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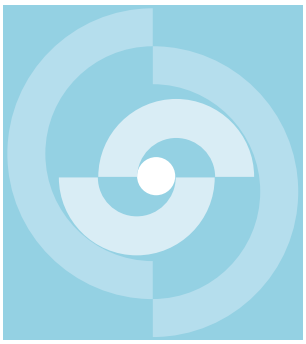

TECHNICAL REPORT 02-07

Partitioning of Radionuclides in Swiss Power Reactor Fuels

August 2002

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ABSTRACT

The potential preferential release of some fission and activation products from spent nuclear fuel into porewater after canister breaching in a deep repository in Switzerland is discussed. Data from studies of fission gas release from UO_2 and mixed oxide (MOX) fuels that are representative of fuel from Swiss nuclear power reactors are used to estimate the average fission gas release (FGR) for spent fuel. The evaluations are performed for average burnup UO_2 and MOX fuel (48 GWd/tIHM) as well as for higher burnup fuels (MOX 65 and UO_2 55, 65 and 75 GWd/tIHM). For the case of UO_2 fuel, the estimates include an analysis of the fraction of FGR present in the rim region, which is particularly important at higher burnup. Information from a variety of leaching studies on LWR fuel are then reviewed and compared to FGR 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 breaching of the fuel cladding. For higher burnup fuels, where leaching data are largely unavailable, IRF values are based on extrapolations of FGR behaviour observed at lower burnup and on an assessment of the impact of fuel restructuring at higher burnup on FGR. The IRF thus includes releases from the fuel/cladding gap and grain boundaries, as well as from the rim region. The expected release rates of radionuclides from Zircaloy cladding are also discussed, based on review of low temperature corrosion and radionuclide release data relevant to repository conditions.

ZUSAMMENFASSUNG

Im vorliegenden Bericht wird die mögliche bevorzugte Freisetzung von Spalt- und Aktivierungsprodukten aus abgebrannten Brennelementen in das Porenwasser nach Versagen der Abfallbehälter in einem geologischen Tiefenlager in der Schweiz diskutiert. Zu diesem Zweck wurden Daten aus Studien über die Freisetzung von gasförmigen Spaltprodukten aus den für Schweizer Kernkraftwerke typischen UO_2- und Mischoxid-(MOX)-Brennstoffen verwendet, um die durchschnittliche Freisetzung von gasförmigen Spaltprodukten (Fission Gas Release FGR) für abgebrannte Brennelemente abzuschätzen. Solche Abschätzungen werden sowohl für UO_2- und MOX-Brennstoffe mit durchschnittlichem Abbrand (48 GWd/tIHM; tIHM = tonne Initial Heavy Metal) als auch für Brennstoffe mit höherem Abbrand (MOX 65 sowie UO_2 55, 65 und 75 GWd/tIHM) durchgeführt. Für UO_2-Brennstoff enthalten die Schätzungen eine Analyse des im äusseren Randbereich vorhandenen FGR-Anteils, der für Brennstoffe mit höherem Abbrand besonders wichtig ist. Als Basis für die Abschätzung des Inventaranteils der wichtigsten Radionuklide, die beim Versagen der Brennstoffhüllrohre bevorzugt freigesetzt werden könnten (the Instant Release Fraction IRF), werden Informationen aus einer Vielzahl von Auslaugungsversuchen an LWR-(Leichtwasserreaktor)-Brennstoff überprüft und mit dem FGR-Anteil verglichen. Für Brennstoffe mit höherem Abbrand, für die weitgehend keine solche Daten aus Auslaugungsversuchen verfügbar sind, basieren die IRF-Werte auf der Extrapolation des bei niedrigerem Abbrand beobachteten FGR-Verhaltens und auf einer Beurteilung des Einflusses der Brennstoff-Restrukturierung auf den FGR-Anteil bei höherem Abbrand. Somit schliessen die IRF-Werte die Freisetzung aus dem Brennstoff/Hüllrohr-Zwischenraum, aus den Korngrenzen sowie aus dem äusseren Randbereich des Brennstoffs ein. Ausserdem werden die erwarteten Radionuklid-Freisetzungsraten aus den Zirkaloy-Hüllrohren basierend auf einer Studie der für ein Tiefenlager relevanten Niedrigtemperaturkorrosion und Radionuklidfreisetzungsdaten diskutiert.

RÉSUMÉ

Ce rapport aborde le relâchement potentiel préférentiel dans l'eau de certains produits de fission et d'activation, consécutif à une rupture des canisters contenant du combustible nucléaire irradié dans un site de stockage géologique profond en Suisse. Le taux moyen de libération de gaz de fission (LGF) pour les combustibles irradiés est estimé à partir de données correspondantes pour des combustibles UO_2 et MOX, représentatifs des assemblages utilisés dans les réacteurs nucléaires suisses. Les estimations concernent aussi bien des combustibles UO_2 et MOX à taux d'irradiation moyen (48 GWd/tIHM) que des combustibles à taux d'irradiation élevé (MOX 65 et UO_2 55, 65 et 75 GWd/tIHM). Dans le cas du combustible UO_2, les estimations comprennent une analyse du taux de LGF près du rebord, particulièrement important dans le cas d'une irradiation élevée. Des données émanant de plusieurs études de lixiviation pour le combustible des réacteurs à eau légère sont passées en revue et comparées au taux de LGF, de manière à estimer le pourcentage de l'inventaire des radionucléides principaux qui pourraient être libérés de manière préférentielle (Taux de Libération Immédiat ou TRI) lors de la rupture des gaines de combustible. Pour les combustibles à taux d'irradiation plus élevé, où il existe peu de données sur la lixiviation, les valeurs du TRI sont basées sur des extrapolations du comportement de la LGF observé à un taux d'irradiation plus bas et sur l'évaluation de l'impact sur la LGF de la restructuration du combustible à un taux d'irradiation plus élevé. Le TRI concerne donc des relâchements de radionucléides se produisant dans l'espace entre le combustible et la gaine, près des joints de grains, ainsi que près du rebord. Le taux de libération des radionucléides à partir des gaines en zircaloy est également calculé en se fondant à la fois sur l'étude de la corrosion à basse température et sur des données relatives au relâchement des radionucléides dans un site de stockage.

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1 INTRODUCTION

The most recent safety assessment studies for the disposal of high-level waste in Switzerland focused on evaluation of the disposal of vitrified waste (NAGRA 1994). The direct disposal of spent nuclear fuel is also now receiving attention and this option requires information on the quantities and rates of release of the various radionuclides from the fuel upon potential contact with groundwater in the repository. At burnups below 50GWd/tIHM and low linear power ratings, the majority of the radionuclide inventory is uniformly distributed throughout the UO_2 matrix, with a small percentage of the inventory of a few radionuclides located at the fuel/cladding gap and at grain boundaries in the fuel. 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/cladding 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 & TAIT 1997). The radionuclides present at grain boundaries in the fuel dissolve more slowly, but still rapidly in comparison to those released during the much slower dissolution of the UO_2 matrix. 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). The term IRF can lead to some confusion, because the release of fission products from the grain boundaries of spent fuel in leaching experiments can be very slow on a laboratory time scale. In spite of this, the concept is useful from a performance assessment perspective, because it acknowledges that segregation of some fission products from the matrix has occurred and the possibility that subsequent phenomena (slow penetration of water along fission gas tunnels or along microcracks developing at grain boundaries) may leach them out over periods of hundreds or thousands of years.

A further source of radionuclides is the Zircaloy cladding, where nuclides are produced by neutron activation of alloying constituents or trace elements in the Zircaloy. Slow uniform corrosion is expected to be the main mechanism controlling release of most radionuclides from cladding. A conceptual representation of the distribution of some important radionuclides in spent fuel rods, based on many studies of fuel rod chemistry and spent fuel leaching, is shown in Figure 1, based on JOHNSON & TAIT (1997).

Although the total number of fuel rods studied to determine the quantities of radionuclides in the gap and at grain boundaries are small, it is nonetheless possible to estimate the average IRF for all the fuel in a 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 for burnups up to about 50 GWd/tIHM. This approach has been used in Canadian (JOHNSON et al. 1996) and Swedish (JOHNSON & TAIT 1997) assessment studies for CANDU (Canada deuterium uranium) and BWR (boiling water reactor) fuels, respectively. The approach can be used for Swiss BWR and PWR (pressurised water reactor) UO_2 fuels as well, although the higher average burnup (~50 GWd/tIHM[1] compared to ~40 GWd/tIHM for Swedish spent fuel (SKB 1999)) requires that the trend of increased FGR at higher burnup be examined to see if the conclusions of JOHNSON AND TAIT (1997) also hold for such fuels. Furthermore, there is some possibility that considerably higher burnups (up to 75 GWd/tIHM) may be achieved for some fuel assemblies. The absence of data on leaching of fission products from higher burnup fuel (> 50 GWd/tIHM) makes it more difficult to evaluate IRF values for such fuel. Nonetheless, because of the trend towards higher burnups, the impli-

[1] tonnes initial heavy metal

cations in relation to the IRF for such fuel deserves some attention. A method for evaluating average IRF values for UO_2 fuel with higher burnups is proposed here based on review of information on FGR and restructuring of high burnup fuel.

