Physics-based modeling of High Burnup Structure porosity in UO₂ fuel: Implementation and validation in SCIANTIX

Author: Ulisse Frattini

Advisor: Prof. Lelio Luzzi

Co-advisor: Davide Pizzocri, Giovanni Zullo

Academic year: 2024-2025

1. Introduction

The high burnup structure (HBS) is a distinctive microstructural transformation that occurs in UO₂ nuclear fuel subjected to extended irradiation. It typically forms in the outer rim of the pellet, where the combined effects of high irradiation damage, associated with local burnups exceeding 45–50 MWd/kgU, and low operating temperatures (below 1000°C) hinder defect recovery processes. This restructuring results in a fine-grained microstructure with submicron grains, high porosity and marked depletion of intra-granular fission gases (FG). Such a process has significant implications for nuclear fuel performance. Its associated porosity contributes to additional fuel swelling, alters key material properties such as thermal conductivity and elastic modulus, and critically affects the fission gas behavior (FGB). In particular, the large amount of gas stored in HBS pores plays a key role during transients like Loss-of-Coolant Accidents (LOCA) and Reactivity-Initiated Accidents (RIA), where rapid gas release and fragmentation may compromise safety. As nuclear programs aim to extend the in-reactor lifetime of fuel by increasing target burnup, developing advanced models for HBS behavior becomes essential. These models must be integrated into fuel performance codes (FPCs) to predict fuel behavior under both steady-state and accident conditions. Despite significant advancements over the years, current FPCs still face major limitations in modeling HBS formation and evolution. Empirical models (e.g., FAST, TRANSURANUS) are efficient but rely on burnup-based correlations fitted on experimental data and have limited predictive capability outside their calibration range. They often impose arbitrary thresholds for restructuring onset, reducing physical accuracy and transferability. Semi-empirical approaches (e.g., DIONISIO, FALCON, BISON) improve generality by combining empirical and mechanistic elements, yet still depend on fitted parameters and phenomenological assumptions. Mechanistic models (e.g., MARGARET, MFPR/R) offer higher physical fidelity through rate theory or cluster dynamics (CD), but at the cost of computational complexity and often include simplifying assumptions that limit realism in capturing gradual and heterogeneous microstructural changes.

This work introduces a new physics-based model for inter-granular pore evolution in the HBS region, developed and implemented within

the SCIANTIX 2.0 code. The model extends the original framework by introducing a novel coupling between intra-granular and grain boundary gas behavior, and by explicitly distinguishing between restructured (i.e., HBS) and non-restructured (NR) fuel domains—two key improvements enhancing gas conservation and physical consistency. By doing so, the model overcomes key limitations of existing approaches, such as the use of an empirical correlations for porosity and arbitrary burnup thresholds for restructuring onset. Additional mechanisms such as fission gas sweeping between NR and HBS regions and vacancy absorption under overpressure are incorporated. At the same time, phenomena like pore nucleation, gas precipitation, re-solution and coalescence are retained and reformulated using a moment-based approach derived from CD. The model's performance is assessed through comparisons with experimental data and reference models, focusing on pore number density, average pore radius, and HBS porosity. A separate-effect validation supports the model's accuracy and robustness, enabling improved prediction of fuel behavior at high burnup.

2. Model

HBS formation has been modeled following the work of Barani et al. [1] describing the progressive restructuring of the fuel matrix as a function of local effective burnup exploiting a Kolmogorov–Johnson–Mehl–Avrami (KJMA) formalism.
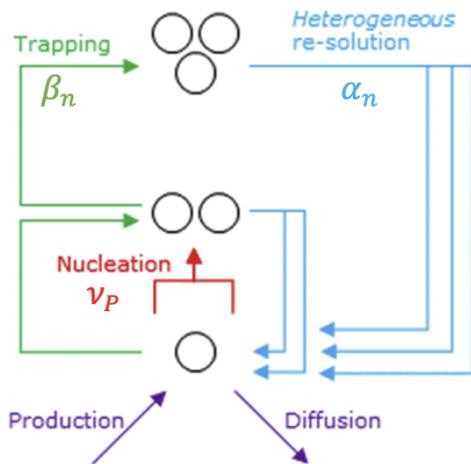

