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Modelling and assessment of inert gas behaviour in UO_2 nuclear fuel for transient analysis

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List of acronyms

AGR	Advanced Gas Reactor
ASTM	American Society for Testing and Materials
BC	boundary condition
DOE	Department of Energy
EFPD	equivalent fuel power day
EPMA	electron probe microanalysis
FD	finite difference
FGB	fission gas behaviour
FGR	fission gas release
FPC	fuel performance code
FUMAC	Fuel Modelling in Accident Conditions
HBS	high burnup structure
IAEA	International Atomic Energy Agency
IC	initial condition
IFPE	International Fuel Performance Experiments
IGB	inert gas behaviour
INERI	International Nuclear Energy Research Initiative
INL	Idaho National Laboratory
INSPYRE	Investigations Supporting MOX Fuel Licensing for ESNII Prototype Reactors
KJMA	Kolmogorov-Johnson-Mehl-Avrami
LOCA	loss-off-coolant accident
LWR	light water reactor
NEA	Nuclear Energy Agency
ODE	ordinary differential equation
OECD	Organisation for Economic Co-operation and Development
PCMI	pellet-cladding mechanical interaction
PDE	partial differential equation
PIE	post irradiation examination
POLIMI	Politecnico di Milano
RIA	reactivity-initiated accident
SEM	scanning electron microscopy
TEM	transmission electron microscopy

List of symbols

at: *atom*

bub: *bubble*

ff: *fission fragment*

fiss: *fission*

a	(m)	grain radius (assumed as spherical)
a_{∞}	(m)	grain radius, asymptotic value
B	(K)	parameter of μ
B	(m)	$(3\Omega/4\pi)^{1/3}$
c_1	(at m ⁻³)	concentration of single gas atoms
c_2	(at m ⁻³)	concentration of dimers
c_n	(at m ⁻³)	concentration of clusters of n atoms
c_{tot}	(at m ⁻³)	total intra-granular gas concentration, $c_1 + m$
bu	(GWd tU ⁻¹)	burnup
bu_{eff}	(GWd tU ⁻¹)	effective burnup
bu_{τ}	(GWd tU ⁻¹)	parameter of u
c_{tot}	(at m ⁻³) or (wt.%)	intra-granular gas concentration
D	(m ² s ⁻¹)	diffusion coefficient
$\mathbf{D}$	(s ⁻¹)	diffusion operator
dt	(s)	time step
$\mathbf{E}$	(s ⁻¹)	exchange operator (among gas concentrations)
E_{RIA}	(J (gUO ₂) ⁻¹)	injected specific energy during a RIA-transient
F	(fiss m ⁻³ s ⁻¹)	fission rate
F_c	(/)	fractional coverage
F_{sat}	(/)	saturation fractional coverage
H	(/)	Heaviside function
h	(-)	micro-crack healing parameter
k_T	(/)	sensitivity coefficient for μ
k_B	(/)	sensitivity coefficient for μ
k_Q	(/)	sensitivity coefficient for μ
M	(#)	number of modes, PolyPole
m	(at m ⁻³)	concentration of gas atoms in intra-granular bubbles
N_{ig}	(bub m ⁻³)	intra-granular bubble density
N_{gb}	(bub m ⁻²)	grain-boundary bubble density
$\mathbf{P}_n$	(-)	correction matrix, PolyPole
P_{RIA}	(W g ⁻¹)	specific power of a RIA pulse
p_j, q_j	(s ⁻¹)	polynomial coefficients
Q	(-)	parameter of μ
q	(/)	validation metric
R_{at}	(m)	radius of a gas atom in the lattice
$\mathbf{S}$	(at m ⁻³ s ⁻¹)	gas production operator

s	(-)	parameter of μ
R	(m)	intra-granular bubble radius
R_{ff}	(m)	radius of a fission spike
T	(K) or ($^{\circ}\text{C}$)	temperature
T_0	(K)	initial temperature
T_{end}	(K)	final temperature
T_{max}	(K)	maximum temperature
T_{infl}	(K)	inflection temperature, parameter of μ
T_{th}	(K)	temperature threshold
t_{end}	(s)	end time
u	(at m^{-3})	vector of gas concentrations, [c_1 m]
y	(at fiss^{-1})	fission yield
z_n	(at m^{-3})	time coefficients, PolyPole
α	(s^{-1})	re-solution rate
β	(s^{-1})	trapping rate
$\Delta V/V$	(/)	swelling
η	(bub ff^{-1})	nucleated bubbles per fission fragment
μ	(/)	micro-cracking parameter
μ_{ff}	(m)	length of a fission spike
ν	(bub $\text{m}^{-3} \text{s}^{-1}$)	nucleation rate
ρ	(/)	non-dimensional radius
τ_{RIA}	(ms)	FWHM of a RIA power pulse
χ	(/)	intra-granular fission gas release
ψ	(/)	spatial modes, PolyPole
Ω	(m^3)	atomic gas volume in bubbles

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1. Introduction

This thesis work concerns the modelling and the assessment of inert gas behaviour (IGB) in UO_2 nuclear fuel, with application to integral fuel performance codes (FPC) and emphasis on transient behaviour. The proper modelling of IGB during transients is fundamental for the performance (and safety) analysis of nuclear fuel rods, since IGB affects the thermo-mechanical condition of the fuel rods and can represent a limiting life factor for their permanence in reactor [1].

For the sake of effectively and efficiently modelling IGB at the engineering scale, i.e., at the scale of FPCs (pellet and fuel rod level), it is fundamental to find a compromise between physical description of meso-scale processes concerning IGB (occurring at the scale of fuel grains) and the computational effort they impose to a fuel performance code. I adopted a modelling strategy mixed between physics-based and semi-empirical, always aiming at obtaining a conveniently simple description of IGB. Similar approaches to IGB modelling have been proven effective in the past [2], [3]. The widely adopted schematization of IGB in UO_2 defines several subsequent stages in the evolution of IGB. Namely, after inert gas (IG) are produced (xenon and krypton by fission, and helium mainly from alpha decay) within the matrix of nuclear fuel grains, the evolution of IGB is conceptually divided into two main stages [1], [4]. First, the formation of a population of *intra-granular* bubbles which exchange gas with the matrix through the trapping and the resolution mechanisms, along with the net diffusion of single gas atoms from the matrix towards the grain boundaries. Second, the formation of a population of *inter-granular* bubbles at the grain boundaries, which coalescing may form an interconnected path and allow inert gas venting to the fuel exterior (i.e., FGR, fission gas release). Grain-boundary gas venting and FGR may also occur through the alternative mechanism of grain-boundary separation due to micro-cracking¹. The volume occupied by both intra- and inter-granular bubbles is responsible of the *gaseous swelling*² of

¹ FGR can occur also via other mechanisms (e.g., *a-thermal* release). Nevertheless, in particular during transients, these other mechanisms are responsible of a limited amount of FGR compared to those herein considered.

² Gaseous swelling refers to the isotropic increment of nuclear fuel volume caused by the precipitation of gas into bubbles. Other swelling mechanism can occur (e.g., *solid swelling* is caused by the accumulation of solid fission products within the fuel lattice).

the fuel. In addition, in UO_2 fuel, the external region of the fuel (*rim zone*) can experience severe irradiation damage at high burnup (50-100 GWd/tU), resulting in a radical change of the polycrystalline structure [5]. This high burnup structure (HBS) presents smaller grains than the rest of the fuel, together with characteristic large inter-granular pores collecting the gas, and shows a peculiar IGB. Given the large amount of gas generated in this part of the fuel (due to the higher local fission density), it is of particular importance to model in FPCs the formation of the HBS and the peculiar IGB of this region.

The combination of these IGB mechanisms is responsible of critical thermo-mechanical feedbacks on the fuel rod in transient conditions as well as in design basis accidents. During loss-of-coolant accidents (LOCA, time-scale of minutes/seconds, indicatively [6]), gas release from the fuel towards the rod free volume may cause an increase in the thermal resistance of the gap, leading to an increase in the fuel temperature, positively feed-backing more FGR (*thermal feedback*)³. During reactivity-initiated accidents (RIA, time-scale of milliseconds for the initial power pulse [7]), a significant gas release is observed, together with a sudden increase in the swelling coming from the contribution of the HBS pores.

In view of its crucial importance, IGB modelling is the main topic in several international projects. In particular, this thesis work is grafted in the FUMAC Project [8], coordinated by the International Atomic Energy Agency (IAEA), the INSPYRE and COMBATFUEL H2020 Projects [9], supported by the European Commission, and the SciDAC Project on fission gas behaviour of the US DOE.

The model-development strategy followed in this thesis work is made of several steps (Fig. 1). First, the critical IGB-related phenomena were identified in the framework of the above mentioned international projects. These phenomena were selected based on their impact on fuel performance analysis (from an engineering point of view, with a special focus on transient conditions). The considered phenomena are:

- The evolution of the intra-granular bubble population, which can be responsible of up to half of the gaseous swelling during transients and

³ Several irradiation experiments showed that the impact of the thermal feedback depends on the burnup, i.e., whether there is a sufficient amount of gas to produce a serious effect.

strongly interacts with the diffusion of single gas atoms towards the grain boundaries [8], [10], [11] (Chapter 2).

- The burst release of gas occurring during temperature transients and ascribed to the micro-cracking of grain boundaries [8], [12] (Chapter 3).
- The formation of the high burnup structure with a combined description of the grain recrystallization and of the intra-granular gas depletion [13] (Chapter 4).

In order to model these three phenomena (intra-granular bubble evolution, burst release, and HBS formation), I developed three sub-modules, independent but connected. All these sub-modules have been implemented, verified and validated in a 0D stand-alone computer code (referred to as SCIENTIX⁴). I also developed, implemented and verified dedicated numerical algorithms (i.e., PolyPole, for the treatment of PDEs in time-varying conditions). After this fundamental step, I implemented the developed models and algorithms in fuel performance codes. In particular, I used TRANSURANUS [14], [15] (developed by JRC-Karlsruhe)⁵ and BISON [16]–[18] (developed by Idaho National Laboratory)⁶, in the frame

⁴ SCIENTIX is a 0D stand-alone computer code developed as part of this thesis work. It models IGB at the scale of a fuel grain, represented as a point (i.e., 0D). Since it is 0D, no spatial discretization is required, and all variables are treated as average in space, but evolving in time. All the models developed in this thesis work (and other correlations/models available in the open literature) are implemented in SCIENTIX for testing, verification and validation purposes. SCIENTIX is designed for coupling with existing FPCs, effectively acting as an IGB module in the frame of a code. The open source release of the code is planned in the near future.

⁵ TRANSURANUS [15] is an integral fuel performance approximating the fuel rod behaviour with an axisymmetric, axially-stacked, one-dimensional radial representation (often referred to as 1.5D). The code can be employed for both steady-state and transient analyses, and incorporates models accounting for the different and interrelated phenomena occurring in the fuel rod [14], [15]. In TRANSURANUS, the fuel rod is discretised in axial slices, or sections, and in radial coarse zones for the evaluation of the material properties. The coarse zones are in turn divided into finer zones to perform the numerical integrations needed for the thermo-mechanical analysis.

⁶ BISON [16] is a finite-element based nuclear fuel performance code. The code can be applied to steady-state and transient fuel simulations, and allows for 1D spherically symmetric, 2D axisymmetric or generalized plane strain, or 3D geometry representations. BISON's governing relations consist of fully-coupled partial differential equations for species conservation and thermo-mechanics and include

of a POLIMI-INL-JRC international collaboration (recently formalized in the framework of an INERI Project [19]).

The models developed and assessed in this thesis overcome critical limitations of state-of-the-art models currently used in FPCs. First, the new models are physics-based (even if conserving a certain degree of empiricism) instead of being purely correlation-based. They are thus more reliable outside the validation range of correlations and more suited for transient analysis. Second, the new models consider critical IGB phenomena that are currently neglected in FPCs (e.g., evolution of intra-granular swelling during transients), despite their importance for the overall fuel performance analysis. Third, the new models are applicable with minor modifications to different fuel materials, since the main physical phenomena driving IGB are common to many fuel materials (in this thesis, I focused on UO_2). Fourth, the new models are paired with dedicated numerical algorithms (developed as part of this work), which ensure their effective applicability in FPCs. These algorithms, i.e., PolyPole-1 and PolyPole-2, are described in Chapter 5. The structure of this thesis follows this division, with each Chapter dedicated to one of the developed models/algorithms. The detailed description of the state of the art of the modelling of all the phenomena considered in fuel current FPCs is discussed in the introduction of each Chapter.

A consequence of the introduction of a meso-scale physics-based model in the framework of an engineering-scale FPCs is that the accuracy of model predictions depends not only on the intrinsic characteristics of the model itself, but also on calculated variables of the code (e.g., local temperature, local burnup) that are input for FGB calculations. Uncertainties exist that ensue from material data and physics-based models, but also from the basic structure of the code with the relative simplifications in representing the actual domain (e.g., dimensionality) and solving the thermo-mechanics equations (e.g., numerical technique) [20]. Recently, international benchmarks of fuel performance codes [21], [22] pointed out the difficulties in predicting FGR and fuel dimensional changes due to swelling, and demonstrated that, given identical conditions, different codes can provide a broad range of outputs. It follows that, when assessing the predictions of a

constitutive laws for nonlinear kinematics and nonlinear material behaviour. Models are available to describe the various interacting processes involved in fuel rod behaviour. [16]–[18].

specific meso-scale physics-based model incorporated in a fuel performance code, the information inevitably contains an unknown bias associated with uncertainties in the global thermo-mechanical analysis that interacts with the model. In other words, it is difficult to evaluate the source of the deviation between the code results and the experimental data, or of an improvement in predictions obtained with a new model. Improvements may indeed arise from a better representation of the specific process, but also from compensating errors between the physical model and code's calculated global variables that are input for the model.

For these reasons, the validation strategy pursued in this thesis work is twofold (Fig. 1). When possible (i.e., fully physics-based models and availability of local measurements arising from separate effect experiments) the validation is performed with the SCIANTIX code, by simulation of local behaviour of the fuel. In this way, the focus is on the specific output of a specific model, avoiding the uncertainty brought about by its integration in a fuel performance code. When this approach is not feasible (e.g., for semi-empirical models derived from separate effects experimental data), a way to increase the confidence in the assessment of a model in fuel rod analysis is applying the same model in multiple fuel performance codes (i.e., both BISON and TRANSURANUS). Comparisons between predictions from the model applied in different codes would isolate, to some degree, the effect of the specific model from compensations related to the uncertainties in the global fuel rod analysis. This twofold validation strategy adds value to the assessment of the models and confidence in conclusions drawn about the predictive capability of each specific meso-scale model. Each chapter details which validation strategy is pursued for each of the models developed. It is worth pointing out that, whenever possible, experimental data concerning transients have been used for the validation of the herein developed models (and as well for the verification of the numerical algorithms). Moreover, a sensitivity analysis to the model parameters is performed, in order to evaluate their impact on specific figures of merit (e.g., the swelling).

To summarize, the goal of this thesis is twofold:

- The development of three physics-based meso-scale models describing specific aspects of IGB in UO_2 . The models are designed and developed to work together as an overall consistent model describing the evolution of gas at the scale of fuel grains and to be suitable for transient analysis.
- The introduction of the developed, tested, verified and validated IGB models in BISON and TRANSURANUS, in compliance with the peculiar characteristics of these codes. BISON and TRANSURANUS including

the new models have been validated against integral irradiation experiments data, when this validation strategy is required.

Moreover, the development of the 0D stand-alone SCIANTIX code is already a result of considerable importance, since it represents a POLIMI-owned software that can be included as an IGB module in existing FPCs, according to a multi-physics/multi-scale coupling strategy.

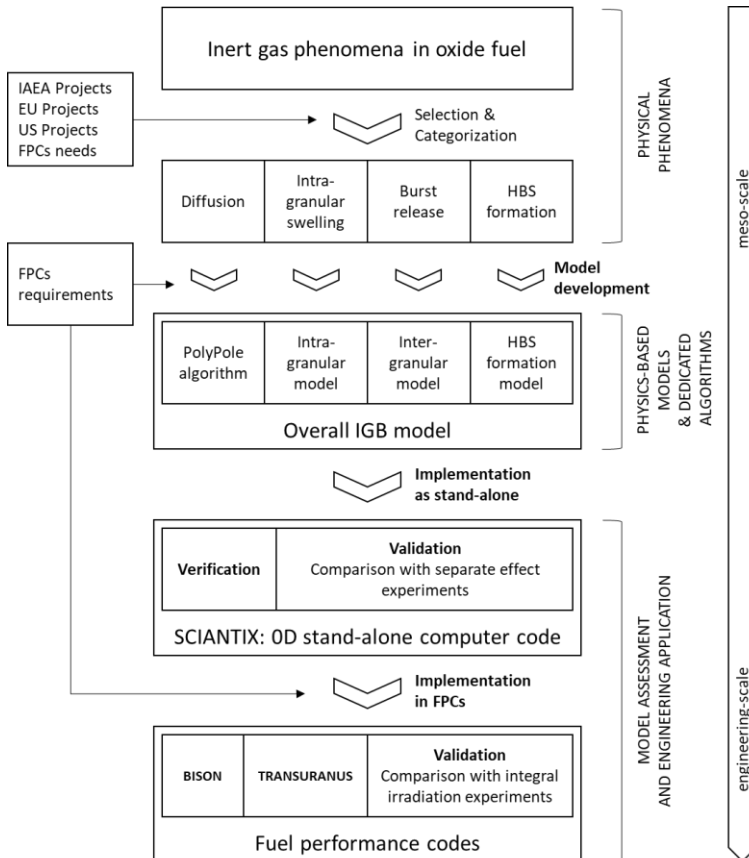


Figure 1. Concept map summarizing the organization of the thesis work.

2. Intra-granular inert gas behaviour

Abstract

In this chapter, I present a model describing the evolution of intra-granular inert gas bubbles in terms of bubble number density and average bubble size. The model considers the fundamental processes of single gas atom diffusion, gas bubble nucleation, bubble re-resolution, and gas atom trapping at bubbles. I derived the model from a detailed cluster dynamics formulation, under the experimentally corroborated assumption that free vacancies are of secondary importance compared to gas atoms in particular conditions. By rigorous simplification, I reduced the master equations of cluster dynamics to three differential equations, being efficiently applicable in engineering fuel performance codes while retaining a physical basis. After implementation in the SCIENTIX code, I validated the model against separate effect experimental data, both in terms of bubble number density and average bubble radius finding an improved agreement compared to other models currently available in fuel performance codes. I performed an uncertainty analysis by propagating the uncertainties in the parameters to the model results, and a sensitivity analysis showing the importance of each model parameter as well.

The content of this Chapter is based on:

D. Pizzocri, G. Pastore, T. Barani, A. Magni, L. Luzzi, P. Van Uffelen, S.A. Pitts, A. Alfonsi, J.D. Hales, 2018, *A model describing intra-granular fission gas behaviour in oxide fuel for advanced engineering tools*. Journal of Nuclear Materials, 502, 323-330.

2.1 Introduction

As discussed in the Introduction of this thesis, given the fundamental role played by fission gas swelling and release in the thermo-mechanical behaviour of nuclear fuel rods during irradiation [1], [23], models of fission gas behaviour (FGB) must be included in fuel performance codes. In this first Chapter, I address the modelling of intra-granular gas behaviour, by developing a model for the evolution of the population of intra-granular bubbles. Bubbles nucleate in fuel grains due to the low solubility of the fission gas and evolve through trapping of dissolved gas atoms and the counteracting process of irradiation-induced bubble re-resolution. Gas diffusion to grain boundaries is mainly due to dissolved atoms, with nucleation, trapping and re-resolution determining the coupling between bubble evolution and gas atom diffusion [1], [2], [4], [23]–[25]. Intra-granular gas diffusion to grain boundaries provides the source term for the inter-granular processes, ultimately leading to grain-boundary gaseous swelling and fission gas release [2], [23], [26], [27]. Intra-granular bubbles not only affect the gas diffusion rate to grain boundaries, but also contribute to fuel swelling, although intra-granular gaseous swelling becomes significant compared to grain-boundary swelling only at high burnup or during transients to high temperatures [10], [28]. Therefore, models are needed to describe the evolution of the intra-granular bubble population. In particular, in order to estimate the intra-granular swelling and the bubble effects in intra-granular diffusion, the bubble number density and average bubble size need to be calculated [14], [18], [24], [29].

The traditional approach adopted in engineering-scale fuel performance codes describes the evolution of intra-granular fission gas bubbles by relying on empirical correlations. These correlations estimate the mean bubble size and number density as a function of macroscopic parameters, (e.g., the local temperature [2], [30]). Indeed, the inherent simplicity of the correlation-based approach entails the advantage of a small computational burden. The application of these correlations is limited to their interpolation range, hence to conditions for which experimental data are available. Furthermore, these empirical correlations are material-specific, while physically based models can be applied to different materials by adapting the fundamental parameters. Finally, fundamental parameters can be derived using lower-length scale modelling, then, physically based engineering models have the additional advantage of allowing for scale bridging in multiscale modelling approaches [31].

A complete and accurate description of the bubble population evolution can be accomplished by employing cluster dynamics approaches (e.g., [32]–[35]). Cluster dynamics models calculate the entire bubble (clusters of atoms and vacancies) size distribution and the distribution evolution over time by solving coupled rate equations for the number densities of clusters of different sizes. Clearly, these advanced modelling approaches are computationally expensive and cannot be used directly for fuel performance code applications [24]. On the other hand, advanced modelling can be used to inform engineering-scale models with improved parameters.

Representing the bubble size distribution as only the mean size and the total number density (single-size models) allows for a compromise between simplicity and accuracy, surpassing the inherent physical limitations of the correlation-based approaches and, at the same time, avoiding the high computational cost of cluster dynamics [24].

One of the most recent and significant work on single-size models for intragranular bubble evolution is the paper of Olander and Wongsawaeng [24]. They reviewed the mechanisms of bubble nucleation, growth and re-solution and derived single-size formulations by applying in the first place the assumption of all bubbles having the same size. While this approach captures the main physical mechanisms and surely represent an improvement compared to correlation-based approaches, it has also important limitations [24]. For instance, it implies that bubbles nucleated at a given time grow instantaneously to the mean size and that bubble destruction only occurs at the mean size, which introduces a bias in the bubble size calculation. Capturing such effects requires consideration of a non-uniform bubble size distribution. The model presented in this Chapter aims at obtaining an improvement of this state of the art for single-size models suitable for application in fuel performance codes. I start from a detailed cluster dynamics physical representation, then I derive a formulation for the evolution of the bubble mean size and number density that preserves fundamental size-distribution effects.

