Modeling high burnup structure in oxide fuels for application to fuel performance codes. Part I: High burnup structure formation

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1. Introduction

In nuclear fuels, where substantial irradiation damage (e.g., local burnups above 45/50 MWd kgU−1) is accompanied by a limited possibility of recovering the damage (i.e., local temperatures below 1000 °C), a dramatic change occurs to the as-fabricated microstructure. The initial microstructure, usually featured by micrometric, re-crystallized grains alongside micrometric pores, of roughly spherical shape. This phenomenon is referred to as High Burnup Structure (HBS), or rim effect, since it was historically observed and postulated to be limited to the restructuring observed in the outer region of Light Water Reactor (LWR) fuel pellets [1–5]. Indeed, this phenomenon has been observed in fuel types other than UO2, e.g., in Pu-rich islands of heterogeneous U–Pu mixed oxides (MOX) fuel [3,6–8], in the rim zone of Fast Breeder Reactor (FBR) U–Pu oxide [9,10], in U–Pu carbides [11], and in U–Mo fuels [12].

Despite intensive experimental and modeling activities for decades, the formation mechanisms of HBS are still debated [3,4,13–15]. HBS formation is ascribed either to a re-crystallization [16–23] or a polygonization [24–29] process. Indeed, commonly observed features encompass the build-up of dislocations [20], the depletion of intra-granular fission gas [3,7,30], the formation of pristine, sub-micrometric grains [26], and development of a novel porosity [31,32].

The (economically) appealing interest in increasing the target burnup of LWR rods constitutes a strong driving force in developing models describing HBS behavior to be included in fuel performance codes (FPC). In facts, the development of HBS porosity brings about an additional fuel swelling source, which must be properly represented to predict UO2 thermo-mechanical performance at high burnups [15,33]. Moreover, HBS development contributes to determine both fuel thermal conductivity [34] and elastic modulus [35]. Finally, the fission gas stored in HBS porosity plays a major role in determining fission gas release (FGR) and fuel fragmentation (and consequent relocation) during Loss of Coolant Accident (LOCA) [36–39] and Reactivity Initiated Accident (RIA) [40] conditions.

Several models, either semi-empirical or mechanistic, have been...
conceived to describe HBS effects in the framework of FPCs. Since this work is focused on modeling HBS formation and intra-granular fission gas depletion – leaving the description of porosity development to a forthcoming, second part – we limit our discussion to the modeling of these two aspects.

Lassmann et al. [41] developed a pragmatic, empirical model to account for intra-granular xenon depletion, decreasing its concentration with an exponential law as a function of burnup (not considering the effect of temperature) and needing as an input parameter the HBS formation threshold. This approach represents the legacy treatment of HBS in the TRANUSURANUS FPC code [42]. In a recent work, Jernvikst [43] adopted the same concept to describe xenon depletion, while combining it to a threshold value estimated by the theoretical model of Rest [44]. The model has been made available to the FRAPCON/FRAPTRAN codes [45]. Lemes et al. [46] extended the model by Lassmann, including the treatment of Kr and complementing it with a mixed empirical and mechanistic description of the porosity development. The model is implemented in the DIONISIO code [47].

The model by Khvostov and coworkers [48,49] describes the HBS restructuring through a Kolmogorov-Johnson-Mehl-Avrami (KJMA) relationship, introducing also the concept of “effective burnup.” In the description of HBS to weight differently the burnup accumulation at high and low temperatures and thus consider thermal annealing of defects in the description of HBS formation. The semi-empirical description of HBS formation is then paired to a mechanistic description of intra- and inter-granular gas behavior, grafted in the GRSW-A model [49] and integrated in the FALCON code [50]. The concept of effective burnup was integrated into the Lassmann model by Holt et al. [51], introducing a higher temperature threshold in the definition of effective burnup with respect to the one originally proposed by Khvostov. This model is the standard treatment of the HBS formation/depletion in TRANUSURANUS.

