.0
ATM-101[e] (PWR)	28	0.2	2						4	2-7

a) Forsyth and Werme (1992)
b) Gray et al. (1992)
c) Oversby and Shaw (1987); Wilson (1987); Data at 25°C
d) Wilson (1987); Wilson and Gray (1990); Data at 85°C
e) Neal et al. (1988); Data at 200°C for 9 months, results likely represent IRF plus some matrix dissolution

Table 3. IRF Values for Key Radionuclides

Nuclide	T ½(year)	IRF(%) best estimate	IRF(%) pessimistic estimate	Rationale
C-14	5730	5	10	see text
Cl-36	$3 \cdot 10^5$	6	12	see text
Co-60	5.3	-	-	activation product in Zircaloy cladding
Ni-59	$7.5 \cdot 10^4$	-	-	activation product in Zircaloy cladding
Ni-63	100	-	-	activation product in Zircaloy cladding
Se-79	$6.5\ 10^4$	3	6	Cs_2Se assumed to be comparable to CsI in volatility in fuel during irradiation (Cubicciotti and Sanecki 1978)
Kr-85	10.8	2	4	see text
Sr-90	28.5	0.25	1	see text
Zr-93	$1.5 \cdot 10^6$	-	-	mainly dissolved in fuel matrix
Nb-94	$2.0 \cdot 10^4$	-	-	mainly dissolved in fuel matrix
Tc-99	$2.1 \cdot 10^5$	0.2	1	see text
Pd-107	$6.5 \cdot 10^6$	0.2	1	alloyed with Tc in metal inclusions
Ag-108m	127	3	6	present as metal, somewhat volatile during irradiation (Cubicciotti and Sanecki 1978)
Cd-113m	14.6	3	6	present as metal, somewhat volatile during irradiation (Cubicciotti and Sanecki 1978)
Sn-126	$1.0 \cdot 10^5$	2	4	relatively nonvolatile during irradiation (Cubicciotti and Sanecki 1978)
I-129	$1.6 \cdot 10^7$	3	6	see text
Cs-135	$2.0 \cdot 10^6$	3	6	see text
Cs-137	30.2	3	6	see text
Sm-151	93	-	-	present in solid solution in fuel matrix
Eu-154	8.8	-	-	present in solid solution in fuel matrix
Ho-166m	1,200	-	-	present in solid solution in fuel matrix
actinides		-	-	present in solid solution in fuel matrix

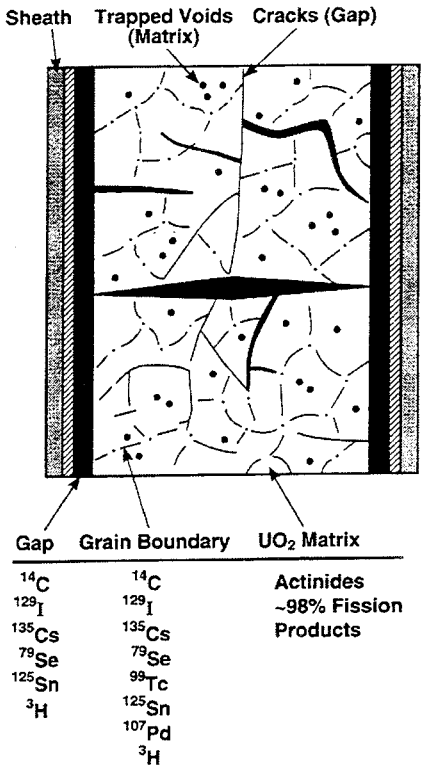

Figure 1. Conceptual Distribution of Some Fission and Activation Products Within a Spent Fuel Element.

