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Models for fission products release from spent nuclear fuel and their applicability to the First Nuclides project

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ABSTRACT

This paper gives an overview of conceptual and mathematical models regarding the *Instant Release Fraction* (IRF) under repository conditions. To this end a variety of models, including those for fission products release under in-reactor conditions, are reviewed. Correlation models are used to give direct prediction of IRF based on the measured or estimated release of fission gases (FGR). Semi-empirical models applied to experimental results allow conceptual models of radionuclide release to be constructed and verified against measured data. In addition, physics-based mechanistic models for fission products release under in-reactor conditions are being continually developed. These models are often included into larger coupled codes for the assessment of fuel performance, and can be used to accurately predict the release of fission gases and some other fission products during reactor operation. Such information is valuable for estimating IRF under the conditions expected for repository of spent nuclear fuel.

1. INTRODUCTION

Safety assessment of the repository for spent UO_2 fuel requires that the potential release of radioactivity upon breaching of the engineered barriers system due to degradation process be quantified. From the point of view of performance assessment (PA) of the repository for spent fuel, the instant release fraction (IRF) is the fraction of the radioactive inventory that will be released from the waste "immediately" after the fuel rod cladding fails, and the waste containment is compromised. The IRF may have important implications for PA because some of the preferentially released radionuclides are characterised by both relatively long half-lives and high degrees of mobility (e.g. ^{129}I and ^{36}Cl [1]). As a result a great deal of effort has been directed into improving the understanding and quantifying the physico-chemical processes involved in the instant release of radionuclides from SNF. These goals are being achieved through a combination of experimental and modelling work [1, 2, 3, 4, 5, 6, 7, 8].

In-reactor irradiation with neutrons causes a continuous production of fission products within the fuel. The fission products include isotopes of noble gases (mainly Xe and Kr) and others, such as ^{3}H, ^{14}C, ^{79}Se, ^{99}Tc, ^{107}Pd, ^{125}Sn ^{129}I, ^{135}Cs, and ^{137}Cs. Fission products may occur in the fuel as volatiles (I, Br, Cs, Rb, Te), metallic precipitates (Mo, Tc, Ru, Rh, Pd, Ag, Cd, In, Sn, Sb, Te), oxide precipitates (Rb, Cs, Sr, Ba, Zr, Nb, Mo, Te) or oxides in the in the UO_2 structure (Cs, Nb, Te, Y, Zr, the earth alkaline elements Sr, Ba, Ra and the lanthanides La, Ce, Pr, Nd, Pm, Sm, Eu) [9].

In a rod containing stacked pellets of polycrystalline UO_2 fuel during reactor operation, concentration and thermal gradients drive diffusive redistribution of some of the fission products into the inter-granular space, and outside the pellet towards the inner-rod void space.

It is thought, that under repository conditions, breaching of the cladding would rapidly release the inventory accumulated in the void space of the rod. Also relatively quickly would be released some radionuclides segregated at the fuel grain boundaries. Both of these inventory fractions are typically considered to constitute the IRF. In contrast, a much slower radionuclide release would occur over long periods of time due to dissolution of the UO_2 matrix [8, 10].

Typically, the rapid release of radioactivity when spent UO_2 fuel is contacted by water is assessed through laboratory experiments. These, however are technically and economically challenging due to the high levels of radioactivity retained by the spent fuel. The measured IRF depends on the type and in-reactor history of the fuel, which requires that a large number of varied spent UO_2 materials be studied. This work progresses steadily and simple models of IRF have been developed based on the available data.

In this contribution a review of available conceptual and mathematical models related to the IFR from spent UO_2 fuel is given. A special focus is placed on modelling of the release of fission gases from UO_2 under reactor operation conditions, which has been investigated for a long time and is relatively well understood. The modelling approaches developed for fission gas release and the results obtained using them can be helpful in estimating the IRF from UO_2 under repository conditions.

2. MODELLING APPROACHES

2.1 Empirical and Semi-empirical Models for IRF Release

The release of fission products from spent nuclear fuel has been investigated experimentally [1, 3, 10, 11]. The fission gas inventory in the inner-rod void can be collected and analysed, the release of fission product radionuclides from the spent UO_2 fuel can be investigated through leaching experiments, and the results obtained compared with reactor operating conditions and various fuel properties. Despite the limited amount of experimental data, useful correlations of this type have been obtained, which form the basis of empirical models used to estimate the IRF.

Most empirical models rely on the observation that the quantities of radionuclides in the inner-rod void and those leached from the fuel grain boundaries correlate with FGR [3, 8, 10, 11]. Examples of correlation with FGR for ^{137}Cs release into the cladding gap, and for ^{137}Cs release into the cladding gap and leached from the grain boundaries are shown in Figure 1 and Figure 2, respectively. Similar correlations for other radionuclides, such as, ^{14}C, ^{36}Cl, ^{79}Se, ^{90}Sr, ^{99}Tc, ^{107}Pd, ^{126}Sn, ^{129}I and ^{135}Cs have been determined [3, 8, 10, 11]. These correlations can be used inversely to estimate the IRF (within the calibration range) if the FGR is known.

With respect to the fission radionuclides for which there are no experimental data and no such empirical correlations exist (e.g. Cd), bounding IRF values can be defined based on consideration of their diffusion coefficient during irradiation relative to other radionuclides for which such data are available [8].