Blair and coworkers [52] developed a steady-state model describing the xenon depletion in HBS grains, considering in a physically based manner the main intra-granular behavior processes and focusing on the effects of grain boundary diffusivity and re-solution. The main limits of the model are the lack of description of HBS formation and the steady-state formulation, which prevents its application to transient conditions.

L. Noirot proposed a model accounting for HBS formation based on an empirical estimation of the dislocation density evolution [53]. In this model, the evolving dislocation density is fitted to the data by Nagita and Une [54]. The size-distribution of dislocation density is assumed to be a square pulse, and two thresholds of dislocation densities are employed to estimate the fraction of HBS restructuring. The description is plugged in the MARGARET code, which provides a mechanistic description of fission gas behavior, and is integrated into the ALCYONE code [55,56].

Pizzocri et al. [57] proposed a semi-empirical model describing HBS formation and xenon depletion, representing HBS development as a progressive shrinkage of the average grain size of fuel from the original micrometric to the sub-micrometric size as a function of the local effective burnup. Xenon depletion is reproduced solving an intra-granular diffusion problem featured by a decreasing domain size, resulting in an accelerated diffusion towards the grain boundaries. The model is available in the SCIANITIX code [58].

Besides the aforementioned empirical and semi-empirical models, more mechanistic models have been developed to describe HBS formation. Rest [44] proposed a mechanistic model considering the evolution of cellular dislocation networks as re-crystallization nuclei and their interaction with intra-granular bubbles. Based on thermodynamics considerations, i.e., comparing the free energies of the original and re-structured phases, he derived a threshold for HBS formation as a function of the local fission density. It must be noticed that the proposed model cannot account for the observed restructuring starting from the grain boundaries and accompanied by the formation of high-angle grain boundary grains [59]. Veshchunov and Shestak [60] proposed a model accounting for the evolution of point, line, and volume defects under irradiation. The key parameter determining HBS formation is the predicted dislocation density, which is compared to a threshold inferred from experimental data [54] to establish HBS formation. Albeit featuring a consistent description of defects evolution, the transition from original to restructured microstructure is step-wise, thus likely failing to properly describe the gradual xenon depletion experimentally observed. The model is available in the MFPR code [61]. Whereas the previous models deal mostly with UO2, combining the predictions by ALCYONE and image analysis techniques, Boulou et al. [8] developed a probabilistic model to estimate HBS formation in MIMAS (Micronized MASterblend) MOX fuel for LWRs, based on xenon EPMA measurements. This model allows one to estimate the portion of restructured fuel at a certain burnup, as well as the retention of fission products in the HBS region. This work is a unique example in the open literature of application of HBS modeling in MIMAS MOX fuel.

In this work, we propose a novel model describing HBS formation, accounting for the progressive Xe depletion and its feedback on the fuel matrix swelling. The HBS formation – i.e., the increase of fuel volume that underwent restructuring as a function of the local effective burnup – is described through the KJMA formalism for phase transitions [62,63]. We derived the coefficients of the KJMA expression fitting the functional form to experimental data on the restructured volume fraction as a function of the local effective burnup. The data employed are a combination of available data in the literature [64] and novel data on UO2 that we extracted through image analysis and fuel performance simulations of recently published experimental results [59]. The HBS formation is paired to a mechanistic model describing intra-granular fission gas behavior in UO2 published by some of the present coauthors [65], providing the evolution under irradiation of the intra-granular bubble population, accounting for bubble nucleation, gas atom trapping into and irradiation-induced re-solution from bubbles, along with diffusion to the grain boundaries. Estimating the evolving concentrations of retained gas into the grain allows us to consistently calculate the fuel matrix swelling, i.e., the swelling due to solid fission products and xenon atoms found in the fuel matrix and in intra-granular bubbles, up to high burnups. The contribution of the swelling from the large HBS pores will be dealt with separately in a future publication. We implemented the model in the SCIANITIX code [58], a stand-alone, open-source, meso-scale computer code developed at Politecnico di Milano, aimed at simulating fission gas behavior in nuclear fuel and conceived for coupling/inclusion in fuel performance codes.

