High Burn Up Structure formation and growth and fission product release modelling: new simulations in the mechanistic code MFPR-F

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ABSTRACT

A model for the High Burn up Structure (HBS) formation and growth has been developed and implemented in the MFPR-F code. It is based on the model of dislocation density simulation already presented [16] and modified for the specific conditions of high fission rate. This model gives correct results when compared to experimental data from [13] and [15] but still underestimates the restructuring at temperature>1000K. On this basis, a description of the HBS zone was performed with a fit of the pores characteristics on data from [12]. Finally a simplified model for fission product behaviour based on the pores pressure was implemented and tested on LOCA type conditions [4] with encouraging results. In high temperature annealing conditions the model underestimates the release because the significant effects of the transient on microstructure are not yet considered in the model.

1. Introduction

1.1 The context of the HBS modelling

The high burnup structure (HBS) is formed in the external part of irradiated nuclear fuel pellets when the conditions of local burnup (>~50GWd/t) and local temperature (<1300K) [10] are fulfilled. This zone is constituted by huge, micron-sized and over-pressurized porosities, surrounded by submicronic grains, with polyhedral or round shape. A very detailed characterization of this structure is given in [1]. The HBS zone has high gas retention capacities and does not evolve towards an interconnected channels system even at high porosities up to 24% [24]. Higher irradiation temperatures favour growth of the newly formed grains, but no increase at higher burnup has been measured [11]. The porosity and the average pore size increase with both local irradiation temperature and burnup, but upon exceeding a local burnup threshold of ≈100 GWd/tHM a decrease in the pore number density has been observed [11,12]. This structure has received special attention because of its mechanical interaction with the metallic cladding, namely during off-normal transients. The fuel thermal conductivity degradation was another issue of interest due to its potential effects on the maximum temperature at the centre line of the fuel pellet. At last, the gas and fission product behaviour in annealing transients was a further motivating topic as their release from the HBS zone was found to be very significant for both high temperature [3] or LOCA [4] transients.

The proper description and modelling of the mechanisms responsible for the HBS formation is still lacking, due to the complexity of the phenomena involved. A review of the main corresponding hypothesis is given in [2]. Recently, very detailed experiments with ion beams were performed to simulate the HBS formation [5] and to understand the processes at the very basic level. In the fuel performance codes, the description of the HBS formation and

behaviour remains often empirical or semi-empirical [6-9], even if based on data obtained from separate effects experimental studies [10-12]. In this paper, a mechanistic approach for the HBS formation and growth in uranium oxide fuel will be proposed and discussed. A simplified modelling of the fission product behaviour in the HBS for normal and off-normal conditions will also be proposed. Both models will be compared to available experimental data on HBS formation [10] and growth in LWR [13] and to data on fission gas and fission products release in high temperature [4] or LOCA [5] transients.

1.2 Main hypothesis of the models

The formation and growth of the HBS is described according to Nogita and Une [14] who observed that the accumulation of radiation damage results in the formation of tangled dislocation networks which form sub-grains, further acting as recrystallization nuclei. These low-angle grain boundaries networks were found to appear at 44GWd/t for a dislocation density ~$6.10^{14}m^{-2}$ and evolved into a subdivided grains structure with high angle boundaries completely formed at 83GWd/t, corresponding to a dislocation density of around $10^{15}m^{-2}$ [1]. Baranov et al [17] have recently confirmed this by means of dislocation dynamics simulations. On this basis, the simulation of the dislocation network formation and growth might be the basis of a model for HBS formation based on a dislocation density criterion. This model must also reproduce the effect of temperature on the HBS formation, clearly evidenced in the HBRP analytical experiments reported in [10, 15] in which 1300K seems to be the highest possible temperature for the HBS formation.

Such model for dislocation density evolution was already described in details in [16] and implemented in the MFPR code [22]. This model constitutes the basis for the present work with particular attention on the effect of some model key parameters and on some modifications implemented in the new MFPR-F to take into consideration the particular effects which could become important for high fission rate, which is typical of the HBS location in the fuel pellet.

The MFPR code was developed by IBRAE in collaboration with IRSN [27] (IBRAE: models and code development; IRSN: code application to interpretation of FP behaviour and benchmarking), and since 2011, the code has been developed independently by both institutes. IRSN is developing its own version MFPR-F, which has been coupled with TRANSURANUS.

The description of the HBS is based on the main experimental observations reported in [1] for the grain size and the pores pressure and in [1, 11, 12] for the pores size and concentration.