Figure 1: Schematization of the phenomena accounted by the CD master equations

Then a CD model has been adopted with suitable simplifications to account for some of the main phenomena in HBS porosity evolution (Fig.1): pore nucleation (ν_P), gas precipitation into (β_n) and re-solution from (α_n) HBS pores. Such a model has been reduced in its dimensionality to allow a suitable application in engineering codes through the first three central moments of the cluster size distribution:

$$N_p = \sum_{n=2}^{\infty} c_n \qquad (1)$$

$$A = \sum_{n=2}^{\infty} c_n n \qquad (2)$$

$$B = \sum_{n=2}^{\infty} c_n (n - \bar{n})^2 \qquad (3)$$

where c_n ($atoms\ m^{-3}$) is the number density of clusters composed by n gas atoms, $\bar{n}$ ($atoms\ pore^{-1}$) is the mean of pore size distribution. A ($atoms\ m^{-3}$) and B ($atoms^2\ m^{-3}$) represent the Xe concentration in HBS pores and its variance, respectively. Finally, the resulting system of ODEs has been coupled with a model of gas transfer across the NR–HBS interfaces that accounts for the sweeping phenomenon and the depletion of restructured grains. The former is shaped through an exchange term L, proportional to the time evolution of restructured volume fraction α_r, as proposed by Zullo [2] that connected both the NR and HBS grain boundaries and the NR and HBS grain interiors:

$$L = \frac{1}{1 - \alpha_r} \frac{\partial \alpha_r}{\partial t} \qquad (4)$$

While the latter is modeled via simple geometric considerations, defining a coefficient w for the quantity of gas atoms that directly diffuses in HBS pores instead of HBS grain boundaries:

$$w = \frac{4\pi (R_{\bar{n}}^P)^2 N_p}{\frac{3}{a}} \qquad (5)$$

where $R_{\bar{n}}^P$ (m) is the number-averaged radius of the distribution, a (m) is the HBS grain radius, and the numerator and the denominator represent the surface-to-volume ratios of HBS pores and HBS grains, respectively. The resulting system of differential equations, reported below, integrates all the physical mechanisms described

above and represents the final formulation of the proposed model.

$$\begin{cases} \dfrac{dN_p}{dt} = \nu_P - \alpha_n N_p \\[2mm] \dfrac{dA}{dt} = 2\nu_P - \alpha_n A + \beta_n N_p + wU_2 \\[2mm] \dfrac{dB}{dt} = \nu_P(\bar{n} - 2)^2 - \alpha_n B + \beta_n N_p \\[2mm] \dfrac{dc_{gb,1}}{dt} = -Lc_{gb,1} + U_1 \\[2mm] \dfrac{dc_{gb,2}}{dt} = (1 - w)U_2 - (2\nu_P - \alpha_n A \\[2mm] \qquad\qquad + \beta_n N_p) + Lc_{gb,1} \end{cases} \quad (6)$$

Note that a holistic approach to FG conservation has been applied, ensuring that the gas released from the grains is consistently redistributed between the grain boundaries and the inter-granular pores. To achieve this, two source terms U_1 and U_2 ($atoms\ m^{-3}\ s^{-1}$) representing the intra-granular gas release from NR and HBS grains respectively, have been introduced and explicitly partitioned between the recipient regions.

HBS pores are known to be strongly over-pressurized due to the continuous inflow of FG, primarily xenon, from the surrounding depleted grains. This internal pressure provides a strong driving force for vacancy absorption from the matrix, acting as a mechanism of pressure relief. To limit computational complexity, the CD model was deliberately formulated as a single-species framework, as it's shown in Eq.(6). As a result, vacancy absorption is not treated explicitly for each cluster size but is instead modeled through the average pore radius, which provides a physically consistent and computationally efficient approximation. The rate of vacancy absorption into the pores is expressed as:

$$\frac{dn_{vp}}{dt} = \frac{2\pi D_{gb}^v \rho_P}{k_B T \zeta}(p^P - p_{eq}^P) \qquad (7)$$

where n_{vp} ($/$) is the number of vacancies per pore, D_{gb}^v ($m^2\ s^{-1}$) is the vacancy diffusion coefficient at grain boundaries, ρ_P (m) is the radius of the Wigner-Seitz cell assigned to each pore, k_B (J K^{-1}) is the Boltzmann constant, T (K) is the local temperature and ζ is a dimensionless factor depending on the ratio between average pore radius and the radius of Wigner-Seitz cell.

The total pore volume variation over time includes contributions from both FG absorption and vacancy incorporation, and is described by:

$$\frac{dV_p}{dt} = \omega\frac{d\bar{n}}{dt} + \Omega\frac{dn_{vp}}{dt} \qquad (8)$$

where ω and Ω (m^3) are the volume occupied by each gas atom and the vacancy volume respectively.

