



## Two-phase modelling for fission gas sweeping in restructuring nuclear oxide fuel

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### ABSTRACT

In this work, we propose a modelling approach for the intra-granular fission gas behaviour in  $\text{UO}_2$  under restructuring process. Leveraging the definition of restructured volume fraction, we consider the fuel matrix transition from the non-restructured to the restructured phase, together with the evolution of the corresponding fission gas concentrations retained in the fuel matrix. Firstly, we derive a sweeping term that exchanges fission gas atoms from the non-restructured to the restructured fuel region. The sweeping term is then included in the conventional intra-granular fission gas diffusion problem. Secondly, the spectral diffusion algorithm is employed to solve two spatially-dimensionless problems, properly representing the non-restructured region with micrometric grains and the restructured region with sub-micrometric grains. The model developed is implemented in SCIANTIX, a 0D meso-scale code for physics-based modelling of fission gas behaviour in nuclear oxide fuel and compared with experimental data and semi-empirical models.

### 1. Introduction

It is known that in the rim region of  $\text{UO}_2$  nuclear fuel pellets, the combination of high local burnup and low temperature induces a  $\text{UO}_2$  phase transition, or restructuring process, with the formation of the HBS (Walker et al., 1992; Cunningham et al., 1992; Khvostov et al., 2005; Veshchunov and Shestak, 2009; Pizzocri et al., 2017; Wiss et al., 2017; Rest et al., 2019; Barani et al., 2020, 2022; Cappia et al., 2022). Typical thresholds for initiating the HBS identify local burnups higher than 45–50 GWd/tU (connected to high radiation damage and fission product concentration) and temperatures lower than the recovery threshold temperature of 1273.15 K (Wiss et al., 2017; Cappia et al., 2022; Spino et al., 2006; Cappia et al., 2016; Gerczak et al., 2018; McKinney et al., 2023). As reported in previous experimental (Wiss et al., 2017; Cappia et al., 2022; McKinney et al., 2023; Spino et al., 2005; Noirot et al., 2008) modelling (Khvostov et al., 2005; Veshchunov and Shestak, 2009; Pizzocri et al., 2017; Barani et al., 2020, 2022), the HBS formation involves an accumulation of dislocation defects, the polygonization/recrystallisation of micrometric grains into sub-micrometric grains<sup>1</sup> without extended defects (Cappia et al., 2022;

Spino et al., 2006; McKinney et al., 2023), the decrease of the intra-granular fission gas concentration (also known as depletion), and the formation of a novel population of inter-granular spherical pores that accumulate fission gas. Properly modelling the formation and evolution of the HBS is critical for fuel rod fuel performance since it impacts the material properties (e.g., thermal conductivity, elastic modulus). This represents a potential concern for the safe operation of nuclear fuel to extended burnups. For instance, slow temperature changes at burnup values above 60 MWd/kgU can trigger fine fuel fragmentation phenomenon, posing safety issues during design-basis accidents, such as reactivity-initiated accidents and loss-of-coolant accidents (Jernkvist, 2019, 2020).

In particular, in this brief work, we deal with the modelling of the fission gas depletion problem, this being the fundamental starting point to provide a solid representation of (i) the distribution of fission gas that accumulates inside inter-granular cavities and HBS porosity, and (ii) the smooth phase transition from the non-restructured  $\text{UO}_2$  matrix to the restructured one. Both semi-empirical (Lassmann et al., 1995a; Lemes et al., 2014; Pizzocri et al., 2017) and mechanistic models (Barani et al.,

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<sup>1</sup> It is worth recalling that the detailed High Burnup Structure (HBS) formation mechanisms are still debated. As explained in Barani et al. (2020), recrystallisation implies the formation and growth of sub-grains (Burke and Turnbull, 1952). Polygonization is the subdivision of original grains into sub-grains. Recent experimental analyses support the polygonization theory, with the increasing concentration of high-angle grain boundaries in the HBS region (Gerczak et al., 2018; McKinney et al., 2023).