We present the stand-alone validation of model predictions, comparing the predicted xenon intra-granular concentration as a function of local burnup to available Electron Probe Micro Analysis (EPMA) data [3,7,30] and to a number of models available in the open literature and meant for application in FPCs [41,46,57]. Moreover, we showcase the capabilities of the developed modeling approach to account for the experimentally observed modification of fuel matrix swelling as HBS forms. In particular, we compare the results obtained considering the present model to the experimental data by Spino and coauthors [56]. The comparison to more refined and mechanistic models, e.g., the model by Veshchunov and Shestak available in the MFPR code [60], is of sure interest in perspective, but is left as a part of an extensive comparison between SCIANITIX and MFPR codes.

The present work presents an original model for HBS formation
due to polygonization, which is described in a continuous and smooth manner, consistently accounting for the intra-granular fission gas behavior during the formation process. The consistent description of the kinetics of intra-granular fission gas behavior and fuel gaseous swelling, together with the formulation grounded on a physical basis for both formation and fission gas depletion results in a model which founds a wider applicability, in terms of operating conditions and fuel types, than state-of-the-art models available in fuel performance codes. These features are reconciled with the needs of models to be included in fuel performance codes by industries and research, i.e., ensuring an acceptable computational burden and an optimal numerical stability. Finally, the inclusion of the developed model in the SCIANTIX code will make it available as an open-source tool to the interested public.

The outline of the paper is hereafter briefly outlined. In Section 2, we present the extraction of new data on HBS formation, which are exploited in Section 3, where we present the formulation of the model for HBS formation and intra-granular depletion. The comparison to experimental results and available model predictions are presented in Section 4, whereas we draw the conclusions in Section 5.

2. Derivation of data on High Burnup Structure formation

In this Section, we present the extraction of novel data on the progressive formation of HBS based on the experimental results by Gerczak and coauthors [59]. In particular, we are interested in quantifying the restructured portion of fuel volume and correlating it to the local effective burnup. As a complementary and independent data-set, we report the experimental results on HBS-restructured volume fraction available in the open literature, and published by J. Noirot and coauthors [64].

2.1. Simulation of the analyzed fuel sample

The fuel sample considered in Ref. [59] was taken from a commercial fuel rod irradiated in the H.B. Robinson PWR. The mother fuel rod characteristics, listed in Table 1, together with the irradiation history reported in Refs. [59,67] were used to build the input for a TRANSURANUS [42] simulation. The calculated fuel central and outer temperatures for the analyzed specimen are reported in Fig. 1, together with the input linear heat rate, as a function of the specimen average burnup.

The calculations of radial burnup and effective burnup exploits the TUBRNP burnup model of TRANSURANUS [41,68] and are reported in Fig. 2 as a function of the relative pellet radius. The effective burnup is a concept introduced by Khvostov and coauthors [48] to account for the build up of irradiation damage at “low temperatures”, i.e., at temperatures at which defects annealing is suppressed. In this work, rather than adopting the original formulation as proposed in Ref. [48], we chose the definition of effective burnup as proposed more recently by Holt and coauthors [51].

\[ b_{\text{eff}} = \int f(T - T_{\text{dbu}}) \, dT \]  

(1)

where \( f(T) \) is the Heaviside step function, \( T \) (°C) the local temperature, and \( T_{\text{dbu}} \) is a threshold temperature assumed equal to 1000 °C. The estimation of the local (effective) burnup would allow us to complement the information regarding the analyzed experimental data, i.e., associating each analyzed radial position with the corresponding burnup value.