2.2 Model description

2.2.1 Physical processes

Three fundamental processes control the evolution of small intra-granular bubbles in nuclear fuel, i.e., bubble nucleation, gas atom trapping at the bubbles, and irradiation-induced gas atom re- solution from bubbles back into the lattice. Nucleation can be considered as driven by either diffusion [36], [37] or by irradiation [2], while re- solution is (primarily) irradiation-driven [30], [38] and trapping is (mainly) diffusion-driven [39]. For an extensive overview of the modelling of these processes, the work of Olander and Wongsawaeng is recommended [24].

I herein adopt here the terminology of Olander and Wongsawaeng [24] for the different possible mechanisms of nucleation and re- solution, i.e., *heterogeneous* and *homogeneous*. *Heterogeneous* nucleation and re- resolution refer to the nuclei of the new bubbles being created as a direct consequence of the interaction of fission fragments with the lattice and the bubbles being destructed en-bloc by passing fission fragments, respectively. The *homogeneous* mechanisms refer to bubbles being nucleated by diffusion-driven interactions of dissolved gas atoms and re- resolution occurring gradually by ejection of individual atoms. There are several experimental results and theoretical considerations on the *heterogeneous/homogeneous* nature of nucleation, re- resolution and trapping in oxide fuel. Each of these processes is modelled by introducing a specific rate of event occurrence. Here, I denote the nucleation rate as ν , the re- resolution rate as α , and the trapping rate as β .

Focusing first on nucleation, several TEM observations show intra-granular bubbles lying in straight lines [24], [28], [40]. Turnbull's early conclusion was that bubbles were nucleated *heterogeneously* along the track of fission fragments, with nucleation being driven directly by irradiation [41]. Nevertheless, several further observations did not show aligned bubbles, hindering the generality of this conclusion [24], [42]. Additionally, the typical thickness of TEM samples is lower than the range of fission fragments, therefore only portions of actual fission tracks are observed [24]. The other nucleation mechanism, which competes with the *heterogeneous* mechanism, is the *homogeneous*, diffusion-controlled production of dimers (diatomic fission gas clusters) [36], [37]. Presumably, both these mechanisms are active and competing in the fuel grains, but major experimental and theoretical works agree on the dominance of the *heterogeneous* mechanism [41]–[43]. I

thus assume that the *heterogeneous* nucleation mechanism is dominant in oxide fuels. The expression for the nucleation rate is thus

$$\nu = 2\eta F \quad (1)$$

where ν (nucleated bubbles $\text{m}^{-3} \text{s}^{-1}$) is the nucleation rate, η (bubbles per fission fragment) is a number generally considered in the range 5–25 based on experimental observations [41], [42], F is the fission rate ($\text{fiss} \text{m}^{-3} \text{s}^{-1}$), and the factor of 2 accounts for the number of fission fragments per fission. The use of this expression for nucleation rate underlies the assumption that the formation of bubbles occurs in the wake of fission fragments as gas atoms rapidly fill void clusters. Clearly, this is a simplified representation of the nucleation mechanism, consistent with the overall modelling approach.

As for re-resolution, I also assume a dominating *heterogeneous* mechanism. The re-resolution rate is expressed as a function proportional to the fission rate [2], [30], [41]

$$\alpha = 2\pi\mu_{\text{ff}}(R + R_{\text{ff}})^2 F \quad (2)$$

where α (re-solved bubbles $\text{bubble}^{-1} \text{s}^{-1}$) is the re-resolution rate, μ_{ff} (m) and R_{ff} (m) are the average length and radius of a fission spike (the subscript ff corresponds to fission fragment), respectively, and R (m) is the radius of the gas bubble. The assumption of *heterogeneous* re-resolution of small intra-granular bubbles is supported by the molecular dynamics work performed by Govers et al., [44]. They investigated both the *homogeneous* re-resolution mechanism (as originally described by Nelson [45]) and the *heterogeneous* one (originally proposed by Turnbull [41] and in agreement with other contemporary work [46], [47]). Their analysis demonstrated that the *heterogeneous* re-resolution mechanism is more effective in re-solving fission gas bubbles compared to the *homogeneous* mechanism. However, they showed that for a wide variety of bubble radii and gas densities, no clear dependence of the re-resolution rate on the bubble size emerges [44]. Other combined Monte Carlo and molecular dynamics approaches indicated the predominance of the *homogeneous* re-resolution mechanism [48]–[50]. Further molecular dynamics work could shed further light on the re-resolution mechanism details and enable a more accurate consideration of the re-resolution rate, which in perspective can be incorporated in the present model by replacing Eq. 2.

As for the trapping rate, following the formulation derived by Ham [39]

$$\beta = 4\pi D(R + R_{\text{at}})c_1 \quad (3)$$

where β (trapped atoms bubble⁻¹ s⁻¹) is the trapping rate, D (m² s⁻¹) is the diffusion coefficient of single gas atoms, R (m) is the radius of a bubble, R_{at} (m) is the radius of a single gas atom in the lattice, and c_1 (atoms m⁻³) is the concentration of single gas atoms. These three rate equations (Eqs. 1, 2, 3) are used in the present intra-granular gas behaviour model for computing the bubble nucleation, re-resolution and gas atom trapping rates.

2.2.2 Model derivation

The herein derived model results from the combination of the intra-granular bubble population evolution, through the physical processes described in Section 2.2.1, and the gas diffusion towards grain boundaries.

Following the assumptions of Clement and Wood [51] and Fell and Murphy [52], I limit the scope of the analysis to clusters made of only gas atoms and neglect vacancy absorption at the clusters. This assumption is in line with the observation that because of the small size of intra-granular bubbles, the surface-tension stress is large enough to keep the gas density near that of solid xenon [24]. Several experimental observations of over-pressurized intra-granular bubbles corroborate this assumption [28], [30], [41], [42], [53]. The assumption of single-species (i.e., atoms-only) cluster dynamics implies that the gas atoms considered completely occupy available vacancies, with no free vacancies available in the lattice. Clearly, this is a hypothesis, and not meant to be fully representative of the underlying physics that would require the use of an equation of state. Confirming this modelling assumption by performing a two species (gas atoms and vacancies) cluster dynamics analysis is of interest in perspective.

In the cluster dynamics model, a cluster is defined by its size (the number of gas atoms contained within the cluster) and the cluster concentration for each species is governed by a partial differential equation. The master equations of cluster dynamics consider that (a) the processes of cluster evolution occur at the rates described in Section 2.2.1, and (b) the nucleation and re-resolution processes are *heterogeneous*. For the sake of clarity, Fig. 2 depicts the processes of cluster evolution for the first three cluster sizes (single atoms, dimers and trimers). The master equations of the cluster dynamics model are

$$\begin{aligned}
\frac{\partial c_1}{\partial t} &= +yF + D\nabla^2 c_1 - 2\nu - \sum_{n=2}^{\infty} \beta_n c_n + \sum_{n=2}^{\infty} n\alpha_n c_n \\
\frac{\partial c_2}{\partial t} &= +\nu - (\beta_2 + \alpha_2)c_2 \\
&\dots \\
\frac{\partial c_n}{\partial t} &= +\beta_{n-1}c_{n-1} - (\beta_n + \alpha_n)c_n
\end{aligned} \tag{4}$$

where c_1 (atoms m^{-3}) is the concentration of single gas atoms, and $c_2, c_3, \dots, c_n$ (clusters m^{-3}) are the number densities of clusters of atomic size 2, 3, ..., n , respectively. Other than the fission yield y (atoms fiss^{-1}), the other quantities in Eq. 4 have been defined in Section 2.2.1.

I include in Eqs. 4 the diffusion term only for single gas atoms, assuming that larger clusters (bubbles) are immobile [38], [42]. On the same basis, the interactions between clusters (bubble coalescence) are neglected. The assumption if immobile bubbles may be not valid at high temperatures above 1500-1800°C [28], [30], [54], [55]. The other assumption in Eqs. 4 is that bubbles nucleate as dimers. If nucleation is heterogeneous, it is possible that the size of the nucleated bubbles depends upon the concentration of dissolved gas [24], hence this is an arbitrary assumption and it is of interest to further improve this modelling aspect as more data/information become available.

The simultaneous solution of Eqs. 4 provides the complete cluster size distribution. Nevertheless, Eqs. 4 define a system of a large number (thousands) of coupled differential equations whose solution is impractical for direct application to engineering codes, both in terms of computational cost and memory requirement, even with the application of grouping schemes [24], [52]. In order to reduce the dimension of the cluster dynamics system, I introduce two broader cluster categories, i.e., clusters of size $n = 1$ (single atoms) and clusters of size $n > 1$ (bubbles). I define the number density of bubbles (clusters of size $n > 1$), N_{ig} (bub m^{-3}), and the total concentration of gas in bubbles, m (at m^{-3}), as

$$N_{ig} = \sum_{n=2}^{\infty} c_n \quad (5)$$

$$m = \sum_{n=2}^{\infty} n c_n \quad (6)$$

Combining Eqs. 5, 6 with the cluster dynamics master equations (Eqs. 4) one obtains a system of three partial differential equations

$$\begin{aligned} \frac{\partial N_{ig}}{\partial t} &= +\nu - \sum_{n=2}^{\infty} \alpha_n c_n \\ \frac{\partial m}{\partial t} &= +2\nu + \sum_{n=2}^{\infty} \beta_n c_n - \sum_{n=2}^{\infty} n \alpha_n c_n \\ \frac{\partial c_1}{\partial t} &= +yF + D\nabla^2 c_1 - 2\nu - \sum_{n=2}^{\infty} \beta_n c_n + \sum_{n=2}^{\infty} n \alpha_n c_n \end{aligned} \quad (7)$$

The series in Eqs. 7 can be approximated by applying the Taylor expansion of $\alpha_n = \alpha(n)$ and $\beta_n = \beta(n)$ in conjunction with the definitions given in Eqs. 5, 6. For the re-solution rate defined by Eq. 2, under the assumption that $R_n = Bn^{1/3}$ [24], with $B = (3\Omega/4\pi)^{1/3}$ (Ω being the atomic gas volume in bubbles) [24], one obtains

$$\begin{aligned} \alpha_n &= \alpha_{\bar{n}} + \left[\frac{d\alpha_n}{dn} \right]_{\bar{n}} (n - \bar{n}) + o(n) \approx \\ &\approx \alpha_{\bar{n}} \left(1 + \frac{2 B n \bar{n}^{-2/3} - B \bar{n}^{1/3}}{B \bar{n}^{1/3} + R_{ff}} \right) = \alpha_{\bar{n}} \varphi_{n, \bar{n}} \approx \alpha_{\bar{n}} \end{aligned} \quad (8)$$

As for the trapping rate, considering that it is proportional to the bubble radius (Eq. 3) and again, with $R_n = Bn^{1/3}$ [24], one obtains

$$\begin{aligned} \beta_n &= \beta_{\bar{n}} + \left[\frac{d\beta_n}{dn} \right]_{\bar{n}} (n - \bar{n}) + o(n) \approx \\ &\approx \beta_{\bar{n}} \left(\frac{2}{3} + \frac{1}{3} \frac{n}{\bar{n}} \right) = \beta_{\bar{n}} \psi_{n, \bar{n}} \approx \beta_{\bar{n}} \end{aligned} \quad (9)$$

The final approximations in Eqs. 8, 9 are based on the coefficients $\varphi_{n, \bar{n}}$ and $\psi_{n, \bar{n}}$ being close to unity for a size distribution that peaks near the average value. This kind of size distribution is typical of experimental observations

of intra-granular bubbles [41], [42]. It would be of interest to confirm the validity of this assumption by comparing to detailed cluster dynamics simulations deriving the full-size distribution of intra-granular bubbles, although this is out of the scope of the present work. It is worth noting that it is possible to refine the above approximation including more terms in the Taylor expansions, if needed. With the approximations of Eqs. 8,9 and the definitions given in Eqs 5, 6, Eqs. 7 become

$$\begin{aligned}\frac{\partial N_{ig}}{\partial t} &= +\nu - \alpha_{\bar{n}} N_{ig} \\ \frac{\partial m}{\partial t} &= +2\nu + \beta_{\bar{n}} N_{ig} - \alpha_{\bar{n}} m\end{aligned}\tag{10}$$

$$\frac{\partial c_1}{\partial t} = +yF + D\nabla^2 c_1 - 2\nu - \beta_{\bar{n}} N_{ig} + \alpha_{\bar{n}} m$$

I close the system by defining the average number of atoms in a bubble and the average bubble radius (according to [24]), respectively, as

$$\begin{aligned}\bar{n} &= m/N_{ig} \\ R_{\bar{n}} &= B\bar{n}^{1/3}\end{aligned}\tag{11}$$

where $B = (3\Omega/4\pi)^{1/3}$ with Ω being the atomic volume. For spherical bubbles, the intra-granular swelling (fractional volume increase of fuel due to intra-granular bubbles) is at last

$$\left(\frac{\Delta V}{V}\right)_{ig} = \frac{4}{3}\pi R_{\bar{n}}^3 N_{ig}\tag{12}$$

Note that neglecting vacancy absorption at the bubbles implies that the total swelling due to fission gas atoms does not vary with transfer of atoms from the solution to the bubbles, and that the total intra-granular swelling is given by the sum of the contributions due to bubbles (Eq. 12) and dissolved gas.

The resulting final formulation of the model, Eqs. 10-12, leads to a number of considerations. The bubble number density, N , varies according to the rate of nucleation of new bubbles and the re-solution rate, with the latter corresponding to the bubble destruction rate in the heterogeneous interpretation. Note that the derivation from the equations of cluster dynamics leads to the same rate equation for the total bubble number density in the heterogeneous model of Olander and Wongsawaeng [24]. Rather than by

starting from the detailed physical description of cluster dynamics, the model in [24] was derived by applying in the first place the assumption of all bubbles having the same size and, consistently, of the re-resolution and trapping rate being the same for all bubbles. The calculation of the average number of atoms in a bubble, $\bar{n}$, differs from the heterogeneous model of Olander and Wongsawaeng [24] in that it naturally includes the effects of nucleation and re-resolution through the first two of Eqs. 10. In particular, nucleation and re-resolution affect the average bubble size because in general bubbles are created/destroyed at sizes different from the mean. These effects can be considered only if a size distribution of bubbles is considered and cannot be naturally included through a pure single-size derivation. The herein developed model based on the detailed cluster dynamics approach introduces these important bubble-distribution related effects in a formulation that in its final form only includes equations for the average quantities. In [24], this problem is circumvented by introducing the effects of nucleation and re-resolution on the average bubble size *a posteriori* through consideration of the average bubble lifetime.

Furthermore, the present model overcomes the assumption in [24] of gas diffusion to grain boundaries being neglected. Eqs. 10-12 describe intra-granular gas behaviour in terms of bubble evolution coupled to gas diffusion. The diffusion equation is solved numerically using a recently developed algorithm [56] (Chapter 5).

In line with [24], the present model does not treat the gas equation of state in the bubbles in detail (Eq. 11), considering that atom density in small intra-granular bubbles approaches that of solid xenon.

The new model is a description of intra-granular bubble evolution coupled to diffusion that depends on a reduced number of equations and parameters, and can be applied directly to engineering codes. The procedure applied to derive the formulation drastically reduces model complexity relative to the reference cluster dynamics model, while retaining important physical details that are associated with the presence of a non-uniform bubble size distribution and are therefore inherent in the cluster dynamics model, but not fully compatible with traditional single-size approaches.

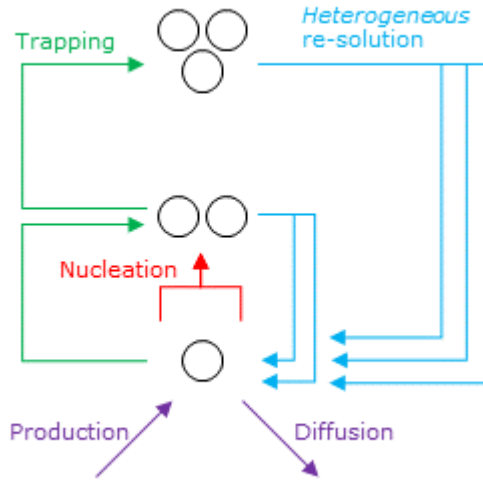


Figure 2. Sketch of the mechanisms of cluster dynamics. Nucleation is assumed to create only dimers ($n = 2$). *Heterogeneous* re-solution destroys entire bubbles, transferring n atoms from the bubble to the single gas atoms bin. It is assumed that production (from fission reactions) and diffusion towards the grain boundaries affect only single gas atoms.

2.3 Model validation

2.3.1 Comparison with experimental data

In this section, I perform the comparison of the present model (Eqs. 10, 11) with experimental data. The reference values for each model parameter used are collected in Table 1. All the calculations are performed with the model implemented in the 0D stand-alone computer code SCIANTIX.

The available experimental databases for intra-granular bubble size and number density as a function of well-defined irradiation conditions include the data from Baker [42] and Kashibe et al. [28]. In this work I excluded the data from Kashibe et al., [28] since they reported a significant coarsening⁷ of the bubbles occurring at high burnup. Modelling intra-granular bubble coarsening requires considering additional bubble growth mechanisms to those governing the evolution of smaller bubbles. Such mechanisms are not yet fully understood, but plausibly involve significant vacancy absorption at the bubbles and/or bubble coalescence [28], [57], [58], which the present model does not represent. The extension of the model to bubble coarsening is out of the scope of this chapter, however, first developments are presented in the Appendix A.

The data from Baker [42] consist of nine measurements of average bubble size and bubble number density. These measurements were performed on samples irradiated at various temperatures (from 1273 to 2073 K) up to a burnup of 23 GWd t⁻¹. In the simulations, to reproduce fuel irradiation up to this burnup I assume a typical fission rate $F = 1 \cdot 10^{19}$ fiss m⁻³ s⁻¹ kept constant for an irradiation time of 5500 hours.

Figure 3 shows the results of the new model in comparison with the experimental data. Results are provided both in terms of average bubble radius and bubble number density. A non-dimensional validation metric q is

⁷ I use *coarsening* as general term to indicate the significant growth of a subpopulation of intra-granular bubbles. This should not be confused to other processes referred to as coarsening in the literature, e.g., grain growth during sintering.

introduced, defined as the logarithm of the ratio between a simulation result y_i and the corresponding experimental data x_i , i.e.

$$q = \text{Log} \left(\frac{y_i}{x_i} \right) \quad (13)$$

This validation metric is zero for $y_i = x_i$, whereas values of $q = \pm 1, 2, \dots$ represent one, two, ... order of magnitude difference between simulation results and experimental data. The simulation results obtained with the model parameters listed in Table 1, the experimental data and the relative values for the validation metric are collected in Tables 2, 3 for the bubble radius and number density, respectively. Overall, there is a good agreement between the simulation results and the experimental data. The average values of q over the entire database are -0.12 and -0.06 for the bubble radius and number density, respectively.

2.3.2 Uncertainty and sensitivity analysis

I performed an uncertainty analysis of the model to the nucleation rate (Eq. 1), the re-resolution rate (Eq. 2), and the trapping rate (Eq. 3). The purpose of this uncertainty analysis is to provide an indication of the propagation of uncertainties from the fundamental model parameters to the model results. I randomly sampled scaling factors for the above model parameters in ranges representing the uncertainties associated to these rates (see Eqs 1, 2, and 3). In particular, for the nucleation rate I use a scaling factor in the range $[0.2, 1]$. This range is justified by several experimental observations reporting from 5 to 25 bubbles per fission track [41], [42]. As for the re-resolution rate, I use a scaling factor in the range $[0.5, 2]$ ⁸. The range I use for the trapping-rate scaling factor is $[0.2, 5]$. This range mainly reflects the experimental

⁸ This range is lower than the range $[0.1, 10]$ used by Pastore et al., for their uncertainty analysis work [17]. Nevertheless, the wider range proposed by Pastore et al. is meant to account also for the difference in the parameters according to different interpretations of the process, i.e., *heterogeneous* and *homogenous* re-resolution [17], whereas in this work we consider one mechanism only and assume pure *heterogeneous* re-resolution. Under this assumption, reducing the uncertainty from a factor of ten to a factor of two is deemed appropriate.

uncertainty on the diffusion coefficient [40], [55]. For each one of the nine experimental points by Baker [42] I performed 2,000 simulations with random sampling of the scaling factors, i.e., adopting a Monte Carlo approach. Figure 3 summarizes the results of this uncertainty analysis, showing the range of variation of the model results associated with the variation of the model parameters, for both the bubble number density and the bubble radius. I also include the experimental uncertainty associated with the data. Note that the resulting uncertainty associated with the predicted bubble radius is comparable to the experimental one (Fig. 3). The accurate calculation of the bubble radius is of particular relevance because the bubble radius affects the intra-granular bubble swelling with a power of three, compared to a power of one for the bubble number density (Eq. 12).

In order to investigate the sensitivities of the model to each of the uncertain parameters used in the uncertainty analysis (nucleation rate, re-resolution rate, trapping rate), I performed a sensitivity analysis. I considered the same variation range for the parameters as in the uncertainty analysis, and the intra-granular bubble swelling (Eq. 12) as the figure of merit for the sensitivity analysis. The results are collected in Figure 4, in terms of Pearson coefficients and normalized sensitivity coefficients as a function of the temperature. Both the Pearson coefficients and the normalized sensitivity coefficients are monotonically varying with temperature, and no strong dependence of the relative importance of the parameters on the temperature is observed. The results indicate that the trapping rate is the dominant parameter over the entire temperature range considered (with respect to the intra-granular bubble swelling). Because the uncertainty range used for the trapping rate mainly reflects the uncertainties in the gas atom diffusion coefficient, (Eq. 3), such a result confirms the importance of the uncertainty in this model parameter.