2020) are available in the literature, representing the fission gas depletion of HBS in  $\text{UO}_2$ . The most mechanistic approach was provided in the work of Barani et al. (2020), in which the authors started from a proper re-fitting of the restructured volume fraction and then solved two intra-granular problems, one for each phase, considering two different integration domains, characterised by their grain size (e.g., micrometric in the as-fabricated region and sub-micrometric in the restructured region). The two intra-granular problems were solved concomitantly with an HBS sweeping condition, given by the conservation of the total concentration of gas in the considered control volume.

This brief work aims to develop a more natural description of the intra-granular gas evolution in uranium dioxide during the restructuring process. Firstly, we derive a sweeping term that exchanges mass from the non-restructured  $\text{UO}_2$  to the restructured  $\text{UO}_2$ . The sweeping term can be included in the intra-granular gas diffusion problem, avoiding further constraints. Secondly, by leveraging the spectral diffusion algorithm for the intra-granular fission gas problem, we avoid dealing with two different integration domains (micrometric and sub-micrometric grains) and solve a single spatial dimensionless problem (i.e., considering the same eigenfunctions). The model developed is implemented in SCIANITX, a 0D meso-scale code for physics-based modelling of fission gases in nuclear oxide fuel (Zullo et al., 2023; Pizzocri et al., 2020), and compared with experimental data (Walker, 1999) and semi-empirical models (Lassmann et al., 1995a).

The work is structured as follows: Section 2 illustrates the model derivation, Section 3 presents the model application to the fission gas depletion process, Section 4 deals with the results and corresponding discussion, and Section 5 draws the conclusion and points out future applications of the model.

## 2. Model derivation

The model derivation stems from using the restructured volume fraction  $\alpha$  (/) to model the HBS formation rate in uranium dioxide. Hence, we consider a unidirectional phase change from a non-restructured phase (N) to a restructured one (R), with the evolution of two fission gas concentrations,  $C_N$  and  $C_R$ .

### 2.1. Phase evolution without exchange term

Given a control volume  $V$ , we label  $V_R = \alpha V$  as the restructured (i.e., developed HBS) volume region, and  $V_N = (1 - \alpha)V$  as the non-restructured one (as-fabricated microstructure). Then,  $C_T$  is the total concentration of fission gas atoms (i.e., gas in restructured and non-restructured regions) over the total volume  $V$ . Similarly,  $C_N$  is the concentration of gas atoms in the non-restructured volume  $V_N$ , and  $C_R$  is the concentration of gas atoms in the restructured volume  $V_R$ . The link among these quantities is expressed by:

$$C_T = \alpha C_R + (1 - \alpha)C_N \quad (1)$$

Afterwards, we define the following concentrations:

- $C_1 = (1 - \alpha)C_N$ , representing the concentration of gas in the non-restructured region, referred to the total volume  $V$ .
- $C_2 = \alpha C_R$ , representing the concentration of gas in the restructured region, referred to the total volume  $V$ .

We compute the balance of gas atoms  $N$  and  $R$  in their respective volumes  $V_N$  and  $V_R$ , invoking the mass conservation principle:

$$\frac{\partial}{\partial t} \int C_N dV_N = \int D_N \nabla^2 C_N dV_N + \int S dV_N \quad (2)$$

$$\frac{\partial}{\partial t} \int C_R dV_R = \int D_R \nabla^2 C_R dV_R + \int S dV_R \quad (3)$$

In Eqs. (2) and (3), we assume a uniform production rate  $S$  of gas atoms per unit volume. Also, we assume that in the restructured region

$V_R$ , the diffusivity  $D_R$  differs from the one characterising the non-restructured region  $D_N$ . It must be noted that Eqs. (2) and (3) are valid in the (initial) assumption of no mass exchanges between the two regions. In particular, from the non-restructured region  $N$  to the restructured region  $R$ . This assumption will be removed later in the text after deriving a suitable exchange term (Section 2.2).