2.2. Extraction of novel data on HBS volume coverage

Gerczak and coworkers [59] investigated the HBS formation

### Table 1

| Characteristic | Value/Material |
|---------------|----------------|
| Cladding material | Zircaloy-4 |
| Cladding outer diameter (mm) | 10.77 |
| Cladding inner diameter (mm) | 9.25 |
| Pellet-cladding gap (µm) | 95 |
| Pellet diameter (mm) | 9.06 |
| Pellet height (mm) | 6.93 |
| Dish volume (%) | 1 |
| 235U enrichment (%) | 2.9 |
| Active fuel height (mm) | 3660 |
| Fuel pin height (mm) | 3860 |
| Specimen (radial average) burnup (MWd kgU⁻¹) | 72 |
| Measured Fission Gas Release (%) | 2.1 |

Fig. 1. Linear heat rate and calculated fuel pellet outer and central temperatures as a function of burnup for the considered specimen.

Fig. 2. TRANSURANUS estimation of local and effective burnup as a function of the relative radial position for the considered fuel pellet.
through advanced electron microscopy techniques. In particular, their analysis was focused on the correlation between progressive polygonization and grain boundary surfaces orientations. They showed how low-angle grain boundaries form starting from intra-granular patches and/or from the original high-angle grain boundaries of the as-fabricated microstructure. Moreover, they showed that low-angle grain boundaries are gradually transforming into high-angle grain boundaries moving towards the pellet periphery, i.e., where HBS development is complete. In the aforementioned work, the authors reported Electron Backscatter Diffraction (EBSD) scans of a fuel pellet microstructure, taken at various distances from the pellet outer radius.

In this work, we present the results of further image analysis applied to the aforementioned experimental results, to quantify the HBS surface and volumetric coverage in the examined samples. The image analysis has been performed on the images taken from Figure 10 of the mentioned paper [39], and reported in Fig. 3 for the sake of completeness. We considered a subset of the reported images, focusing on the data obtained at relative radii equal to 0.63, 0.82, 0.94, and 0.95. We assume the image at 0.99 relative radius as representative for the complete restructuring process. This assumption is corroborated by the observed grain size, whose measurement yields approximately 200 nm, roughly corresponding to the observed, asymptotic grain size in the HBS [26,31].

We measured the area covered by HBS in the analyzed locations by the ImageJ software. In particular, we converted to a binary image each analyzed image and quantified the area enclosed by low- and high-angle grain boundaries in the visible sub-domains. The ratio of the selected area to total area of the sample is presumed to be below 1000.

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This would not account for the formation of HBS following intra-granular patches, which has also been reported in several experimental works [3,14,29,64]. Indeed, the value obtained in this work accounts for all the mentioned contributions, being adherent to the different experimental findings.

For the sake of completeness, we must discuss the validity of the hypotheses underlying the KJMA formalism for phase

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**Fig. 3.** Grain boundary misorientation map overlaid on image quality map for locations \( \frac{r}{r_0} \) 0.99 to 0.35, (a) through (g). Taken from Ref. [59].

LAGBs from 2-5° are shown in green, LAGBs 5-15° in yellow, and HAGBs 15-65° in purple.

| Min | Max |
|-----|-----|
| 15° | 65° |
| 5°  | 15° |
| 2°  | 5°  |
transformation, namely, constant temperature during the transformation, random and homogeneous formation of secondary phase nuclei. The experimental data considered in this work [59], coherently with other recent results [71], corroborate the hypothesis of homogeneous formation of the HBS grains in the original microstructure volume, whereas the random disposition of the nuclei is questionable. In fact, the grain boundaries of the as-fabricated microstructure, as well as the intra-granular fission gas bubbles, appear to be a preferred source for the formation of grain sub-domains. The hypothesis of constant temperature is respected to the extent that in our formulation, the rate of transformation only depends on the effective burnup (Eq. (3)), which in turn is independent of temperature below 1000 °C (Eq. (1)).

3.2. Modeling fission gas depletion in the HBS

As the model is conceived to be included in fuel performance codes, we adopt the classical representation employed in such codes to describe fission gas diffusion, i.e., we represent the single grain as an equivalent spherical domain, employing the concept of an “effective” diffusion equation [15,33,65,72,73].