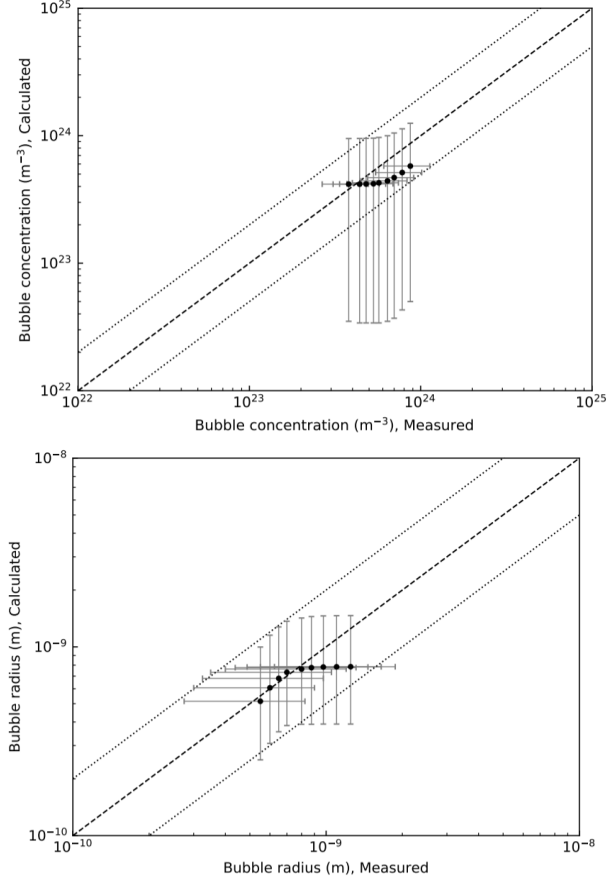


Figure 3. Comparison between the present model results and the data from Baker [42]. Each black full symbol represents a simulation obtained with the reference model parameters compared to the corresponding experimental value. The vertical error bars span the maximum and minimum results obtained in the 2,000 simulations performed for each experimental case. The horizontal error bars represent the experimental uncertainties, i.e., 30% for the bubble number density and 50% for the bubble radius [28], [42]. The distance from the 45° dashed black line is a graphical measure of accuracy. The dashed grey lines correspond to deviations of a factor of 2 from measured data, as an indication of the model accuracy.

Table 1. List of model parameters used for the validation of the intra-granular bubble evolution model.

Symbol	Definition	Value	u.o.m.	Ref.
η	Number of bubbles nucleated per fission fragment	25	bubbles ff ⁻¹	[2]
μ_{ff}	Range of a fission fragment in UO ₂	6	μm	[24]
R_{ff}	Radius of influence around a fission fragment track	1	nm	[24]
D	Diffusion coefficient of fission gas in UO ₂ $D_1 + D_2 + D_3$ $D_1 = 7.6 \cdot 10^{-10} \exp(-4.86 \cdot 10^{-19}/kT)$ $D_2 = 4 \cdot 1.41 \cdot 10^{-25} \sqrt{F} \exp(-1.91 \cdot 10^{-19}/kT)$ $D_3 = 2 \cdot 10^{-40} F$		m ² s ⁻¹	[40]
R_{at}	Radius of a gas atom in the lattice	0.2	nm	Est. [24]
Ω	Atomic gas volume in bubbles	$4.09 \cdot 10^{-2}$	nm ³	[59]

Table 2. Intra-granular bubble radius (nm) simulation results compared to Baker data [42]. The results of Olander and Wongsawaeng's model, taken from [24], are reported for additional comparison.

Temperature (K)	Exp. [42]	Olander and Wongsawaeng [24]		Present model	Validation metric ^a
		Heterogeneous	Homogeneous		
1273	0.55	0.4	1.0	0.515	-0.03
1373	0.60	-	-	0.607	0.005
1473	0.65	-	-	0.681	0.02
1573	0.70	-	-	0.733	0.02
1673	0.80	-	-	0.763	-0.02
1773	0.88	-	-	0.776	-0.05
1873	0.98	1.6	5.1	0.782	-0.01
1973	1.10	-	-	0.784	-0.15
2073	1.25	-	-	0.785	-0.20

^a The validation metric adopted in this work is defined as Log(calculated value / experimental value) (Eq. 13). For brevity, only the values of this metric for the results of the present model are reported.

Table 3. Intra-granular bubble number density (10^{23} bub m^{-3}) simulation results compared to Baker data [42]. The results of Olander and Wongsawaeng's model, taken from [24], are reported for additional comparison.

Temperature (K)	Exp. [42]	Olander and Wongsawaeng [24]		Present model	Validation metric ^a
		Heterogeneous	Homogeneous		
1273	8.7	7	3	5.78	-0.18
1373	7.8	-	-	5.14	-0.18
1473	7.0	-	-	4.69	-0.17
1573	6.4	-	-	4.42	-0.16
1673	5.7	-	-	4.27	-0.13
1773	5.3	-	-	4.20	-0.10
1873	4.8	2	0.03	4.18	-0.06
1973	4.4	-	-	4.17	-0.02
2073	3.8	-	-	4.17	0.04

^a The validation metric adopted in this work is defined as $\text{Log}(\text{calculated value} / \text{experimental value})$ (Eq. 13). For brevity, only the values of this metric for the results of the present model are reported.

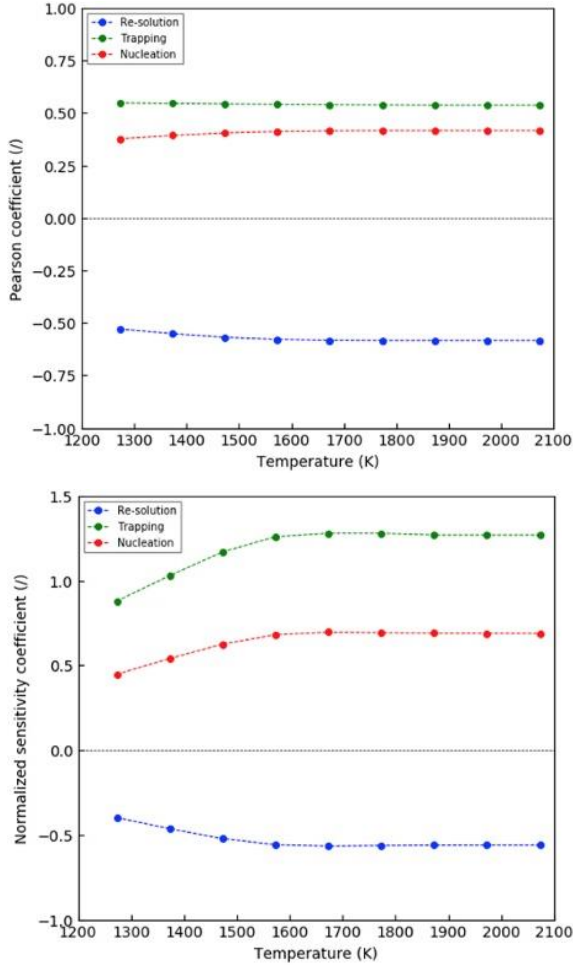


Figure 4. Results of the sensitivity analysis performed on the re-resolution, trapping and nucleation rates, using the intra-granular bubble swelling as the figure of merit. The results are shown in terms of Pearson coefficients (top) and normalized sensitivity coefficients (bottom), as a function of temperature. The trapping rate is the dominant model parameter over the considered temperature range.

2.4 Closing remarks

In this Chapter, I developed a model for the description of the intra-granular behaviour of fission gas in oxide fuel. In particular, the model computes the evolution of intra-granular bubble number density and size coupled to intra-granular gas atom diffusion to grain boundaries. The developed model finds its foundations in cluster dynamics but provides a single-size representation of the bubble distribution. As opposed to previous single-size models for intra-granular bubble evolution, this derivation from the detailed cluster dynamics description leads to consideration of important effects related to the presence of a non-uniform bubble size distribution. This advantage comes at no cost in terms of additional model complexity since the final formulation includes only three coupled differential equations for the bubble mean size, number density, and the concentration of dissolved gas. Therefore, this model can be efficiently used in fuel performance codes. Summarizing, the main outcomes of this part of the work are:

- The derivation of a new physics-based single-size model starting from the master equations of cluster dynamics.
- The implementation of the model equations in the SCIENTIX code and its validation as a stand-alone against experimental data obtained in separate effect experiments for a wide temperature range. This comparison with experimental data shows a good predictive capability of the new model, both in terms of bubble number density and average bubble radius.
- The uncertainty and sensitivity analysis performed with respect to the model parameters (nucleation rate, re-solution rate, and trapping rate) corroborates the validation, quantifying the impact of the uncertainties in the model parameters on the model results and providing useful information to prioritize lower-length scale research aimed at characterizing the model parameters.

Being based on theory rather than empirical relations, the new model can be applied to fuel materials other than UO_2 by adapting the model parameters. Potential future applications include mixed oxide (MOX) fuel, in the framework of the INSPYRE Project [9]. Furthermore, the model can be informed by fundamental parameters extracted at the lower-length scale with advanced modelling techniques, which make it potentially transferable to new fuel materials for which experimental data are unavailable. Finally, by means of an analogous derivation, a model can be obtained that considers *homogeneous* nucleation/re-solution mechanisms. This may add generality to

the developed approach as potentially applicable to fuel types characterized by a different nature of the nucleation/re-solution mechanisms (e.g., metallic fuels).

The new model includes the fundamental mechanisms of intra-granular fission gas behaviour and is also suited as a basic framework for future developments. For instance, the description of the rapid growth of a second population of larger bubbles (tens to hundreds of nm) observed at high burnup or during transients to high temperatures, referred to as intra-granular bubble coarsening is of great interest. Preliminary results of this development and first comparison with experimental data are reported in the Appendix A.

Considering its good predictive capability and in view of its interesting features, I implemented this intra-granular gas behaviour model in the BISON code. Implementation in the TRANSURANUS code is planned in the near future, in the framework of both INSPYRE and INERI Projects.

3. Inter-granular inert gas behaviour

Abstract

In this Chapter, I present a model describing the sudden release of gas from the grain boundaries during transients. Experimental observations indicate that substantial fission gas release (FGR) can occur on a small-time scale during temperature transients (burst release). To accurately reproduce the rapid kinetics of the burst release process in fuel performance calculations, I developed a model accounting for non-diffusional mechanisms such as fuel micro-cracking along grain boundaries. The concept and governing equations of the model are herein presented together with the sensitivity of results to the newly introduced parameters. The model is assessed for application to integral fuel rod analysis by implementation in both BISON and TRANSURANUS (and in SCIENTIX as well). Since the developed model is semi-empirical and derived directly from separate effects experiments, the model validation is based on the analysis of 19 light water reactor fuel rod irradiation experiments from the OECD/NEA IFPE (International Fuel Performance Experiments) database, all of which are simulated with both BISON and TRANSURANUS. The results point out an improvement in both the quantitative predictions of integral fuel rod FGR and the qualitative representation of the FGR kinetics with the transient model relative to the canonical, purely diffusion-based models of the codes. The overall quantitative improvement of the integral FGR predictions in the two codes is comparable.

The content of this Chapter is based on:

Barani T., Bruschi E., Pizzocri D., Pastore G., Van Uffelen P., Williamson R.L., Luzzi L., 2017. *Analysis of transient fission gas behaviour in oxide fuel using BISON and TRANSURANUS*. Journal of Nuclear Materials, 486, 96–110.

3.1 Introduction

In the previous Chapter, I presented a model for the evolution of intra-granular bubbles. I briefly discussed how the bubble population interacts with the single gas atoms diffusing towards the grain boundaries. Bearing in mind that modelling of fission gas behaviour in oxide fuel under irradiation calls for the treatment of various interactive phenomena [1], [23], [27], in this Chapter I discuss the release of gas from the grain boundaries occurring during transients. In fact, diffusion to the grain boundaries of the gas generated within the fuel grains needs to be described along with intricate grain boundary processes. The latter involve precipitation and growth of lenticular bubbles at the grain faces contributing to fuel swelling, and eventual gas venting from the grain faces leading to thermal fission gas release [3], [60]. Venting occurs after extensive gas bubble growth and interconnection, driven by gas atom and vacancy diffusion to the bubbles [1], [60].

In addition to this diffusion-based process, experimental observations suggest that gas release from the grain boundaries may occur through a mechanism of grain-face separation due to micro-cracking [10], [61]–[63]. Such mechanism is thought to be responsible for the high FGR observed in transient tests, which is characterized by a rapid kinetics that cannot be explained as purely diffusion-controlled [61], [62], [64]–[67]. Such rapid FGR observed in transient tests is often referred to as burst release. Carroll et al. [61] ascribed an increase in the FGR rate observed during UO_2 irradiation experiments to micro-cracking along grain boundaries due to a combination of thermal stresses and stresses generated by the accumulation of fission gas. Rest and Gehl [68] experimentally observed planar separations on the grain surfaces in UO_2 fuel specimens after transient heating. They noted that cracking along a given grain-grain contact can only result in release of the gas stored at the grain surfaces. Examinations of transient-tested UO_2 fuel rods performed by Hastings et al. [62] and by Walker et al. [63] also demonstrated the presence of grain-boundary separations. In both papers, micro-cracking was postulated to be responsible for the high FGR observed during transient tests. Micrographs of UO_2 specimens from the AGR/Halden Ramp Test Programme [10], which underwent power transient testing covering various power and temperature conditions, also showed separations of a fraction of the grain surfaces as well as associated grain-face gas depletion.

Models have been developed in the past to describe fission gas burst release during transients in fuel performance codes [69]–[73]. Generally, an additional gas release contribution from the grain boundaries during transients is introduced, considering that micro-cracking implies loss of gas storing capacity and instantaneous gas venting from cracked grain faces. The approaches from [69], [70], [73] consider an additional release of gas from the grain boundaries due to micro-cracking, invoked when specific empirical conditions that define the transient are fulfilled. In particular, discrete thresholds are introduced in terms of power variation and absolute power. Also, a burnup-dependent temperature threshold is considered in [70], [73]. The treatment proposed in [71] allows for an exponential reduction with time of the gas concentration at grain boundaries after the beginning of a transient. The model by Hering [69] allows also for the healing process of micro-cracks. Specifically, a restoration of the gas storing capacity of cracked grain faces after the transient event is considered and is described as linear with burnup. For the sake of completeness, it is worth mentioning that other models are available, attempting to explain burst release without relying on any description of micro-cracking (e.g., [74]). The review of these models is out of the scope of this work.

In this Chapter, I present a new physics-based, yet semi-empirical, model describing burst release. The model represents an extension of the current diffusion-based model available in both BISON [18] and TRANSURANUS [14], i.e., the model developed by Pastore et al. [3]. In this thesis work, I implemented the same transient release model in the two codes as an extension of the canonical FGB models. Several light water reactor (LWR) fuel rod irradiation experiments are simulated with both codes and results of FGR are compared to experimental data in order to assess the predictive capability of the developed transient model.

3.2 Model description

I developed a new semi-empirical model for transient gas release due to grain-boundary micro-cracking in oxide fuel. Such capability is thought as an extension for purely diffusion-based models that introduces the effect of micro-cracking on local gas retention capacity of the fuel. Based on experimental evidence, the model includes a dependency upon temperature variations that results in higher FGR during transients. Compared to previous ones, this model overcomes the usage of discrete thresholds in power and/or temperature for triggering an additional release contribution. This allows for a continuous kinetics of FGR, which is a more physically sound representation. Furthermore, as the underlying mechanisms of local gas retention and release are described directly, the model preserves the physical coupling between FGR and gaseous swelling. This model considers the dependency of grain-boundary micro-cracking on fuel burnup, consistently with experimental observations.

3.2.1 Physical processes

Fission gas transport from within the fuel grains (assumed to be spherical) to the grain boundaries is generally computed via the solution of the diffusion equation in spherical geometry

$$\frac{\partial c_{tot}}{\partial t} = \frac{\alpha}{\alpha + \beta} D \nabla^2 c_{tot} + yF \quad (14)$$

where c_{tot} (at m^{-3}) is the intra-granular gas concentration (both as single atoms and gas in bubbles), t (s) the time, β (s^{-1}) the rate of gas atoms trapping into intra-granular bubbles, α (s^{-1}) the rate of gas atom resolution from bubbles back into the lattice, D ($\text{m}^2 \text{ s}^{-1}$) the intra-granular single atom diffusion coefficient, y (at fiss^{-1}) is the gas fission yield and F ($\text{fiss m}^{-3} \text{ s}^{-1}$) the fission rate (i.e., yF is the gas generation rate). The term $\alpha/(\alpha + \beta)D$ represents the effective intra-granular diffusion coefficient according to Speight [25]. From Eq. 14, is clear how fundamental is the description of the intra-granular bubble population to assess the diffusion of single gas atoms towards the grain boundaries (Chapter 2). The grain-boundary gas behaviour analysis entails a physics-based and consistent treatment of both fission gas swelling and release through a direct description of the grain-face bubble development [3]. The major features of the analysis are the following

- The absorption rate at the grain-face bubbles is assumed to be equal to the arrival rate of gas at the grain boundaries [1], [2].
- An initial number density of grain-face bubbles is considered, and further nucleation during irradiation is neglected (one-off nucleation, e.g., [27]).
- All grain-face bubbles are considered to have, at any instant, equal size and equal lenticular shape of circular projection with semi-dihedral angle of 50° [2], [58].
- Grain-face bubbles grow (or shrink) by inflow of gas atoms from within the grains and concomitant absorption (or emission) of vacancies from the grain boundaries. Bubble size is also affected (reduced) by the gas escape term as FGR occurs.

The gas atom inflow rate is obtained solving Eq. 14, while the absorption/emission rate of vacancies at the bubbles is derived from the model of Speight and Beere [75]. Grain-face bubble coalescence is considered through an improved model of White [3], [27]. The time evolution of the grain-boundary bubble number density, N_{gb} (bub m⁻²), due to coalescence, is represented as a function of the time evolution of the bubble projected area on the grain face, A_{gb} (m²). The total relative volume of the lenticular grain-face bubbles represents the grain-boundary contribution to fission gas swelling [3], [76]. Thermal fission gas release is considered as a consequence of bubble growth and interconnection, and is represented based on a principle of grain face saturation. More precisely, when the fractional coverage of grain faces, $F_c = N_{gb} A_{gb}$ (/), attains a fixed saturation value, F_{sat} , further bubble growth is compensated by gas release from the grain faces in order to maintain the saturation coverage condition

$$\frac{dF_c}{dt} = 0 \text{ if } F_c = F_{sat} \quad (15)$$

The constant saturation coverage, F_{sat} , value is equal to 0.5 [3], [27]. Note that fission gas release and swelling are therefore described as inherently coupled phenomena, as fission gas release from the grain faces counteracts bubble growth and thereby fission gas swelling.

I developed an extension of this just outlined diffusion-based model accounting for the burst release effect during transients [77], [78]. The transient model is developed based on the available experimental evidence of transient fission gas release in oxide fuel. From relevant experimental work reported in [10], [61], [62], [64], [65], [67], [68], [79]–[82], the following common characteristics of the burst release process emerge:

1. FGR during transients is characterized by a rapid kinetics (burst release).
2. Burst release is triggered by temperature variations.
3. Burst release occurs during both heating and cooling transients.
4. Burst release occurs in a limited temperature range, with the maximum release rate around a certain temperature.

As for the underlying mechanism of burst release, as discussed in Section 3.1, experimental evidence and consensus exist towards micro-cracking along grain boundaries [10], [61], [62], [68]. This is also consistent with characteristic 1, which indicates a non-diffusional mechanism as responsible for the process.

Based on this evidence, in this thesis work, I model burst release as if caused by fuel micro-cracking along the grain boundaries. However, in view of the uncertainties that would affect a detailed model for the micro-cracking mechanism, it has been chosen to develop the transient model on a largely empirical basis. The effect of micro-cracking is considered as a loss of both gas inventory (depletion) and storing capacity of cracked grain faces. Modifications in the grain-face gas inventory and storing capacity can be represented macroscopically through the fractional coverage and the saturation coverage, respectively. Gas depletion of a fraction of the grain faces is modelled as a reduction of the fractional coverage, F_c , which corresponds to the fraction of grain surface occupied by gas bubbles, in the sense of an average over the fuel volume associated with a macroscopic calculation point. In particular, F_c is scaled by a factor, f (/), which can be interpreted microscopically as the fraction of non-cracked (intact) grain faces. The reduction of the fractional coverage effectively leads to a decrease of the amount of gas retained in the fuel (and, as a consequence, of fission gas swelling) and to a corresponding increase of fission gas release. This contribution to fission gas release adds to the diffusion-interconnection mechanism considered in the diffusion-based model outlined before. The saturation coverage, F_{sat} , corresponds to the average grain-face gas storing capacity at a given macroscopic calculation point. The value $F_{sat} = 0.5$ is considered in absence of micro-cracking. In a microscopic sense, I model the grain-face gas storing capacity as dropping to zero for a cracked grain face. Then, the loss in average storing capacity due to micro-cracking is represented in the model by scaling the saturation coverage, F_{sat} , down from 0.5 by the factor f . Finally, a healing process of cracked grain faces is considered as a progressive restoration of the grain-face gas storing capacity.

It follows that, if the transient model is applied, the fractional coverage and the saturation coverage obey the following relationships

$$\begin{aligned}\frac{dF_c}{dt} &= \left[\frac{dF_c}{dt}\right]_{\text{diff}} + F_c \left[\frac{df}{dt}\right]_{\text{micro-cracking}} \\ \frac{dF_{\text{sat}}}{dt} &= F_{\text{sat}} \left(\left[\frac{df}{dt}\right]_{\text{micro-cracking}} + \left[\frac{df}{dt}\right]_{\text{healing}} \right)\end{aligned}\tag{16}$$

where the subscript diff stands for diffusion-controlled processes. The initial (maximum) value for the saturation coverage, corresponding to all intact grain-faces, is $F_{\text{sat},i} = 0.5$. The transient model introduces a non-diffusional contribution to the evolution of gas at grain boundaries, which represents the effect of micro-cracking. Such contribution is considered through a generalized calculation of the fractional coverage and saturation coverage parameters relative to the purely diffusion-based model. In particular, the fractional coverage and saturation coverage are effectively reduced to represent the fact that a fraction of the grain faces become unable to retain any gas as micro-cracking occurs. In the following Section, the formulations of the micro-cracking and healing terms, governing df/dt , in Eq. 16 are detailed.