The fission products (FPs) behaviour is described following the observations reported in [1, 10] for the relative amount of gas in the grains and in the pores (or Cs and other so-called volatiles FPs) in normal in-pile operations. The authors reported a very significant migration of those species from the grains into the porosities. For the annealing conditions, the models are derived from the observations from [3, 4] which showed that the fission gas and fission product release is likely based on over pressurization of the pores during the transient. This is further supported by experimental observations from the NFIR program [25], and forms the basis of a specific model for the fragmentation of the high-burnup structure during LOCA [26]. This model assumes that the release occurs when the tangential stress at the pore surface exceeds the fracture strength of the fuel.

2. Model for the HBS formation and growth

As explained above, the model is based on the dislocation density evolution during irradiation which is described in detail in [16]. Only the main features are presented below.

2.1 The dislocation density evolution modelling

The process starts with the nucleation of uranium di-interstitials clusters during irradiation, giving the interstitial dislocation loops concentration evolution as expressed by equation 1 in the mean field approximation:

$$\frac{dC_{il}}{dt} = \frac{\alpha_l D_i c_i^2}{\Omega}\theta\left(D_i c_i - D_v\left(c_v - c_v^{eq}\right)\right) \qquad (1)$$

With C_{il} the dislocation loop concentration, D_i and D_v corresponding to the uranium interstitial and vacancy self-diffusion coefficient respectively, α_l the dislocation loop nucleation constant which is approximated by the interstitial-vacancy recombination constant, c_i represents the uranium interstitial concentration and θ the Heaviside function in which the argument means that dislocation loops can only nucleate during the initial period of irradiation, when this argument is positive. This corresponds to the fact that the dislocation bias for interstitial takes place for finite loops, but not for small di-interstitials clusters. Then, when the argument will become negative, the new formed small clusters will disappear and only the already formed dislocation loops will be stable and grow due to the non-zero bias factor for interstitials, ε, following the expression (2) implemented in the MFPR-F code :

$$\frac{dR_{il}}{dt} = \frac{2\pi}{B}\left(\left(1+2\varepsilon\right)D_i c_i - D_v\left(c_v - c_v^{eq}\right)\right)\begin{cases}\dfrac{1}{\ln\left(8R_{il}/R_d\right)}, & \text{if } R_{il} < L_{il};\\ \dfrac{1}{\ln\left(L_d/R_d\right)}, & \text{if } R_{il} \geq L_{il},\end{cases} \qquad (2)$$

Where R_d is the dislocation core radius estimated as $R_d \cong 3B$, with B the Burgers vector length, while L_d and L_{il} characterize the mean distance between dislocation and interstitial loops respectively

$L_d = 1/\sqrt{\pi\rho_d}$,

$L_{il} = \left(3/4\pi C_{il}\right)^{1/3}$

Then, the dislocation density evolves following the relation (3)

$$\rho_d^{(t)} = \rho_d^0 + 2\pi R_{il} C_{il} \qquad (3)$$

where $\rho_d^{(0)}$ is the initial network dislocation density. In this approach, it is assumed that when dislocation loops are large compared to the sphere of influence, i.e. $R_{il} \geq L_{il}$, the interstitial loops may be best approximated as straight dislocations of the length equal to their circumferences by using the relationship that the corresponding dislocation line density is $2\pi R_{il} C_{il}$. In this approximation, the growth rates of dislocation loops are given by the second part of Eq.(2) corresponding to the case $R_{il} \geq L_{il}$.

This model gives results in good agreement with the experimental observations reported in [14] for dislocation density evolution vs burn up in irradiated fuel, as illustrated by the following figure 1.

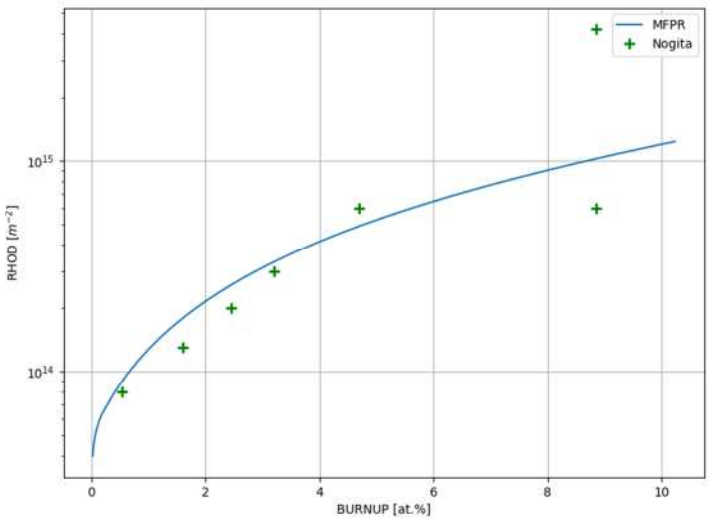

Figure 1: dislocation density evolution vs mean fuel pellet burnup at 1073K simulated with the MFPR-F model compared to experimental data [14]. The simulation has been carried out for a fission rate density of 10^{19} $m^{-3}s^{-1}$, an initial grain diameter of 9 µm, and as-fabricated porosity of 5%.