After introducing the restructured volume fraction  $\alpha$ , we can refer the previous balance over the total volume  $V$  by substituting  $dV_N = (1 - \alpha) dV$  and  $dV_R = \alpha dV$ :

$$\frac{\partial}{\partial t} \int C_N (1 - \alpha) dV = \int D_N \nabla^2 C_N (1 - \alpha) dV + \int S(1 - \alpha) dV \quad (4)$$

$$\frac{\partial}{\partial t} \int C_R \alpha dV = \int D_R \nabla^2 C_R \alpha dV + \int S \alpha dV \quad (5)$$

The arbitrariness of  $V$  allows us to convert the integral equations into the local relations, obtaining the following rate equations that are referred to the total control volume  $V$ :

$$\frac{\partial}{\partial t} ((1 - \alpha)C_N) = D_N \nabla^2 ((1 - \alpha)C_N) + (1 - \alpha)S \quad (6)$$

$$\frac{\partial}{\partial t} (\alpha C_R) = D_R \nabla^2 (\alpha C_R) + \alpha S \quad (7)$$

or equivalently:

$$\frac{\partial C_1}{\partial t} = D_N \nabla^2 C_1 + (1 - \alpha)S \quad (8)$$

$$\frac{\partial C_2}{\partial t} = D_R \nabla^2 C_2 + \alpha S \quad (9)$$

Eqs. (8) and (9) represent the evolution of the concentrations  $C_1$  and  $C_2$  over the total volume  $V$ . As anticipated, these two equations lack an exchange term that should influence both.

Additionally, it is possible to work out Eqs. (6) and (7) by considering that restructured volume fraction  $\alpha$  varies with time, to obtain the following differential equations for  $C_N$  and  $C_R$ . First, we develop the product derivatives:

$$\frac{\partial}{\partial t} ((1 - \alpha)C_N) = -\frac{\partial \alpha}{\partial t} C_N + (1 - \alpha) \frac{\partial C_N}{\partial t} \quad (10)$$

$$\frac{\partial}{\partial t} (\alpha C_R) = \frac{\partial \alpha}{\partial t} C_R + \alpha \frac{\partial C_R}{\partial t} \quad (11)$$

Then, we combine Eqs. (6), (7) with Eqs. (10), (11), to get:

$$\frac{\partial C_N}{\partial t} = D_N \nabla^2 C_N + S + \frac{1}{1 - \alpha} \frac{\partial \alpha}{\partial t} C_N \quad (12)$$

$$\frac{\partial C_R}{\partial t} = D_R \nabla^2 C_R + S - \frac{1}{\alpha} \frac{\partial \alpha}{\partial t} C_R \quad (13)$$

where the last terms represent:

- $+\frac{1}{1 - \alpha} \frac{\partial \alpha}{\partial t} C_N$ : the increase of  $C_N$  after the decrease of volume  $V_N$ .
- $-\frac{1}{\alpha} \frac{\partial \alpha}{\partial t} C_R$ : the decrease of  $C_R$  after the increase of volume  $V_R$ .

Nevertheless, Eqs. (12) and (13) have one singularity each,  $\alpha$  and  $(1 - \alpha)$ , that may pose some problems in a rigorous numerical implementation. For this reason, later in the text, we will show that it is preferable to work with the variables  $C_1$  and  $C_2$ .

We also compute the time derivative of the total concentration  $C_T$  to analyse its behaviour:

$$\frac{\partial C_T}{\partial t} = \frac{\partial}{\partial t} ((1 - \alpha)C_N) + \frac{\partial}{\partial t} (\alpha C_R) \quad (14)$$

Resulting in the sum of Eqs. (10) and (11):

$$\frac{\partial C_T}{\partial t} = -\frac{\partial \alpha}{\partial t} C_N + (1 - \alpha) \frac{\partial C_N}{\partial t} + \frac{\partial \alpha}{\partial t} C_R + \alpha \frac{\partial C_R}{\partial t} \quad (15)$$