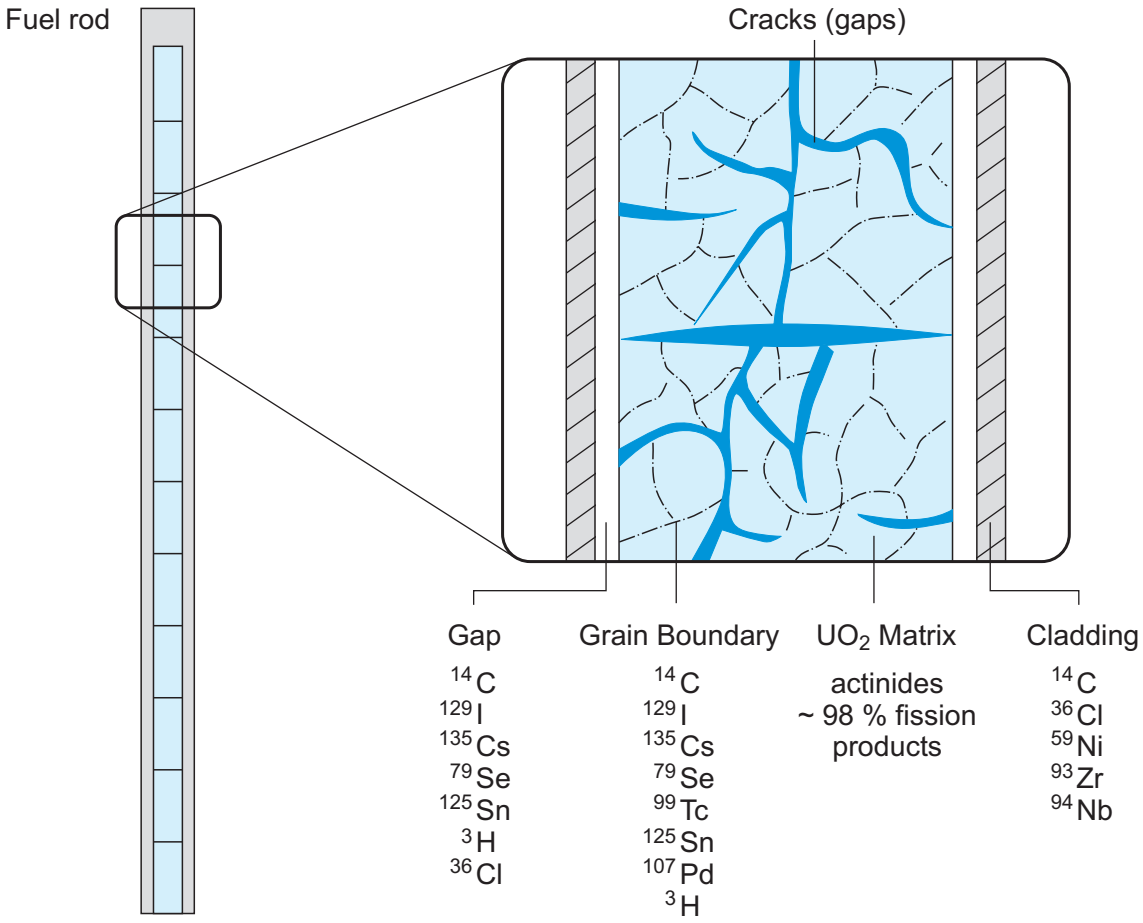

Figure 1: Conceptual distribution of some fission and activation products within a spent fuel element

An additional important question addressed here is the FGR and associated IRF for MOX fuels, which have not been previously examined. Although considerable FGR data is available for MOX fuel for burnups of < 40 GWd/tIHM, relatively little data is available for higher burnups. Furthermore, there is little published data on the leaching of fission products from MOX fuel.

The present study derives average IRF values for both UO_2 and MOX fuel, including estimates for higher burnup fuels. It is noted that what is required in performance assessment calculations is the average IRF for the various relevant radionuclides for the entire population of spent fuel. There is considerable variability in discharge burnups, as well as in FGR at any given burnup. Use of bounding or conservative FGR values will tend to lead to overestimates of derived IRF values, thus where possible, average data are used. Nonetheless, in deriving IRF values, the problem of uncertainties must be given some attention, in particular because there is considerable scatter in FGR data, as well as in data from fission product leaching studies. Furthermore, there are cases where almost no FGR or fission product leaching data are available, thus more

bounding estimates of IRF values must be derived. In order to clarify the discussion on the subject of uncertainties, the following approaches and definitions are adopted in this report:

Best estimate – One based on a good understanding of the mechanism and a good quality database (e.g. FGR in moderate burnup UO_2 fuel and rim restructuring in moderate to high burnup UO_2 fuel).

Bounding estimate – An estimate based on data and process understanding that provides a maximum for the range of derived values; this will result in significant overprediction of average IRF values. Here this applies particularly to cases where data is extremely limited or unavailable, thus understanding of the processes or chemical analogues must be used to derive estimates.

In this report, the approach taken is to develop best estimate IRF values for moderate burnup UO_2 fuel, because it is judged that the understanding and data are sufficient to support this, and to derive only bounding IRF values for the case of MOX fuel and higher burnup UO_2 fuel.

The report also discusses a model for radionuclide release from Zircaloy cladding for use in performance assessment calculations, which is based on review of a number of relevant low temperature corrosion studies.

2 SPENT FUEL TYPES AND AVERAGE BURNUPS

McGINNES (2002) has summarized the types and quantities of spent fuel that may eventually accumulate in Switzerland over 60 years of operation of the existing power plants. BWR UO_2 fuel is expected to comprise ~50 % of the total inventory, PWR UO_2 fuel about 45 % and PWR MOX about 5 %. The projected average burnup in all cases is 48 GWd/tIHM. The trend to higher burnups suggests that values in the range of 50 to 60 GWd/tIHM may become more common and that burnups up to 75 GWd/tIHM should also be considered (McGINNES 2002).

Detailed radionuclide inventories in reference spent UO_2 fuels and Zircaloy cladding are reported by McGINNES (2002).

3 FISSION GAS RELEASE FROM BWR AND PWR UO_2 FUEL

The release of fission gas from UO_2 fuel is strongly correlated with the linear heat rating, which is dependent on fuel temperature, as illustrated in Figure 2 (KAMIMURA 1992). Optimization of fuel assembly designs and irradiation conditions help to ensure that linear heat ratings are kept low and thus FGR is minimized. As a result, FGR values are typically <1 % at burnups below 40 GWd/tIHM (Figure 3, VESTERLUND & CORSETTI 1994). At higher burnups, a reduction of thermal conductivity increases the fuel temperatures, thus FGR tends to increase (SPINO 1998). A great many studies of FGR have been published, but selecting data that is representative of "typical" fuel that has experienced normal irradiation conditions is problematic. The majority of results are from power ramp test rods or from special fuel performance studies. In reviewing the literature on FGR, we have attempted to avoid data from the latter types of studies and have focused in large part on results from typical fuel. In spite of this, it is likely that the results presented still lead to a slight overestimate of the average FGR, because there is a tendency in some of these studies to focus on fuel rods with higher than average power ratings, e.g., corner rods on fuel assemblies.

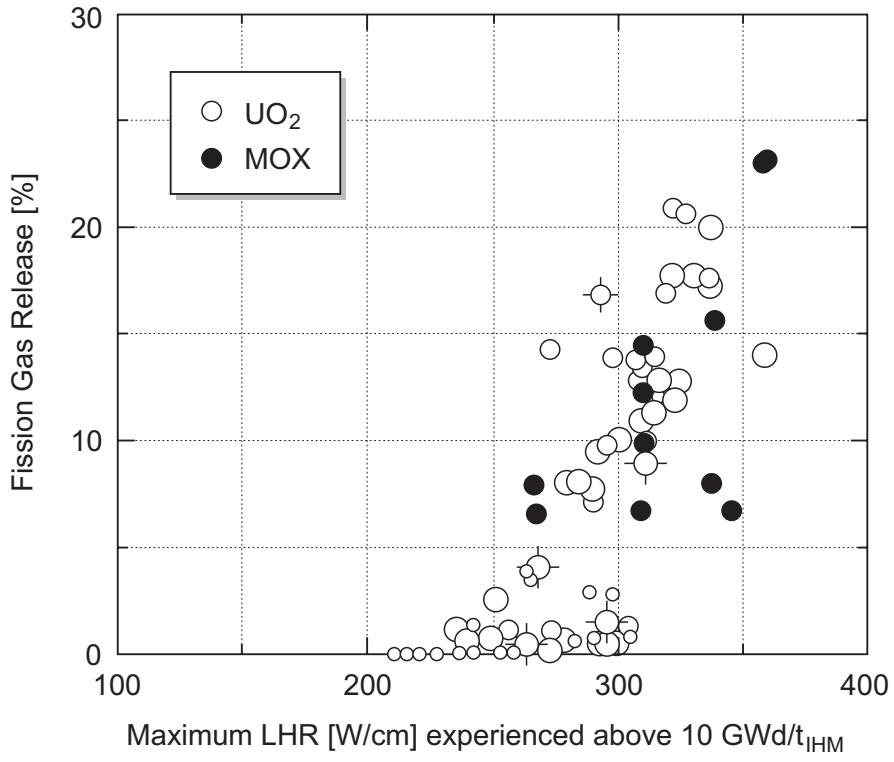

Figure 2: Fission gas release of BWR-MOX and UO_2 fuels as a function of the linear heat rating (in W/cm), from KAMIMURA (1992).