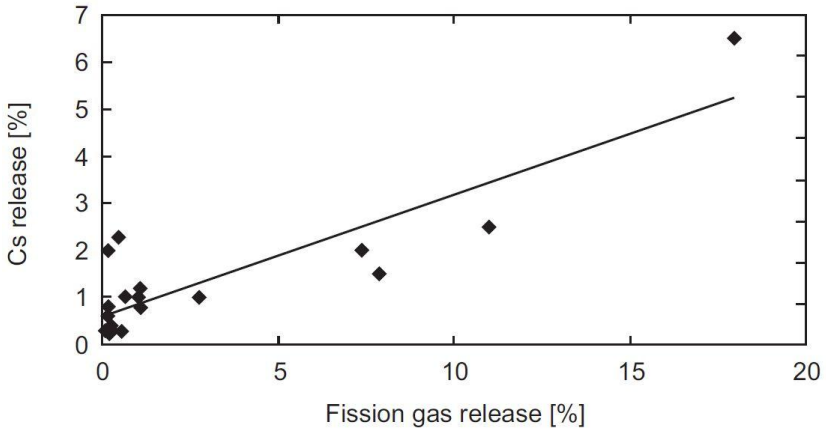

Figure 1: ^{137}Cs release into the cladding gap as a function of FGR (from [3])

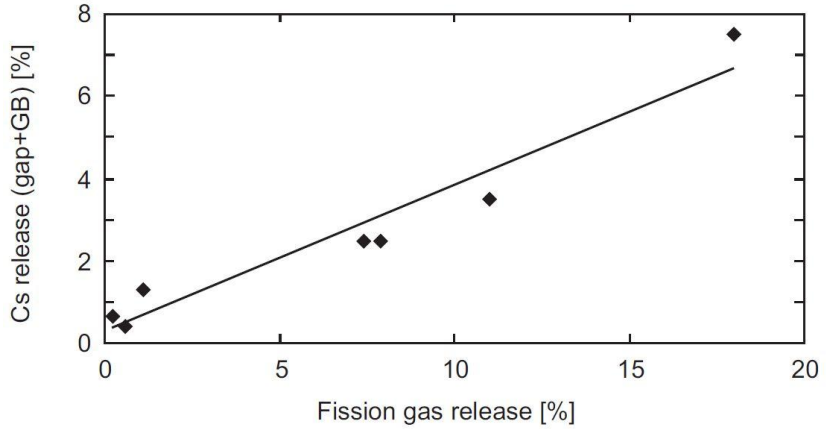

Figure 2: ^{137}Cs release into the cladding gap and leached from grain boundaries as a function of FGR (from [3])

The main limitation of correlative models is that they are only valid under certain conditions (fuel type and in-reactor history). Johnson and co-workers [1] point out that empirical models may have some mechanistic basis given that the diffusion coefficients of some IRF elements in UO_2 (e.g. I and Cs) at reactor-operating temperatures are relatively high. For example, it has been observed [12] that the ratio of diffusion coefficients of Cs/Xe is about 0.33, which is consistent with the observed Cs to fission gas release ratio [3].

Semi-empirical models have been developed based on experimental leaching data with the aim to provide insight into the underlying processes that control the release of fission products from UO_2 spent fuel. For example, Casas and co-workers [13] analyse the results of static

(batch) and dynamic (flow-through) leaching experiments performed on powdered sample of high burn-up, and they develope a kinetic dissolution model whose parameters where fitted to the experimental data (Equation 1).

Equation 1

$$m_{RN}(t) = m_{RN,1\infty} \cdot (1 - e^{-k_1 t}) + m_{RN,2\infty} \cdot (1 - e^{-k_2 t}) + m_{RN,3\infty} \cdot (1 - e^{-k_3 t})$$

Where, $m_{RN(t)}$ is the cumulative mass of the radionuclide as a function of time, $m_{RN,1\infty}$, $m_{RN,1\infty}$ and $m_{RN,1\infty}$ are the cumulative mass of the radionuclide released during stages *1*, *2* and *3*, respectively, and k_1, k_2 and k_3 are constants. $m_{RN,1\infty}$, $m_{RN,1\infty}$, $m_{RN,1\infty}$, k_1, k_2 and k_3 are the fitting parameters (with the added constrain that the sum of $m_{RN,1\infty}$, $m_{RN,1\infty}$ and $m_{RN,1\infty}$ is known from the total leached inventory).
Analysis and interpretation of the model allowed for an operational conceptual model to be constructed. According to the model, radionuclides studied (Cs, U and Tc) are released simultaneously during the experiment from distinct locations within the fuel: the gaps and cracks, grain boundaries, and the fuel matrix.

2.2 Empirical and Semi-empirical Models for Fission Gas Release

FGR can be correlated with the release of other fission products, thus providing useful information on IRF. FGR can be measured during post-irradiation inspection (PIE), but this is costly, impractical for large numbers of rods and does not offer predictive capability. Because FGR from fuel during reactor operation is an important safety and economic consideration, predictive models for this process have been in development since the early days of the nuclear power generation. In general, these models fall into two broad categories: (1) empirical and semi-empirical correlations and (2) process-oriented mechanistic models, often are incorporated into fuel performance assessment codes.
It has been observed that in-reactor FGR from UO_2 fuel correlates strongly with linear heating rate, which depends on fuel temperature [14]. Increasing degree of burn-up causes a reduction of thermal conductivity of the fuel, hence a correlation of FGR with the degree of burn-up is also observed (at burn-up > 40 GWd/t_{iHM}) – Figure 3 [15].