Equation (3) is adopted to evaluate the volume fraction that underwent HBS restructuring. Besides, the model considers a “two phases” material, one pertaining to the original microstructure and one to HBS, as sketched in Fig. 5.

These two “phases” are featured by different grain sizes, i.e., the original microstructure usually by a micrometric size, whereas the HBS is assumed to be formed at a radius equal to 150 nm (in line with experimental data, e.g., by Ray and coworkers [26]). It is worth to underline that this modeling approach can be naturally extended to heterogeneous U–Pu mixed oxide fuels, featured by “regions” with different Pu content, resulting in local higher fission densities along the fuel pellet.

In both domains, the intra-granular gas behavior is described through the mechanistic model presented by Pizzocri and co-authors [65]. This model allows for the calculation of intra-granular fission gas bubble nucleation, growth due to gas atom trapping, destruction due to the interaction with fission fragments, along with considering a net diffusion of gas atoms towards the grain boundaries. The equations – which are applied in both the original and HBS domains – read

\[
\begin{align*}
\frac{\partial c}{\partial t} & = D \nabla^2 c - g_0 c + b_n m - 2v + yF \\
\frac{\partial m}{\partial t} & = 2v + g_0 c - b_n m \\
\frac{\partial N}{\partial t} & = v - b_n N
\end{align*}
\]

(4)

where \(D \text{ (m}^2\text{s}^{-1})\) is the single gas atoms diffusion coefficient, \(c \text{ (atm)}\) is the concentration of gas retained in dynamic solution, \(m \text{ (atm)}\) is the concentration of gas in the bubbles, \(N \text{ (bubble m}^{-3})\) is the bubble number density, \(g_0 \text{ (s}^{-1})\) is the trapping rate, \(b_n \text{ (s}^{-1})\) is the re-solution rate, \(v \text{ (bubble m}^3\text{s}^{-1})\) is the bubble nucleation rate, and \(F \text{ (fission m}^3\text{s}^{-1})\) is the fission gas yield, and \(F \text{ (fission m}^3\text{s}^{-1})\) is the fission gas yield. The average number of atoms in a bubble, \(n \text{ (atom)}\), and the associated radius, \(R \text{ (m)}\), and the resulting swelling, \(\text{(atomic)}\), are defined as

\[
\left( \frac{\alpha R}{v} \right)_{\text{ig}} \text{(1)}
\]

Table 2

| Relative radius | Effective burnup (MWd kgU\(^{-1}\)) | HBS-covered area (Estimated) | HBS-covered volume (Estimated) |
|-----------------|-------------------------------------|-----------------------------|-------------------------------|
| 0.63            | 64.7                                | 0.15                        | 0.22                          |
| 0.82            | 71.2                                | 0.41                        | 0.54                          |
| 0.94            | 88.4                                | 0.54                        | 0.69                          |
| 0.95            | 96.8                                | 0.61                        | 0.76                          |
| 0.99            | 129.4                               | 1.00                        | 1.00                          |

Table 3

| Relative radial position | Effective burnup (MWd kgU\(^{-1}\)) | HBS-covered area (Estimated) | HBS-covered volume (Estimated) |
|-------------------------|-------------------------------------|-----------------------------|-------------------------------|
| 0.30                    | 72.4                                | 0.41                        | 0.55                          |
| 0.66                    | 77.3                                | 0.46                        | 0.60                          |
| 0.97                    | 83.8                                | 0.49                        | 0.62                          |