3.2.2 Model derivation

To characterize the micro-cracking mechanism, I introduce an empirically based micro-cracking parameter, μ . Considering that micro-cracking can only affect intact grain faces, I write

$$\left[\frac{df}{dt}\right]_{\text{micro-cracking}} = -f \frac{d\mu}{dt}\tag{17}$$

where f is again the fraction of intact grain faces. The functional form of the parameter μ is chosen as a temperature-dependent sigmoid function. Based on the available experimental evidence, the following expression for μ is developed [77], [78]

$$\mu(T) = 1 - \left[1 + Q \exp \left(s \frac{T - T_{\text{infl}}}{B} \right) \right]^{-1/Q}\tag{18}$$

where T (K) is the local temperature, T_{infl} (K) the value for the temperature at the inflection point of the function $\mu(T)$ (inflection temperature), and B (K),

s (-) and Q (-) are parameters related to the temperature-domain width of the phenomenon and the deviation from symmetric behaviour during heating/cooling transients, respectively. The values $B = 10$ K and $Q = 33$ are used [77], [78]. I investigated the sensitivity of model predictions to parameters through a sensitivity analysis (Section 3.3), demonstrating that the sensitivity of results to Q and B is low compared to T_{infl} . Eq. 17 combined with the formulation for $\mu(T)$ (Eq. 18) implies

$$\left[\frac{df}{dt}\right]_{\text{micro-cracking}} = 0 \text{ if } \frac{dT}{dt} = 0 \quad (19)$$

which conforms to the experimentally observed characteristic 2 of burst release as triggered by temperature variations [79]–[82]. The value of s (-) in Eq. 18 is set to $+1$ during heating transients and to -1 during cooling transients, so that $\mu(T)$ increases during both heating and cooling, in conformity with experimental observations (characteristic 3). According to Eq. 18, the absolute value of the temperature derivative for μ is maximum at T_{infl} (see also Fig. 5, bottom), thus the combined Eqs. 17 and 18 reproduce the maximum rate of burst release at T_{infl} as temperature varies in time. Thus, the present model with the so constructed micro-cracking parameter also allows for characteristic 4 of the process. The concept of a temperature of maximum burst release rate, or inflection temperature, is corroborated by experimental work [79]–[82], showing peaks of release rate occurring as the fuel reaches a certain temperature, and exhausting within a limited temperature range. Moreover, the experiments indicate that a dependency exists of the inflection temperature on fuel burnup. For the calculation of the parameter T_{infl} in the model, the following correlation is used

$$T_{\text{infl}} = 1773 + 520 \exp\left(-\frac{bu}{10}\right) \quad (20)$$

where bu (Gwd tU⁻¹) is the burnup at the current time. Eq. 20 is derived from the best-estimate fit of quantitative experimental data (Fig. 5, top).

The micro-cracking parameter, μ , and the temperature derivative, $d\mu/dT$, are reported in Fig. 5 (bottom) for a value of $T_{\text{infl}} = 1773$ K. The inflection point (maximum absolute value of derivative) for μ at T_{infl} corresponds to the maximum rate of burst release at the inflection temperature. The asymmetry around the inflection point conforms to experimental observations indicating that burst release during heating transients is higher in the temperature region above the inflection temperature than below, and conversely [65].

A physically based description of micro-cracking would take into account localized stresses at grain boundaries. Here, the choice was to adopt an empirical description in consideration of the uncertainties involved in the calculation of localized stresses, which may outweigh the improved accuracy from the added physical detail. Rather than being an attempt to capture the complex underlying physics of burst release, the transient model represents a pragmatic approach to allow for this important aspect of FGB in fuel performance codes in an effective way. Nevertheless, the empirical basis of the model entails limitations in terms of flexibility of application and insight into the underlying physical processes. Further developing the present preliminary approach to a more physically-based description is advisable in perspective in order to further improve model predictive capability. Namely, the development of a more physics-grounded formulation of μ is of interest.

The effect of micro-crack healing is accounted for in the model via an empirical treatment. Following Hering [69], the effect of the healing process is represented as a progressive restoration with time of the gas storing capacity of cracked grain faces and is purely burnup-dependent. Analogous to the treatment of micro-cracking, to characterize the process I define an healing parameter, h (-). Since the mechanism can only affect cracked grain faces

$$\left[\frac{df}{dt}\right]_{\text{healing}} = (1 - f) \frac{dh}{dt} \quad (21)$$

The functional form of the healing parameter is $h(bu) = bu/bu_\tau$, with $bu_\tau = 1 \text{ GWd tU}^{-1}$.

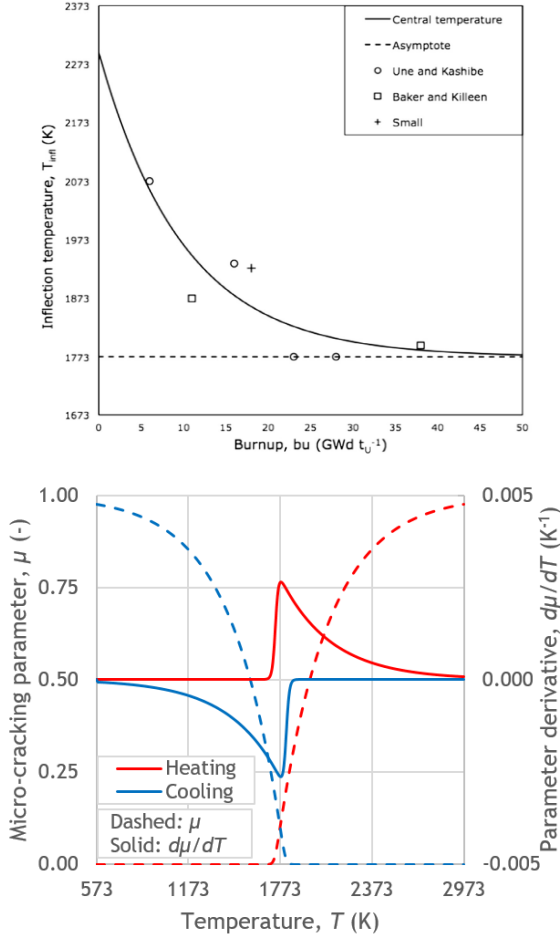


Figure 5. Top: Experimental data for the temperature of maximum burst release rate from Une and Kashibe [79], Baker and Killeen [81] and Small [82] as a function of burnup and best-estimate fitting curve. Bottom: Micro-cracking parameter, μ , and temperature derivative, $d\mu/dT$, as a function of temperature. The asymptotic value of 1773 K for the inflection temperature is considered for this plot.

3.3 Model validation

Several fuel rod irradiation experiments are simulated in this work using the BISON and TRANSURANUS fuel performance codes with the FGB models described in Section 3.2. Moreover, I performed a sensitivity analysis (analytically) to assess the impact of each model parameter on the overall fission gas behaviour.

3.3.1 Comparison with experimental data

For the analyses with the BISON and TRANSURANUS codes, I considered 19 LWR fuel rod irradiation experiments from the OECD/NEA International Fuel Performance Experiments (IFPE) database [67]. The power histories of the selected experiments are comprised of a base irradiation under normal operating conditions followed by a ramp test at high power. The considered cases are listed in Table 4. Some features of the fuel rods are also summarized, focusing on data that are directly related to FGB modelling. Some of these experiments were included in the IAEA Coordinated Research Projects FUMEX-II [21] and FUMEX-III [22]. I carried out the simulations coherently with the pre-irradiation characterization data, power histories and coolant conditions provided in the IFPE database. Experimental data available from these experiments include Post-Irradiation Examination (PIE) measurements of integral fuel rod FGR at the end of irradiation (generally from rod puncturing). In some cases, the time evolution of integral FGR inferred from on-line rod pressure measurements and PIE of radial xenon profiles in the fuel are also included.

Calculations were performed using the BISON and TRANSURANUS codes with the relative physical models (see Chapter 1, Introduction, for a brief description of the two codes). In addition, the FGB models presented in Section 3.2 were applied for the calculation of fission gas release and fuel gaseous swelling. For the present simulations, I used the single atom diffusion coefficient based on Turnbull et al. [83] as in [17]. To obtain the initial grain radii (Table 4), the measured linear intercepts were multiplied by a factor of 0.78 [84]. Grain growth during irradiation is accounted for using the model of Ainscough et al. [85]. The AN3, AN4, I13 and IFA 597.3 experiments involved re-fabrication of the fuel rod prior to the ramp test, which is considered in the simulations by re-defining specific parameters (fill gas pressure and composition) and employing the restart capabilities of the codes.

The Risø-3 AN3 and AN4 fuel rods were equipped with a fuel centreline thermocouple and a pressure transducer in the plenum, so that the time evolution of FGR could be inferred based on the online pressure measurements. Moreover, PIE data for the diametric xenon concentration profiles are available for these two experiments. In particular, the diametric profile of the total xenon concentration (summation of intra-granular and inter-granular contributions) was determined by X-Ray Fluorescence (XRF) analysis. These experiments allow for a detailed investigation of model fidelity, since comparisons of calculations to time-dependent fuel rod FGR (as opposed to the sole end-of irradiation values) as well as retained gas in the fuel as a function of diametric position in the fuel pellet are possible.

Fig. 6 shows results from the BISON and TRANSURANUS calculations along with experimental measurements of integral FGR during the AN3 ramp test. Measured and calculated fuel central temperature is also shown. As illustrated in the figures, burst release during transients is experimentally observed during the AN3 test (note the stepwise release at about 48 and 70 h from the beginning of the test). Results point out a significant improvement in FGR predictions with the transient model compared to the canonical diffusion-based models (w/o transient model), for both codes. Improvements are in terms of both representation of the FGR kinetics and end-of-irradiation FGR values. In particular, differently from the canonical models, the transient model accounts for the burst release effect, with stepwise release during transients being reproduced. However, the magnitudes of the experimental (from pressure measurements) and calculated FGR steps are different, with an under-prediction being observed. Indeed, part of the rapid increase in the measured inner pressure and FGR may be ascribed to a delayed detection of gas actually released before the transient and related to gap and cracks reopening during power reductions [86]. Such hypothesis may partly explain these discrepancies. When the transient model is used, the deviation between the end-of-irradiation FGR calculated with both codes and the measured one appears satisfactory, in view of the intrinsic modelling uncertainties [17]. With the canonical models, burst release is not reproduced, indicating that the phenomenon cannot be represented through a purely diffusion-based interpretation of fission gas behaviour.

Fig. 7 shows results from the BISON and TRANSURANUS calculations along with experimental measurements of fuel central temperature and integral FGR during the AN4 ramp test. Significant improvements of the FGR predictions are observed when applying the transient model compared to the canonical models, for both codes. In this case as well, improvements

are both in the predicted kinetics of the process and the end-of-irradiation FGR value. From the presented results, it is noted that the calculated FGR with the canonical models (w/o transient model) from the two codes is significantly different, with BISON tending to compute a higher FGR than TRANSURANUS. In addition, the impact of the transient model on predictions is different. It is postulated that this would be ascribed to the effect of compressive hydrostatic stress. In the canonical FGB model of TRANSURANUS, hydrostatic stress acts to compress grain-face bubbles, delaying the saturation of the grain faces [1], [3], [58] and reducing FGR. This effect is currently neglected in BISON [18]. Correspondingly, in the TRANSURANUS canonical representation, a higher amount of gas is retained in the fuel compared to BISON. Hence, more gas is available for release via micro-cracking at the grain faces, which is considered if the transient model is applied. This peculiarity will be the subject of further investigation once a stress-dependent FGB model is included in BISON. Other contributions to differences between predictions from the two codes include differences in the calculated fuel temperatures, as well as some differences between the formulations of the canonical FGB models of the codes [3], [14], [17], [18].

In the analysis of both AN3 and AN4 experiments, the temperatures calculated by the codes are compared with the measured temperature during the ramp tests. Calculations with the transient FGB model applied are considered. For both the BISON and TRANSURANUS simulations, the results point out a good agreement between calculated and experimental temperatures for the AN3 experiment. As for the AN4 experiment, the deviation between the calculated temperatures and the experimental data is more significant. However, for both BISON and TRANSURANUS the error is within 10% of the experimental values, which is in line with the intrinsic uncertainties [15], [16].

Finally, I present comparisons of calculations to experimental data of local xenon concentrations in the fuel. In particular, I consider profiles of retained xenon concentration across the pellet diameter, measured by XRF analysis in a fuel sample obtained from the AN3 rod [67]. The comparisons between the experimental profile and the BISON and TRANSURANUS calculations, both with the canonical diffusion-based models and with the transient model extension applied, are illustrated in Fig. 8. The plots display local gas retention in the fuel, which is coupled to gas release, as reproduced by the fission gas behaviour models in the codes. Direct modelling of the physical mechanisms of fission gas diffusion and bubble evolution underlies

calculated concentrations [3], [17]. The transient model acts on such physical representation introducing a reduction, dependent on local conditions, of the retained gas concentration. This affects (reduces) local fuel gaseous swelling and leads to a corresponding increase in FGR. In general, a good agreement is observed between the calculated and the measured xenon concentration profiles, although some discrepancies are observed. These may be ascribed to uncertainties inherent in fission gas behaviour modelling [17], as well as uncertainties in the measured concentrations, which are associated with the experimental technique [87]. For both codes, the transient model tends to improve the results compared to the canonical model. For the TRANSURANUS calculation with the canonical model, a higher amount of gas is stored in the fuel inner region compared to the BISON calculation. This is consistent with the differences in calculated FGR mentioned above. As discussed, such different behaviour may be ascribed to the role of hydrostatic stress in compressing grain-face bubbles and limiting release, which corresponds to higher gas retention and is considered only in TRANSURANUS at this time. In the outermost region of the fuel pellet, red-marked in Fig. 8, the potential formation of the High Burnup Structure (HBS) may affect the xenon concentration profile [49,50]. Fission gas behaviour in the HBS is different than in the regular fuel structure, requiring a different modelling approach. This topic is covered in Chapter 4.

Lastly, I present simulation results in terms of integral FGR at the end of fuel rod irradiation for all 19 experiments considered in this work (Table 4). The comparisons of calculations to experimental data are shown in Fig. 9 for BISON (left) and TRANSURANUS (right). Data are tabulated in Table 5. In Fig. 9, both results obtained with the canonical diffusion-based models (Section 3.2) and those obtained applying the transient model extension (Section 3.2) are shown. Comparisons demonstrate that, when the transient model is applied, an improvement of the overall results is obtained for both codes. To provide a quantitative estimation of the improvement brought about by the transient model, I computed the relative error of each simulation result with respect to the measured fission gas release, namely, $(FGR_{\text{calculated}} - FGR_{\text{measured}}) / FGR_{\text{measured}}$. The average of the absolute values of this error is a measurement of the overall accuracy.

With the canonical models, the average relative error over all considered cases is of about 50% and 52% for BISON and TRANSURANUS, respectively. When applying the transient model, errors reduce to approximately 35% for both codes. Hence, the transient model improves the overall predictive accuracy for both codes.

Furthermore, the overall improvements for the two codes are similar. For both BISON and TRANSURANUS calculations, the higher relative error is observed for the Super-Ramp PK6 rods. The FGR values for these cases (Table 5) are lower. In this region of lower FGR, higher relative simulation errors are indeed expected due to (i) higher inherent modelling uncertainties [30] and (ii) higher numerical errors related to the solution of the intra-granular fission gas diffusion equation with the current algorithm [56], [88].

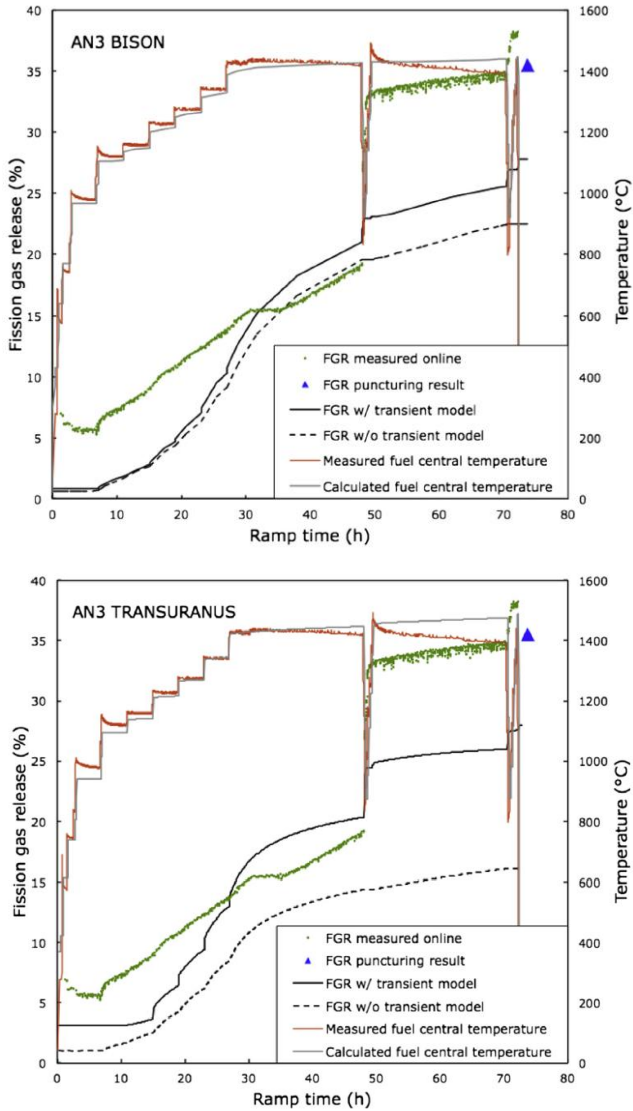


Figure 6. FGR and fuel central temperature as a function of time for the AN3 fuel rod during the ramp test. Experimental measurements are compared to calculations with BISON (top) and TRANSURANUS (bottom). FGR results obtained with (w/) and without (w/o) the transient model applied are presented.

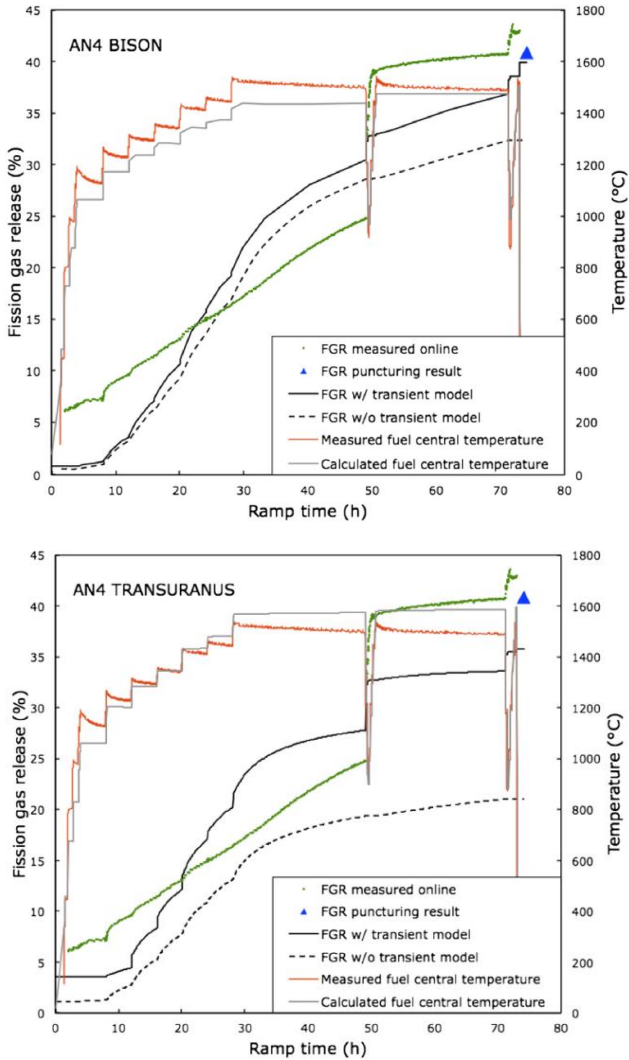


Figure 7. FGR and fuel central temperature as a function of time for the AN4 fuel rod during the ramp test. Experimental measurements are compared to calculations with BISON (top) and TRANSURANUS (bottom). FGR results obtained with (w/) and without (w/o) the transient model applied are presented.

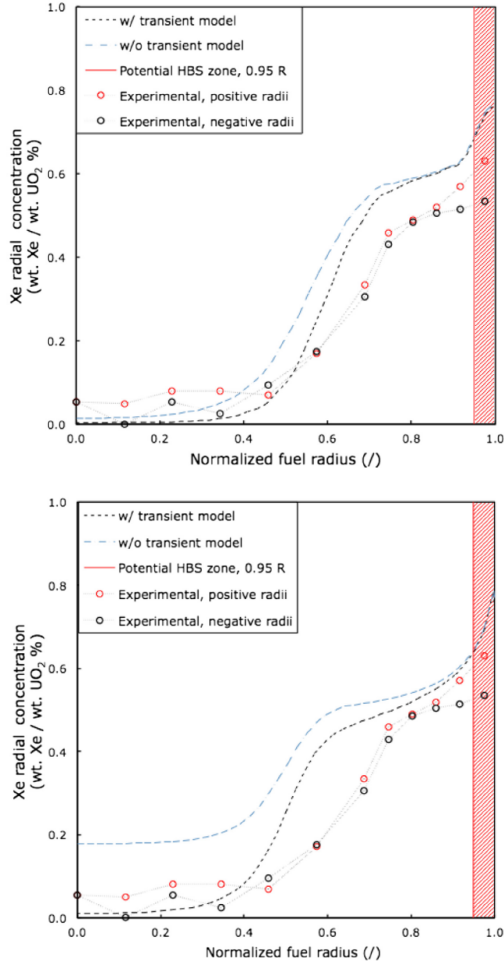


Figure 8. Radial profiles of xenon concentration in the CB-2R-5 fuel sample from the AN3 rod. Experimental data are compared to calculations with BISON (top) and TRANSURANUS (bottom). Since the experimental data were measured along the fuel diameter, but our calculations are axially symmetric, a positive and a negative radius are defined in reporting measurements. The origin is assumed in the centre of the pellet. The red region represents the potential HBS formation zone, which is not accounted for in the simulation and for which the comparison is less meaningful.

Table 4. Summary of the irradiation experiments from the OECD/NEA IFPE database [67] analysed in this work.