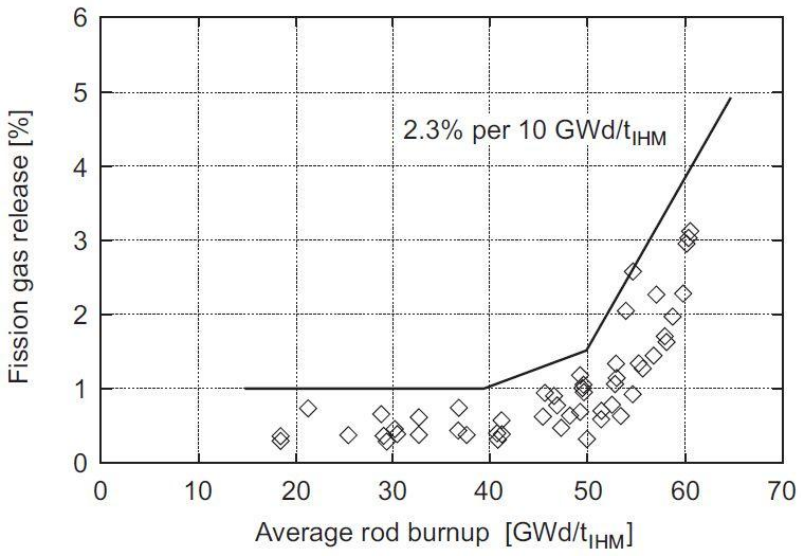

Figure 3: Fission gas release from PWR fuel as a function of burn-up (from [8] after [15])

In UO_2 fuels characterised by high degree of burn-up (>40 GWd/t_{iHM}) restructuring of the peripheral parts of the fuel is observed [8]. The rim region of the fuel pellet undergoes recrystallization resulting in smaller grain sizes and increased porosity filled with over-pressurised fission gas bubbles [8]. Experimental data indicate that the thickness of the rim is a function of the degree of burn-up experienced by the rim area Figure 2 [16]. Koo et al. [16] have developed a model to calculate the fraction of the total amount of Xe produced in the fuel pellet which is retained in the rim pores (Figure 5). In radionuclide release models, the inventory contained in these bubbles can either be treated as part of the IRF or can be considered to be released during matrix dissolution only [8]. Johnson et al. [8] argue that fission gas retained in rim pores can be considered to be released from the fuel matrix and should therefore be considered to be part of the IRF even though it is not released into the cladding gap. Other fission products, which do not form a solid solution with UO_2, are expected to be released from the matrix into the inter-grain space during recrystallization of the rim and could be released if contacted by water [8].

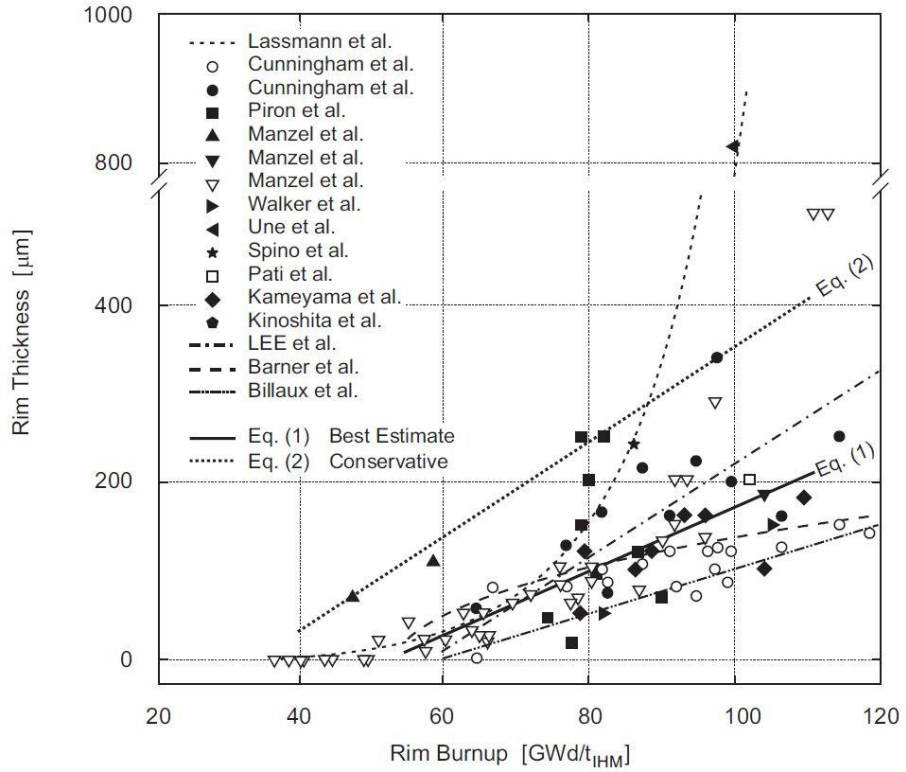

Figure 4: Rim thickness (in µm) as a function of burn-up (in GWd/t_U), from [16]. Eq (1) and Eq (2) represent best- and "conservative" (pessimistic) fit to data, respectively.

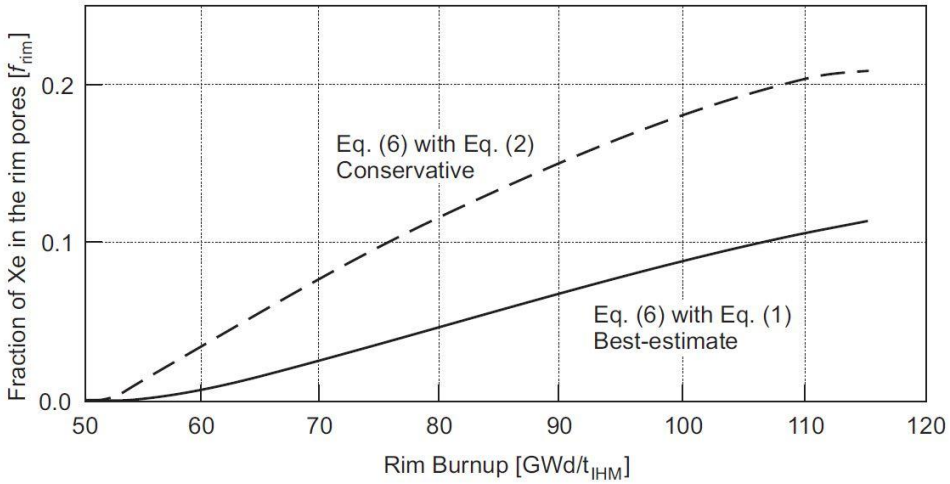

Figure 5: Fraction of the total Xe produced within a pellet retained in the rim pores (from [16]). Equations are given in the original paper.