Considerable data are now available summarizing typical values of FGR for PWR UO_2 fuel irradiated to burnups of at least 60 GWd/tIHM and for BWR UO_2 fuel to 50 GWd/tIHM. The irradiation conditions of the fuel for which data are published are similar to those for Swiss power reactor fuel, and include fuel assembly designs used in Swiss power reactors, thus the data should provide a suitable basis for estimating the average FGR for performance assessment of the disposal of Swiss fuel.

Several data sets for BWR fuel that appear to be reasonably representative of normal irradiation conditions were discussed by JOHNSON & TAIT (1997) in their review of the FGR of Swedish BWR fuel. The data of HALLSTADIUS & GRAPENGIESSER (1990) are shown in Figure 4. There is a clear trend of increasing FGR with burnup, but considerable scatter arising from results from high power rods. An average of the data for the 40 to 50 GWd/tIHM range of particular interest in the present study gives a FGR value of ~6 %. The data of SCHRIRE et al. (1997), shown in Figure 5, appear to represent normal irradiation conditions and suggest an average FGR of ~ 3 to 5 % at 50 GWd/tIHM. This data more clearly illustrates the increase in FGR that occurs above about 40 GWd/tIHM. Overall, the results suggest that 5 % is a reasonable best estimate of the average FGR for BWR fuel at a burnup of 45-50 GWd/tIHM, with a pessimistic FGR value being ~10 %.

For the case of PWR fuel, several studies suggest that at high burnups FGR values are somewhat lower than for BWR fuel. The data of GUERIN et al. (1999) for French PWR fuel (Figure 6) illustrates that FGR values for UO_2 fuel are typically 1 to 2 % at 45 to 50 GWd/tIHM, increasing to only ~3 % at 60 GWd/tIHM. The data of VESTERLUND & CORSETTI (1994) indicate that average FGR values are ~1 % at 45 to 50 GWd/tIHM (Figure 3). Similar results are reported by YOKOTE et al. (1996) for Japanese PWR fuel. Based on all these data, it can be concluded that a best estimate of the average FGR for PWR fuel at the reference average burnup of 48 GWd/tIHM is 1 % and that a bounding estimate is 2 %.

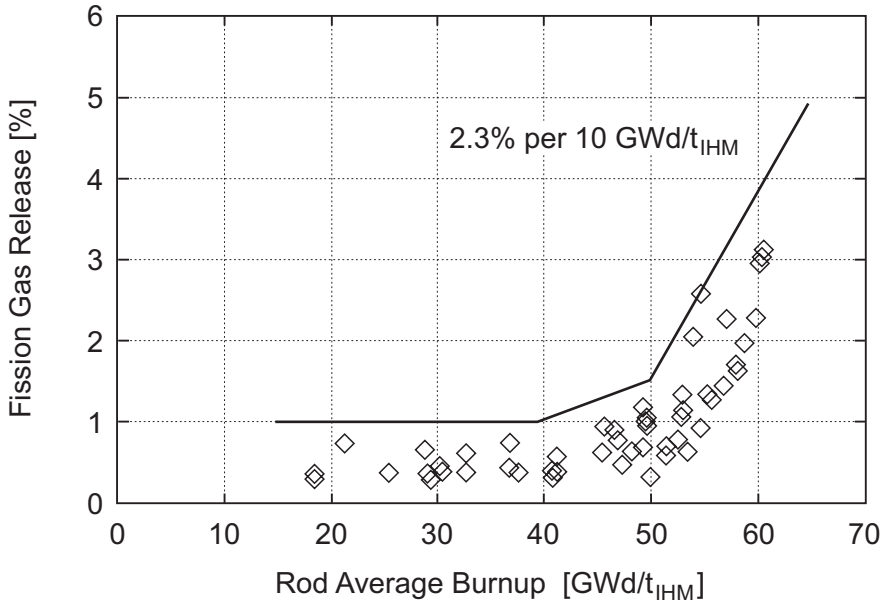

Figure 3: Fission gas release from PWR fuel as a function of burnup (VESTERLUND & CORSETTI 1994). The line represents the bounding values of all the data.

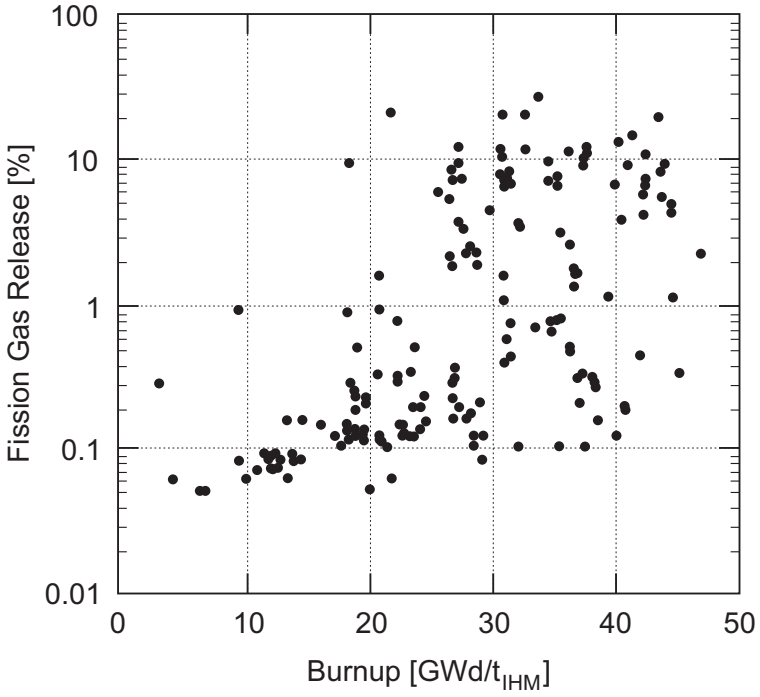

Figure 4: Fission gas release as a function of burnup for ABB BWR fuels (HALLSTADIUS & GRAPENGIESSER 1990)

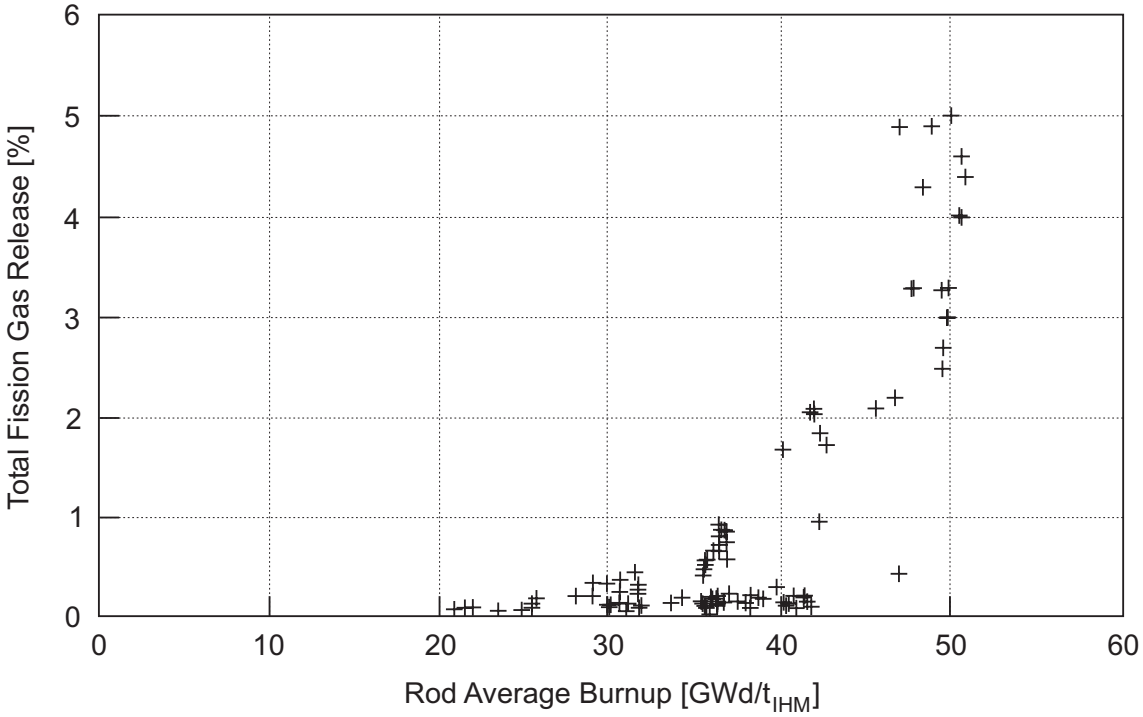

Figure 5: Fission gas release of BWR fuel as a function of average rod burnup (SCHRIRE et al. 1997)