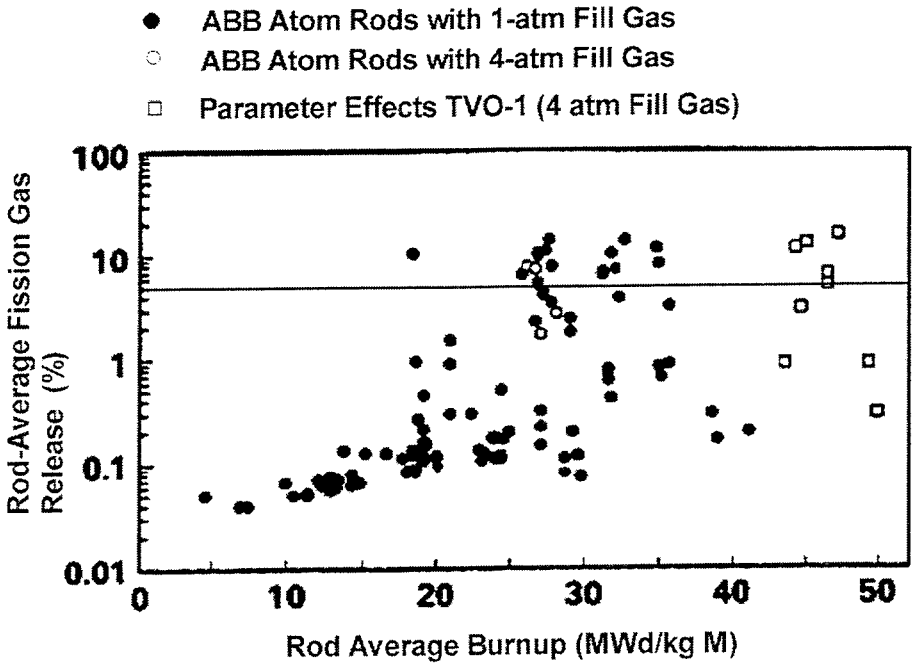

Figure 2. Comparison of "Parameter Effects" TVO-1 Fission Gas Release Data from Table 1 with other ABB Atom Fission Gas Release Data (Barner et al. 1993). The horizontal line is arbitrarily drawn at 5% gas release.

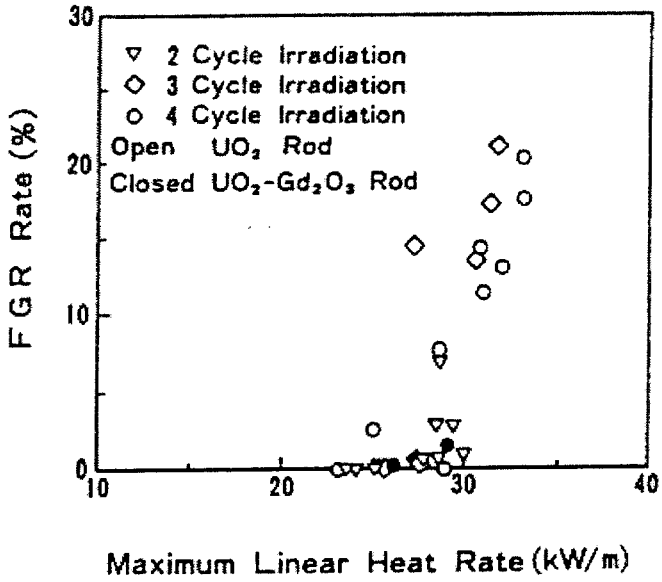

Figure 3. Power Dependence of Fission Gas Release Rate for BWR fuels with burnups >10 MWd/kg M (Koizumi et al. 1987).

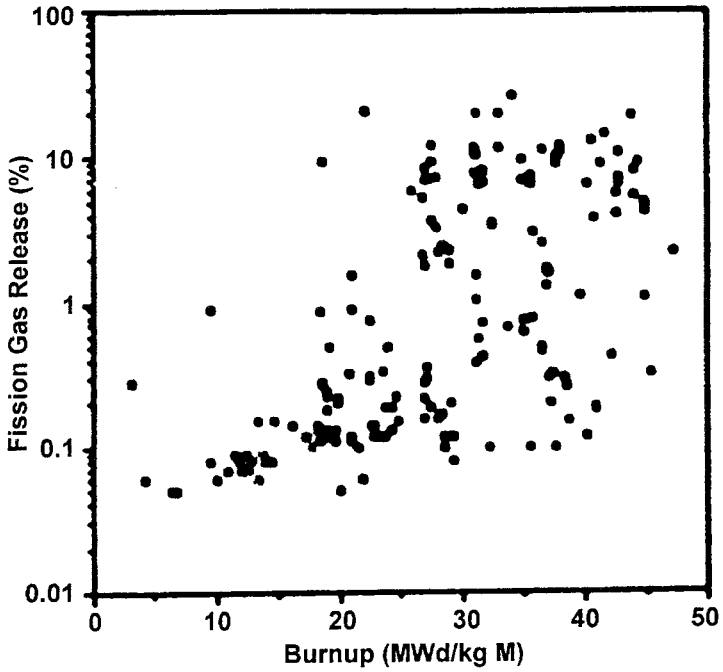

Figure 4. Fission Gas Release as a Function of Burnup for ABB fuels (Hallstadius and Grapengiesser 1990)