By substituting Eqs. (12) and (13) into the previous Eq. (15), it results that:

$$\frac{\partial C_T}{\partial t} = (1 - \alpha) D_N \nabla^2 C_N + \alpha D_R \nabla^2 C_R + S \quad (16)$$

Eq. (16) states that the rate of change of the total concentration  $C_T$  over the control volume  $V$  is governed by two diffusional leakage terms (one per volume region), and the global production rate  $S$ . In particular:

- $\frac{\partial C_T}{\partial t}$  is referred to the total volume  $V$ .
- $(1-\alpha)D_N \nabla^2 C_N$  is referred to the volume  $V$ , thanks to the  $(1-\alpha)$  term.
- Likewise,  $\alpha D_R \nabla^2 C_R$  is referred to the volume  $V$  thanks to the  $\alpha$  term.
- The source  $S$  is the production rate per unit volume.

## 2.2. Sweeping exchange term

In this Section, we apply a discrete approach to derive an exchange term of gas atoms from the non-restructured region  $N$  to the restructured one  $R$ . Given the control volume  $V$ , we suppose it is formed by a number  $n$  of volumes  $V_N$  and a number  $m$  of volumes  $V_R$ . Each volume  $V_N$  contains  $N_N$  atoms, and each  $V_R$  containing  $N_R$  atoms. We simplify the derivation by neglecting previously considered production and leakage terms.

Then, we assume a discrete increase of  $\alpha$  during a single time step. Labelling with  $i$  the initial values:

$$C_{Ni} = \frac{nN_N}{nV_N} = \frac{N_N}{V_N} \quad (17)$$

and

$$C_{Ri} = \frac{mN_R}{mV_R} = \frac{N_R}{V_R} \quad (18)$$

We suppose that during the considered time step, a certain number  $k$  of volumes, together with the corresponding atoms, have transformed from  $V_N$  to  $V_R$ . Hence, labelling  $f$  the final values:

$$C_{Nf} = \frac{(n-k)N_N}{(n-k)V_N} = \frac{N_N}{V_N} \quad (19)$$

and

$$C_{Rf} = \frac{mN_R + kN_N}{mV_R + kV_N} \quad (20)$$

This discrete representation clearly shows how a pure restructuring operation does not influence  $C_N$  but only  $C_R$ . Eq. (20) can be worked out giving:

$$C_{Rf} = \frac{C_{Ri} + \frac{k}{m} \frac{N_N}{V_N}}{1 + \frac{k}{m} \frac{V_N}{V_R}} \quad (21)$$

In the reasonable hypothesis that during a single time step, the restructuring process transforms a limited portion of the control volume  $V$ , we can consider  $k \ll n$  and  $k \ll m$  and linearise the previous term to get  $dC_R = C_f^R - C_i^R$  as:

$$dC_R = -C_{Ri} \frac{k}{m} \frac{V_N}{V_R} + \frac{k}{m} \frac{N_N}{V_R} - \frac{k^2}{m^2} \frac{N_N V_N}{(V_R)^2} \quad (22)$$

where again, the last term can be neglected, providing:

$$dC_R = -C_{Ri} \frac{k}{m} \frac{V_N}{V_R} + \frac{k}{m} \frac{N_N}{V_R} \quad (23)$$

By grouping  $\frac{k}{m} \frac{V_N}{V_R}$ , the following equation is obtained:

$$dC_R = \frac{k}{m} \frac{V_N}{V_R} (C_N - C_{Ri}) \quad (24)$$

If we introduce the discrete  $\alpha$  terms.

$$\alpha_i = \frac{mV_R}{V}, \quad \alpha_f = \frac{mV_R + kV_N}{V} = \alpha_i + k \frac{V_N}{V} \quad (25)$$

we get:

$$\frac{\alpha_f - \alpha_i}{\alpha_i} = \frac{k}{m} \frac{V_N}{V_R} \quad (26)$$