2.3 Mechanistic Models

The main limitation of empirical models, such as those presented above, is that their application is restricted to a specific fuel type used under specific reactor operation conditions. In addition, such models offer little in the way of system behaviour understanding.

This deficiency is however remedied by more mechanistic, process-oriented models, which are being increasingly developed. Such models enable the approximation of the grain-boundaries and cladding gap fission gas inventories, thus providing an estimate of the IRF.

Conceptual models for Fission Products

Fission gas generation and transport during irradiation contributes to fuel swelling and has the potential to cause damage to cladding due to strong mechanic pellet-cladding interactions. As such it is a significant safety consideration and considerable amount of effort has been devoted to better understanding these processes.

Underlying mechanistic models are conceptualisations of fission gas generation and transport within the fuel. For high burn-up UO_2 fuel, the following processes are typically considered [17]:

- *Gas generation due to the fission of* ^{235}U. This is mainly the production of the stable isotopes of Xe and Kr, which, due to their mass, have the highest probability of being created.
- *Recoil and knock-out.* Due to high kinetic energy (60-100 MeV) and momentum conservation, a fission gas nucleus can be directly recoiled from the fuel surface if the fission fragment is created sufficiently closely (i.e. less than 6-7 μm) to the surface. Alternatively, elastic collisions with neighbouring nuclei may cause them to be knocked-out across the fuel surface. Overall, however, these two processes are considered to contribute little to the global fission gas release from the fuel.
- *Lattice (intra-granular) diffusion.* This is the transport from within the grain towards the grain's edge and is driven by concentration and thermal gradients.
- *Grain-boundary diffusion.* This is typically much faster than the lattice diffusion due to the atomic jump frequency on a grain boundary being significantly larger than the jump frequency of the lattice atoms.
- *Trapping*
  - Intra-granular. Bubbles of fission gases are formed either spontaneously (homogenous nucleation) or due to the presence of (natural or radiation-induced) crystal imperfections (heterogeneous nucleation).
  - Inter-granular. Two different conceptual models for the grain boundary have been postulated:
    - The grain boundary is considered as a high-diffusivity pathway allowing fast gas diffusion into the inter-granular space or the cladding gap.

    - The grain boundary is considered to be a perfect trap for fission gases, that form bubbles at the boundary.

- *Irradiation-induced resolution*. Some gaseous atoms trapped in intra-granular bubbles can be re-dissolved into the fuel matrix through bubble interaction with fission fragments.
- *Intra-granular bubble motion*. This is an additional mechanism (to lattice diffusion) of gas transport towards the grain boundary due to bubble motion.
- *Grain growth*. Larger fuel crystals grow at the expense of smaller. This causes a moving grain boundary that sweeps and collects fission gases, but also, due to increased mean diffusion distance to the grain boundary, reduces the rate of gas transport to the boundary.
- *Grain boundary bubble development.* This is the process, caused by grain boundary bubbles interact, resulting in bubble coalescence and the formation of continuous pathways through which the gas may be released to the free volume (released from the fuel).

Mathematical Implementation

Adequate modelling of the fission gas generation and release requires considering a number of mutually interdependent processes (described above). A more detailed description of how these processes can be represented mathematically is given in [17, 18, 34]. In what follows, a brief overview is given.

The first attempt to model intra-granular diffusion of fission gas out of a fuel element is that of Booth [19]. To simplify calculations, the Booth model considers a sinter composed of a collection of uniform spheres (idealised grains) with an equivalent radius. This model considers the single process of atomic diffusion in the spherical grain using the Fick's law and assuming constant, single-atom diffusion coefficient. The fission gases are considered to diffuse from the grain towards the grain's boundary. Depending on the boundary conditions, the solution (radial concentration of fission gas in time) to the model can be a closed analytical solution or expressed by an infinite series. By integrating the amount of the gas passing through the grain boundary surface and dividing by the total amount initially in the sphere initially, the fraction of the gas diffused out of the grain at a given time can be obtained [19]. The original Booth model ignores fission gas release due to recoil and knock-out near grain boundary surfaces. This limitation is overcome in some codes by, for example, the implementation of an empirical release term, which is a function of burn-up [20].

The model of Booth was later extended by Speight [21] (Equation 2), by further accounting for the presence of (immobile) intra-granular gas bubbles into which diffusing gas atoms can be trapped and re-dissolved due to irradiation (with constant rates, *g* and *b*, in Equation 2,

respectively). The model is described by a single diffusion equation and in the absence of bubbles is mathematically equivalent to the Booth model. In the Speight model, gas atom trapping and re-solution is expressed by multiplying the diffusion coefficient by a constant expression ($b/(b+g)$), which renders the single-atom diffusion coefficient an effective diffusion coefficient.

Equation 2

$$\frac{\partial C_t}{\partial t} = \frac{b}{b+g} D_s \nabla^2 C_t + \beta$$

Where, C_t is the fission gas concentration as a function of time (t) and space, D_s is the single-atom diffusion coefficient, β is the fission gas generation rate, and g and b are the trapping and re-solution parameters, respectively.

Van Uffelen and collaborators [22] have further extended the Speight model by considering the mobility of intra-granular fission gas bubbles. The mobility of fission gas trapped in bubbles is accounted for by adding an extra term to the effective diffusion coefficient of Speight (Equation 3):

Equation 3

$$D_{eff} = \frac{b}{b+g} D_s + \frac{g}{b+g} D_b$$

Where, D_b is the bubble diffusion coefficient.