Fuel rod	Database	Reactor type	Average burnup (GWd t _U ⁻¹)	FGR, measured (%)	Grain radius (µm)
PK1-1	Super-Ramp	PWR	35.4	8.5	4.68
PK1-2	Super-Ramp	PWR	35.6	13.5	4.68
PK1-3	Super-Ramp	PWR	35.2	22.1	4.68
PK1-4	Super-Ramp	PWR	33.1	13.0	4.68
PK2-1	Super-Ramp	PWR	45.2	28.0	4.29
PK2-2	Super-Ramp	PWR	45.1	32.1	4.29
PK2-3	Super-Ramp	PWR	44.6	44.9	4.29
PK2-4	Super-Ramp	PWR	41.4	9.5	4.29
PK6-2	Super-Ramp	PWR	36.8	3.5	17.16
PK6-3	Super-Ramp	PWR	36.5	6.7	17.16
PK6-S	Super-Ramp	PWR	35.9	6.1	17.16
AN2	Risø3	PWR	41.5	29.7	4.68
AN3	Risø3	PWR	44.0	35.5	4.68
AN4	Risø3	PWR	41.9	40.9	4.68
AN8	Risø3	PWR	43.8	13.7	4.68
GE7	Risø3	BWR	41.7	14.4	9.4
II3	Risø3	BWR	14.5	17.4	9.5
Rod 8	IFA 597.3	BWR	70	15.8	6.1
L10	Regate	PWR	53.4	10.2	7.5

Table 5. Summary of integral FGR results obtained with BISON and TRANSURANUS for the fuel rod irradiation experiments analysed in this work.

Fuel rod	FGR(%), measured	BISON		TRANSURANUS	
		FGR (%), w/o burst model	FGR (%), w/ burst model	FGR (%), w/o burst model	FGR (%), w/ burst model
PK1-1	8.5	5.6	8.1	6.7	14.2
PK1-2	13.5	8.4	11.8	9.0	18.0
PK1-3	22.1	14.4	18.5	13.2	24.0
PK1-4	13.0	12.6	16.5	12.2	22.0
PK2-1	28.0	8.8	12.6	10.0	19.0
PK2-2	32.1	16.9	22.3	15.9	27.5
PK2-3	44.9	23.5	28.2	19.0	31.9
PK2-4	9.5	2.1	5.0	5.7	14.9
PK6-2	3.5	0.9	1.3	0.9	2.2
PK6-3	6.7	1.3	1.8	1.3	2.8
PK6-S	6.1	0.9	1.3	0.8	2.1
AN2	29.7	23.3	26.8	15.8	28
AN3	35.5	22.5	27.8	16.2	28.1
AN4	40.9	32.4	39.9	21.0	35.8
AN8	13.7	6.0	8.3	7.4	16.6
GE7	14.4	8.9	12.3	6.0	11.3
II3	17.4	8.8	12.7	3.2	6.8
Rod 8	15.8	3.7	8.4	12.1	20.4
L10	10.2	3.3	6.2	3.5	8.1

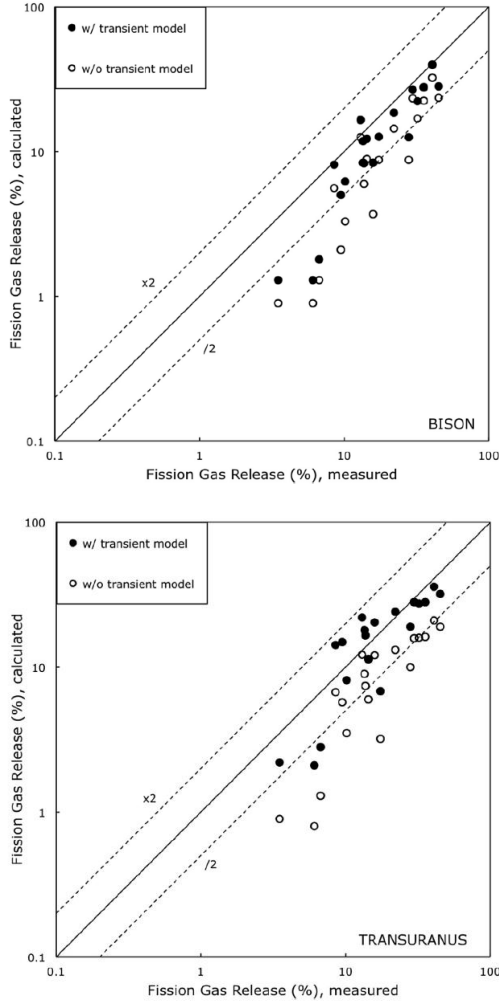


Figure 9. Simulation results of integral FGR at the end of irradiation from BISON (top) and TRANSURANUS (bottom) compared to experimental data. Each data point corresponds to one of the 19 irradiation experiments from the OECD/NEA IFPE database [67] analysed in this work. The open symbols represent the results obtained with the canonical models, whereas the full symbols are the results obtained with the transient model applied. The distance from the 45° line is a measure of the accuracy. Dashed lines correspond to deviations of a factor of 2 from measured data [17].

3.3.2 Sensitivity analysis

In order to assess the sensitivity of calculations to model parameters, I perform an analytic sensitivity analysis of the developed representation of the micro-cracking effect, in particular, Eq. 18. The model parameters of interest in this sensitivity analysis are B , Q , T_{infl} . As representative figure of merit, the fractional coverage, F_c , is selected. The reason for this selection is the fractional coverage being the physical quantity that carries the information from the transient model to the overall FGB model through Eq. 16.

Table 6 reports the definitions and the analytic expressions of the sensitivity coefficients (k_T , k_B , and k_Q) as defined in this analysis for the three parameters of interest (T_{infl} , B , Q , respectively). The results of the analysis are reported in Fig. 10. The sensitivity coefficient k_T , associated with the parameter T_{infl} , is characterized by the higher average and maximum over the considered temperature and burnup range (Fig. 10a). Hence, it clearly emerges from the sensitivity analysis that T_{infl} is the dominant parameter. Fig. 10b shows the temperature-burnup 2D map of the k_T sensitivity coefficient, which is maximum close to the value of T_{infl} and rapidly drops away from it. This is consistent with the fractional coverage (hence, the overall FGB model results) being significantly affected by the transient model only in the neighbourhood of the inflection temperature (Eqs. 16, 17). Instead, the sensitivity of the figure of merit to the B and Q parameters is low (coefficients k_B and k_Q , respectively, in Fig. 10a), which points out that the choice of the values for Q and B has a limited impact on model predictions.

Table 6. Definitions and analytic expressions of the relative sensitivity coefficients defined in this analysis. The analytic expressions can be derived by differentiation from Eqs. 16, 17.

Relative sensitivity coefficient	Definition	Analytic expression
k_T	$\frac{dF_c}{F} = k_T \frac{dT_{\text{infl}}}{T_{\text{infl}}}$	$f \frac{s}{B} T_{\text{infl}} \exp\left(s \frac{T - T_{\text{infl}}}{B}\right) \left[1 + Q \exp\left(s \frac{T - T_{\text{infl}}}{B}\right)\right]^{-\frac{1}{Q}-1}$
k_B	$\frac{dF_c}{F} = k_B \frac{dB}{B}$	$k_T \frac{T - T_{\text{infl}}}{T_{\text{infl}}}$ $-fQ \exp\left(-\frac{1}{Q} \log\left(1 + \exp\left(s \frac{T - T_{\text{infl}}}{B}\right)\right)\right)$
k_Q	$\frac{dF_c}{F} = k_Q \frac{dQ}{Q}$	$\left[\frac{1}{Q^2} \log\left(1 + \exp\left(s \frac{T - T_{\text{infl}}}{B}\right)\right) - \frac{\exp\left(s \frac{T - T_{\text{infl}}}{B}\right)}{Q \left(1 + \exp\left(s \frac{T - T_{\text{infl}}}{B}\right)\right)} \right]$

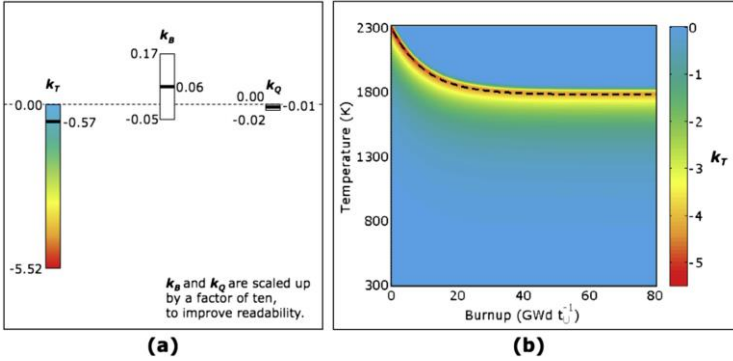


Figure 10. Results of the analytic sensitivity analysis of the transient model. (a) Bar plot reporting the upper, mean and lower value of the relative sensitivity coefficients analysed over the temperature-burnup range of [300, 2300 K] x [0, 80 GWd tU⁻¹]. (b) Detailed temperature-burnup 2D map of the relative sensitivity coefficient associated with the inflection temperature. The dashed line represents the inflection temperature as a function of burnup.

3.3 Closing remarks

In this Chapter, I presented and assessed a model for transient FGB in oxide fuel that accounts for the burst release process associated with fuel micro-cracking. The model conforms to the experimentally observed characteristics of the process and extends conventional diffusion-based interpretations of FGB in fuel performance codes. Even if still semi-empirical, the new model connects to physical quantities such as the fractional coverage of grain boundaries and its saturation value. Therefore, it represents a compromise between the physical description of the phenomena occurring at the meso-scale and the requirements of fuel performance codes. Summarizing its main characteristics, the new model:

- Assumes micro-cracking of the grain boundaries as the leading phenomenon causing burst release during temperature transients.
- Provides a semi-empirical description of micro-cracking as a function of temperature and burnup, and a description of burnup-dependent micro-crack healing as well.
- Does not require any discrete threshold in temperature and/or burnup, thus allowing for a continuous description of fission gas release and gaseous swelling (no step-like behaviour).

I implemented the transient model in two structurally different fuel performance codes (i.e., BISON and TRANSURANUS) as an extension of the canonical, purely diffusion-based FGB models of the codes. I assessed the model by performing simulations of 19 LWR fuel rod irradiation experiments from the OECD/NEA IFPE database and comparing code predictions from both codes to the available experimental data. The simulation results pointed out an improvement of the overall FGR description by employing the developed transient model, compared to the canonical models. More in detail:

- A better accuracy in the predictions of integral FGR at the end of irradiation was observed for both BISON and TRANSURANUS calculations. In particular, the average relative error of FGR predictions over the 19 considered cases improved from about 50% with the canonical models to approximately 35% with the transient model applied, for both codes.
- A more consistent representation of the FGR kinetics during transients was observed for both codes, as pointed out by comparisons of calculated FGR time evolution to on-line measurements for the Risø-3 AN3 and AN4 experiments. In particular, unlike simulations with the canonical

models, the burst release effect was reproduced in calculations applying the transient model.

- Satisfactory capabilities of both codes in predicting diametric xenon concentration profiles were observed, as pointed out by comparison of calculations to PIE for the Risø-3 AN3 experiment. The transient model tends to improve the results of local xenon concentration compared to the canonical models.

In short, the present results indicated that the transient FGB model applied to fuel rod simulations allows for both an improved qualitative representation of the kinetics of FGR during transients and an improved quantitative accuracy of FGR predictions.

Furthermore, by applying the model in two structurally different fuel performance codes, it was verified that such improvements hold for both codes. As discussed in Chapter 1, such cross comparison provides an argument in favor of the improvements, being related to the specific model itself, rather than to possible compensating errors between the model and the global fuel rod thermo-mechanical analysis. The successful outcome of model assessment in two different fuel performance codes thus suggests that the transient fission gas behaviour model indeed provides a better representation of the relevant physics (compared to purely diffusion-based models).

As a further development, a description of transient fission gas release as triggered by micro-cracking, similar to the model considered in this work, could be developed also for MOX fuel if enough data become available. This development is planned in the framework of the INSPYRE Project. Moreover, it is of sure interest to achieve a more physics-based and less empirical formulation of the model, in order to better describe the physical evolution of micro-cracks.

4. High burnup structure inert gas behaviour

Abstract

In the previous Chapters 2 and 3, I presented new models describing the evolution of inert gas behaviour within the fuel grains and along the grain boundaries. In this Chapter, I focus on IGB peculiar to the high burnup structure. The high burnup structure (HBS) forms at the rim zone of UO_2 nuclear fuel pellets, due to the combination of high burnup and low temperature driving a microstructural change. I propose a semi-empirical model to describe the formation of the HBS, which embraces the polygonisation/recrystallization process and the depletion of intra-granular fission gas, describing them as inherently related. For this purpose, I performed grain-size measurements (by image analysis) on samples at radial positions in which the restructuring was incomplete (at JRC-Karlsruhe, in the frame of GENTLE Project). Based on these new experimental data, I infer an exponential reduction of the average grain size with local effective burnup, paired with a simultaneous depletion of intra-granular fission gas driven by diffusion. The comparison with currently used models indicates the applicability of the herein developed model within integral fuel performance codes.

The content of this Chapter is based on:

D. Pizzocri, F. Cappia, L. Luzzi, G. Pastore, V.V. Rondinella, P. Van Uffelen, 2017. *A semi-empirical model for the formation and depletion of the high burnup structure in UO_2* . Journal of Nuclear Materials, 487, 23–29.

4.1 Introduction

In the rim zone of UO_2 nuclear fuel pellets, the combination of high local burnup ($>50\text{--}60 \text{ GWd tU}^{-1}$, i.e., high radiation damage and high fission product concentration) and low temperature ($<1000^\circ\text{C}$, i.e., limited thermal recovery of the radiation damage) drives a microstructural change, leading to the formation of the high burnup structure (HBS). This process always includes four characteristic phenomena [5], [89]–[92]: (i) dislocations pile-up forming an entangled network, (ii) the polygonisation/recrystallization of the original micrometric grains into sub-micrometric grains free of extended defects [93], [94], (iii) the decrease of the intra-granular fission gas concentration (depletion, [95]), and (iv) the formation of a novel population of inter-granular spherical bubbles [96]. These processes are not strictly sequential, but may be thought as (partially) concomitant.

The proper modelling of the formation and evolution of the HBS is a critical and challenging aspect of nuclear fuel performance analysis. The increase of porosity characteristic of the HBS formation contributes to decrease both thermal conductivity and elastic modulus, thus affecting the fuel rod thermo-mechanical performance. This, in turn, represents a potential concern for the safe operation of nuclear fuel to extended burnups, especially under design basis accident conditions.

Several semi-empirical models describing the formation and the evolution of the HBS are available in fuel performance codes [97]–[99]. In addition, Veshchunov and Shestak [100] provided a mechanistic model for the evolution of dislocations under irradiation up to high burnups. This model allows for a mechanistic interpretation of the temperature and burnup conditions under which the high burnup structure develops. Nevertheless, with the exclusion of the model by Veshchunov and Shestak [100], these models do not allow for the physical coupling between the phenomena occurring during the restructuring (*i-iv*).

A considerable number of experiments have been performed, strongly enlightening the physical processes involved in the formation of the HBS (see [92] for an overall review). Nevertheless, previous grain-size measurements [93], [94] only focused on the fully recrystallized/polygonised region. They concluded that the grain size of the recrystallized/polygonised structure is reasonably independent on the local burnup when the HBS formation is complete, while the extension of the HBS region depends on the local burnup and the details of the power history experienced by the sample.

In this Chapter, I focus on the development of a model describing the formation of the HBS. In particular, I consider a reduction of the grain size paired with the depletion of the gas from the fuel grains. The model is based on new measurements of grain sizes as a function of the local burnup which I performed at JRC-Karlsruhe on already available scanning electron microscopy (SEM) images.

I compared the results of the new model with experimental data⁹ and with the model currently employed in the TRANSURANUS code. Considering its promising results, the model has been recently implemented in the BISON code. The implementation in TRANSURANUS is planned in the near future in the framework of both INSPYRE and INERI Projects.

⁹ For this particular model, the validation strategy outlined in the Introduction had to be modified. In fact, the current model is semi-empirical and thus would require validation in the frame of fuel performance codes (i.e., comparison with integral irradiation experiments). Nevertheless, with minor exceptions, the available data at this engineering-scale arise from not well characterized irradiation histories and are thus impossible to be reliably simulated by FPCs. As a way around, I performed the simulations presented in this Chapter with a 0D stand-alone code representing only the average conditions of the considered integral irradiation experiments. This clearly hinders the validation of the model, which is thus referred to as a comparison to experimental data. More extensive validation of the model is envisaged as more (and better characterized) integral data become available.

4.2 Model description

4.2.1 Experimental measurements

This Section presents the experimental measurements of grain-size in the high burnup structure I performed at JRC-Kalrsruhe based on scanning electron microscopy (SEM) images.

In this work, the focus is on the formation process of the HBS. Thus, I measured the average grain size and its size-distribution in our samples, without limiting the measurement to the fully recrystallized/polygonised region. The characteristics of the fuel sample considered in this work are detailed by Manzel and Walker [101]. According to the fuel rod characteristics (Table 7), and the power history shown in Fig. 11, the average burnup and its radial profile (Fig. 12) were calculated with the TRANSURANUS code [14]. In addition, I calculated the radial profile of the effective burnup bu_{eff} (GWd tU⁻¹). This is defined as the burnup integrated under 1000°C [98], [102], i.e.,

$$bu_{\text{eff}} = \int H(T_{\text{th}}) dbu \quad (22)$$
$$H(T_{\text{th}}) = \begin{cases} 1 & T \leq T_{\text{th}} = 1000^\circ\text{C} \\ 0 & T > T_{\text{th}} \end{cases}$$

where bu (GWd/tU) is the burnup, and H is the Heaviside function.

The effective burnup is intended to account for the accumulation of radiation damage only when the temperature is low enough to thermally recover radiation damage. Hence, the effective burnup it is a more suitable quantity for defining a threshold for the HBS formation compared to the local burnup.

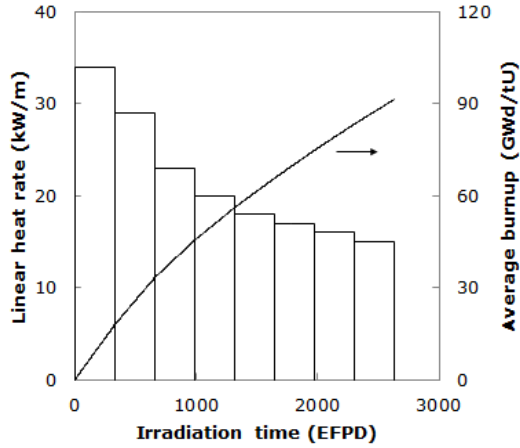


Figure 11. Power history and average burnup evolution in the fuel rod [101].

Table 7. Fuel pellet and rod design characteristics [101].

Fuel type	UO ₂
Pellet diameter (mm)	9.3
Enrichment (wt.%)	3.5
Fuel density (%TD)	95
Grain-size* (μm)	9-12
Diametrical gap (μm)	160-190
He fill gas pressure (MPa)	2.25

* 2D linear intercept

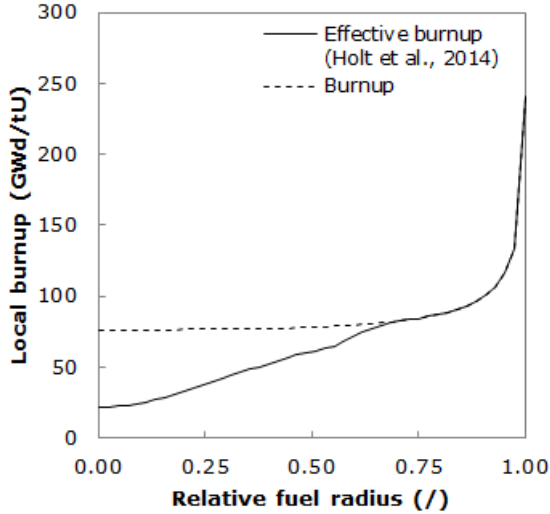


Figure 12. Effective burnup and burnup radial profiles of the fuel sample [101] calculated with the TRANSURANUS code using Holt et al. [102] correlation.

The average grain size was measured by means of the linear intercept method [103], [104] according to ASTM [105] recommendations. Considering the high density of grain boundaries, no etching of the samples was possible. Therefore, the SEM images used for the grain-size measurements are directly the fresh fractures of the fuel samples (Fig. 13). The use of non-etched images limits the accuracy of the grain-size measurement. Thus, the relative error assumed is the maximum suggested by ASTM [105] for linear intercept methods, i.e., $\pm 25\%$. The measured 2D mean linear intercept is converted to the 3D average spherical grain radius by multiplying the former by a factor $1.5/2$ [84], [105], [106].

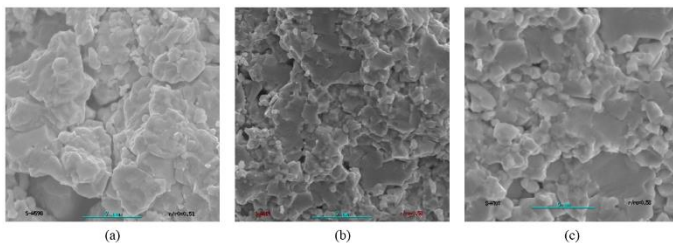


Figure 13. Fresh fracture SEM images of partially restructured UO_2 fuel samples.

Table 8 reports the results of the performed measurements. It is worth underlining that the measured grain size is not limited to a specific area of the image (e.g., the restructured HBS area), but represents an average of the entire area.

Fig. 14 reports the grain-size distributions of the measured samples. The histograms refer to the measured 2D linear intercepts, with no stereological correction applied. The reported Q-Q plot compares the experimental distributions with the theoretical log-normal distribution by plotting their quantiles against each other. Considering the uncertainties involved in the measurements, the agreement is considered satisfactory. Moreover, the experimental distributions show a very limited bimodality (bimodality coefficient 0.553, 0.608, and 0.531, for samples (a), (b), and (c), respectively, with threshold for bimodality being >0.555 [107]).

Table 8. Summary of the results of the grain-size measurements performed in this work. The measuring procedure is detailed in ASTM [105].