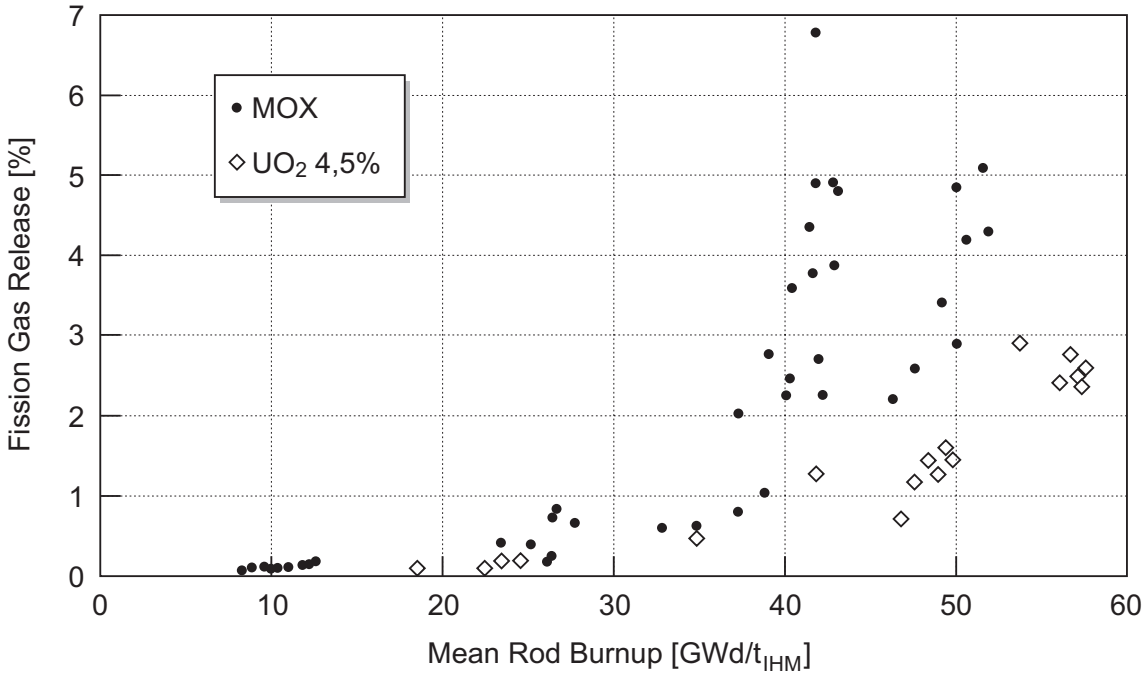

Figure 6: Fission gas release for French LWR UO_2 and MOX fuel as a function of burnup (GUERIN et al. 1999)

4 FISSION GAS RELEASE BEHAVIOUR OF HIGH BURNUP FUEL

Several phenomena occurring in the rim region of fuel pellets result in restructuring of fuel grains. These include:

- high fission density as a result of high yields of ^{239}Pu arising from capture of epithermal neutrons,
- increased porosity,
- reduction in grain size, and
- increased athermal release of fission gas from the grains.

From the perspective of assessing the release of fission products from spent fuel under disposal conditions, the restructuring process may be important, thus it is briefly reviewed here based on studies summarised by KOO et al. (2001). The discussion is based on the assumption of LWR fuel rods with a fuel pellet diameter of ~ 9 mm.

Fission of ^{239}Pu in the rim region, present at increased concentrations as a result of capture of epithermal neutrons by ^{238}U, leads to a sharp increase in local burnup. Although the outer few μm may reach as much as twice the average pellet burnup, the mean local burnup within the entire rim region is about 1.3 times the average pellet burnup. As illustrated in Figure 7, the best estimate of the rim thickness at an average burnup of 50 GWd/tIHM is 50 μm, increasing to 120 μm and 170 μm at 65 and 75 GWd/tIHM using a best fit (Equation 1 in KOO et al. (2001)) of all the data. A 'conservative' function (bounding, according to the definition given earlier), based on Equation 2 of KOO et al. (2001), that encompasses almost all the data, yields significantly larger rim widths. These functions are combined with an expression for the Xe distribution in the fuel to calculate the fraction of the total Xe produced in the pellet that is retained in the rim pores assuming no release to the gap during restructuring. This is shown in Figure 8, which illustrates that for a pellet average burnup of 50 GWd/tIHM (rim burnup of 62 GWd/tIHM), the best estimate fraction of Xe trapped in rim pores is ~1 to 2 %. This increases to 8 % at an average burnup of 75 GWd/tIHM (rim burnup of 98 GWd/tIHM). As indicated in Figure 7, the scatter in the data used to derive these estimates is large, thus the 'conservative' curve proposed by KOO et al. (2001) gives significantly larger rim widths at higher burnups.

Within the rim region, the structure is characterised by an average pore size of around 1 μm and an average grain size of 0.5 μm (POINSSOT et al. 2001). In spite of the high degree of restructuring, fission gas is typically effectively retained in the new pore structure (MOGENSEN et al. 1999), which explains the low overall fission gas release for PWR fuels at high burnups (Figure 6). Nonetheless, the fission gas in this region can be considered released from the fuel matrix, even though it is not released to the void space in the fuel rod. Similarly, other fission products that are not in solid solution in UO_2 can be expected to be released from the grains during restructuring. As a result, from the perspective of release under disposal conditions, such fission products can be considered to belong to the grain boundary inventory of the fuel and be potentially available for rapid release even if matrix dissolution does not occur.

The fraction of the FG produced that is present in rim pores for various burnups of UO_2 fuel, based on Figure 8 is summarised in Table 1. The total fractional fission gas release from the grains of the fuel to both the free void and closed pores at a given burnup can be considered to be the sum of the measured fission gas releases given earlier and the values given in Table 1. The question of uncertainties and the impact of using the bounding data in Table 1 is discussed in Section 6.4.

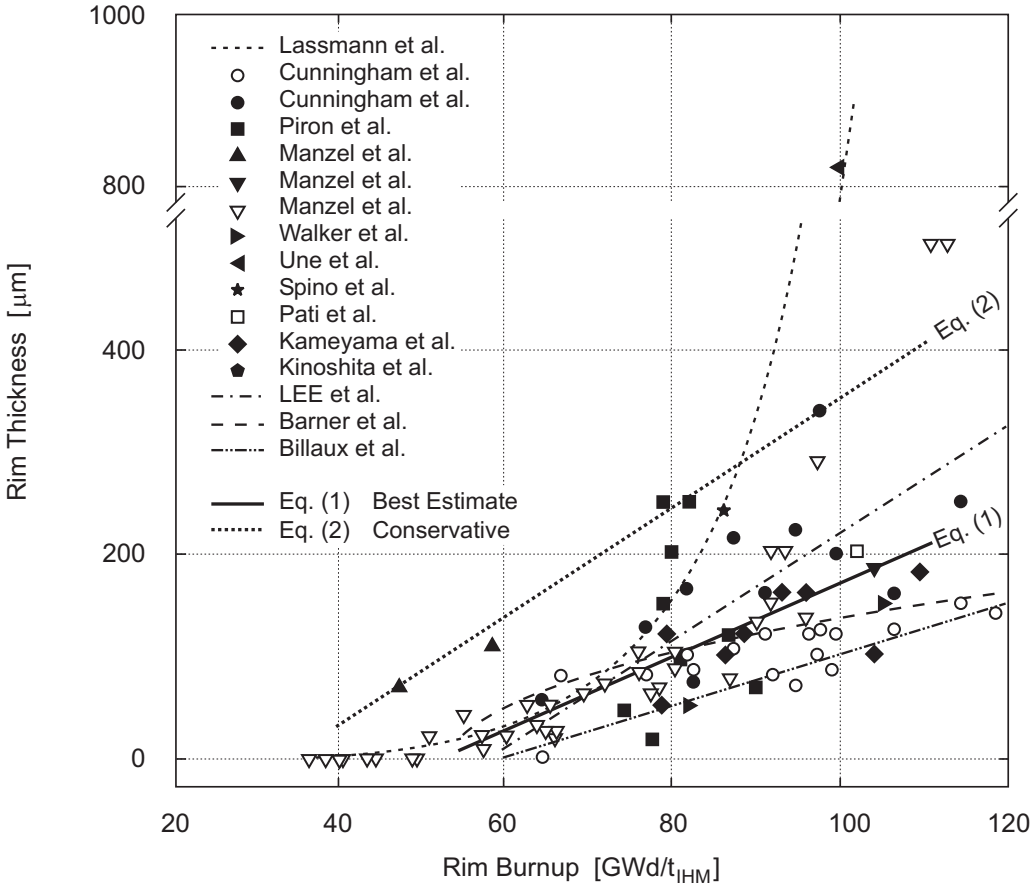

Figure 7: Rim width in μm as a function of burnup (GWd/tU), from KOO et al. (2001). Equation 1 represent a best fit of the data, while Equation 2 is a conservative or bounding expression that encompasses almost all the data. The references and equations referred to are in the paper by KOO et al. (2001).