Resulting in the following exchange term:

$$dC_R = \frac{d\alpha}{\alpha_i} (C_N - C_i^R) \quad (27)$$

Or, in the continuous limit:

$$\frac{\partial C_R}{\partial t} = \frac{1}{\alpha} \frac{\partial \alpha}{\partial t} (C_N - C_R) \quad (28)$$

Hence, Eq. (28) represents the pure sweeping term that transfers gas atoms from the non-restructured region  $N$  to restructured one  $R$  after a virtual step of the restructuring process. To express the exchange term (Eq. (28)) for  $C_1$  and  $C_2$ , we must consider that during the restructuring process  $dC_N = 0$  ( $C_{Ni} = C_{Nf}$ ), while  $dC_R \neq 0$ . Namely:

$$\begin{cases} \frac{\partial C_N}{\partial t} = 0 \\ \frac{\partial C_R}{\partial t} = \frac{1}{\alpha} \frac{\partial \alpha}{\partial t} (C_N - C_R) \end{cases} \quad (29)$$

In terms of  $C_1$  and  $C_2$ , we obtain:

$$\begin{cases} \frac{\partial C_1}{\partial t} = 0 \\ \frac{\partial C_2}{\partial t} = \frac{1}{\alpha} \frac{\partial \alpha}{\partial t} \left( \frac{C_1}{1-\alpha} - \frac{C_2}{\alpha} \right) \end{cases} \quad (30)$$

After expanding the product derivatives, the isolated exchange terms are:

$$\begin{cases} \frac{\partial C_1}{\partial t} = -\frac{1}{1-\alpha} \frac{\partial \alpha}{\partial t} C_1 \\ \frac{\partial C_2}{\partial t} = \frac{1}{1-\alpha} \frac{\partial \alpha}{\partial t} C_1 \end{cases} \quad (31)$$

Because of the linearity of the operations involved so far, we can use the superposition principle and incorporate Eq. (29) into Eqs. (12) and (13):

$$\frac{\partial C_N}{\partial t} = D_N \nabla^2 C_N + \frac{1}{1-\alpha} \frac{\partial \alpha}{\partial t} C_N + S \quad (32)$$

$$\frac{\partial C_R}{\partial t} = D_R \nabla^2 C_R - \frac{1}{\alpha} \frac{\partial \alpha}{\partial t} C_R + \frac{1}{\alpha} \frac{\partial \alpha}{\partial t} (C_N - C_R) + S \quad (33)$$

Or similarly, we can include Eq. (31) into Eqs. (8) and (9):

$$\begin{cases} \frac{\partial C_1}{\partial t} = D_N \nabla^2 C_1 - \frac{1}{1-\alpha} \frac{\partial \alpha}{\partial t} C_1 + (1-\alpha)S \\ \frac{\partial C_2}{\partial t} = D_R \nabla^2 C_2 + \frac{1}{1-\alpha} \frac{\partial \alpha}{\partial t} C_1 + \alpha S \end{cases} \quad (34)$$

The last system of equations gives a simple and consistent representation of the mass balances due to production, leakage, and gas sweeping from phase  $N$  to phase  $R$  in the volume  $V$ .

## 3. Application to fission gas depletion in HBS

In this Section, Eq. (34) is applied in the SCIANITX code (Zullo et al., 2023; Pizzocri et al., 2020) to assess the prediction of the fission gas depletion process. In particular, with the SCIANITX code, we consider the state-of-the-art representation of the fission gas diffusion problem (Rest et al., 2019; Barani et al., 2020; Booth, 1957; Pastore et al., 2013). Summarising, the working hypotheses are the following:

- The single fuel grain is modelled as a spherical domain.
- In line with the new SCIANITX 2.0 code structure, we consider two phases of the  $\text{UO}_2$ . The non-restructured and restructured  $\text{UO}_2$  (i.e.,  $\text{UO}_2$ -HBS). Each phase features its properties, e.g., different grain sizes.
- Each  $\text{UO}_2$  phase forms a dedicated system (e.g., Xe-in- $\text{UO}_2$  and Xe-in- $\text{UO}_2$ -HBS), each one with its specific properties (e.g., the diffusivity of xenon in the fuel matrix).