	UO ₂ sample (a)	UO ₂ sample (b)	UO ₂ sample (c)
Relative radial position (/)	0.51	0.58	0.58
Local burnup (GWd/tU)	78	79	79
Local effective burnup (GWd/tU)	61	69	69
2D average grain-size* (μm)	2.21	1.50	1.28
3D av. spherical grain radius (μm)	1.65	1.13	0.96
Uncertainty (%)	±25	±25	±25

* linear intercept

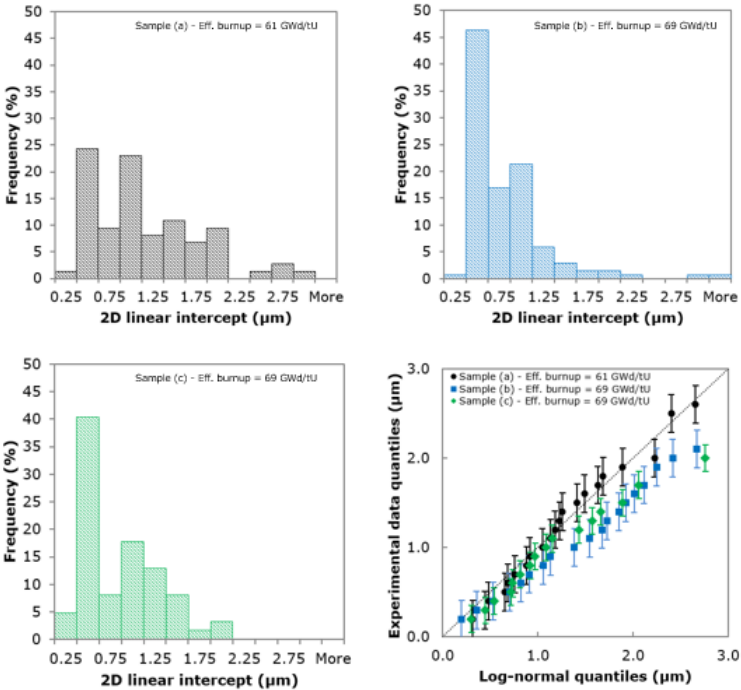


Figure 14. Grain-size distributions (2D linear intercept) of the measured samples. The Q-Q plot shows the agreement of the present data with a log-normal distribution.

4.2.2 Model derivation

The grain-size experimental data detailed in Section 4.2.1, together with data from Ray et al. [93] and Spino et al. [94] are herein used to derive an empirical relation between the fuel grain radius and the local effective burnup (defined by Eq. 22). In particular, the data (Fig. 15) are fitted with a first-order differential model with proper initial conditions (IC)

$$\frac{da}{dbu_{\text{eff}}} = -\frac{1}{\tau}(a - a_{\infty}) \quad (23)$$

$$\text{IC } a(bu_{\text{eff},0}) = a_0$$

where a (μm) is the 3D equivalent spherical grain radius, bu_{eff} (GWd/tU) is the local effective burnup, and $\tau = (7.0 \pm 0.8)$ GWd/tU and $a_{\infty} = (0.15 \pm 0.03)$ μm are determined by least-square method data fitting (Fig. 15). The initial condition given at $bu_{\text{eff},0} = 50$ GWd/tU [102] accounts for the effect of the initial grain-size a_0 (μm) on the formation of the high burnup structure [108].

As regards the thermal mechanism of grain growth, I adopt the model proposed by Botazzoli [106] (which is based on data from [85], [109]). According to the observation that no substantial grain growth occurs at high burnup and low temperature [85], the thermal grain growth is considered negligible in the HBS. Thus, the thermal grain growth model is effectively switched-off when $bu_{\text{eff}} > bu_{\text{eff},0}$ under normal operating conditions.

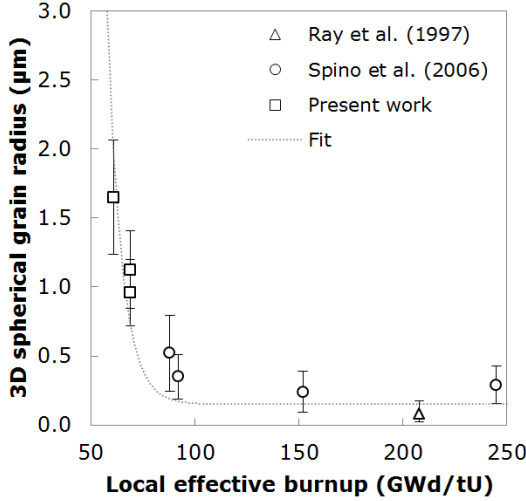


Figure 15. 3D spherical grain radius as a function of local effective burnup. The 3D spherical grain radius was measured considering both the non-restructured and the HBS region. Knowing the irradiation histories of the samples, the effective burnup was calculated by means of the TRANSURANUS code. The fitting curve is a decreasing exponential with initial value $9.0 \mu\text{m}$ [101] and asymptotic value $0.15 \mu\text{m}$. Data at higher local burnup from Ray et al., [93] and Spino et al., [94].

In fuel performance codes, intra-granular fission gas diffusion is usually modelled via an effective diffusion equation solved in the equivalent spherical domain [56], [88], [110].

For the purpose of modelling the fission gas behaviour in the high burnup structure, the present model also applies diffusion theory for a spherical grain. I adopt a relative radial coordinate, $\rho = r/a$, instead of the dimensional radial coordinate r (μm). Accordingly, the perfect sink boundary conditions (BC) is expressed at $\rho = 1$, rather than at $r = a(bu_{\text{eff},0})$. In addition, the time coordinate is represented by the effective burnup. Therefore, writing the fission gas diffusion equation in the spherical grain as

$$\frac{\partial c_1}{\partial bu_{\text{eff}}} = \frac{D}{a^2} \frac{1}{\rho^2} \frac{\partial}{\partial \rho} \rho^2 \frac{\partial}{\partial \rho} c_1 + yF$$

IC $c_1(\rho, bu_{\text{eff},0}) = c_1^0(\rho)$ (24)

BC $c_1(1, bu_{\text{eff}}) = 0$

$[\partial c_1 / \partial \rho]_0 = 0$

where c_1 (wt.%) is the intra-granular fission gas concentration, D ($\mu\text{m}^2 \text{GWd/tU}^{-1}$) is the effective diffusion coefficient, y (wt.% fiss $^{-1}$) is the fission yield, and F (fiss GWd/tU^{-1}) is the fission rate.

Brémier and Walker [111] discussed the applicability of single gas atom diffusion coefficients in the HBS zone (e.g., [4], [83]). They also considered the possibility of including fission gas trapping into and re-solution from intra-granular bubbles as lumped in the diffusion coefficient (i.e., using an effective diffusion coefficient, [25]). Considering the very limited knowledge available about intra-granular fission gas behaviour in the HBS [112], I choose a simple a-thermal diffusion coefficient in agreement with Brémier and Walker [111], namely $D = 7.8 \cdot 10^{-3} F$. Combining Eqs. 23, 24 leads to a model for the coupled grain size evolution and depletion of intra-granular fission gas (xenon) in the HBS. In this model, gas depletion results from the reduction of grain radius with increasing burnup (Eq. 23) as the diffusion rate D/a^2 (Eq. 24) correspondingly increases.

4.3 Comparison with experimental data

In Fig. 6, the intra-granular concentration of xenon calculated with the present model is compared to the EPMA data discussed by Walker [95]. Considering the uncertainties in both the experimental data (obtained from 38 different UO_2 fuel samples, with different irradiation histories) and in the model parameters, the agreement appears to be satisfactory. For comparison, also the results of the model by Lassmann et al. [97] are shown.

Figure 6 also shows the impact of the initial grain size. If higher initial grain radii are considered, the model predicts a delayed depletion of the intra-granular xenon, thus a delayed formation of the HBS. This trend is coherent with the one experimentally observed by Noirot et al. [113].

These results indicate that gas depletion in the HBS can be described by diffusion theory as grain size decreases during recrystallization/polygonisation. In a semi-empirical approach, the present model combines an empirical relation for grain size evolution to physics-based calculation of diffusion.

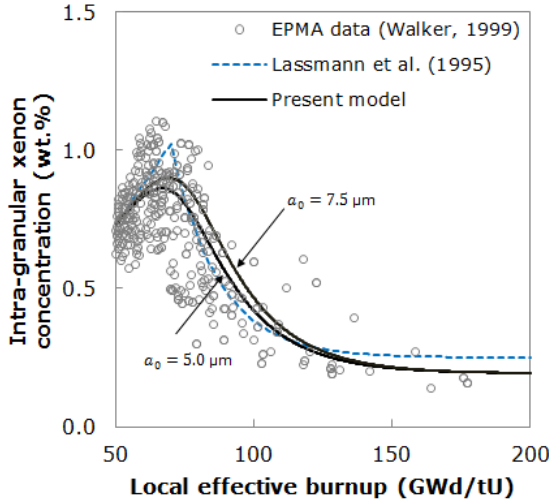


Figure 16. Comparison of the intra-granular xenon concentration measured by EPMA (Walker, 1999, [95]) on several samples with the calculation of the present model, as a function of local effective burnup. The impact of assuming a different initial grain radius, a_0 , is also shown, with bigger radii delaying the xenon depletion. The comparison with the model from Lassmann et al., [97] is reported.

To corroborate the model assessment, I comparatively discuss the present model with those available in the open literature, with focus on those models suitable to be used in fuel performance codes. Among the others, four models are here discussed. In chronological order: Lassmann et al. [97], Khvostov et al. [98], Veshchunov and Shestak [100], and Lemes et al. [99].

The model by Lassmann et al. [97] describes the depletion like a decay process, without relating it to the recrystallization/polygonisation of the grains, essentially because very limited quantitative knowledge about the recrystallization/polygonisation processes was available. Moreover, it considered a purely burnup dependent threshold for the formation of the HBS, neglecting the influence of temperature. This pragmatic model is thus derived directly from the depletion data [95]. Apart from these limitations, the model results were satisfactory (Fig. 16), and it is used in the TRANSURANUS code up to now [14].

The model of Khvostov et al. [98] describes the formation of the HBS with a generic KJMA (Kolmogorov-Johnson-Mehl-Avrami) approach, which is

applicable to any structural change. The process is started based on the effective burnup. The KJMA is based on a phenomenological description of the HBS formation as starting at the grain boundaries. This is a reasonable assumption in general, but is not the case for fuel with big initial grain size [92], [108]. In addition, the temperature limits applied to assess the effective burnup are questionable [102]. The depletion of xenon is treated as in the present model (Eq. 24) but applying a different diffusion coefficient. This model represents a significant improvement in the description of the HBS formation, and is grafted in the overall fission gas behaviour model of the START-3 and the FALCON codes [72].

The mechanistic model from Veshchunov and Shestak [100] describes the combined evolution of intra-granular point defects and extended defects (i.e., dislocations, bubbles, and pores) under irradiation up to high burnups. In particular, the dislocation density predicted by this model is in agreement with the values required for the recrystallization/polygonisation of the HBS grains [114]. The application of this mechanistic model is limited by the underlying mean-field assumption for the concentration of vacancies and interstitials. This assumption is valid for intra-granular defects, but may be inadequate in the HBS, where the concentration of sinks for point defects is much higher. The model is used in the MFPR code [115].

The model from Lemes et al. [99] is an extension of the pragmatic approach from Lassmann et al. [97], empirically describing several phenomena occurring in the HBS. As regards the formation process, the model tracks back to Lassmann et al. [97] work. The model is used in the DIONISIO code.

The model developed in this work is based on the assumption that a reduction in the average grain size can represent both the dislocation pile-up and the actual polygonisation/recrystallization. In fact, dislocations and grain boundaries can be considered as perfect sinks for fission gas [92], [112]. The reduction of the average grain size is calculated as a function of the effective burnup. The underlying physical interpretation is that HBS formation can be seen as a phase transition from a structure with low surface to volume ratio (i.e., big grain size $\approx 10\ \mu\text{m}$) to a structure with much higher surface to volume ratio (i.e., small grain size $\approx 300\ \text{nm}$), the second being able to endure more radiation damage (represented by the effective burnup). In this picture, the depletion of intra-granular fission gas and the formation of new bubbles can be related to the increased surface to volume ratio, which enhances gas diffusion from within the grains, thus promoting HBS pore growth. This physical consideration is accounted in Eq. 23.

Moreover, the two parameters appearing in Eq. 23 are both directly determined from experimental measurements of the grain size. Such simplicity of the model represents an attractive feature in regard to the effective application to integral fuel performance codes. It will be complemented with a model for the evolution of the HBS pore size, which is under development.

The present model represents the depletion of intra-granular fission gas as triggered by the reduction of the average grain size (Eq. 24). Applying a typical value of the diffusion coefficient [111], the model is able to reproduce the experimental EPMA results with a satisfactory degree of accuracy (Fig. 16). The slight over-prediction of the intra-granular xenon concentration may be due to other intra-granular release mechanisms not represented by this purely diffusion-based model (e.g., dislocation sweeping), as well as to the uncertainties pertaining to the diffusion coefficient evaluation itself.

The model is also able to predict the delayed HBS formation observed in fuel with big initial grain-size [108]. In view of the encouraging results, the model has been implemented in the BISON code. The implementation in TRANSURANUS is planned in the near future in the framework of both INSPYRE and INERI Projects.

4.4 Closing remarks

In this Chapter, I proposed a model to describe the formation of the HBS which embraces the polygonisation/recrystallization process and the depletion of fission gas intra-granular concentration, describing them as inherently related.

I performed new grain-size measurements on UO_2 samples in which the restructuring was not complete. Based on these measurements, I describe the reduction of the average grain size as an exponential function of the effective burnup. Such empirical-based component of the model pairs with a physics-based description of the simultaneous intra-granular fission gas depletion driven by diffusion. The comparison of model results to EPMA experimental data of intra-granular gas concentration points out a satisfactory prediction of the fission gas depletion.

Simplicity of the presented model (limited amount of equations and parameters) is an attractive feature for its effective application to integral fuel performance codes. In particular, the model has been included in the BISON code and its implementation in the TRANSURANUS code is planned.

Further extension of this model will consider the evolution of the HBS pore population, retaining a reasonable complexity along with a physically-sound description of the processes involved. Moreover, application of a similar model for the formation of HBS in MOX fuel is of interest in perspective.

5. Diffusion algorithm

Abstract

In this Chapter, I present PolyPole, a numerical algorithm developed to solve the partial differential equations describing the diffusion of gas towards the grain boundaries in time-varying conditions. The diffusion process involves single gas atoms diffusion in conjunction with trapping in and re-resolution from intra-granular bubbles, and is described mathematically by a system of two partial differential equations (PDE). For applications in fuel performance codes, the assumption of instantaneous equilibrium between trapping and resolution (equilibrium hypothesis or quasi-stationary approximation) is generally adopted. Under this assumption, the process can be described by a single diffusion equation with an effective diffusion coefficient, thereby simplifying the numerical treatment. Existing numerical algorithms that offer computational costs sufficiently low for application to integral fuel performance codes rely on the quasi-stationary approximation. In this Chapter, I present a new numerical algorithm (i.e., PolyPole) capable of accurately solving the diffusion problem with the quasi-stationary approximation (PolyPole-1) and without (i.e., solving the system of coupled PDEs, PolyPole-2). The PolyPole algorithms are verified by comparing the results to a reference finite difference solution over a large number of randomly generated operation histories including postulated RIA transients.

The content of this Chapter is based on:

D. Pizzocri, C. Rabiti, L. Luzzi, T. Barani, P. Van Uffelen, G. Pastore, 2016. *PolyPole-1: An accurate numerical algorithm for intra-granular fission gas release*. Journal of Nuclear Materials, 478, 333-342.

G. Pastore, D. Pizzocri, C. Rabiti, T. Barani, P. Van Uffelen, L. Luzzi, *An effective numerical algorithm for intra-granular fission gas release during non-equilibrium trapping and resolution*. Accepted by Journal of Nuclear Materials (2018).

5.1 Introduction

The first and basic step of fission gas release (FGR) and gaseous swelling is gas atom transport to the grain boundaries. It follows that modelling of this process is a fundamental component of any fission gas behaviour model in a fuel performance code. Intra-granular fission gas transport occurs by thermal and irradiation-enhanced diffusion of single gas atoms, coupled to trapping in and irradiation-induced resolution from intra-granular bubbles. Diffusion of intra-granular bubbles becomes relevant at high temperatures, above $\sim 1800^\circ\text{C}$ [23], [116]. Thus, modelling the process of gas transport to the grain boundaries calls for the treatment of different concomitant processes, namely, diffusion coupled with trapping and resolution of gas atoms. Extensive literature deals with the evaluation of the parameters characterizing these mechanisms, both experimental and theoretical work, e.g., [23], [36], [117]–[119]. Rather, in this Chapter, I deal with the numerical problem associated with the computational solution of the equations describing the process. Clearly, this problem has an enormous practical importance for fission gas behaviour calculations in fuel performance analysis. In fact, the solution of these equations is required in each mesh point of the engineering FPCs. Efficient solutions are thus mandatory to guarantee acceptable overall computational times.

Speight [25] proposed a simplified mathematical description of intra-granular fission gas release. He lumped the trapping and re-resolution rates into an effective diffusion coefficient, $D_{\text{eff}} = \alpha / (\alpha + \beta) D$, restating the mathematical problem as purely diffusive, i.e., going simplifying the system

$$\frac{\partial c_1}{\partial t} = D \nabla^2 c_1 - \beta c_1 + \alpha m + yF \quad (25)$$

$$\frac{\partial m}{\partial t} = +\beta c_1 - \alpha m$$

to

$$\frac{\partial c_{\text{tot}}}{\partial t} = \frac{\alpha}{\alpha + \beta} D \nabla^2 c_{\text{tot}} + yF \quad (26)$$

Such simplification implies the assumption of equilibrium between trapping and re-resolution (quasi-stationary approach). The formulation of Speight is universally adopted for models employed in fuel performance codes, e.g., [27], [59], [60], [88], [110], [120], [121]. In addition, the assumption of spherical grain geometry [122] is applied. The solution of the diffusion

equation for constant conditions is well known. Nevertheless, time-varying conditions are involved in realistic problems. Therefore, the solution for time-varying conditions is the issue of interest for applications to fuel performance analysis, which calls for the development of dedicated numerical algorithms. As briefly stated above, given the very high number of calls of each local model (such as the fission gas behaviour model) in a fuel performance code there is a requirement of low computational cost (in addition to the requirement of suitable accuracy for the numerical solution). Clearly, the numerical solution of the diffusion equation in time-varying conditions may be obtained using a spatial discretization method such as a finite difference scheme. However, the high associated computational effort can make a space-discretization based solution impractical for application in fuel performance codes. Several alternative algorithms, which provide approximate solutions for Eq. 26 at high speed of computation and can be used in fuel performance codes, have been developed [88], [110], [120], [123].

In this Chapter, I propose a new numerical algorithm for the accurate and fast solution of both Eqs. 25, 26 in time-varying conditions, which I refer to as PolyPole. I verify the algorithms for Eqs. 25 and Eq. 26 with specifically designed numerical experiments. The verification results of PolyPole-1 have been compared to state-of-the-art algorithms available in FPCs. On the other hand, PolyPole-2 represents the first available algorithm for the solution of Eqs. 25.

As demonstrated in the following, the solution of this general system of PDEs (Eqs. 25) is of high engineering importance related to modelling fission gas behaviour in the HBS during RIA accidents. Fission gas retention in the form of bubbles leads to fuel swelling which promotes pellet-cladding mechanical interaction (PCMI), and the concomitant fission gas release (FGR) to the fuel rod free volume increases the rod internal pressure, both processes affecting the mechanical behaviour of the cladding. Moreover, gas release and precipitation in bubbles affect the thermal conductance of the fuel-cladding gap and the fuel thermal conductivity, respectively, and consequently the temperature distribution in the fuel pellet [1], [23]. Besides, beyond a certain burnup level, the fuel swelling rate increases due to the build-up of gaseous porosity associated with the formation of the high burn-up structure (HBS) [5], [36], [116], [124]. Ultimately, the increased fuel gaseous swelling can severely impact the fuel-cladding contact pressure during PCMI [125], [126]. In particular, PCMI can reduce the safety margin for cladding failure during reactivity-initiated accident (RIA) transients, and

is considered by regulators in acceptance criteria for fuel behaviour under RIAs in light water reactors (LWR) [126], [127]. Furthermore, considerable FGR has been observed in RIA simulation tests, which would increase cladding loading and the risk of creep-induced cladding rupture by ballooning during a RIA transient [117], [128].

5.2 Algorithm development

In this Section, I present the new numerical algorithm solving Eqs. 25 in time-varying conditions. I refer to this new algorithm as PolyPole-2. A simplified version of the same algorithm is also presented, referred to as PolyPole-1, solving Eq. 26. PolyPole-1 represents a specific case of PolyPole-2. To simplify the notation, it is convenient to re-write Eqs. 25 in a vectorial form as

$$\frac{\partial}{\partial t} u = (\mathbf{D} + \mathbf{E})u + \mathbf{S} \quad (27)$$

in which $u = [c_1 \ m]$ and $\mathbf{S} = [yF \ 0]$ are vectors containing the gas concentrations and the fission source term, respectively. The diffusion $\mathbf{D}$ and exchange $\mathbf{E}$ operators are defined as

$$\mathbf{D} = \begin{bmatrix} D\nabla^2 & 0 \\ 0 & 0 \end{bmatrix} \quad (28)$$

$$\mathbf{E} = \begin{bmatrix} -\beta & +\alpha \\ +\beta & -\alpha \end{bmatrix} \quad (29)$$

The boundary conditions of Eq. 25 are $u(r = a, t) = 0$ and $\partial u / \partial r|_{(r=0)} = 0$. The initial condition is $u(r, t = 0) = u_0(r) = [c_0(r) \ m_0(r)]$.

With this notation it is clear that PolyPole-1 is a particular case of PolyPole-2. In fact, under the quasi-stationary approximation, the diffusion-exchange operator $(\mathbf{D} + \mathbf{E})$ is replaced by $D\alpha/(\alpha + \beta)\nabla^2$, $\mathbf{S}$ is replaced by yF and u is replaced by $c_{tot} = c_1 + m$ [25].

Here, the objective is to find an approximate solution u^* for the case of time-varying $(\mathbf{D} + \mathbf{E})$ and $\mathbf{S}$. With the *ansatz* that the solution can be approximated as

$$u^*(r, t) \approx \sum_{n=1}^{\infty} z_n^*(t) \psi_n(r) = \sum_{n=1}^{\infty} \mathbf{P}_n z_n(t) \psi_n(r) \quad (30)$$

in which $z_n(t)$ and $\psi_n(r)$ are the time coefficients and the spatial modes, respectively, of the solution for constant conditions (exponential in time and cardinal sine in space). The operator $\mathbf{P}_n$ embodies the correction for time-varying conditions and is applied to each mode of the solution. The

formulation adopted in this work for this operator is made of two distinct polynomials of second order J

$$\mathbf{P}_n = \begin{bmatrix} 1 + \sum_{j=1}^J p_j dt^j & 0 \\ 0 & 1 + \sum_{j=1}^J q_j dt^j \end{bmatrix} \quad (31)$$

This definition of $\mathbf{P}_n$ reduces to a single polynomial correction factor for the PolyPole-1 algorithm [56].