The migration of fission gases in the UO_2 lattice can be treated using diffusion theory, where the effect of temperature on the diffusion coefficient can be approximated using the Arrhenius theory. However, this situation is complicated by several factors, including [17]:

- Radiation induced damage to the crystal lattice, which acts both to retard (by trapping in intra-granular bubbles) and to speed up (by re-solution and radiation-enhanced diffusion) diffusion
- Chemical effects related to the production of fission products (e.g. changes in oxygen to metal ratio)

In order to represent the above-mentioned effects the single-atom diffusion coefficient is typically defined as a sum of three component coefficients, each expressing a different contribution (e.g. the intrinsic diffusivity and the thermal and athermal impact of irradiation). The components of the diffusion coefficient are represented using analytical expressions based on empirical correlations [17, 22, 30].

Bubble diffusion coefficient (D_b) can be approximated based on the assumption that bubble diffusion is controlled by volume (lattice) diffusion [22]:

Equation 4

$$D_b = \frac{3\Omega}{4\pi R^3} D_v$$

Where, Ω is the volume occupied by fission gas inside the bubble, R is the mean radius of bubbles within a grain and D_v is the volume self-diffusion coefficient (expressed using experimental correlations) [22].

In most codes, swelling of the fuel (fractional volume, ΔV, normalised to the unit fuel volume, V) is represented by means of semi-empirical expressions relating gaseous swelling with temperature and burn-up, for example [23]:

Equation 5

$$\left(\frac{\Delta V}{V}\right)_{gFP} = A(1 - \alpha_{FG} FGR - \alpha_{Cs} CSR) bu$$

Where, A is a constant for solid swelling, FGR is the local fraction fission gas release from the grain, CSR the fractional release of volatile fission products from the grain, bu is the local fuel burn-up, and α_{FG} and α_{CS} are constants.

Considering a number of assumptions and simplifications, more mechanical models relying on fission gas bubble and fuel grain size and geometry have been developed, too. For example, intra-granular swelling (fractional volume, ΔV, normalised to the unit fuel volume, V) can be calculated following Pastore and collaborators [24]:

Equation 6

$$\left(\frac{\Delta V}{V}\right)_{ig} = N_{ig}\left(\frac{4}{3}\pi R_{ig}^3\right)$$

Where, N_{ig} is the number density of intra-granular bubbles, R_{ig} is the intra-granular bubble radius. Following the approach of Pastore et al. [24] the fractional grain-face swelling normalised to the unit volume of fuel can be calculated according to:

Equation 7

$$\left(\frac{\Delta V}{V}\right)_{gf} = \frac{1}{2}\frac{N_{gf}}{(1/3) r_{gr}}\left(\frac{4}{3}\pi\varphi(\theta) R_{gf}^3\right)$$

Where, N_{gf} is the number density of grain-face bubbles per unit surface area, r_{gr} is the grain radius, θ is the bubble semi-dihedral angle, $\phi(\theta)$ is a geometric factor relating the volume of lenticular-shaped bubble to that of a sphere, and R_{gf} is the radius of curvature of grain-face bubbles.

As mentioned earlier, the growth of fuel grains has the effect of fission gas sweeping at the grain boundaries, which provides an accumulation mechanism. In addition, the diffusion

distance towards the grain boundary surface increases which slows down diffusive transport. Incorporating the growth of fuel grain size requires that the differential diffusion equation be solved with a moving boundary thus increasing the level of complexity of the model. Some models disregard the impact of grain size growth on diffusive transport and consider the sweeping effect only, for instance the model of Hargreaves and Collins [25]. Other models attempt to represent both the radionuclide sweeping and a moving boundary diffusion transport, such as the model of Ito and collaborators [26]. However, most models neglect the grain-size distribution, which has been shown to over-estimate the fission gas release fraction [27].

White [28] proposed a model for calculating intra-granular fission gas bubble growth rate based on a calculation of the number of vacancies and gas atoms present.

The process of bubble intersection, merging and growth is termed coalescence. This can be modelled, for example, after White [28], Veshchunov [29] or Pastore and co-workers [34] based on consideration of change in bubble surface area when bubbles merge.

The release of fission gas from the grain boundaries is usually modelled by assuming a threshold value of fission gas concentration at the grain boundary (termed saturation) over which all the gas reaching the grain boundary is released [30]. This concentration can be calculated following White and Tucker [31]. Nevertheless, this model relies on the assumption that the gas is ideal and that the bubble is in mechanical equilibrium. This problem can be circumvented by considering mechanistic kinetic models, where coupling between hydrostatic pressure, temperature, gas swelling and release is represented on a physical basis [24, 31, 34].

Fuel Performance Assessment Codes

The process-oriented FGR models described above can either be coded into stand-alone software or, typically, implemented as modules into the so-called fuel performance assessment codes, briefly described below.

The need to understand in-reactor fuel behaviour is dictated by both safety and economic considerations. As the possibility to study the performance of nuclear fuel through experiments in reactor and post-irradiation examinations (PIE) is limited, it has been recognised that adequate models capable of representing relevant processes affecting nuclear fuel during reactor operation were needed. Fuel performance predictions are required by safety-based calculations, for design purposes and performance assessment. In addition, such codes are also useful in optimising the reactor operation conditions in order for the reactors to be run economically. Many fuel performance codes characterised by varying degrees of complexity have been developed over the decades to simulate fuel behaviour in different

types of reactors. The ultimate goal of these codes is to reliably predict the in-reactor behaviour of a reactor rod during both normal and abnormal operation conditions. Such codes are now being extensively used across the nuclear industry by fuel manufacturers, power plant operators, regulatory and licensing authorities, and by research centres and universities [32, 33]. A brief list of selected commonly used fuel performance assessment codes with the capability to model fission gas release is given in Table 1.

Table 1: Selected fuel performance assessment codes with the capability to model fission gas generation and release.