With the conventional symmetry boundary conditions at the centre of the spherical domain and perfect sink boundary conditions at the border of the spherical domain (Booth, 1957; Speight, 1969; Forsberg and Massih, 1985a,b; Pastore et al., 2013; Pizzocri et al., 2016; Pastore et al., 2018; Zullo et al., 2022). On top of the fission gas behaviour operated by SCIANITX (e.g., the evolution of the intra-granular bubble

**Table 1**  
Values of the model parameters.

Matrix	Parameter	Value	Reference
UO <sub>2</sub>	$D$	$D = D_1 + D_2 + D_3$	Turnbull et al. (1989)
UO <sub>2</sub>	$a_1$ , grain size	5 $\mu\text{m}$	Olander (1976)
UO <sub>2</sub>	$g$ , trapping rate	$g = 4\pi DN_{\text{ig}}R_{\text{ig}}$	Ham (1958)
UO <sub>2</sub>	$b$ , re-solution rate	$b = 2\pi\mu_{\text{ff}}\dot{F}(R_{\text{ig}} + R_{\text{ff}})^2$	Olander and Wongsawaeng (2006)
UO <sub>2</sub> -HBS	$D$	$D = 4.5 \times 10^{-42}\dot{F}$	Pizzocri et al. (2017), Barani et al. (2020)
UO <sub>2</sub> -HBS	$a_2$ , grain size	150 nm	Ray et al. (1997), Barani et al. (2020)

population (Zullo et al., 2023; Pizzocri et al., 2020, 2018)), we consider the following fission gas diffusion problem:

$$\begin{cases} \frac{\partial C_1}{\partial t} = \frac{D_1}{a_1^2} \nabla^2 C_1 - gC_1 + bm_1 - \frac{1}{1-\alpha} \frac{\partial \alpha}{\partial t} C_1 + (1-\alpha)S \\ \frac{\partial m_1}{\partial t} = gC_1 - bm_1 - \frac{1}{1-\alpha} \frac{\partial \alpha}{\partial t} m_1 \\ \frac{\partial C_2}{\partial t} = \frac{D_2}{a_2^2} \nabla^2 C_2 + \frac{1}{1-\alpha} \frac{\partial \alpha}{\partial t} (C_1 + m_1) + \alpha S \end{cases} \quad (35)$$

With  $a_1$  and  $a_2$  the radius of the fuel grain in non-restructured UO<sub>2</sub> and in restructured UO<sub>2</sub>, respectively. Numerical values are given in Table 1. Eq. (35) represent the extended intra-granular problem that we solve in SCIENTIX, tailored to represent a smooth transition from UO<sub>2</sub> to UO<sub>2</sub>-HBS phase. In particular,

- $C_1$  represents the concentration of fission gas in dynamic solution within the non-restructured matrix.
- $m_1$  represents the concentration of fission gas trapped in intra-granular bubbles within the non-restructured matrix.
- $C_2$  represents the concentration of fission gas in dynamic solution within the restructured matrix.

Since the intra-granular diffusion problem we consider with SCIENTIX is discretised with a spectral diffusion algorithm, we avoid solving the first two differential equations on the non-restructured micron-sized fuel grain and the third one on the restructured nanometric fuel grain by non-dimensionalising the Laplace operator. In other words, we solve the diffusion problem on the unit-radius sphere without changing the system eigenvalues  $\frac{\pi^2 D}{a^2} k^2$  (Zullo et al., 2022).