Subsequently, the HBS porosity is evaluated using the following expression:

$$\xi = \frac{4\pi}{3}N_p(R_{\bar{n}}^P)^3 \qquad (9)$$

thus overcoming the limitations of the empirical formulation implemented in SCIANTIX 2.0. In addition to growth by gas and vacancy absorption, pores may also grow through interconnection by impingement, which occurs when neighboring pores coalesce due to expansion.

All the parameters used in the present model have been selected based on values commonly reported in the literature for the system under investigation, except the vacancy diffusion coefficient at the grain boundaries, which has been chosen through a dedicated sensitivity analysis.

3.   Results and discussion

The simulations performed using this model have been compared with the model developed by Barani and coworkers [3] and the previous model implemented in SCIANTIX 2.0 [4]. For the sake of comparison, many experimental data sets have been included in the following plots. The calculated quantities have been obtained considering an irradiation history representative for the conditions met in the periphery of a fuel pellet in Pressurized Water Reactors (PWRs), namely, a temperature of $723K$, a fission rate density equal to $2 \cdot 10^{19}\ fiss\ m^{-3}\ s^{-1}$ and a hydrostatic stress equal to $20MPa$ representative for pellet-cladding mechanical interaction (PCMI). The considered experimental databases consist of samples coming from a homogeneous set of material composition (i.e., uranium dioxide) and irradiation (i.e., commercial PWRs).
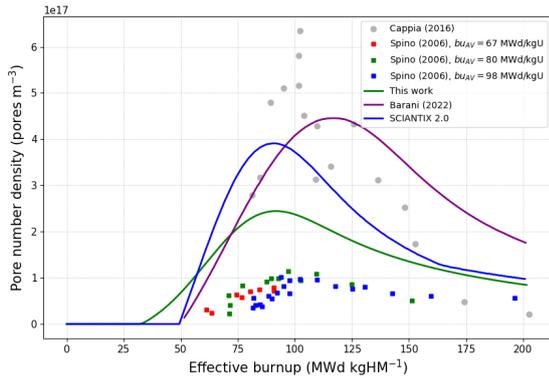
Figure 2: Comparison of experimental and predicted HBS pore number densities from current, previous, and state-of-the-art models

Fig. (2) shows that the model developed in this work reproduces the same trend as the other curves. It shows a peak around 100 MWd/kgU, but remains slightly conservative in the burnup range between 80 and 150 MWd/kgU. At low burnup levels, all models predict negligible pore density, in agreement with experimental observations, while at high burnup, the present model offers a better estimate of pore number density, unlike Barani's model, which tends to overestimate experimental data.
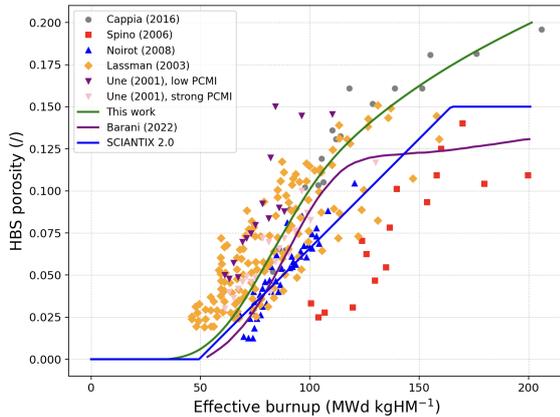

Figure 3: Comparison of experimental and predicted HBS porosity from current, previous, and state-of-the-art models

HBS porosity evolution (Fig.3) predicted by the model aligns well with experimental data at high burnups (above 130 MWd/kgU), unlike SCIANTIX 2.0, which underestimates porosity due to a fixed 15% plateau. The new model reflects the continuous growth observed in experiments more realistically. Barani's

model underpredicts porosity across most of the burnup range, especially beyond 100 MWd/kgU, suggesting that it may not fully account for coalescence. At low burnup values (<50 MWd/kgU), all models predict negligible porosity, in agreement with experimental observations. Notably, the proposed model reproduces the onset of porosity at the same burnup level of SCIANTIX 2.0 without relying on any predefined threshold. Overall, the new model delivers a more consistent and physically grounded description of porosity evolution in the HBS, particularly at high burnup levels. In contrast to benchmark models, it improves prediction accuracy in a regime where reliable modeling of swelling and gas release is essential.