Program Name	Developer
COMETHE	Belgonucleaire, Belgium
COPERNIC	AREVA NP, Germany
COSMOS	KAERI, South Korea
ENIGMA	British Energy, BNFL, UK
FALCON	EPRI, USA
FASTGRASS	NRC, USA
FEMAXI	JAEA, Japan
FRAPCON	PNNL, USA
FRED	Kurchatov Institute, Russian Federation
LIFE	ANL, USA
MACROS	SCK•CEN, Belgium
MFPR	IBRAE (Russian Federation) & IRSN (France)
ORIGEN	ORNL, USA
PARFUME	INEEL, USA
SPHERE	PSI, Switzerland
START	VNIINM-Bochvar, Russia
TOUTATIS	CEA, France
TRANSURANUS	ITU, Germany
VICTORIA	NRC, USA

The in-reactor fuel behaviour is affected by a range of processes including: thermal, mechanical, irradiation, densification, swelling, fission gas generation and release. Modelling of processes related to fission gas generation, transport and release during irradiation in reactor are an important component adequate analysis of the thermo-mechanical behaviour of the fuel. One reason for this is that fuel swelling due to fission gas generation promotes pellet-cladding interactions. Moreover, FGR into the free volume inside the rod causes pressure build-up and degrade thermal properties of the rod-filling gas. Therefore, the thermal, mechanical and fission gas release processes, and their impact on the fuel performance are closely linked. The thermal and mechanical processes are described in terms of dimensional equations, thus defining the dimensionality of the model. When simulating the behaviour of a

complete fuel rod, efforts are made to decrease the dimensions of the model in order to reduce computational costs. Most codes are 1-D or quasi-2-D (so called 1.5-D), where radial calculations are coupled axially by the coolant energy equation, common internal rod gas pressure and, sometimes, by a 1-D model for the axial friction forces [34].

Although specific capabilities and numerical implementations vary broadly between the different codes, the following are calculated by a typical modern fuel performance assessment code (such as the TRANSURANUS code):

- ***Thermal Analysis***
  - Axial heat transfer in the coolant
  - Heat transfer in the cladding
  - Heat transfer from cladding to fuel
  - Heat transfer in the fuel
- ***Mechanical Analysis***
  - Elastic strains
  - Non-elastic strains: swelling
  - Non-elastic strains: plasticity and creep
  - Non-elastic strains: pellet fragment relocation
- ***Fission gas generation, transport and release***
  - Swelling due to gaseous fission products
- ***Thermal and irradiation induced densification of the fuel***
- ***Volume changes due to phase changes***
- ***Neutronics***

Several codes are currently being developed with a special focus on the FGR analysis. MFPR (Model for Fission Products Release) represents a state-of-the-art implementation of such coupled mechanistic models. Recent developments in the code take account of the influence of microscopic defects (vacancies, interstitial and fission atoms, bubbles, pores and dislocations) in the UO_2 structure on the transport of fission gases. The code provides a mechanical description of the behaviour of chemically active elements based on the multi-phase and multi-component thermo-chemical equilibrium at the grain boundary and fuel oxidation state calculation [35]. MFPR incorporates an advanced model for inter-granular bubble mobility in irradiated UO_2, which considers three mechanisms: volume diffusion, evaporation/condensation and surface diffusion [36]. The code also includes an advanced model for grain face transport of gas atoms that consistently considers the effects of atom diffusion, trapping and irradiation-induced re-solution [37, 38].

Modern FGR models are capable of reproducing experimentally measured data with good accuracy. Figure 6, Figure 7 and Figure 8 show the results of the validation oft he FRAMATOM FGR model (implemented in the COPERNIC fuel performance assessment code) against experimental data [39]. In the exercise, the results obtained using the

FRAMATOM FGR model were compared with a database that covers over 400 irradiated rods and includes high burn-ups, high powers, short to long transients, UO_2, UO_2-Gd_2O_3 and MOX fuels (with burn-ups of up to 76, 55 and 53 GWd/tM for UO_2, UO_2-Gd_2O_3 and MOX fuels, respectively).

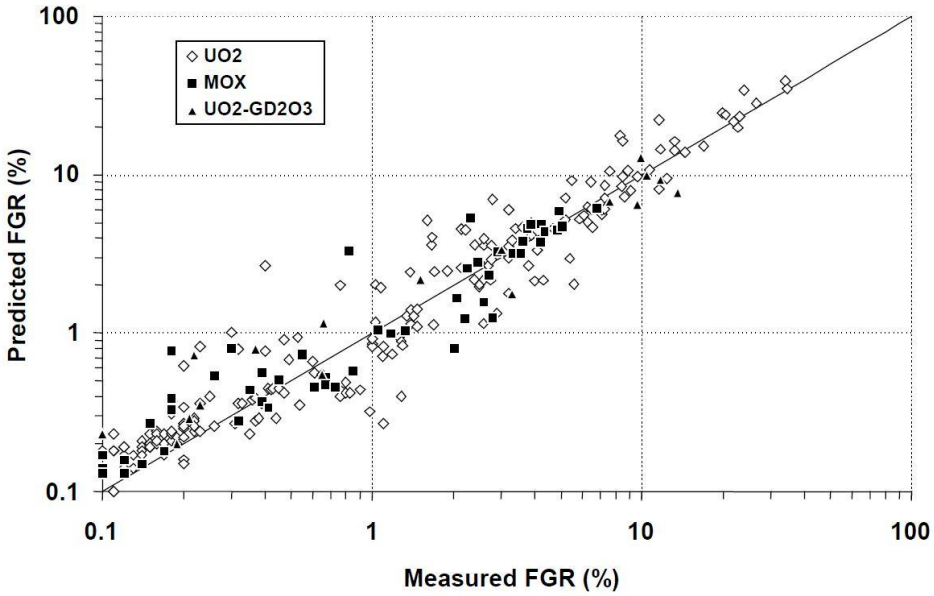

Figure 6: Validation of the FRAMATOM FGR steady-state model against experimental data (from [39]).