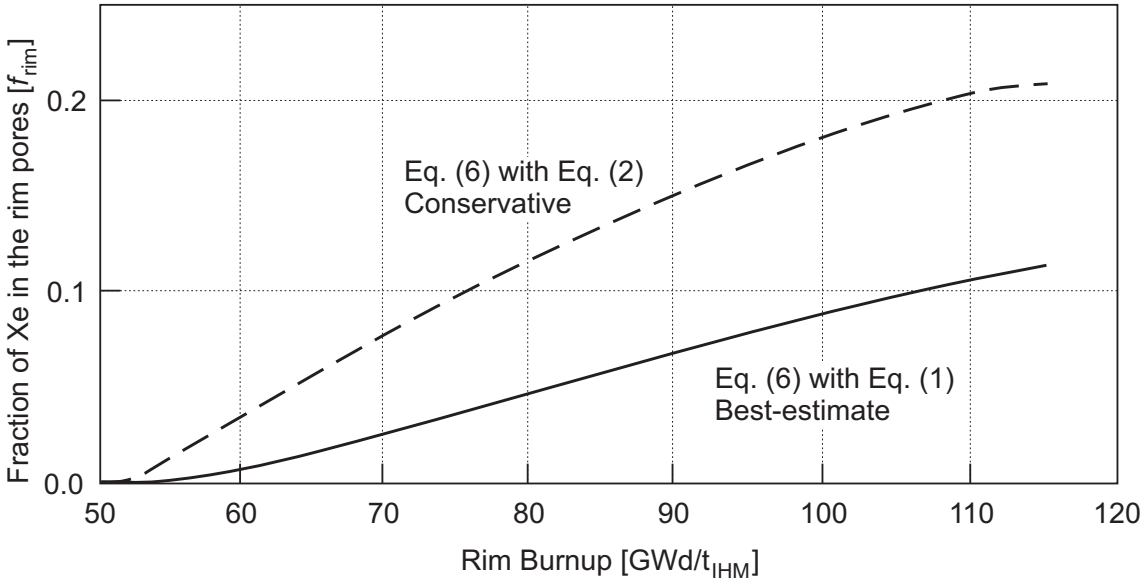

Figure 8: Fraction of total Xe inventory trapped in rim pores as a function of rim burnup, from KOO et al. (2001). Equations 1, 2 and 6 are given in the original paper.

Table 1: Fraction of the total fission gas inventory in a fuel rod that is present in the pores in the rim region of UO_2 fuel with burnups of 48, 55, 65 and 75 GWd/tIHM, based on the best estimate and 'conservative' curves in Figure 8 (KOO et al. 2001).

Average burnup (GWd/tIHM)	Rim burnup (GWd/tIHM)	Fraction of total FG produced present in pores in the rim (best estimate)	Fraction of total FG produced present in pores in the rim (bounding)
48	65	2	4
55	72	3	8
65	84	6	13
75	98	8	17

5 FISSION GAS RELEASE FROM MOX FUELS

The irradiation of MOX fuel on a routine basis in commercial power reactors began in the late 1980s. As a result, compared to FGR data for UO_2 fuels, data for MOX fuel is rather limited. The most comprehensive FGR data set for MOX fuel appears to be that of GUERIN et al. (1999) (Figure 6), which shows that FGR for MOX PWR fuel rods increases rapidly above 40 GWd/tIHM. The limited data above 45 GWd/tIHM suggests that the FGR reaches about 5 % at 50 GWd/tIHM. Increased FGR relative to UO_2 fuel at higher burnups arises from higher reactivity and higher power/temperatures compared with UO_2 fuel. As seen in Figure 2, MOX and UO_2 fuel perform similarly at the same linear heat rating.

This trend of higher FGR for MOX fuel relative to UO_2 fuel at higher burnups is less apparent for BWR MOX fuel, as shown in Figure 9, taken from the study of HAAS & LIPPENS (1997). At a burnup of 50 GWd/tIHM, the data suggest an average FGR of ~25 %.

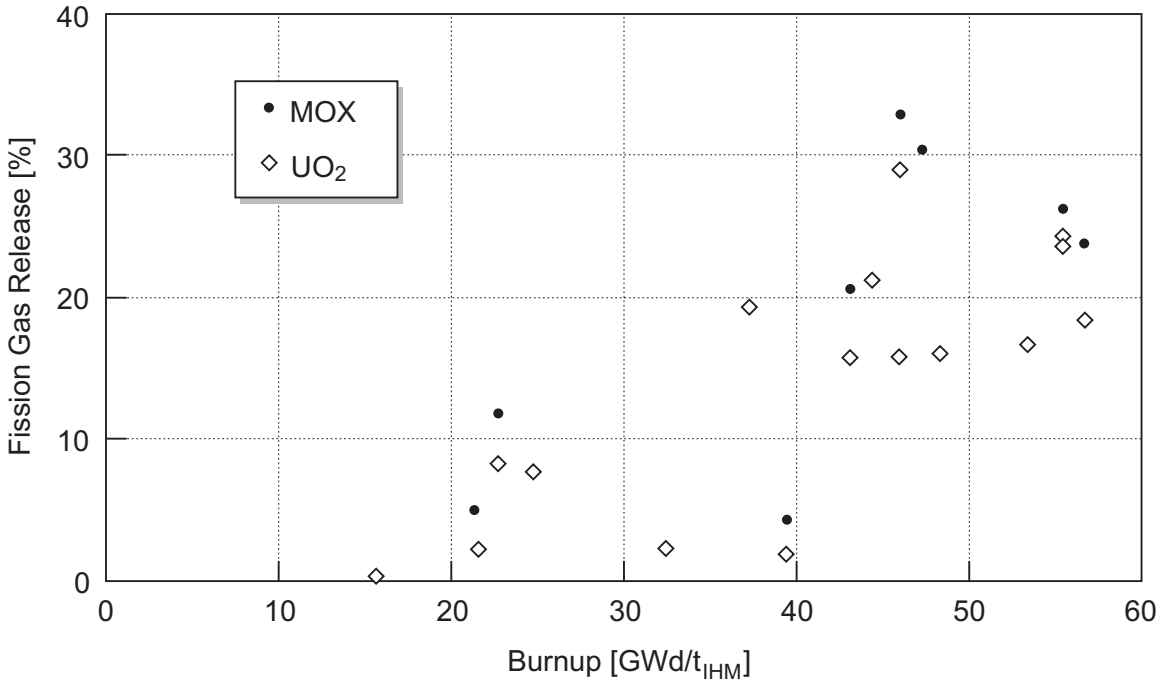

Figure 9: Fission gas release from BWR MOX and UO_2 fuel (HAAS & LIPPENS 1997)

GATES et al. (1998) present FGR data for a MOX database that includes fuel irradiated to burnups of ~50 GWd/tIHM (Figure 10). At 45 to 50 GWd/tIHM, the best estimate average FGR is ~5 %, with a bounding value of 10 %. There is considerable scatter in the data and the increase in FGR with burnup apparent in Figure 6 is not evident. There is vitally no published data on fission gas release from MOX power reactor fuel at burnups above 50 GWd/tIHM. The rim effect is not so pronounced with MOX fuel, because the Pu concentration throughout the fuel is already high as a result of the fabrication method, which involves mixing of PuO_2 and UO_2 powders (POINSSOT et al. 2001). Nonetheless, the development of high fission gas porosity in the PuO_2 grains is expected to increase the releases to the grain boundaries significantly.

Based on the limited data, the best estimate average FGR for PWR MOX fuel with a burnup of 45 to 50 GWd/tIHM appears to be ~ 5 %. In the absence of data for PWR MOX fuel at burnups exceeding 55 GWd/tIHM, and noting the increasing FGR trend in Figure 6, a bounding value of 20 % for a burnup of 65 GWd/tIHM is proposed for use in performance assessment calculations.

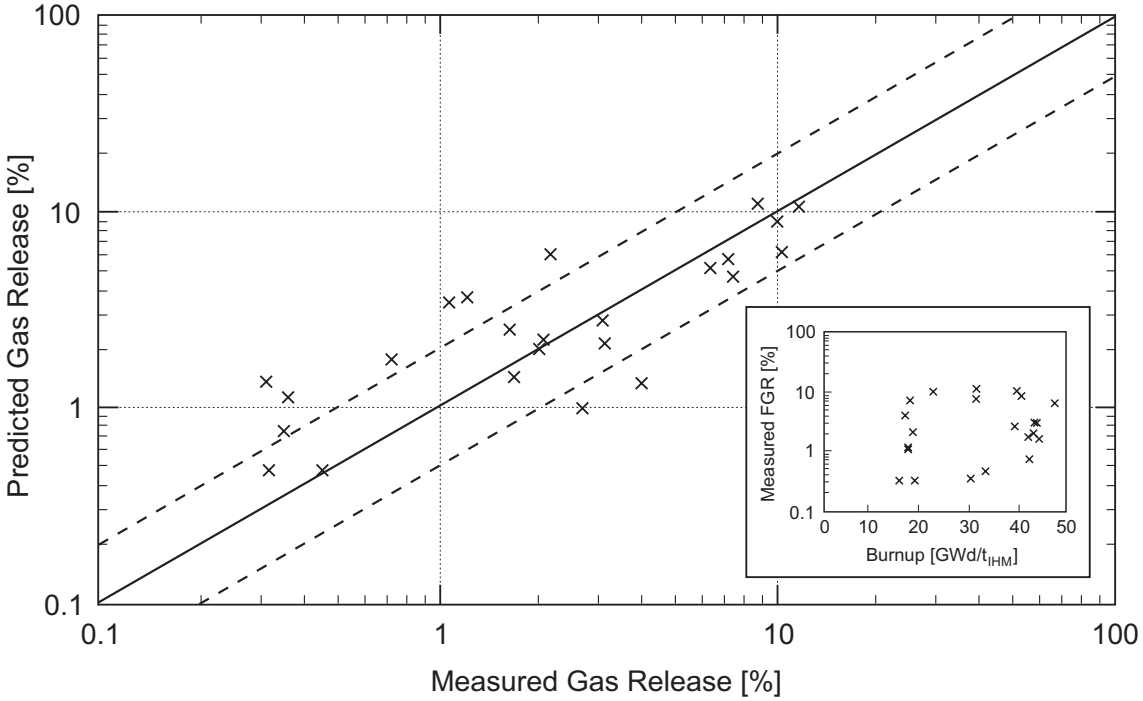

Figure 10: Predicted vs. measured fission gas release for MOX fuel (from GATES et al. 1998)

6 LEACHING OF RADIONUCLIDES FROM SPENT FUEL AND ESTIMATED IRF VALUES

The relationship between FGR and leaching of a number of fission products from UO_2 fuel has been reviewed recently by JOHNSON & TAIT (1997), to which the reader is referred for background on the behaviour in spent fuel of various fission and activation products of interest in performance assessment studies. In the present study we present only a brief summary of the observations derived from various leaching studies. Results of fission product leaching and FGR studies for various PWR and BWR fuels are shown in Table 2, taken from JOHNSON & TAIT (1997), with additions incorporated from the study of GRAY (1999). It is apparent that the data in Table 2 on FGR from ATM-106 PWR fuel for a burnup of 50 GWd/tIHM lies far above the range of values in Figures 3 and 6 or seen in other studies (e.g., YOKOTE et al. 1996). The reasons for this are not known (GUENTHER et al. 1988), but it is emphasized that review of a large body of FGR data suggests that the result is highly atypical. The data are nonetheless retained in the present study, because of the importance of the associated results on fission product leaching.