Fig. 4. Experimental measurements derived from Refs. [59,64] on the fraction of restructured fuel volume and KJMA relationship as a function of local effective burnup.
In engineering-scale fuel performance codes, fission gas behavior models are called in each integration point of the fuel computational mesh (e.g., Refs. [53, 42, 76]).
where \( \frac{\Delta V}{V_0} \) is the swelling rate, \( \frac{\Delta V}{V_0} \)\(_{ig}\) refers to the swelling due to intra-granular bubbles, \( \frac{\Delta V}{V_0} \)\(_s\) refers to the swelling due to solid fission products, and \( \frac{\Delta V}{V_0} \)\(_{sg}\) refers to the swelling due to solid fission products. The equation for swelling due to solid fission products is calculated as

\[
\frac{\Delta V}{V_0} = c_1 \cdot q^2
\]

where \( c_1 \) is the overall concentration of gas as single atoms retained in dynamic solution with the matrix (i.e., not in intra-granular bubbles) and \( q \) is the \( \{\text{UO}_2\} \) lattice parameter. The previous value obtained by Olander entails considering that all the created Mo is in metallic inclusions, while it is known that as burnup increases Mo can be found as MoO\(_2\) partly dissolved in the fuel matrix. The model proposed by Pizzocri et al. [57] determined higher fuel swelling rates (evaluated on the basis of various atomic scale calculations (Midbry Olander as 0.45% per at.%). A more recent and detailed assessment is three times the average atomic volume in \( \{\text{UO}_2\} \), equal to \( a^3/12 \).

As for the swelling due to solid fission products, we rely on the theoretical considerations by Olander [80], who proposed a swelling rate due to fission product compounds not soluble in the \( \{\text{UO}_2\} \) fluoride structure equal to 0.32% per atom percent burnup. Nonetheless, it must be noticed that solid swelling rate strongly depends on the local chemical speciation, which is governed by the local oxygen potential, whose variation with burnup determines the phases in which elements are found [81]. For example, the previous value obtained by Olander entails considering that all the created Mo is in metallic inclusions, while it is known that as burnup increases Mo can be found as MoO\(_2\) partly dissolved in the fuel matrix [81], determining higher fuel swelling rates (evaluated by Olander as 0.45% per at.%).

A more recent and detailed assessment on the basis of various atomic scale calculations (Middleburgh et al. for \( \{\text{UO}_2\} \), Ducher et al. for UC [83] and Klipfel et al. for UN [84]) accounts for the local environment of each fission product that is also dependent on the evolving stoichiometry. Although we consider a constant value as a function of burnup, in line with state-of-the-art approaches (such as in the MATPRO FSWELL correlation [45]), a shift towards higher values of the swelling rate would be more adequate to the behavior of fission products such as molybdenum. Finally, it must be underlined that state-of-the-art models for fuel matrix swelling do not take into consideration the contribution brought about by irradiation damage, such as the formation of dislocation loops. This contribution is expected to contribute to the swelling rate decreasing associated with HBS formation.

4. Results and discussion

In this Section, we present the results of the presented model, comparing the predictions, in terms of local xenon retention as a function of effective burnup, to available experimental data and to several models available in the open literature and conceived for application to FPCs. Moreover, we showcase the capability of the presented model to account for matrix (macroscopic) swelling modification as HBS formation occurs, due to fission gas atom depletion.

The presented results were obtained implementing the model presented in Section 3 into the SCIANTIX code [58], a stand-alone, meso-scale computer code developed at Politecnico di Milano, aimed at simulating fission gas behavior in nuclear fuels. The expressions of parameters mentioned in Section 3 and employed in SCIANTIX are reported in Table 4.

### 4.1. Xenon depletion

In Fig. 6, we compare the intra-granular concentration of xenon predicted by SCIANTIX simulations, considering the present model, to several experimental data obtained by different authors [3,7,30] via EPMA. Considering the huge scattering of the experimental data, due to different irradiation conditions and initial fuel specifications, the agreement is deemed satisfactory. For the sake of completeness, it must be underlined that xenon is accompanied by other gases in determining the overall gas behavior (namely, krypton and helium). In this work, we draw a special attention on xenon since it was the subject of the most intensive experimental investigations.