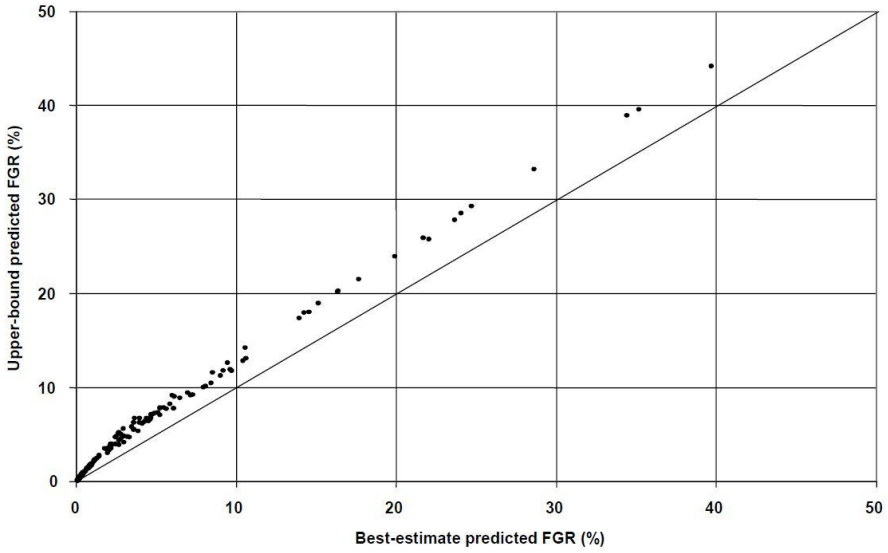

Figure 7: Validation of the FRAMATOM FGR steady-state model – upper-bound uncertainty limit of the model at the 95% level (from [39]).

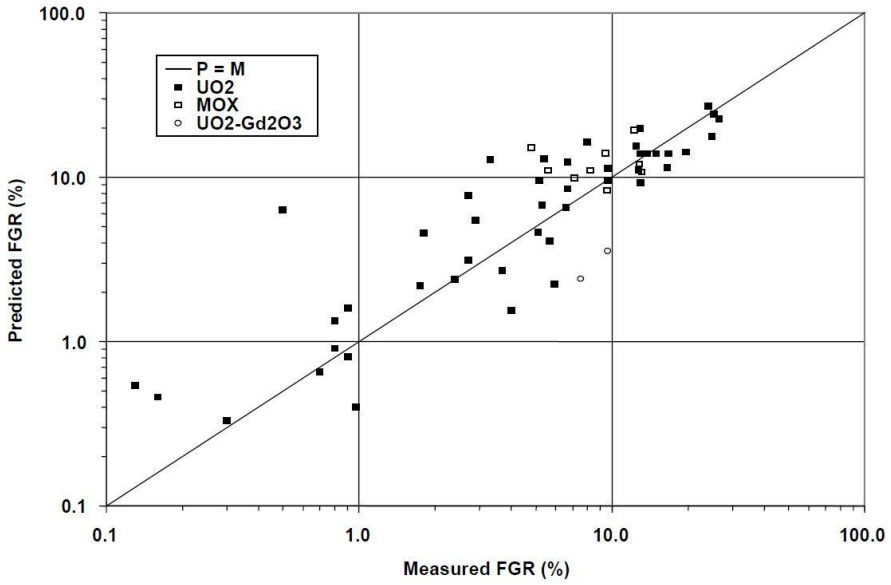

Figure 8: Validation of the FRAMATOM FGR transient model against experimental data (from [39]).

FASTGRASS [40] is an example of a fuel performance code with the capability to simulate the release of not only fission gases, but also other fission products: volatiles (I, Te, Cs) and alkaline earth elements (Sr and Ba).

Developments in Fuel Performance Assessment Codes

As of today simplified correlations are still used in many fuel assessment codes to represent complex material behaviour [34]. However, recent efforts in computational algorithms, computer hardware and parallel computing are progressively replacing these simplified approaches by more realistic (and computationally expensive) coupled, pure physics-based models. There are currently several codes under development that take the advantage of the recent advances in the field, these include: AMP (ORNL, USA [44]), Bison (INL, USA [41]), BACO (CNEA, Argentina [42]) and PLEIADES/ALCYONE (CEA-EDF, France [43]). These codes rely on state-of-the-art solution methods to simulate in 2- and 3-D the behaviour the fuel nominal operation and expected transients [44].

Recently, simulations based on statistical physics concepts, such as the phase-field theory, have been successfully applied to modelling the 3-D structure evolution of irradiated fuels. Processes modelled using the phase-field approach include: solidification, grain growth, martensitic transition, precipitation, ferroelectric/ferromagnetic transition, dislocation dynamics, deformation twin, sintering, and microstructure evolution in irradiated materials [45, 46]. Li and co-workers [47] developed a phase-field model to simulate intra-granular fission gas bubble evolution in a single crystal during post-irradiation thermal annealing. In a follow-up work, Li and co-workers [48] presented a phase-field model of intra-granular gas atom and bubble behaviour extended to incorporate thermodynamic and kinetic properties at

grain boundaries, to simulate fission gas bubble growth kinetics in polycrystalline UO_2 fuels. The model takes into account gas atom and vacancy diffusion, vacancy trapping and emission at defects, gas atom absorption and resolution at gas bubbles, internal pressure in gas bubbles, elastic interaction between defects and gas bubbles, and the difference of thermodynamic and kinetic properties in matrix and grain boundaries. The enhanced phase-field model was used to simulate gas atom segregation at grain boundaries and the effect of interfacial energy and gas mobility on gas bubble morphology and growth kinetics in a bi-crystal UO_2 during post-irradiation thermal annealing. The application of phase-field models has the potential to reduce many of the simplifying assumptions used calculate the gas deposition rate at grain boundaries [48].