It should be noted that the data set for leaching of ^{137}Cs, ^{129}I, ^{14}C and ^{36}Cl from CANDU fuel is considerable and is taken into account in estimating IRF values as well, in particular for ^{14}C and ^{36}Cl. A discussion of the CANDU fuel data, which provides much of the basis for the ^{14}C IRF estimates in the present study and provides the only ^{36}Cl leaching data available for spent fuel, is presented in JOHNSON & TAIT (1997). The following discussion summarizes the observations from various leaching studies for each of the relevant radionuclides.

6.1 Low to Moderate Burnup (< 50 GWd/tIHM) UO_2 Fuel

IRF for ^{137}Cs and ^{135}Cs

GRAY (1999) measured both gap and grain boundary inventory values of ^{137}Cs for several PWR and BWR fuels and these data are included in Table 2. A plot of the sum of gap and grain boundary inventories yields a correlation for ^{137}Cs:FGR of 1:3. For low FGR of about 1 %, ^{137}Cs releases are similar to FGR. Grain boundary inventories of ^{137}Cs are relatively small, even for high FGR fuels. Such a result is consistent with the finding that the diffusion coefficient of Cs in UO_2 is lower than that of fission gases (POINSSOT et al. 2001). Studies of high linear power CANDU fuel by STROES-GASCOYNE et al. (1993) also indicate that grain boundary inventories are smaller than gap inventories. For BWR fuel with an average FGR of 5 %, the limited data suggest the released fraction is 3 %. The results of FORSYTH (1997), showed that ^{137}Cs release from clad fuel rod sections increased with burnup from 0.5 % to 1% in the interval from 25 to 43 GWd/tIHM, decreasing slightly in the interval of 43 to 49 GWd/tIHM. This may be associated with the closure of the fuel/clad gap that is observed as burnup increases (POINSSOT et al. 2001), thus the finding does not necessarily contradict the general observation of increased fission gas and IRF with increased burnup. For the best estimate average FGR for PWR fuel at 48 GWd/tIHM of 1 %, the released fraction is assumed to be 2 %. The IRF values for Cs for BWR and PWR fuel are given in Table 3, and represent the sum of the values given above and the fractional release in the rim region, as discussed in Section 4.

Table 2: Gap and grain boundary (GB) leaching data for BWR and PWR fuels.

Fuel I.D.	Burnup (MWd/kgHM)	Fission Gas Release (%)	Cs Gap (%)	Cs GB (%)	Sr Gap (%)	Sr GB (%)	Tc Gap (%)	Tc GB (%)	I Gap (%)	I GB (%)	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-104 [b] (PWR)	44	1.1	1.2	0.1							
ATM-105 [b] (BWR)	31	0.59	0.3	0.1					0.1	2.2	
ATM-105 [b] (BWR)	34	7.9	1.5	1.0					2.5	5	
ATM-106 [b] (PWR)	43	7.4	2	0.5	0.11	0.03	0.13		0.1	8.5	
ATM-106 [b] (PWR)	46	11.0	2.5	1.0	0.02	0.13	0.01	0.01	1.2	8.0	
ATM-106 [b] (PWR)	50	18.0	6.5	1.0	0.1	0.07	0.05	0.12	15	7.6	
PWR-HBR [c]	31	0.2	0.8		0.024		0.03		0.008		0.001
PWR-TP [c]	27	0.3	0.32		0.012		0.04		0.002		
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
MOX [f]	12-25	not reported	10 to 12						1 to 2		

[a] FORSYTH & WERME (1992), FORSYTH (1997)

[b] GRAY (1999); data are estimated from graph as raw data are not presented. Data represent average values of repeat measurements.

[c] OVERSBY & SHAW (1987); WILSON (1988); Data at 25 ^{o}C.

[d] WILSON & SHAW (1987); WILSON & GRAY (1990); Data at 85 ^{o}C.

[e] NEAL et al. (1988); Crushed fuel, includes grain boundary inventory; Data at 200^{o}C for 9 months, results likely represent IRF plus some matrix dissolution.

[f] GRAMBOW et al. (2000)

IRF for ^{129}I

GRAY (1999) suggested that there is a ~ 1:1 correlation between the sum of gap and grain boundary releases and FGR. This result appears consistent with the observation that the diffusion coefficients of I and fission gases are similar (POINSSOT et al. 2001). However, the majority of the fraction of ^{129}I released from the matrix resides in grain boundaries, not in the gap (Table 2), in contrast to the behaviour of ^{137}Cs. A linear regression of all the ^{129}I data (gap plus grain boundary vs. FGR) in Table 2, including the data of NEAL et al. (1988), which is for crushed fuel, yields a slope of 1 and a best estimate release value of 6.5 % at 5 % FGR, as shown in Figure 11. When added to the rim fraction, this gives a best estimate IRF value of 9 % for BWR fuel. For the same FGR, a bounding release of 9 % is estimated based on a plot that bounds all the combined gap and grain boundary data (Figure 11), giving a bounding IRF of 13 %. For PWR fuel with an average FGR of 1 %, the same approach gives a best estimate IRF of 2 % and a bounding IRF of 5 %.

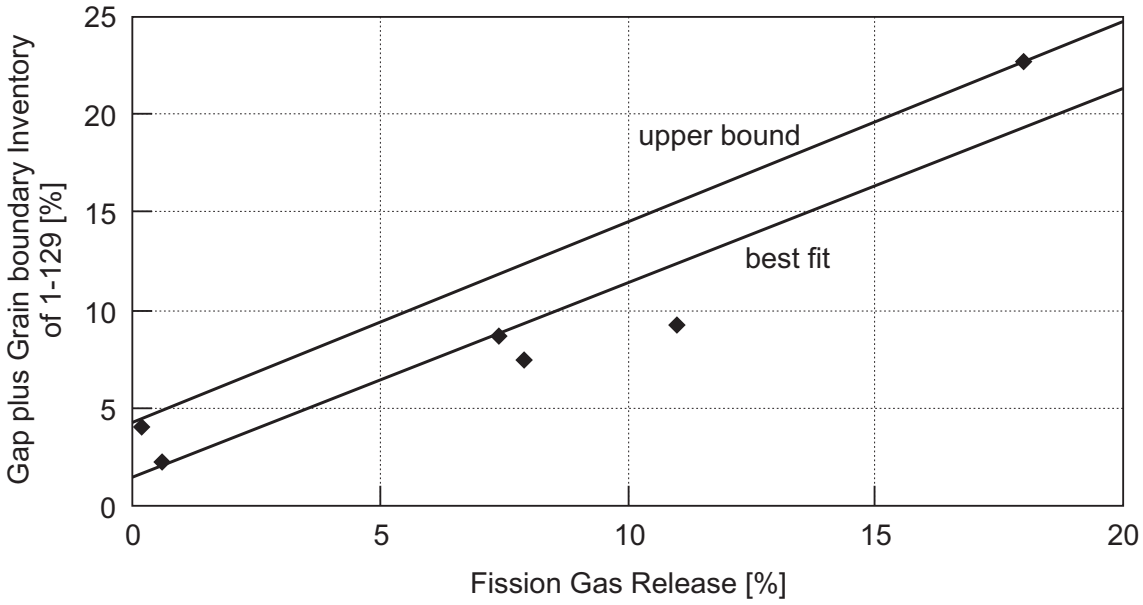

Figure 11: Gap plus grain boundary release of I-129 vs. fission gas release (data from Table 2)

IRF for ^{36}Cl

As noted by JOHNSON & TAIT (1997), there is no data on leaching of ^{36}Cl from LWR fuels. The estimated IRF is thus based on the CANDU fuel data of TAIT et al. (1997), which shows that ^{36}Cl releases increase sharply with FGR, reaching values three times the FGR for high linear power rating fuels. It should be noted that the linear power rating of CANDU fuel is much higher than that of LWR fuel, thus use of this data for LWR fuel is likely to overestimate releases. Only bounding IRF values for ^{36}Cl are estimated, acknowledging the absence of data for LWR fuel.