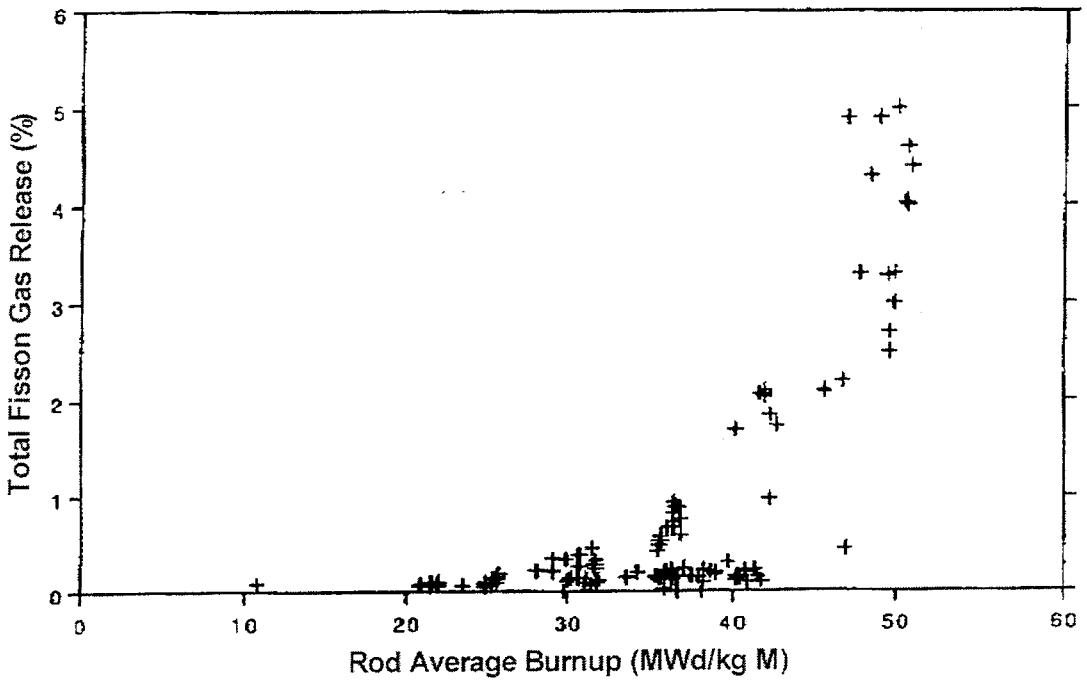

Figure 5. Fission Gas Release as a Function of Average Rod Burnup (Schrire et al. 1997).

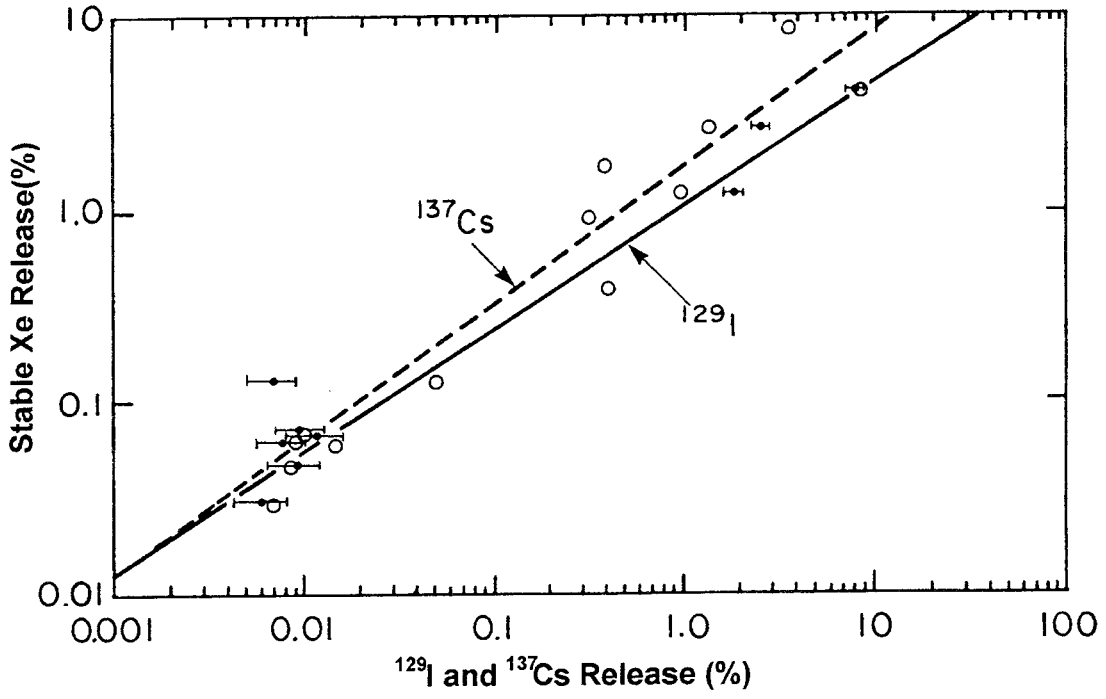

Figure 6. Relationship Between Xenon Release During In-reactor Irradiation and Short-term Leachability of ^{137}Cs and ^{129}I (Johnson and Shoesmith 1988).