All the models shown in Fig.(4) exhibit the expected increasing trend of pore radius with burnup, consistent with the underlying mechanisms of gas and vacancies accumulation and pore coarsening.
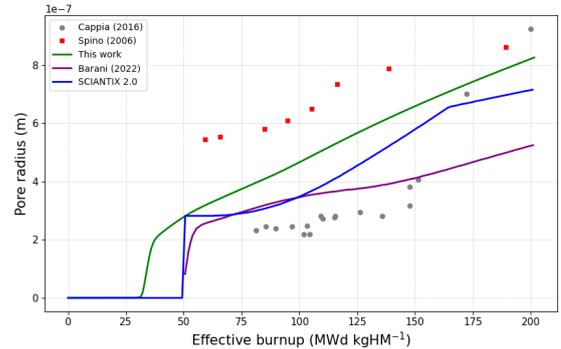

Figure 4: Comparison of experimental and predicted HBS average pore radius, previous, and state-of-the-art models

However, clear differences emerge in both the rate of growth and the absolute values predicted. The present model predicts a more rapid increase in pore size starting from the onset of restructuring, and continues to grow steadily with burnup. This behavior is in good agreement with the upper range of the experimental dataset, particularly in the high burnup regime (above 130 MWd/kgU), where pore coarsening becomes dominant. In contrast, Barani's model underestimates the pore radius at that burnup range, confirming a limited treatment of coalescence. The SCIANTIX 2.0 prediction lies between the two, correctly capturing the initial

pore size but exhibiting slower growth beyond 150 MWd/kgU, which results in an underestimation at higher burnup levels. This behavior may be attributed to the lack of vacancy absorption modeling in SCIANTIX 2.0.

4.   Separate effect validation

In order to assess the physical accuracy and robustness of the proposed model, a separate-effect validation has been carried out. Nine radial positions within the rim region of the fuel pellet ($0.8 \leq \frac{R}{R_{max}} \leq 1$) were simulated using the model developed in this work, by appropriately tuning the fission rate density and local fuel temperature for each location. Specifically, the following ranges were adopted: $1.58 \cdot 10^{19} < \dot{F} < 3.89 \cdot 10^{19}$ fiss $m^{-3}s^{-1}$ and $666 < T < 861$ K. The corresponding average radial burnup was estimated at approximately $79 MWd/kgU$. The resulting radial profile was then compared with experimental datasets and existing reference models, spanning different average burnup values, to validate the model's capability to accurately reproduce porosity evolution across the whole HBS region.
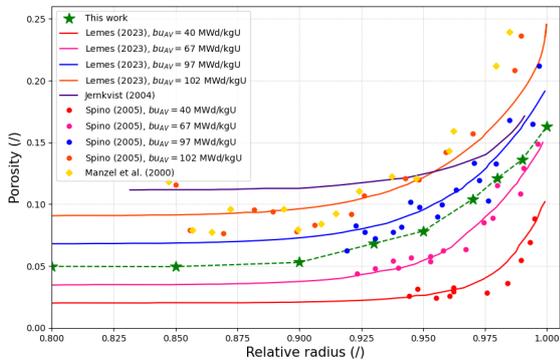
Figure 5:   Comparison between experimental results, simulations performed by state-of-the-art models and radial locations simulated by SCIANTIX for the HBS porosity vs. the relative radial position in the pellets

The results shown in Fig.(5) are satisfactory, as the simulated points lie between both two reference curves and two series of experimental points belonging to average burnups of 67 and $97 MWd/kgU$, which is consistent with the estimated average burnup of $bu_{AV} = 79 MWd/kgU$.
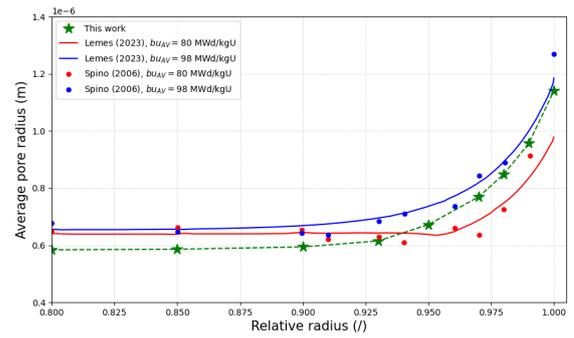
Figure 6: Comparison between experimental results, models and radial locations simulated by SCIANTIX for the radial evolution of average pore radius