Research is being conducted attempting to incorporate multi-scale atomic-level simulations into fuel performance assessment codes [49, 50, 51]. Vega [50] presented a proof-of-principle for the inclusion of atomic-level (molecular dynamics) simulations of thermal expansion and thermal conductivity of UO_2 into the FRAPCON performance code. Vega found that modifying FRAPCON in such a way as to accept input from the atomistic simulations is possible for a number of thermo-mechanical properties. Harp [51] used multi-scale atomistic-level simulations to model macroscopic-level diffusion of fission gases in the UO_2 TRISO fuel (for High Temperature Gas Reactor) and explored the link between the experimentally measured fission gas release and the results of the simulations. The modelling involved molecular dynamics simulation of thermal properties of the UO_2 and its validation against available experimental data. Self-diffusion of O and U in UO_2 was explored using molecular dynamics simulations and included an evaluation of the influence of radiation damage and crystal grain boundaries. The results were used in a kinetic Monte Carlo simulation of Xe and Kr migration in UO_2. This approach allows the calculation of lattice diffusion coefficients for noble gases at different temperatures in the fuel from first principles [51].

3. APPLICATION TO THE FIRST NUCLIDES PROJECT

Most FGR models focus on the release under reactor operation conditions. Nevertheless, such models can provide valuable information regarding the future release of radioactivity from spent fuel assemblies under repository conditions.

For one, the mechanistic FGR models presented here are capable of accurately predicting the amount of fission gases captured within the inner-rod voids, which would constitute a part of the IRF. Secondly, the release of some fission products upon contact of the fuel with water has been demonstrated experimentally to correlate with that of FGR. Therefore, information on FGR obtained from the mechanistic models can be combined with empirical correlations to make predictions of FGR under repository conditions. In addition, mechanistic FGR models can forecast numerous properties of the fuel and cladding (e.g. cracking, porosity,

grain size distribution, etc.), which can be used as boundary conditions for other models, specifically focusing on FGR under repository conditions.

Mechanistic models exist that are capable of quantifying the inventory of fission gases and other fission products segregated at UO_2 grain boundaries, however to what extent this inventory is susceptible to release remain unresolved [1]. As explained earlier, the IRF is typically considered to comprise (1) the release of fission products from the fuel/cladding gap and (2) the release of material segregated at fuel grain boundaries [1]. However, the extent to which preferential release of fission products from grain boundaries occurs is currently unresolved. Leaching experiments do not provide an unambiguous indication of when the transition from rapid release to dissolution-based release takes place, because simultaneous release of fission products present at grain boundaries may also occur during the experiment. Although Johnson and co-workers indicate that experimental data do not support the hypothesis that fission products occupying grain boundaries are easily leached [1], they emphasize that methods for differentiating between fission products released from the inner-rod gap and grain boundaries should be further explored. This is important for the application of FGR models to estimating IRF from spent fuel assemblies under repository conditions.

The release of non-volatile fission products from the fuel grain surfaces requires wetting the surface with water in order to dissolve them and provide the possibility for aqueous diffusion out of the fuel. Under repository conditions this could happen after the engineered barriers are corroded through and the host-rock pore water contacts the fuel. The rate of release of these fission products would therefore be related to the rate of wetting of the fuel grain surfaces. Numerical modelling of spent fuel pellet saturation with water has been attempted as part of the FIRST NUCLIDES project [52]. This work is currently under development and is hoped to provide estimates on the time required for saturation with water spent fuel pellets under different conditions (e.g. experimental leaching of fuel materials in the laboratory and at the repository depth). Ultimately, the water saturation model can be linked with radionuclide transport and relevant retention processes to help better understand the rapid release phase of fission products from UO_2 spent fuel.

4. SUMMARY AND PROSPECTS

The instant release of some radionuclides under repository conditions has important implications for the safety assessment of the spent fuel disposal facility. Therefore, IRF has been studied through a combination of experimental and modelling efforts. As the release of fission gases from fuel into the inner-rod void during in-reactor irradiation is important for safe and economical operation of the reactor, FGR models have been developed to help predict the release. These models can be categorised into two general groups: (1) empirical and semi-empirical correlations, which are based on experimental post-irradiation examination of the spent fuel assemblies, and (2) mechanistic, physics-based models that

simulate the relevant processes of fission gas generation, transport and release into the cladding gap. The former are well established, but suffer from a number of limitations, including: prohibitive economical costs of performing experiments on highly-radioactive materials which results in scarcity of such data, limited applicability of the derived models to a narrow range of calibration conditions, and limited potential of these models to contribute to a more fundamental understanding of the processes involved in FGR. These limitations are however relieved with the help of mechanistic FGR models, many of which are incorporated into fuel performance assessment codes. Many of these models have been validated against experimental data and are known to be capable of accurately predicting FGR during irradiation. The advantage of mechanistic models lies in their flexibility to deal with fuel used under different reactor operation conditions. This stems primarily from the process-oriented nature of such models, which promotes better system understanding and enables extrapolation to a variety of conditions. Mechanistic FGR models are an active field of development and are characterised by varying degrees of sophistication. New generation performance assessment codes are now being introduced which are increasing based on highly coupled multi-physics models, and draw on the power of massively-parallelised computing. Currently, efforts are being made to develop advanced models of fuel behaviour based on the theories of statistical physics that employ the concept of multi-scale modelling. Such models show the potential to succeed the currently used models in the future.

The currently available models for FGR under irradiation conditions can provide information useful for predicting the release of fission products under repository conditions. By combining information from the empirical and mechanistic models it is possible to make a prediction of the IRF from spent fuel under repository conditions. Nonetheless, there exist a number of unresolved issues (such as the availability of grain boundary segregated fission products for rapid release), which need to be further studied and addressed in order for the uncertainty associated with the use of these models to be reduced.

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