For the sake of comparison, we report also the results obtained by several state-of-the-art models employed in FPCs, namely the model by Lassmann et al. [41], by Lemes et al. [46], and by Pizzocri et al. [57]. The models by Lassmann and Lemes, the latter being an extension of the former, represent pragmatic and purely empirical approaches to account for fission gas depletion as HBS forms, considering an exponential decay of retained gas concentration with burnup. Indeed, they are directly fitted on a subset of the reported EPMA data, i.e., those by Walker [30]. Moreover, they consider a threshold for HBS formation solely dependent on burnup, discarding the effect of temperature, which is instead considered in this work via the effective burnup concept. In Fig. 6, predictions of those models considering different burnup thresholds are reported, showing how this parameter impacts the maximum xenon retention and the subsequent depletion. We underline that in this Figure the burnup and effective burnup coincide, since the data were taken from the rim zone of commercial \( \{\text{UO}_2\} \) fuel, where the fuel local temperature remains below 1000 °C.

On the other hand, the model proposed by Pizzocri et al. [57] represents a step forward with respect to the aforementioned ones, as it is not purely empirical. This model represents HBS formation by a gradual reduction of the grain size, which may be a questionable representation of the underlying physical processes. Differently from the present model results, the predictions obtained through this model and shown in Fig. 6 are obtained considering a xenon production rate adjusted to the experimental data at low burnups to Walker data [30], to obtain a maximum xenon retention in line with experimental data. Finally, this model does not consider the evolution of intra-granular fission gas bubbles, employing a fixed value for intra-granular bubble density and radius, and disregards volume changes due to irradiation damage in
the lattice. This results in an inconsistent estimation of gas partition between bubbles and dynamic solution, preventing a consistent calculation of matrix swelling due to single gas atoms and intra-granular bubbles, in addition to affect the “effective diffusion” representation. The present model also accounts for the observed delay in xenon depletion (thus HBS formation) when considering higher grain sizes in the original microstructure, as shown in Fig. 6 and coherently with several experimental observations [20,64].

4.2. Fuel total matrix swelling

In Fig. 7, we compare the predictions obtained on the fuel total matrix swelling (as defined in Section 3.3) as a function of effective burnup (as for Fig. 6, also in this case effective and local burnup coincide) to the data compiled by Spino et al. [66]. The matrix swelling predicted by the MATPRO FSWELL model — which is employed in several state-of-the-art fuel performance codes to evaluate the swelling due to solid fission products (e.g., in TRANSURANUS [42], BISON [76], FRAPCON/FRAPTRAN [45]) — is included for comparison and discussion.

The results obtained through the present model, both with the nominal and upper solid fission products swelling rates, demonstrate the impact of the matrix depletion of fission gas as HBS progressively forms. The depletion of fuel matrix, starting predominantly around 60 MWd kgU⁻¹, is likely causing a decrease of the fuel matrix swelling rate, in accordance with the elaboration presented by Spino et al. [66] and based on EPMA results on Xe retention. The agreement of the predicted trend of matrix swelling with the considered data is encouraging. As for the absolute value, the matrix swelling obtained with the nominal swelling rate ascribed to solid fission products (solid blue line in Fig. 7), i.e., 0.32% per atom percent burnup [80], and considering the volume occupied by xenon in dynamic solution and intra-granular bubbles is somewhat overestimating the data reported and the values predicted by the MATPRO FSWELL model. On the other hand, the results obtained considering the upper bound of the solid swelling rate — 0.45% per atom percent burnup [80] — yields a better agreement with the considered data and with the MATPRO FSWELL model.