A similar separate-effect validation methodology was applied to the other key figures of merit describing the HBS microstructure. For these cases, simulations were carried out using a different average burnup value (i.e., 90 MWd/kgU for the average pore radius and 40, 57, 67 MWd/kgU for the pore number density) to enable a more meaningful comparison with the available data. The results are presented in Figs.(6,7). The model successfully reproduces the trend of increasing average pore radius with burnup, closely following the evolution of porosity and reflecting the underlying restructuring mechanisms. These results confirm the model's ability to capture the key physical processes driving pore development in the HBS.
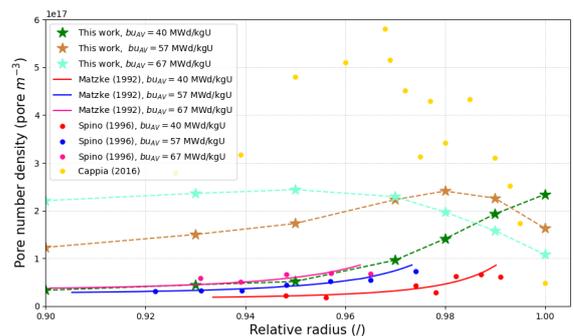
Figure 7: Comparison between experimental results, models and radial locations simulated by SCIANTIX for the radial evolution of pore number density

However, when comparing the predicted pore number density with available reference data, a noticeable deviation emerges. The model predicts a higher pore number density compared

to both experimental measurements and traditional model outputs. At average burnups of 57 and 67 MWd/kgU, simulations show a decreasing trend due to pore coalescence—an effect not captured in Spino's data, but consistent with Cappia's more recent observations. Only the 40 MWd/kgU case aligns with both experimental and reference model results, suggesting that the effective burnup threshold for coalescence has not yet been reached at this level. Conversely, at 57 and 67 MWd/kgU, this threshold appears to be exceeded, at least according to the present model, which predicts the onset of pore coalescence around a local effective burnup of 100MWd/kgU.

The discrepancy with Spino's data may be due to the model predicting pore growth to begin at slightly lower burnups than commonly assumed. This leads to an earlier onset of coalescence, particularly near the pellet edge where burnup peaks. Since experimental and reference data rarely cover these very peripheral regions, coalescence may simply not have been observed due to insufficient local burnup.

5. Conclusions

This thesis presents a physics-based model for the evolution of inter-granular porosity in the HBS of UO$_2$ fuel. Building upon the SCIANTIX 2.0 framework, where porosity was modeled empirically, the new formulation introduces key mechanisms such as vacancy absorption and fission gas sweeping between fuel regions, coupled with a more consistent treatment of grain boundary behavior. Pore coalescence is retained and integrated within a reduced CD scheme, ensuring both physical accuracy and computational efficiency.

The model has been calibrated and validated through separate-effect validation, showing strong agreement with experimental data across a wide burnup range. It also outperforms the previous SCIANTIX 2.0 implementation and Barani's model, particularly in predicting HBS porosity, a key indicator for swelling and fission gas release. The model demonstrates particularly strong predictive performance at burnups exceeding 100 MWd/kgU, where microstructural changes become more significant.

This work advances the predictive capabil-

ity of meso-scale fuel models and provides a more robust basis for future multi-scale simulations. An integral validation within the SCIANTIX–TRANSURANUS coupling is foreseen, along with possible extensions to other fuel systems, supporting safer and more efficient reactor operation at extended burnup conditions.

6. Acknowledgements

References

[1] Tommaso Barani, Davide Pizzocri, Fabiola Cappia, Lelio Luzzi, Giovanni Pastore, and P Van Uffelen. Modeling high burnup structure in oxide fuels for application to fuel performance codes. part i: High burnup structure formation. *Journal of Nuclear Materials*, 539:152296, 2020.

[2] G Zullo, Alessandro Scolaro, T Barani, and D Pizzocri. Two-phase modelling for fission gas sweeping in restructuring nuclear oxide fuel. *Nuclear Engineering and Design*, 429:113602, 2024.

[3] Tommaso Barani, Davide Pizzocri, Fabiola Cappia, Giovanni Pastore, Lelio Luzzi, and Paul Van Uffelen. Modeling high burnup structure in oxide fuels for application to fuel performance codes. part ii: Porosity evolution. *Journal of Nuclear Materials*, 563:153627, 2022.

[4] G Zullo, D Pizzocri, and L Luzzi. The SCIANTIX code for fission gas behaviour: Status, upgrades, separate-effect validation, and future developments. *Journal of Nuclear Materials*, 587:154744, 2023.