IRF for ^{14}C

No new data for ^{14}C has been reported since the study by JOHNSON & TAIT (1997), thus their bounding value of 10 % for all UO_2 fuels is proposed here. It should be noted that the sparse data for LWR fuel in Table 2 are in good agreement with the data for CANDU fuel, for which

STROES-GASCOYNE et al. (1994) report an IRF of 2.7 % based on a large number of measurements. The latter study noted that the IRF of ^{14}C is independent of fuel power rating, thus the CANDU fuel data appear to be directly applicable to the case of the lower rating LWR fuel. The chemical form of released ^{14}C is unknown, because studies of release of ^{14}C from spent fuel normally involve oxidative treatment of the solution to capture $^{14}CO_2$ for chemical analysis (STROES-GASCOYNE et al. 1994).

IRF Values for Other Radionuclides

A number of other radionuclides 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 into groundwater. The approach to estimating IRF values for other radionuclides is discussed in JOHNSON & TAIT (1997), with the exception of ^{3}H, where the value is based on the discussion in JOHNSON et al. (1996). The estimated IRF values for radionuclides with significant IRFs are listed in Table 3 for BWR and PWR UO_2 fuels with a burnup of 48 GWd/tIHM. All other radionuclides are assumed to be homogeneously distributed in the fuel matrix.

Table 3: Recommended best estimate and bounding IRF values of key radionuclides for BWR and PWR UO_2 fuel for a burnup of 48 GWd/tIHM.

Nuclide	t ½ (year)	Best estimate IRF Value (%)		Bounding IRF Value (%)	
		BWR UO_2 Fuel (48 GWd/tIHM)	PWR UO_2 Fuel (48 GWd/tIHM)	BWR UO_2 Fuel (48 GWd/tIHM)	PWR UO_2 Fuel (48 GWd/tIHM)
Fission gas	-	5 (gap) 2 (rim)	1 (gap) 2 (rim)	10 (gap) 4 (rim)	2 (gap) 4 (rim)
^{3}H	1.23×10^{1}	1	1	1	1
$^{14}C^{*}$	5.73×10^{3}	-	-	10	10
^{36}Cl	3.0×10^{5}	-	-	13	10
^{79}Se	1.1×10^{6}	9	4	13	9
^{90}Sr	2.86×10^{1}	1	1	1	1
^{99}Tc	2.1×10^{5}	2	2	4	4
^{107}Pd	6.5×10^{6}	2	2	4	4
^{126}Sn	2.3×10^{5}	9	4	13	6
^{129}I	1.57×10^{7}	9	4	13	9
^{135}Cs	2.3×10^{6}	5	4	7	5
^{137}Cs	3.02×10^{1}	5	4	7	5

* Value given is for release from the fuel matrix. The IRF for ^{14}C in cladding is 20 %, as discussed in Section 7.

For many of the radionuclides in Table 3 for which leaching measurements are not available, the only basis for the estimates are the observations that diffusion coefficients in UO_2 during reactor irradiation decrease in the order FG > I > Cs > other fission products, and the understanding of fission product chemistry, which has identified which fission products form solid solutions with UO_2 and which form secondary phases (JOHNSON & SHOESMITH 1988). An indication that using Cs or I to bound the release of other fission products is a conservative approach can be obtained by considering the case of Cd, one the most volatile of the fission products (after FG,

Cs and I) (CUBICCIOTTI & SANECKI 1978). Quantitative X-ray photoelectron spectroscopy of grain boundaries in CANDU fuel has been performed by HOCKING et al. (1994), who noted that Cd was only occasionally detected and would have been routinely seen if it had experienced the same fractional release as Cs. Iodine was not observed perhaps because of its low fission yield. The IRF for ^{129}I should thus provide a bounding value which would not be exceeded by other fission products. There is some doubt whether Pd and Tc should be included in the category of nuclides with a significant IRF, as these elements are present in UO_2 as insoluble alloy inclusions, thus releases are extremely small, as noted in leaching studies (see Table 2). Nonetheless, because they segregate significantly to grain boundaries, a value is assigned here based on the extent of restructuring in the rim region.

6.2 IRF Values for Higher Burnup UO_2 Fuels

There are no data available on the leaching of fission products from the rim region of higher burnup fuel from which to evaluate the releases under disposal conditions. In the absence of data, one can assume that restructuring controls the segregation to grain boundaries in the same way that it does for fission gas. The rim fraction derived from Figure 8 can thus be added to the fission gas release value to give values for all fission products that are insoluble in UO_2. Using the values in Section 4 for fission gas release from higher burnup PWR fuel, including some extrapolation, along with the bounding rim fractions from Table 1, the bounding IRF values for fission products for fuels with burnups of 55, 65 and 75 GWd/tIHM are summarised in Table 4. Only bounding values are given, as it is difficult to justify any other approach given the complete absence of fission product leaching data.

Table 4: Recommended IRF values of key radionuclides for BWR UO_2 fuel with a burnup of 55 GWd/tIHM and PWR UO_2 fuel for burnups of 55, 65 and 75 GWd/tIHM.

Bounding IRF Value (%)				
Nuclide	BWR UO_2 Fuel (55 GWd/tIHM)	PWR UO_2 Fuel (55 GWd/tIHM)	PWR UO_2 Fuel (65 GWd/tIHM)	PWR UO_2 Fuel (75 GWd/tIHM)
Fission gas	10 (gap) 8 (rim)	3 (gap) 8 (rim)	5 (gap) 13 (rim)	8 (gap) 17 (rim)
^{3}H	1	1	1	1
$^{14}C^{*}$	10	10	10	10
^{36}Cl	18	11	18	25
^{79}Se	18	11	18	25
^{90}Sr	1	1	1	1
^{99}Tc	8	8	13	17
^{107}Pd	8	8	13	17
^{126}Sn	18	11	18	25
^{129}I	18	11	18	25
^{135}Cs	18	11	18	25
^{137}Cs	18	11	18	25

* Value given is for release from the fuel matrix. The IRF for ^{14}C in cladding is 20 %, as discussed in Section 7.

6.3 IRF Values for MOX Fuels

There are few published data available on the leaching of radionuclides from MOX fuel (see Table 2). The limited data available show significant differences from UO_2 fuel, with ^{137}Cs releases greatly exceeding ^{129}I releases. Why such differences exist is not known, but they may be due to differences both in solid state chemistry and in microstructure. For example, MOX fuel is fabricated by blending of PuO_2 and depleted UO_2 powders, followed by cold compaction and high temperature sintering. Because the local burnup in PuO_2 grains is very high, fission gas accumulation leads to high local porosity (GUERIN et al. 1999), making MOX fuel potentially more susceptible to fission product leaching than UO_2 fuel. The understanding of how the differences influence fission product release is poor, thus proposed IRF values in Table 5 can only be considered rough bounding estimates.

Table 5: Recommended bounding FGR and IRF values of key radionuclides for PWR MOX fuel with burnups of 48 and 65 GWd/tIHM.

		IRF Value (%)	
Nuclide	t ½ (year)	PWR MOX Fuel (48 GWd/tIHM)	PWR MOX Fuel (65 GWd/tIHM)
Fission gas*	-	8	15
^{3}H	1.23×10^{1}	1	1
^{14}C#	5.73×10^{3}	10	10
^{36}Cl	3.0×10^{5}	15	20
^{79}Se	1.1×10^{6}	15	20
^{90}Sr	2.86×10^{1}	1	20
^{99}Tc	2.1×10^{5}	2	20
^{107}Pd	6.5×10^{6}	2	20
^{126}Sn	2.3×10^{5}	15	20
^{129}I	1.57×10^{7}	15	20
^{135}Cs	2.3×10^{6}	10	20
^{137}Cs	3.02×10^{1}	10	20

* Gap inventory

\# Value given is for release from the fuel matrix. The IRF for ^{14}C in cladding is 20 %, as discussed in Section7.

6.4 Uncertainties in IRF values for UO_2 and MOX Fuel

The amount of data on leaching of fission and activation products from UO_2 and MOX fuel is very limited, when one considers the different reactor types, the number of different fuel element designs and the variation in fuel operating conditions. Furthermore, the question of whether subsequent microstructural changes, such as increased cracking as a result of He buildup, or increased accumulation of fission products at grain boundaries by solid-state diffusion, might occur during the period over which canisters are likely to remain intact (~10'000 a), has been asked (POINSSOT et al. 2001). Some further assessment of the uncertainties in the recommended values is thus necessary.

Several factors indicate that the best estimate values proposed for normal burnup (48 GWd/tIHM) UO_2 fuel are reasonable and are unlikely to be exceeded. In particular, the study of GRAY & STRACHAN (1991), which involved flow-through tests on fuel samples ground to the original grain size, illustrates that the grain boundary inventory of fission products is released from the surfaces of grains quickly, after which congruent dissolution occurs. The IRF values recommended in the present study are consistent with this observation, i.e. the IRF values implicitly assume the fuel has a network of open pathways connecting all grain boundaries. As a result, no credit is given to the isolation capacity of fragments, thus microcracking, should it occur, will not increase IRF values.