We consider the restructured volume fraction proposed by Barani and co-workers (Barani et al., 2020, 2022):

$$\alpha = 1 - \exp(-2.77 \times 10^{-7} \beta_{\text{eff}}^{3.54}) \quad (36)$$

The restructured volume fraction  $\alpha$  is proportional to the local effective burnup  $\beta_{\text{eff}}$ , used as a virtual threshold to define the HBS formation at temperature lower than the healing temperature threshold ( $T_{\text{th}} = 1273.15$  K):

$$\beta_{\text{eff}} = \int H(T_{\text{th}}) d\beta \quad (37)$$

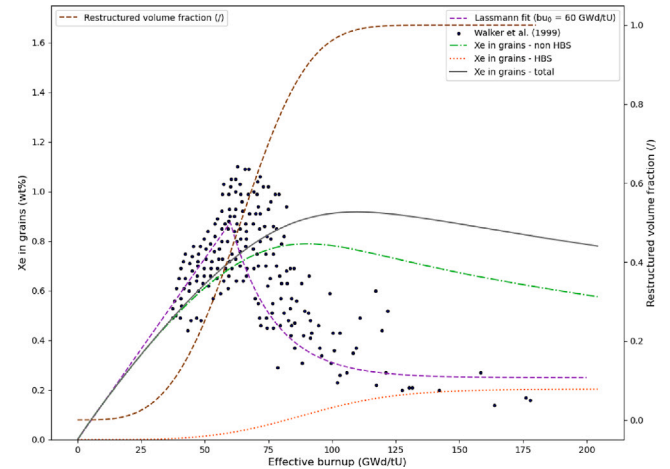
where  $\beta$  (MWd kgU<sup>-1</sup>) is the fuel local burnup, and  $H$  is the Heaviside function (Khvostov et al., 2005; Holt et al., 2014; Pizzocri et al., 2017).

#### 4. Results and discussion

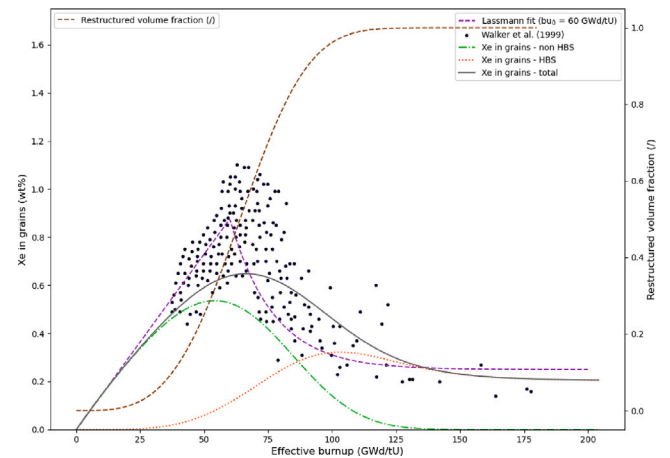
We applied the previous modelling approach to simulate the behaviour of a representative Light Water Reactor (LWR) fuel pellet rim portion with SCIENTIX. Namely, we considered a steady-state irradiation history up to 200 GWd/tU at low temperature (723 K) and high fission rate ( $2 \times 10^{19}$  fission m<sup>-3</sup> s<sup>-1</sup>). Figs. 1 and 2 show the results obtained from solving Eq. (35). To evaluate the impact of the sweeping term (Eq. (31)), Fig. 1 is obtained by neglecting such term while it is considered in Fig. 2.

Moreover, Figs. 1 and 2 include separate contributions for xenon concentrations retained in UO<sub>2</sub> matrix:

- The contribution from the xenon retained in non-restructured UO<sub>2</sub> grains ( $C_1 + m_1$ ).



**Fig. 1.** Intra-granular xenon concentration calculated with the present model, without the sweeping term, as a function of local effective burnup. The comparison with the model from Lassmann et al. (1995b), is reported (purple dotted line), together with experimental data measured by EPMA on several samples (black dots, from Walker, 1999).



**Fig. 2.** Intra-granular xenon concentration calculated with the present model, including the sweeping term, as a function of local effective burnup. The comparison with the model from Lassmann et al. (1995b), is reported (purple dotted line), together with experimental data measured by EPMA on several samples (black dots, from Walker, 1999).