The problem of finding an approximate solution for time-varying conditions is basically shifted to the problem of finding the coefficients of the polynomials in $\mathbf{P}_n$. To calculate the coefficients p_j and q_j , $2J$ equations are needed. This set of equations is obtained by sampling the time-varying operators, $\mathbf{D}(t)$, $\mathbf{E}(t)$ and $\mathbf{S}(t)$, at J uniformly distributed instants along the time-step dt . The sets of sampled values, $\mathbf{D}[j]$, $\mathbf{E}[j]$ and $\mathbf{S}[j]$, contain the information on the variation of the operators along the time step and are used to calculate the corrective polynomials as follows.

The time coefficients $z^*(t)$ defined by Eq. 30 are assumed to satisfy Eq. 27 (here written in vector notation) at the sampling times $t[j]$, $t_i \leq t[j] \leq t_{i+1}$

$$\left. \frac{\partial(\mathbf{P}_n z_n)}{\partial t} \right|_{t[j]} = \langle \psi_n | \mathbf{D}[j] + \mathbf{E}[j] \rangle (\mathbf{P}_n z_n) + \langle \psi_n | \mathbf{S}[j] \rangle \quad (32)$$

Eq. 32 defines a linear system of $2J$ equations for the polynomial coefficients p_j and q_j , and is used to determine the polynomial correction operator $\mathbf{P}_n$. The time coefficients of the analytic solution for constant conditions, z_n , are calculated using averages along the time step of the sampled values $\mathbf{D}[j]$, $\mathbf{E}[j]$ and $\mathbf{S}[j]$.

The PolyPole-2 (and PolyPole-1, identically) solution is then reconstructed as a linear combination of the spatial with the corrected time coefficients, according to Eq. 30. The series is approximated by a finite number of terms (M , number of modes). The value of M is determined based on a D'Alembert-like remainder criterion. Usually, only the spatial average over the fuel grain of u^* is of interest in fuel performance codes, and for verification purpose I consider only this spatial average.

5.3 Algorithm verification

In this Section, I present a numerical experiment with the twofold objective of (i) verifying the PolyPole-2 solution in RIA-like transients and verifying the PolyPole-2 solution in operational conditions, and (ii) verifying the PolyPole-1 solution in operational conditions.

5.3.1 PolyPole-2 numerical verification

The numerical experiment consists of the application of the PolyPole-2 algorithm to the numerical solution of Eqs. 25 for a large number of randomly generated histories. The results from the PolyPole-2 algorithm are compared to the reference solution provided by a finite difference (FD) algorithm. The FD algorithm has been developed as part of this work in order to provide a reference numerical solution of Eqs. 25 and Eq. 26 as well. The developed FD algorithm is fully implicit first order in time and second order in space with an adaptive time step control. The output solution is calculated with a fully implicit scheme (backward Euler). At each time step, also the explicit solution (forward Euler) is calculated, and the absolute difference between the explicit and the implicit solutions is used to obtain an estimation of the second-order error. The next time step is then computed based on this estimation. This time step criterion ensures that the second-order error of the FD solution is kept below a fixed tolerance. Details of this method can be found in [129]. This algorithm can be applied to obtain a reference solution of Eqs. 25, 26 (in principle, up to any tolerance) for piecewise-linear operation histories. The considered histories, both for RIA-like and operational transients, are in terms of temperature and fission rate, from which the time-dependent parameters in the diffusion and exchange operators, $\mathbf{D}(t)$ and $\mathbf{E}(t)$, and in the source term, $\mathbf{S}(t)$, are calculated¹⁰.

The figure of merit chosen for this numerical experiment is the fractional intra-granular fission gas release at the end of the considered history, defined as

¹⁰ I use the expressions for the diffusion coefficient, D , by Turnbull et al., [148], the re-solution rate, α , by White and Tucker [60], and the trapping rate, β , by Ham [39]. A value of 0.3 is considered for the fission yield, y . For the purpose of this numerical experiment, as long as values and dependencies are realistic, the specific choices are arbitrary.

$$\chi = \frac{\int_0^{t_{end}} yF(t')dt' - c_1(t_{end}) - m(t_{end})}{\int_0^{t_{end}} yF(t')dt'} \quad (33)$$

where t_{end} (s) is the end time of each history considered.

In order to verify the PolyPole-2 algorithm in RIA-like conditions, I considered two sets of representative random histories. The first one, which is referred to as centre, is designed to represent the intra-granular gas diffusion during a RIA transient in the centre of the pellet. The second one, referred to as rim, is designed to be representative of the intra-granular gas behaviour during a RIA transient in the HBS at the rim of the pellet. These two sets of histories differ in the grain size a , which is assumed as 5.0 μm for the centre and as 150 nm for the rim, respectively.

Moreover, since the focus of the verification is only on the RIA transient and not on the base irradiation up to the transient itself (during which the quasi-stationary approximation is valid), I consider an initial gas concentration corresponding to the gas generated during an irradiation up to 50 GWd tU^{-1} . This initial gas concentration is assumed to be stored in the grains in the centre (70% as single atoms and 30% as atoms in bubbles) [60]. In the rim, the initial concentration is set to $10^{26} \text{ at m}^{-3}$, corresponding to the asymptotic gas concentration achieved in the HBS grains [13], [111]. The RIA-like random histories have the following characteristics (for both centre and rim):

- The peak width, τ_{RIA} , is a random variable, sampled uniformly over 20-60 ms.
- The specific energy, E_{RIA} , is a random variable, sampled uniformly over 400-800 J $(\text{gUO}_2)^{-1}$.
- The maximum specific power, P_{RIA} (W g^{-1}), is calculated as $P_{\text{RIA}} = 62.5 E_{\text{RIA}}/\tau_{\text{RIA}}$ [7], [130].
- The power pulse shape is calculated according to the Nordheim-Fucks model [7], [130].
- The initial temperature, T_0 (K), is calculated as $T_0 = 883 (5 \cdot 10^{-4} E_{\text{RIA}} + 0.7772)$ (uniform in the range 863-1039 K) [130].
- The maximum temperature, T_{max} (K) is calculated as $T_{\text{max}} = 2564 (5 \cdot 10^{-4} E_{\text{RIA}} + 0.7772)$ (uniform in the range 2506-3017 K) [130].
- The maximum temperature is reached after $2 \tau_{\text{RIA}}$, i.e., at the end of the power pulse. The temperature increase in time is calculated as the integral of the power pulse.

- The end time is fixed at 300 ms after the beginning of the RIA-transient.
- The final temperature, T_{end} (K), is calculated as $T_{end} = 1227 (5 \cdot 10^{-4} E_{RIA} + 0.7772)$ (uniform in the range 1200-1444 K) [130].
- Prior to RIA-transient each history has a plateau of 100 hours at T_0 , to ensure the establishing of equilibrium between single gas atoms and gas in bubbles.

The results of the RIA-like numerical experiment are presented in Fig. 17. Each data point in these figures corresponds to one of the randomly generated operation histories and represents the intra-granular fission gas release (Eq. 33) obtained with the PolyPole-2 algorithm versus the reference FD solution. The deviation from the 45° line is a measure of the accuracy of PolyPole-2. Results appear to be distributed between two clusters of data points. The higher intra-granular fission gas release results (5 to 15%, approximately) are the simulations with the smaller grain radius of 0.1 μm . With a small grain radius diffusion to grain boundaries occurs at a higher rate (considering a pure diffusion problem as a first approximation, the diffusion rate is D/a^2), thus leading to higher intra-granular fission gas release compared to behaviour with larger grains.

In Fig. 17 I also report the results from the FD solution of Eq. 25 for the same input histories. The comparison highlights the effect of the quasi-stationary approximation (implied in Eq. 25) for the analysis of RIA transients. Results demonstrate that the quasi-stationary approximation is inadequate in describing the fast transients to relatively high temperatures (RIAs) considered in this study. In particular, the approximation leads to a strong (nearly 100%) under-prediction of the intra-granular fission gas release.

Besides accuracy, speed of computation is an essential feature for an algorithm to be effectively employed in a fuel performance code. The computational time (i.e., the time taken for the analysis of a single operation history) for the PolyPole-2 and FD algorithms are compared in Fig. 18. The computational time of PolyPole-2 is approximately two orders of magnitude lower than that of the FD algorithm. Such efficiency of computation, combined with the demonstrated accuracy, makes PolyPole-2 suitable for implementation in any fuel performance code.

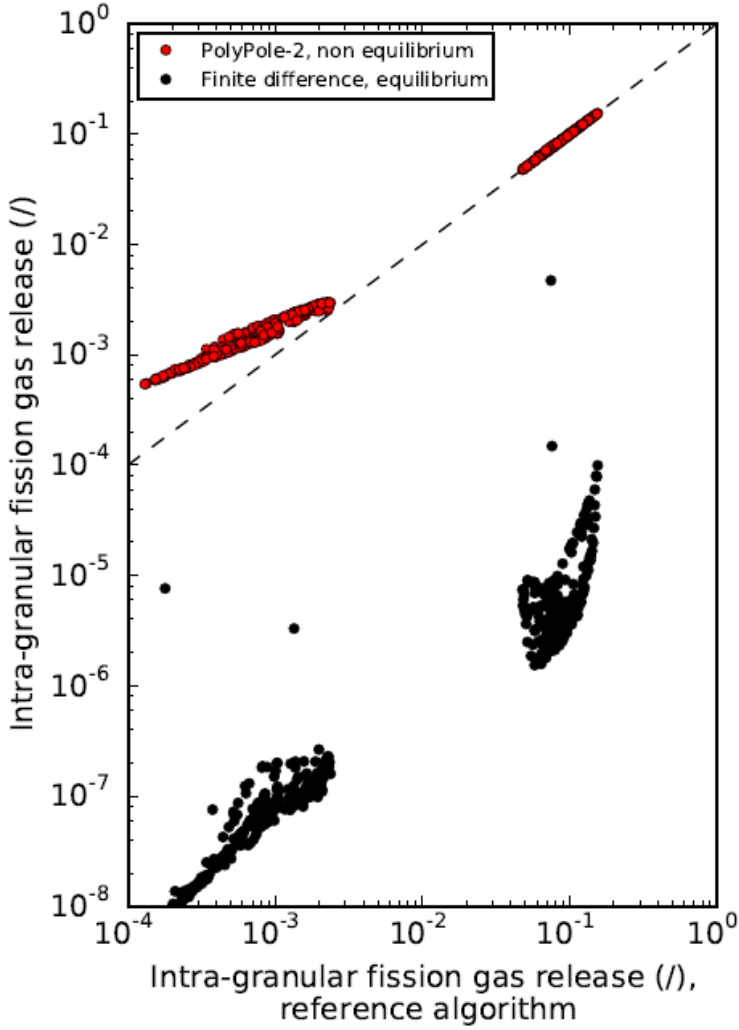


Figure 17. Comparison between intra-granular fission gas release calculated by the reference algorithm for the general PDE system (non equilibrium) and the PolyPole-2 results – red points – and the FD results for the quasi-stationary approximation (equilibrium) – black points –. Each data point corresponds to a calculation with randomly generated RIA conditions. The distance from the 45° dashed line is a graphical measure of accuracy.

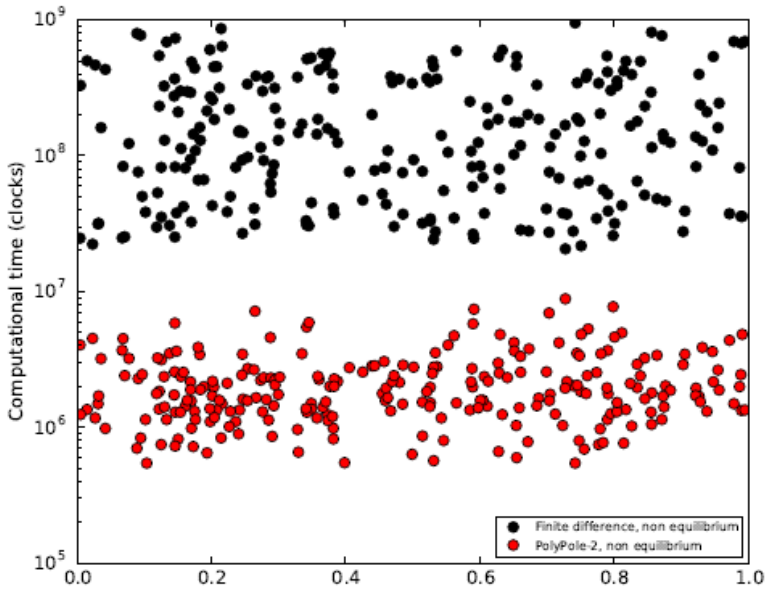


Figure 18. Comparison between the computational times associated with the FD and PolyPole-2 algorithms for the solution of the general PDE system. Each data point corresponds to a calculation with randomly generated RIA conditions.

In order to verify PolyPole-2 in operational conditions, I compare it with the PolyPole-1 algorithm (independently verified, as discussed in Section 5.3.2). The numerical experiment for this comparison is made of 1000 random histories with the following characteristics:

- Each individual history is piecewise-linear with varying temperature and fission rate.
- In each individual history, the following quantities are considered as random variables (sampled from uniform distributions)
 - Number of linear steps (1-50).
 - Time duration of each linear steps (0-1000 h).
 - Temperature (300-2700 K).
 - Fission rate ($0-3 \cdot 10^{19} \text{ fiss m}^{-3} \text{ s}^{-1}$).

Diffusion coefficient, re-solution rate, trapping rate and production rate are the same used in the RIA-transient numerical experiment.

Clearly, the quasi-stationary approximation is a particular case of the general formulation, and under conditions where the approximation is valid, PolyPole-2 needs to reproduce the quasi-stationary solution. The results of the numerical experiment are presented in Fig. 19. Each data point in this figure corresponds to one of the randomly generated operation histories and represents the intra-granular fission gas release (Eq. 33) obtained with the PolyPole-2 algorithm versus the reference (PolyPole-1) solution. The deviation from the 45° line is a measure of the accuracy. Results of the numerical experiment indicate that the solutions are very close for all of the 1000 random histories considered and therefore the PolyPole-2 solution correctly reduces to the quasi-stationary solution for operational conditions.

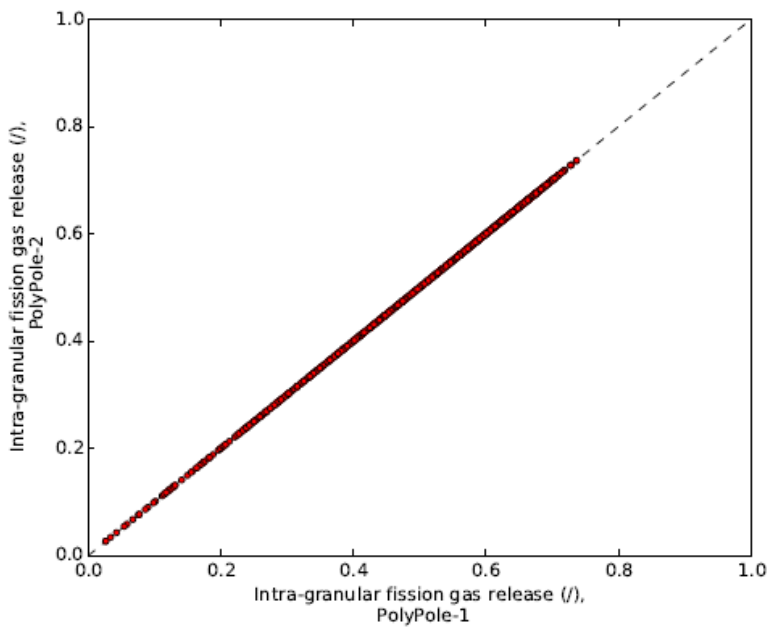


Figure 19. Comparison between the values of intra-granular fission gas release calculated with the PolyPole-2 and the PolyPole-1 algorithms. Each data point corresponds to a calculation with randomly generated operational conditions.

5.3.2 PolyPole-1 numerical verification

For the purpose of verifying PolyPole-1, I developed a dedicated numerical experiment, and applied to PolyPole-1 and other two semi-analytic algorithms for the solution of Eq. 26 currently used in fuel performance codes, namely: (1) URGAS [88]; (2) FORMAS¹¹ [120], [123]¹². The numerical experiment consists of application of each algorithm to the numerical solution of Eq. 26 for 1000 randomly generated operation histories. Results from the three semi-analytic algorithms are compared to the reference FD solution¹³.

The considered operation histories are in terms of temperature and fission rate, from which the time-dependent parameters of Eq. 26, i.e., $D_{\text{eff}}(t)$ and $yF(t)$, are calculated¹⁴ and applied to the numerical algorithms by the program. The figure of merit for testing and comparing the algorithms is the fractional intra-granular fission gas release at the end of the considered operation history, defined by Eq. 33.

The randomly generated operation histories have the following characteristics:

- Each individual history is piecewise-linear with varying temperature and fission rate.
- In each individual history, the following quantities are considered as random variables (sampled from uniform distributions)
 - Number of linear steps (1–11).
 - Time duration of each linear step (0–100 hours).
 - Temperature (500–2000 K).
 - Fission rate ($0\text{--}3\cdot 10^{19}$ fiss $\text{m}^{-3} \text{s}^{-1}$).

¹¹ Among the various versions of the FORMAS algorithm, we use the FORMAS algorithm with four exponential terms from [31].

¹² For this numerical experiment, we consider a second-order corrective polynomial (Eq. 31) and a 10^{-7} limiting value for the D'Alembert remainder (Eq. 30).

¹³ For this numerical experiment, we set the tolerance of the FD scheme to 10^{-6} .

¹⁴ The temperature- and fission rate dependent diffusion coefficient from [11] is used as $D_{\text{eff}}(t)$. $yF(t)$ is calculated as the fission rate times the yield of fission gas atoms ($y = 0.3$). For the purpose of this numerical experiment, as long as dependencies are realistic, the specific choices are arbitrary.

With these principles, the numerical experiment approximately covers the whole range of intra-granular fission gas release.

The results of the numerical experiment are presented in Figs. 20-22 for the URGAS, FORMAS, and PolyPole-1 algorithms. Each data point in these figures corresponds to one of 1000 randomly generated operation histories and represents the intra-granular fission gas release (Eq. 17) obtained by the considered semi-analytic algorithm versus the reference FD algorithm. The deviation from the 45° line is a measure of the accuracy. Results of the numerical experiment indicate that all the three semi-analytic algorithms are adequate for a fuel performance code, also in view of the uncertainties involved in the modelling of intra-granular fission gas diffusion [17], [88].

To investigate in finer detail the accuracy of the three semi-analytic algorithms, Fig. 23 shows the relative error of the solution obtained with each algorithm with respect to the FD reference solution. The efficiency and accuracy of the URGAS and FORMAS algorithms were previously analysed by Lassmann and Benk [88]. Although a more recent version of FORMAS is considered here, the results in Fig. 23 are consistent with the conclusions of Lassmann and Benk [88] that (i) the FORMAS algorithm is superior to the URGAS algorithm at fission gas release above $\chi \approx 0.05$, and (ii) the FORMAS algorithm presents a deficiency for low values of χ , which is ascribed to an approximation involved in the method. Although both algorithms were evaluated as sufficient to be used in a fuel performance code, drawbacks were attributed to each of them in line with the conclusions above. Choice of one of the two algorithms based on the specific applications was recommended.

The results in Fig. 23 indicate that the PolyPole-1 algorithm represents a significant path forward in this respect. First, it is evident that the overall accuracy of PolyPole-1 is vastly superior to both FORMAS and URGAS. Indeed, the accuracy of the FORMAS algorithm may be improved by an increase in the number of parameters [123]. However, the accuracy of the PolyPole-1 algorithm can also be further improved by increasing the order of the polynomial corrective term (Eq. 31) and/or by decreasing the limiting value for the approximation of the series (Eq. 30). Furthermore, PolyPole-1 overcomes the deficiency at low FGR that characterizes the FORMAS algorithm. Remarkably, the relative error associated with PolyPole-1 is highly consistent over the whole range of intra-granular fission gas release.

Besides accuracy, speed of computation is an essential feature for an algorithm to be effectively employed in a fuel performance code. The computational time (i.e., the time taken for the analysis of a single operation

history) for the three semi-analytic algorithms and all histories considered in the numerical experiment is illustrated in Fig. 24. PolyPole-1 requires a computational time similar to the other algorithms, which are successfully used in fuel performance codes. Such efficiency of computation, combined with the demonstrated accuracy, makes PolyPole-1 suitable for implementation in any fuel performance code. The computational time for the finite difference solution is also shown. Clearly, if a spatial discretization method such as a FD method is used to solve the intra-granular fission gas release problem in a fuel performance code, the associated higher computational time can result in significantly decreased speed of computation of fuel rod analysis, even with modern computational resources. This highlights the value of developing numerical algorithms that allow for a faster computation while preserving accuracy, such as PolyPole-1.

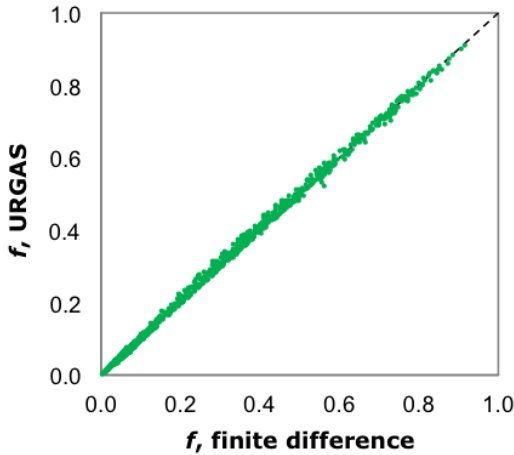


Figure 20. Comparison between the values of intra-granular fission gas release calculated by the URGAS algorithm and by the reference finite difference algorithm. Each data point corresponds to a calculation with randomly generated conditions.