The question of whether or not solid-state diffusion of fission products to grain boundaries will cause the grain-boundary inventories to increase with time is an active area of study (POINSSOT et al. 2001). Thermal diffusion of fission products is completely irrelevant at the low temperatures of interest here. Volume diffusion of species incompatible with the UO_2 lattice is known to occur (e.g. loss of Pb and other U daughters from uraninite) and has been discussed in the context of loss of fission products from Oklo uraninite (COWAN 1978). The diffusion coefficients for elements such as Cs, I and Xe in Oklo uraninite must have been at least 4×10^{-26} m^2/s in order to explain their losses. A value of this magnitude would increase the loss from fuel grains (radius ~5 μm) by approximately 10 % in 10'000 years. Whether such a value is relevant to behaviour of spent fuel in a repository is unclear. In particular, the Oklo reactors experienced high temperatures (~400 ^{0}C) and fission, both of which may have increased the diffusion coefficient relative to values that might be expected for spent fuel in a repository. Radiation-enhanced diffusion in spent fuel as a result of α-recoil, like fission, may also contribute to such migration (POINSSOT et al. 2001), although no definitive answers will be available until experimental measurements of the rates are performed. No account is taken of this loss mechanism in the IRF estimates presented. If the process does occur, it will lead to a slow loss of fission products from fuel grains, but the magnitude is unlikely to be greater than the bounding values given for high burnup fuels. As a result, the latter values can be used in performance assessment calculations to evaluate the significance of this process.

For higher burnup fuel, an improved understanding of IRF values can only come with more laboratory leaching studies.

7 RADIONUCLIDE RELEASE FROM ZIRCALOY CLADDING

Although Zircaloy is highly resistant to uniform corrosion, its susceptibility to localized corrosion in groundwaters is unknown. Combined with considerable uncertainties regarding hydrogen-pickup rates and the possibility of hydrogen-induced cracking, this has led to the conservative assumption in performance assessment studies that cladding provides no barrier to the release of radionuclides from the spent fuel (JOHNSON et al. 1996, SKB 1999). Nonetheless, the corrosion resistance of Zircaloy may result in significant delays in the release of radionuclides entrained within the cladding itself, which is discussed further below.

7.1 Concentrations and Distributions of Radionuclides in Zircaloy Cladding

A variety of radionuclides are present in the Zircaloy cladding and other structural materials in LWR fuel elements. These radionuclides are produced by neutron activation of alloying elements and impurities present in the as-fabricated materials. The most important of these in terms of long-term safety assessment are ^{14}C, ^{36}Cl, ^{59}Ni and ^{63}Ni.

Because the impurities in Zircaloy are uniformly distributed, and the temperature of the cladding during in-reactor irradiation and the associated solid-state diffusion rates are relatively low, the activation products would be expected to be likewise uniformly distributed on a microscopic scale (although there will be larger-scale variations because of flux variation within fuel rods). There is, however, evidence that ^{14}C has a higher concentration in the oxide film than in the underlying alloy. For example, YAMAGUCHI et al. (1999) determined that 17 % of the ^{14}C in cladding is present in the oxide film. Assuming that the oxide film in their study was about 60 μm thick, this represents a ^{14}C concentration in the oxide film that is at least three to four times that in the underlying alloy. A similar ^{14}C enrichment in the oxide film was observed by SMITH & BALDWIN (1993). For other long-lived radionuclides present at trace levels in Zircaloy, e.g., ^{36}Cl, ^{59}Ni and ^{63}Ni, the distributions within cladding are unknown, although within the alloy itself they are likely to be present as grain-boundary phases.

7.2 Release Rates of Radionuclides from Zircaloy Cladding

The release of radionuclides from cladding is expected to be controlled in large part by the uniform corrosion rate of Zircaloy (the case of ^{14}C is discussed further below). At the low temperatures in a repository, the data are rather limited, but the study of ROTHMAN (1984) has dealt with the question of extrapolation of high-temperature (in-reactor) uniform corrosion rates down to150 ^{o}C. At this temperature, he estimated that the rate would be ~1 nm a^{-1}, and significantly lower rates would be expected at lower temperatures as the repository cools. ROTHMAN also notes that the corrosion rate is expected to be the same in low salinity groundwaters. A study of the corrosion rate of irradiated Zircaloy pressure tube material at 90 ^{0}C in aerated Hanford River water by JOHNSON (1977) gave an estimated rate of 3 to 5 nm a^{-1}. VIDEM (1981) reported corrosion rates for unirradiated Zircaloy of 0.1 to 1 nm a^{-1} in aerated KBS groundwater at 85 ^{0}C. It is also worth noting that the uniform corrosion rate of a similar metal, titanium, is reported to be ~2 nm a^{-1} in water-saturated bentonite at 90 ^{o}C. (MATTSSON & OLEFJORD (1984, 1990) and MATTSSON et al. (1990)). For materials such as titanium and zirconium alloys, the very thin passive oxide film is produced as a result of oxidation by water, thus the presence of oxygen in solution is not required for the maintenance of passivity. Based on the above values, and giving particular weight to the rates reported by JOHNSON (1977) for irradiated material, a maximum corrosion rate of 10 nm a^{-1} is proposed for spent fuel cladding for radionuclides entrained in the Zircaloy.

Two other recent studies report the corrosion rate of unirradiated Zircaloy, measured using H_2 evolution methods in alkaline pore water (pH 10 to 13.5) at temperatures of 30 to 50 ^{0}C. These data are particularly relevant to the conditions in a low and intermediate level waste repository. WADA et al. (1999) reported initial corrosion rates of 1 nm a^{-1}, dropping to ~0.1 nm a^{-1} after 300 days. The highest long-term rate (0.3 nm a^{-1}) was observed at pH 10 and 50 ^{0}C. At pH 13.5 and 30 ^{0}C the rate was similar to that at pH values of 10 and 12.5. KURASHIGE et al. (1999) measured the H_2 evolution rate of Zircaloy in alkaline pore water at 30 and 45 ^{0}C. They reported average rates of 1 to 2 nm a^{-1} after 400 to 500 days. Some samples were treated with steam at 400 ^{0}C to thicken the oxide film, but this had no effect on the subsequent low temperature corrosion rate. YAMAGUCHI et al. (1999) observed that a corrosion rate of 0.03 μm a^{-1} appeared to account for the ^{14}C release from a cladding sample with the oxide film removed. The difference between their rate and the lower rate extrapolated from high temperature studies may be a result of an acceleration in the corrosion rate caused by removal of the protective oxide film. Besides the difficulty of estimating an appropriate long-term corrosion rate of the alloy, there remains the observation of a rather high concentration of ^{14}C in the oxide film (YAMAGUCHI et al. (1999)), the release of which occurred at a rate considerably greater than that from the alloy itself. VAN KONYNENBURG et al. (1987) and SMITH & BALDWIN (1993) also reported a rapid release of ^{14}C (<1 % of the total inventory) in air oxidation and water corrosion studies on Zircaloy cladding. For the present study, the release of ^{14}C from the oxide film is assumed to be rapid, and is represented as an IRF for the cladding of 20 %. The remaining ^{14}C is assumed to be released congruently with the uniform corrosion of the Zircaloy, at a corrosion rate of 0.01 μm a^{-1}. Assuming that corrosion occurs from both sides of the cladding and the cladding thickness is ~600 μm, radionuclide release would occur at a constant rate of ~3 x 10^{-5} a^{-1} of the initial inventory.

An additional special consideration for release calculations for ^{14}C is its chemical form. It is likely that ^{14}C is present in cladding as carbide. Studies of the corrosion of steel containing carbides in anoxic water indicate that a range of range of hydrocarbons are produced, including methane, ethene, ethane, propane, propene and butenes (DENG et al. 1997). In the case of leaching in anoxic cement pore water, YAMAGUCHI et al. (1999) observed that ^{14}C was released in organic form, although they detected it in the aqueous phase only. Nonetheless, under the high hydrogen partial pressure conditions expected in the repository, the ^{14}C may be converted to a form such as methane. As a result, radionuclide transport scenarios should consider the possibility of gas phase transport of ^{14}C from the near field.

8 CONCLUSIONS

The data available on FGR for PWR and BWR UO_2 fuel provide a good basis for estimating the average FGR for fuel irradiated in Swiss power reactors for the reference burnup of 48 GWd/tIHM. Based on these data, the best estimate average FGR values for BWR and PWR UO_2 fuel of this burnup are 5 and 1 %, respectively. Estimates of average IRF values for UO_2 fuel for various fission and activation products have been derived by correlating the results from spent fuel leaching studies and FGR measurements. In the case of UO_2 fuel with a burnup of < 50 GWd/tIHM, the IRF values, which are below 10 % for all radionuclides, can be considered reasonably well grounded in terms of experimental data and process understanding.

For UO_2 fuel, the quantities of fission gas released from grains but trapped in rim pores have also been estimated. This process is relatively unimportant at burnups below 50 GWd/tIHM, but becomes increasingly significant above this value. Limited information is available on fission gas release for higher burnup fuel, and no data at all are available in the case of fission product leaching. Based on a bounding estimate of fission gas that is segregated within the rim, which may reach ~ 17 % of the inventory in a PWR UO_2 fuel with a burnup of 75 GWd/tIHM, and FGR measurements of ~ 8 %, a bounding estimate of the IRF is ~ 25 %.

For MOX fuel, there is almost no fission product leaching data and no published fission gas release data for burnups exceeding 50 GWd/tIHM. There are thus substantial uncertainties in estimating IRF values for MOX fuel, in particular at higher burnups (65 GWd/tIHM), where IRF values may be as high as 20 %.

Evidence indicates that Zircaloy cladding will corrode at a rate of less than 10 nm a^{-1}. For an assumed rate of 10 nm a^{-1}, the radionuclide release rate would be 3 x 10^{-5} a^{-1} of the initial inventory. For ^{14}C, an IRF value for the cladding of 20 % is estimated, consistent with the findings of a significantly enhanced release from the oxide film. There are indications that the released ^{14}C may be organic, which may have significant implications for transport.

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