However, it must be noticed that the interpretation by Spino and coworkers of the data by Walker [30] entails a likely overestimation of the volume occupied by xenon atoms in intra-granular fission gas bubbles. To estimate the swelling due to intra-granular bubbles, the authors assume intra-granular bubbles to be in equilibrium and to obey the van der Waals equation of state [66]. The applicability of both the assumptions is questionable. On one hand, intra-granular bubbles forming under irradiation are far from being in equilibrium [86–88], thus, considering equilibrium bubble, the estimated radii are overestimated. On the other hand, the applicability of van der Waals equation of state to intra-granular bubbles is debatable, due to the high gas densities reached in intra-granular bubbles [86,87]. This calls for the consideration of more advanced equations of state, e.g. the modified hard sphere equation of state proposed by Ronchi [89]. Based on these considerations, we postulate that the data presented by Spino and coauthors in [66] and reported in Fig. 7 may somewhat be an overestimation of the actual portion of fuel matrix swelling ascribed to intra-granular gas.

5. Conclusions

In this work, we presented a novel model describing HBS formation and fission gas depletion in UO₂, conceived for application in fuel performance codes. The model is featuring a semi-empirical,
yet physically sound, description of HBS formation, described through the KJMA approach for phase transformation. We fitted the KJMA functional form to two sets of independent experimental data available in the open literature. In particular, the volume fraction of restructured fuel was correlated to the local effective burnup, i.e., the burnup accumulated below 1000 °C, which was chosen as a lumped figure embodying the effect of irradiation damage and prevented annealing of the damage itself.

The model provides a consistent and continuous description of HBS formation and associated intra-granular fission gas behavior. In fact, the intra-granular fission gas behavior in the re-structured and original microstructure is described through a previously developed mechanistic model. This modeling approach allows us to evaluate the evolution under irradiation of the intra-granular concentration of fission gas in the matrix and trapped into bubbles, together with the estimation of the diffusional flux towards the grain boundaries. The consistent, physically-grounded description of the kinetics of gas behavior and fuel restructuring paves the way to a wide applicability of the model in terms of operating conditions and fuel types, especially if compared to other state-of-the-art models used in industrial fuel performance codes, which constitutes the objective of our modeling effort. In this regard, the present model is compatible with the requirements in terms of numerical stability and computational burden for the inclusion in fuel performance codes, while preserving a certain degree of physical basis on the considered phenomena.

A first comparison of the stand-alone model predictions to experimental data indicates a good agreement in terms of retained intra-granular xenon concentration, also when compared to other semi-empirical models available in the open literature. The model can reproduce the experimentally observed delay in xenon depletion — connected to a delayed HBS formation — when the original microstructure is featured by larger grain sizes. Moreover, the results in terms of matrix fuel swelling predicted by the model are well reproducing the experimentally observed change in fuel matrix swelling rate at high burnup, as a consequence of the depletion of the xenon retained in dynamic solution with the matrix.

The model has been implemented into the SCiANTiX code and will be distributed open-source. The present work represents the first part of a modeling effort devoted to the development of a comprehensive model describing HBS formation in UO2 and conceived for inclusion in fuel performance codes. Forthcoming works will tackle the evolution of HBS porosity and consequent fuel gaseous swelling, as well as a more extended and integral assessment of the overall model on fuel performance simulations, namely via the TRANSURANUS/SCiANTiX code suite, of high burnup rods.

As future perspective on the presented work, we envisage to enlarge the fitting data-set of the KJMA relation, analyzing other experimental databases (such as the High Burnup Rim Project [90]). Moreover, the developed modeling approach paves the way to the analysis of HBS in other oxide fuel concepts. Indeed, as new data on this side would be available, a tailoring of the current model formulation would be of interest.

CRediT authorship contribution statement

T. Barani: Conceptualization, Methodology, Software, Validation, Writing - original draft, Writing - review & editing, Visualization. D. Pizzocri: Conceptualization, Methodology, Software, Writing - review & editing. F. Cappa: Conceptualization, Writing - review & editing. L. Luzzi: Writing - review & editing, Funding acquisition, Supervision. G. Pastore: Writing - review & editing, Supervision. P. Van Uffelen: Conceptualization, Methodology, Software, Writing - original draft, Writing - review & editing, Funding acquisition, Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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