- The contribution from the xenon retained in restructured UO<sub>2</sub> grains ( $C_2$ )

and their sum. We stress again that the only purpose of Fig. 1 is to clarify the impact of the sweeping term which ensures a more realistic representation of the xenon depletion process together with advantages of numerical and implementation-wise nature. In particular, moving from Figs. 1 to 2, it can be noted that the mass transfer between the two matrix phases is responsible for the smooth decrease of the retained gas in reasonable agreement with available measured

data (from Walker, 1999). Moreover, the tail of the calculated intra-granular xenon concentration depends on the grain size of the  $\text{UO}_2$ -HBS matrix (i.e., asymptotic gas concentration). We use a value of 150 nm (Table 1), that is in line with experimental observations (Cappia et al., 2016, 2022; McKinney et al., 2023; Noirot et al., 2008; Khvostov et al., 2005) and previous modelling approaches (Lassmann et al., 1995a; Veshchunov and Shestak, 2009; Veshchunov et al., 2006; Pizzocri et al., 2017; Barani et al., 2020), and that agrees with the experimental data considered in Fig. 2. On the other hand, such grain size has its degree of intrinsic uncertainty (Pizzocri et al., 2017; Barani et al., 2020). The experimental data in Fig. 2 comes from different irradiation conditions and initial fuel characteristics. Considering the scattering of the considered data, the agreement can be regarded as acceptable (Lassmann et al., 1995b). Some degree of uncertainty must be necessarily taken into consideration, for instance, in the single-atom diffusivities of the xenon in the fuel matrices (reported in Table 1), regarding the experimental characterisation, which in principle could allow us to consider different (but reasonable) initial fuel grain sizes. Most importantly, some uncertainty must be ascribed to the 0-dimensional simulation we are performing with the SCIANITX code, considering that a constant temperature and constant fission rate are not fully representative of the local temperature and burnup gradients. For instance, uncertainty is brought to the predicted position of the peak of the retained xenon ( $\sim 60$  GWd/tU in the considered simulation), which depends on the restructured volume fraction  $\alpha$ , and on the local burnup itself. Further refinement of this feature is to be considered in a more detailed representation of the restructured volume fraction, possibly including other quantities like the dislocation density as it was done in other works (Veshchunov et al., 2006; Veshchunov and Shestak, 2009). Issues of this kind have been addressed and detailed in previous works (Pizzocri et al., 2017; Barani et al., 2020), and a similar examination and comparison are valid for the present work.

## 5. Conclusions

This work outlines a modelling approach to describe the intra-granular fission gas behaviour in  $\text{UO}_2$ , considering the evolution of the non-restructured and restructured fuel matrices. The model leverages the definition of the restructured volume fraction, which currently depends on the local effective burnup. The primary outcome of the model is a simple yet effective description of two intra-granular gas diffusion problems interconnected by an exchange term, sweeping gas atoms from one phase to the other. In addition, by leveraging the spectral diffusion algorithm for the intra-granular fission gas problem, we avoided dealing with two different integration domains (micrometric and sub-micrometric grains) and solved a single spatial dimensionless problem. The model developed has been implemented in SCIANITX, a 0D meso-scale code for physics-based modelling of fission gases in nuclear oxide fuel and compared with experimental data and semi-empirical models. The model results are consistent with the experimental data and past semi-empirical models, with the sweeping term providing a smooth representation of the xenon depletion in forming HBS.

## CRediT authorship contribution statement

**G. Zullo:** Writing – original draft, Visualization, Software, Methodology, Investigation, Data curation, Conceptualization. **A. Scolaro:** Writing – review & editing, Supervision, Conceptualization. **T. Barani:** Writing – review & editing, Supervision, Conceptualization. **D. Pizzocri:** Writing – review & editing, Supervision, Project administration, Methodology.

## 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.

## Data availability

Data will be made available on request.

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