6 DISCUSSION OF DATA UNCERTAINTIES

6.1 GENERAL

The data for IRF values of radionuclides summarized in the text and in Table 3 are based on two principal bodies of knowledge. The first involves measurements of fission gas release of LWR fuels and release of radionuclides in a variety of spent fuel leaching studies (^{137}Cs, ^{129}I, ^{14}C, ^{36}Cl, ^{99}Tc, ^{90}Sr, lanthanides and actinides). In this case studies have been done on BWR, PWR and CANDU fuel over a range of burnups. The second involves inferences from theoretical studies of solid state chemistry of fuel rods coupled with post-irradiation examination of fuel using scanning electron microprobe analysis. The uncertainties in the data are generally larger for IRF estimates based on the latter, as discussed below.

6.2 USE IN SR 97

The data in Table 3 describe the fractions of the inventory of the various radionuclides in the fuel that are released immediately upon contact of the fuel with groundwater.

Dependencies on Other Parameters, Ranges of Validity, Sensitivities

The IRF is influenced principally by in-reactor irradiation conditions. The values in Table 3 are estimated average values for the entire spent fuel population. Groundwater salinity has little effect on IRF values (Katayama 1979). Redox conditions have a major impact on the IRF of ^{99}Tc (Forsyth and Werme 1992), with anoxic conditions greatly reducing the release. The values for ^{99}Tc in Table 2 are conservatively based on results from experiments performed under oxidizing conditions. The IRF for ^{137}Cs and ^{99}Tc increases only slightly with temperature in the range of 25 to 100 ^{0}C (Wilson and Gray 1990). For ^{14}C and ^{129}I, the IRF increases significantly as the temperature is increased from 25 to 85 ^{0}C (see Table 2). These observations are taken into account in defining the values in Table 3.

6.3 CHARACTERIZATION OF UNCERTAINTIES

The derived IRF values are subject to several types of uncertainty, including:

- those related to the method of obtaining an average fission gas release over the entire spent fuel population,
- those related to radiochemical measurement ,
- those related to estimating the release of a radionuclide in the case of limited data, or in some cases, use of largely qualitative information from post-irradiation examination studies on spent fuel.

Regarding the uncertainties in estimating an average fission gas release for the spent fuel population, several studies on BWR fuel have been examined. The number of rods tested for fission gas release (see Figures 2 to 5) is significant and the average values obtained from these data are similar (1.2 to 3.5 %). It is considered extremely improbable that the average fission gas release could exceed 4%.

Uncertainties arising from radiochemical measurements are very small. The largest possible sources of error arise in determination of ^{14}C, ^{36}Cl and ^{129}I releases, in which loss of volatile chemical forms of these nuclides would lead to underestimates of release. The use of stable carriers during leaching and subsequent radiochemical analysis precludes the possibility that this occurred in the referenced studies.

In some cases, the limited data (e.g., the absence of Cl leaching data for other than CANDU fuels, and the sparse data for ^{14}C and ^{129}I leaching from LWR fuels) raise questions about the uncertainties in the estimated IRF values. The significant data base on release of these nuclides from CANDU fuels and the clear correlations with gas release, however, suggest that the estimated IRF values are conservative. For some radionuclides, there are no data on release during leaching (e.g., ^{79}Se, ^{108m}Ag, ^{113m}Cd, and ^{107}Pd). These elements are likely to be in metallic form in spent fuel and would therefore be expected to have very small IRF values. The uncertainties in the estimated releases are large, however, so the IRF values for ^{129}I and ^{137}Cs , which are generally considered to be more volatile during reactor irradiation, are adopted for these nuclides An indication that this is a conservative approach can be obtained by considering the case of Cd, the most volatile of this group of metals (Cubicciotti and Sanecki 1978). Quantitative X-ray photoelectron spectroscopy of CANDU fuel has been performed (Hocking et al. 1994) and the authors noted that Cd was only occasionally detected and would have been routinely seen if it had experienced the same fractional release as Cs. It thus would appear that while the uncertainties are significant for radionuclides for which no leaching data exist, other analytical data on fuels allows a conservative upper limit of release to be reliably established.

6.4 QUANTIFICATION OF UNCERTAINTIES

Data are sufficiently limited that statistical analysis of IRF values to establish distribution functions is not warranted.

6.5 CORRELATIONS

Not applicable because only best estimate values are proposed.

6.6 TREATMENT IN SAFETY ASSESSMENT

Best estimate and pessimistic values of IRFs are proposed for use in the safety assessment.

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[2] SGU, Lund

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