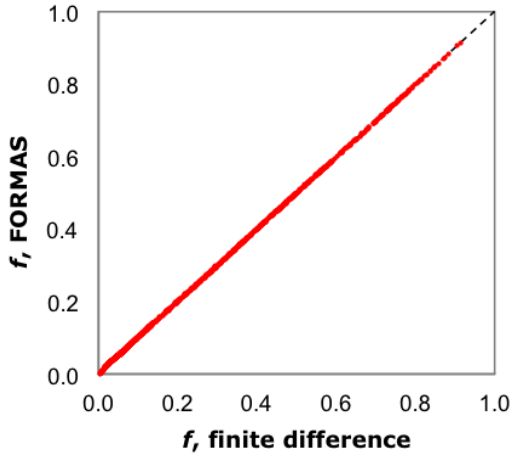


Figure 21. Comparison between the values of intra-granular fission gas release calculated by the FORMAS algorithm and by the reference finite difference algorithm. Each data point corresponds to a calculation with randomly generated conditions.

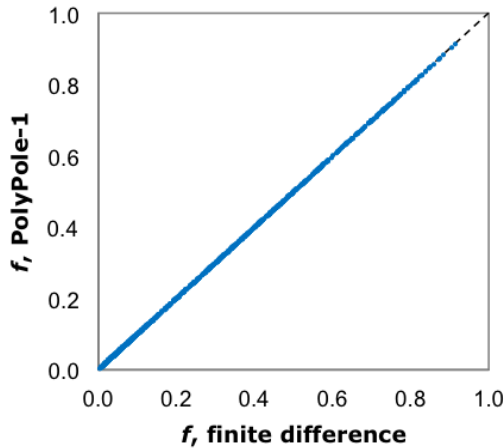


Figure 22. Comparison between the values of intra-granular fission gas release calculated by the PolyPole-1 algorithm and by the reference finite difference algorithm. Each data point corresponds to a calculation with randomly generated conditions.

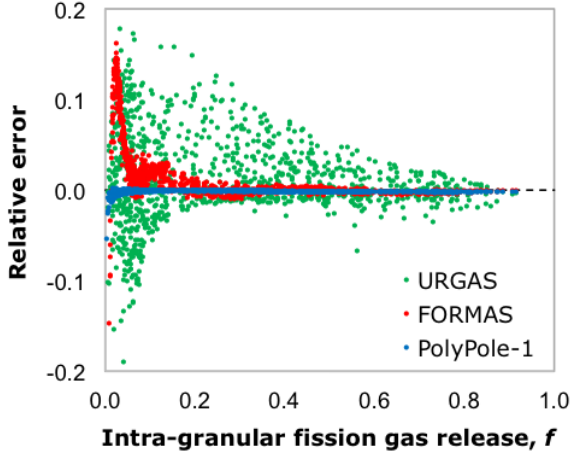


Figure 23. Comparison between the URGAS, FORMAS and PolyPole-1 algorithms in terms of relative error with respect to the reference finite difference algorithm. Each data point corresponds to a calculation with randomly generated conditions.

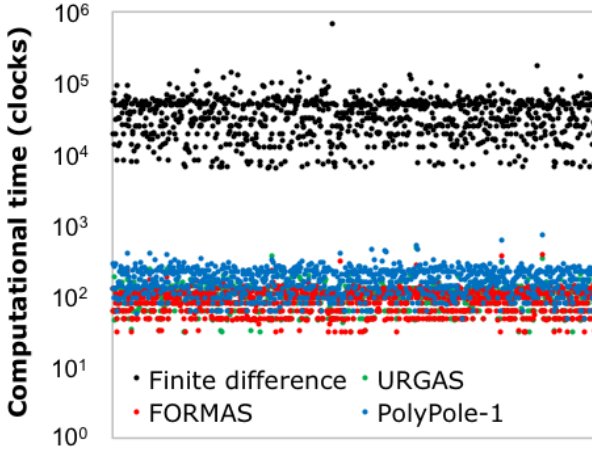


Figure 24. Comparison between the computational times associated with the finite difference, URGAS, FORMAS and PolyPole-1 algorithms. Each data point corresponds to a calculation with randomly generated conditions.

5.4 Closing remarks

In this Chapter, I developed a new algorithm (PolyPole) for the numerical solution of the problem of intra-granular fission gas diffusion in time-varying conditions. The development was aimed at obtaining an accurate solution at low computational cost for application to fuel performance codes.

The PolyPole algorithm is based on the analytic modal solution of the general system of PDEs describing intra-granular diffusion coupled with trapping and re-solution in constant conditions, with the addition of polynomial corrective terms that embody the information on the deviation from constant conditions. This semi-analytic concept is intended to allow for significantly lower computational time than spatial discretization methods, such as finite difference schemes.

Two versions of the algorithm have been verified with numerical experiments involving a high number of randomly generated histories, namely: PolyPole-1 and PolyPole-2. PolyPole-1 (a particular case of PolyPole-2) allows for the solution of intra-granular diffusion PDEs under the quasi-stationary approximation. PolyPole-2 instead allows for the solution of the full general system of PDEs, overcoming the quasi-stationary approximation. The results of the verification pointed out that:

- The accuracy of the PolyPole-1 solution is high and superior to other algorithms (e.g., URGAS, FORMAS) currently adopted in fuel performance codes and relying on the quasi-stationary approximation.
- The computational time associated with PolyPole-1 is similar to the algorithms mentioned above.
- Differently from the current algorithms, the accuracy of the PolyPole-1 solution is highly consistent over the whole range of intra-granular fission gas release.
- Remarkably, PolyPole-2 is the first of a kind algorithm allowing for the solution of the general system of PDEs in fuel performance codes. This overcomes the quasi-stationary approximation enabling for a more accurate representation of intra-granular diffusion in FPCs. The importance of this result is critical for RIA transients, in particular in the HBS.
- The accuracy of the PolyPole-2 solution is high and acceptable within the requirements of fuel performance codes.

The PolyPole-1 algorithm is available open source (under license from INL). The implementation of PolyPole-1 and PolyPole-2 in TRANSURANUS is envisaged in the near future. Both PolyPole-1 and PolyPole-2 have been implemented in the BISON code.

6. Main conclusions and further developments

This PhD thesis pursued the twofold objective of: (1) developing a physics-based IGB modelling approach (embodying all the relevant physical phenomena and able to perform transient analysis), and the related numerical algorithms necessary to handle it, and (2) introducing the developed models in existing engineering-scale FPCs (BISON and TRANSURANUS).

I proceeded by developing three IGB models, dedicated respectively to intra-granular, inter-granular and high burnup structure gas behaviour. Each of these models embodies peculiar physical phenomena (intra-granular bubble nucleation, intra-granular bubble coarsening, grain-boundary micro-cracking, HBS grain recrystallization) which all together allow for representing the complete evolution of inert gas behaviour during transients. This represents a significant step forward with respect to the state of art, in which some of these phenomena are represented by fully empirical correlations (with none or very limited transient capabilities) and others are completely not represented despite their potential impact on the fuel performance. I compared the results of each developed model with experimental data, according to a dedicated validation strategy, always showing an improvement with respect to available state-of-the-art models.

I developed two effective and efficient numerical algorithms dedicated to transient analysis. In particular, PolyPole-1 is developed for operational transients, whereas PolyPole-2 is developed to overcome the quasi-stationary approximation, being therefore applicable to RIA conditions. I verified the algorithms against a reference finite-difference algorithm in dedicated numerical experiments considering randomly generated transient histories.

Overall, the outcome of this thesis encompasses several different aspects in IGB modelling, i.e., both intra- and inter-granular behaviour, both regular fuel structure and HBS, both normal operating and transient conditions, both representation of physical phenomena and numerical aspects.

All the models and the algorithms developed in this thesis have been implemented in FPCs. I personally performed the implementation in BISON and TRANSURANUS, or directly collaborated with the code developers. In addition to providing a means for engineering calculations, thanks to their physical basis the models can be informed with parameters extracted at the smaller scales through atomistic calculations. The models hence provide an interface for scale bridging within multiscale modelling approaches.

The new models are applicable with minor modifications to different fuel materials, since the main physical phenomena driving IGB are present in many fuel materials. The results shown are focused on UO_2 , but extension to other materials is of future interest. For example, in the frame of a collaboration between POLIMI and INL, we are extending the present intra- and inter-granular models for application to uranium silicide fuels.

The main conclusions of this PhD thesis are:

- The development and assessment of a physically-based IGB model (representing intra-granular, inter-granular and HBS IGB) suitable for transient analysis and applicable in FPCs. The development of numerical algorithms capable of solving the equations of the new model with computational requirements in line with FPCs.
- The development and verification of SCIANTIX, a 0D IGB code useful for testing, verification and validation of models and specifically designed for coupling with existing FPCs.
- The actual inclusion of the developed models and algorithms in FPCs, which represents a fundamental achievement from an engineering perspective.

The results obtained pave the way for further developments in several directions.

A first direction is represented by the extension of the IGB model itself. As far as inert gas in oxide fuels is concerned, the extension of the work on helium is crucial (e.g., introducing in FPCs correlations for helium diffusivity and solubility in oxide fuel, see Appendix B) due to the important role played by this gas in MOX fuels, at high burnup, and in storage conditions. More in general, are of sure interest the further development of the HBS model by including the HBS porosity description, the extension to other fuel materials (e.g., MOX, uranium silicide), the extension to include other non-inert fission products (e.g., caesium and iodine), the inclusion of the description of collateral physical phenomena (e.g., restructuring of MOX fuels in fast reactor conditions).

Second fundamental direction is the continuation of the validation of fuel performance codes incorporating the new models against integral irradiation experiments (already performed for the inter-granular gas behaviour model). This will allow quantifying the improvement brought in by the use of these new models. Code-to-code comparison will provide fundamental insights on

code behaviour, pointing out further directions for code development. Moreover, more extensive validation is envisaged with SCIANTIX in order to further assessed the developed models.

Another fundamental direction for further research is represented by the optimization of SCIANTIX. This will lead to the development and licensing of a general tool for the development of new meso-scale models in the field of nuclear fuel performance, allowing for its use as a meso-scale IGB module in the framework of fuel performance codes.

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Appendix A. Intra-granular bubble coarsening

In this Appendix, I present an extension to the intra-granular bubble model detailed in Chapter 2, involving the description of bubble coarsening during transients.

The content of this Appendix is based on:

D. Pizzocri, G. Pastore, T. Barani, A. Magni, S. Lorenzi, L. Luzzi, P. Van Uffelen, S.A. Pitts, J.D. Hales, *A model describing intra-granular bubble evolution in oxide fuel during transients and at high burnup*, Under preparation.

Experimental observations from White et al. [10] indicated that during transients (i.e., power ramps) a small percentage of the intra-granular bubble population experiences a fast and consistent growth. They observed that these coarsened bubbles always lie in proximity of dislocation lines, suggesting that the vicinity to dislocations may play a role in the growth process. This phenomenon, referred herein as *bubble coarsening*, can be responsible of up to half of the gaseous swelling of the fuel [10].

Based on these experimental observations, I developed a preliminary model to describe the evolution of bubble coarsening. The model represents an extension of the intra-granular bubble model described in Chapter 2. I introduced a second population of intra-granular bubbles, which is close to dislocations. During operational conditions, this population is assumed to have an evolution identical to that of the other intra-granular bubbles present in the grain matrix far from dislocations, with the only exception of an increased trapping rate arising from gas atoms trapped along dislocation lines. During transient conditions, if the internal energy of the bubbles close to dislocations equals or overcomes their surface energy, these bubbles grow by vacancy absorption along dislocations. This condition is expressed as

$$G = \frac{kT}{\Omega} \frac{\bar{n}}{n_v} \frac{2\gamma}{R_d} \quad (\text{A.1})$$

where k (J/K) is the Boltzmann's constant, T (K) is the temperature, Ω (m³) is the vacancy volume, n and n_v are the number of atoms and of vacancies per bubble, respectively, γ (J/m²) is the surface tension, and R (m) is the bubble radius. This growth process triggered by Eq. A.1 is not possible for bubbles away from dislocations, which experience a vacancy starvation regime.

I performed a preliminary assessment of the model on the database of White et al., [10]. This database contains 12 UO_2 samples with several radial points considered for each sample, for a total of 52 measurements of bubble density and bubble radius. All the samples have been irradiated at low constant power (base irradiation) and subsequently ramped at high power for a short time (ramp test). Figure 25 reports the results of the validation of the bubble coarsening model against the data from White et al., [10]. Compared to the results reported in Fig. 3 for the bubbles far from dislocations, it is clear that coarsened bubbles are less in number, but present much bigger sizes, thus representing a significant contribution to the overall swelling of the fuel. It is important to notice that this model for bubble coarsening is the first of its kind that has been developed in the literature. State-of-the-art approaches used in FPCs completely disregard this phenomenon. In view of its novelty and of its impact, the development of this model was supported by an ongoing contract between POLIMI and INL, in the frame of the scientific collaboration with INL. Since the finalization is still ongoing both in terms of model development, characterization of parameters and model validation, just the basic concepts behind this bubble coarsening is reported here as Appendix.

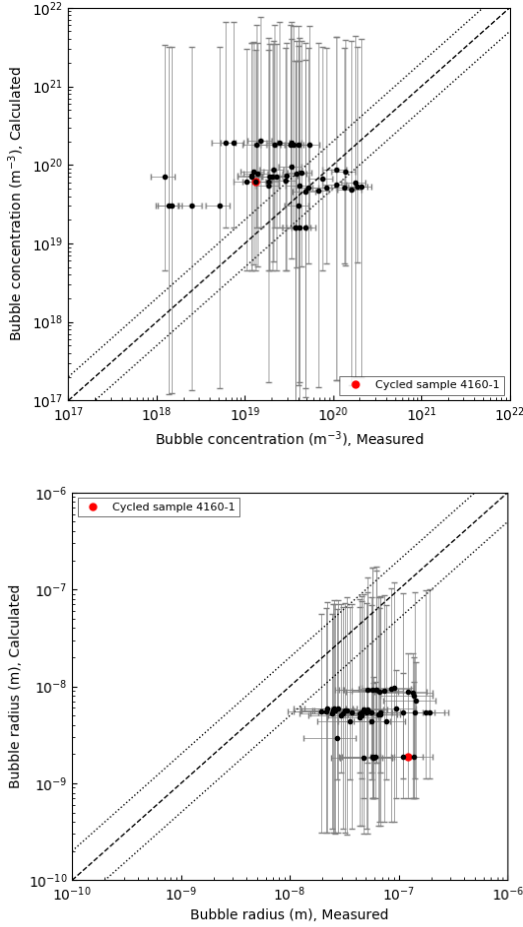


Figure 25. Comparison between the present model results and the data from White et al., [10]. Each black full symbol represents a simulation obtained with the reference model parameters compared to the corresponding experimental value (red and blue mark refer to specific samples with representative power histories, ramped and cycled respectively). The vertical error bars span the maximum and minimum results obtained with an uncertainty analysis performed on the model parameters. The horizontal error bars represent the experimental uncertainties, i.e., 30% for the bubble number density and 50% for the bubble radius. The distance from the 45° full black line is a graphical measure of accuracy. The dashed lines correspond to deviations of a factor of 2 from measured data, to provide a visual indication.

Appendix B. Helium diffusivity

In this Appendix, I present an activity performed as a side of this thesis work, with the aim of improving the helium diffusivity correlations currently used in fuel performance codes. This work has been performed with the strong collaboration of T. Barani and L. Cognini, and is therefore reported as an Appendix.

The content of this Appendix is based on:

L. Luzzi, L. Cognini, D. Pizzocri, T. Barani, G. Pastore, A. Schubert, T. Wiss, P. Van Uffelen, 2018, *Helium diffusivity in oxide nuclear fuel: Critical data analysis, new correlations*. Nuclear Engineering and Design, 330, 265-271.

Helium behaviour is one of the fundamental aspects in determining nuclear fuel performance, both in operation (especially for MOX and at high burnup) and during storage [1]. The production ways of helium in nuclear fuel are ternary fissions, (n,α) -reactions and α -decay [106], [131]. Helium then precipitates into intra- and inter-granular bubbles and can be absorbed (or released) from (or to) the nuclear fuel rod free volume [4]. Helium thus contributes to the fuel gaseous swelling and to the pressure in the fuel rod plenum.

Helium diffusivity and solubility are the properties governing the transport and absorption/release mechanisms [132]–[134]. In particular, helium diffusivity determines its kinetics during transients, and thus it is the fundamental property to consider in simulation of design basis accidents. For this reason, POLIMI activity focused on the evaluation of helium diffusivity, in order to improve the description of helium behaviour in FPCs. In particular, POLIMI worked on the derivation of new and less uncertain correlations for helium diffusivity considering all the available literature data, after a critical analysis of the data themselves (for details, see Luzzi et al. [135]). The analysis evaluates three introduction techniques used experimentally, namely: infusion, ionic implantation, and doping. A considerable amount of experiments has been performed with the goal of determining the diffusivity of helium in nuclear fuel by means of these techniques [38], [132]–[134], [136]–[145]. Table 2 collects all the diffusivity data available in the open literature. Figure 2 shows the experimental data reported in Table 2. Two clusters of data emerge from Figure 2: the measurements performed with the infusion technique (in the lower region),

and those performed with the ion implantation and doping techniques (in the upper region). The overall spread of available diffusivities is extremely large. Therefore, we propose two distinct empirical correlations for the helium diffusivity.

By fitting the two clusters of data independently, the fitting quality results improved compared to a fit of the all data together. Nevertheless, the correlations for the helium diffusivity currently implemented in TRANSURANUS, as well as those available in the open literature, are derived from rough data fitting, or are intended to be engineering upper/lower boundaries to the data [131], [143].

The best estimate correlations we derived are:

$$D = 2.0 \cdot 10^{-10} \exp[-2.12/kT] \quad (34)$$

in ($\text{m}^2 \text{s}^{-1}$), for the cluster of data from infusion, and

$$D = 3.3 \cdot 10^{-10} \exp[-1.64/kT] \quad (35)$$

in ($\text{m}^2 \text{s}^{-1}$), for the cluster from doping and ionic implantation.

For a more extensive discussion (and the related uncertainty analysis) of the derived correlation, see Luzzi et al. [135].

Table 9. Summary of the experimental helium diffusivities in oxide fuel available in the open literature, categorized depending on the technique used to introduce the helium in the samples.

Ref.	Sample	Diffusivity ($\text{m}^2 \text{s}^{-1}$)	Temperature (K)
<i>Infusion</i>			
[136]	UO ₂ powder (0.16 μm)	$9.05 \cdot 10^{-22}$	968
[136]	UO ₂ powder (0.16 μm)	$1.01 \cdot 10^{-20}$	1070
[136]	UO ₂ powder (0.16 μm)	$4.08 \cdot 10^{-20}$	1166
[136]	UO ₂ powder (0.16 μm)	$1.86 \cdot 10^{-19}$	1268
[138], [146]	UO ₂ powder (4 μm)	$1.5 \cdot 10^{-17}$	1473
[139]	UO ₂ single-crystal (1 μm)	$6.14 \cdot 10^{-18}$	1473
[139]	UO ₂ single-crystal (1 μm)	$9.15 \cdot 10^{-18}$	1623
[139]	UO ₂ single-crystal (1 μm)	$12.57 \cdot 10^{-18}$	1773
[133]	UO ₂ single-crystal (18 μm)	$9.50 \cdot 10^{-10} \exp[-2.05/kT]$	range 1170–2110
[133]	UO ₂ single-crystal (18 μm)	$4.88 \cdot 10^{-10} \exp[-1.93/kT]$	range 1390–2070
<i>Ion implantation</i>			
[140]	UO ₂ poly-crystal	$(3.7 \pm 0.74) \cdot 10^{-18}$	1273
[141]	UO ₂ poly-crystal (8 μm)	$6 \cdot 10^{-17}$	1373
[142]	UO ₂ poly-crystal (10 μm)	$8 \cdot 10^{-9} \exp[-(2 \pm 0.1)/kT]$	range 1123–1273
[142]	UO ₂ poly-crystal (10 μm)	$4 \cdot 10^{-10} \exp[-(2 \pm 0.1)/kT]$	range 1123–1273
[38]	UO ₂ poly-crystal (24 μm)	$2.25 \cdot 10^{-17}$	1073
[38]	UO ₂ poly-crystal (24 μm)	$7.6 \cdot 10^{-17}$	1373
[144]	(U _{0.75} , ²³⁹ Pu _{0.25})O ₂ poly-crystal	$9.2 \cdot 10^{-18}$	1123
[144]	(U _{0.75} , ²³⁹ Pu _{0.25})O ₂ poly-crystal	$1.6 \cdot 10^{-16}$	1273
[145]	UO ₂ poly-crystal	$5 \cdot 10^{-10} \exp[-(1.4 \pm 0.2)/kT]$	range 973–1373
<i>Doping</i>			
[143]	(U _{0.9} , ²³⁸ Pu _{0.1})O ₂ poly-crystal	$(8 \pm 2) \cdot 10^{-7} \exp[-(2.00 \pm 0.02)/kT]$	N/A
[147]	(U _{0.999} , ²³⁸ Pu _{0.001})O ₂ poly-crystal (10 μm)	$10^{-7} \exp[-2.59/kT]$	range 1320–1800

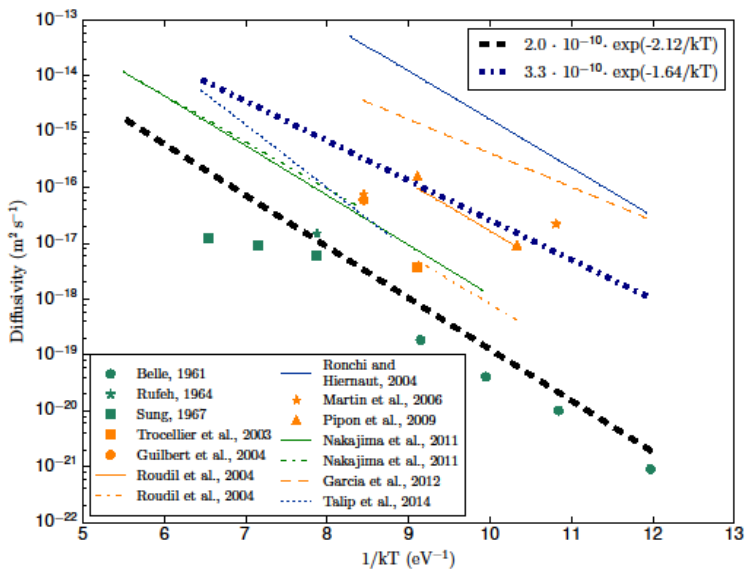


Figure 26. Plot of the experimental helium diffusivity in oxide fuel. The measurements performed via the infusion technique (dark green), those measurements performed via the ion implantation (orange) and doping (navy blue) technique. Each cluster is fitted by a distinct correlation (black dashed and blue dot-dashed).