2.2 The model at high fission rate

The uranium interstitial concentration, c_i, is a key factor for the dislocation loop formation as shown in Eq (1). It is described by the following Eq (4) in MFPR-F:

$$\frac{dc_i}{dt} \sim K - \alpha D_i c_i c_v - K_d - \left(k_i^2 + k_{igb}^2\right) D_i c_i \qquad (4)$$

The first term, K, represents the production term which corresponds to the uranium Frenkel pair production rate K~ Fz_S (F the fission rate and z_S the damage formation in the fission track volume, ~2.10^5).

The second term represents the vacancy-interstitial recombination term (α is the recombination constant and c_v the vacancy concentration).

Recently, [28] from classical molecular dynamics simulations, this constant was investigated in more details by recombination radii re-evaluation. It was shown from these new results that the recombination radii are significantly larger than the widely used values for spontaneous recombination which will impact the recombination rate. As shown in [28] the new increased) values for improves significantly the results for HBS growth when compared to experimental values.

K_d is the term for interstitial loss for dislocations formation (see Eq 1) and the last terms represent the interstitial sink strength in extended defects (bubbles pores and dislocations) and grain boundaries, respectively.

The self-interstitial diffusion coefficient D_i is expressed by an Arrhenius form in which only the thermal part is considered. At low temperature (which is representative of the external part of the fuel pellet), D_i remains low and prevents any significant recombination with vacancies. In addition, the Frenkel pair production rate, K, becomes very high for high fission rate values (again typical in the rim of the fuel pellet). This induces a very quick increase of the dislocation density which can exceed the threshold value for restructuration between 6.10^{14} and $2.10^{15} m^{-2}$) at low, unrealistic, burnup value (10GWd/t or less).

This problem can be solved by considering, as was done in [17], a dislocation dynamics model with radiation-enhanced dislocation climbing mobility. This model gave very interesting features about restructuring. It showed that relative climb/glide dislocation mobility is very important and that dislocation annihilation must be taken into account for significant climb mobility. This can be an interesting direction to limit the dislocation density at high fission rate.

In the present work, we investigated a more tractable approach in the context of the model implemented in the MFPR-F code, close to the approach suggested in [28]. At first, following [18, 19] a saturation of point defects production can be foreseen when collision cascades superposition is considered at high fission rate. In [18], the classical molecular dynamics simulation performed showed that, after the ballistic collision process, a molten core is produced in the damaged volume during a short period (order of picoseconds), favouring a recombination of the majority of defects produced during the ballistic phase. When considering a cascade overlap sequence, the point defects concentration increases and subsequently it levels off, in agreement with previous molecular dynamics simulations [19]. On this basis, a simple correction of the Frenkel pair production rate was formulated by adding a fission rate dependent correction factor, $\varphi(F)$ (<1), $K = Fz_S\varphi(F)$. This gives the new following evolution of the Frenkel pair production rate with fission rate illustrated in Fig. 2

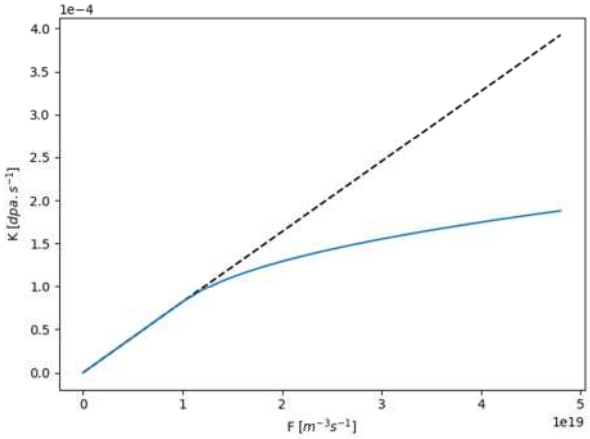

Figure 2: Frenkel pair production rate versus fission rate in the previous linear formulation (black dashed line) and in the new formulation with saturation factor (blue line).

Secondly, the self-interstitial diffusion coefficient formulation was modified by adding an a-thermal part (fission rate dependent): $D_i = A\, e^{(-E/kT)} + BF$. This will significantly increase D_i at low temperature and the vacancy-interstitial recombination term in Eq (4). Again, this corresponds to the results from empirical potential molecular dynamics simulation [20] in which the basic processes of vacancy-interstitial recombination are investigated. These processes are complex, but the distance between point defects is the key parameter for spontaneous recombination. With an irradiation-enhanced interstitial diffusion coefficient, more interstitial atoms will be transported in the region of spontaneous recombination. The following Fig. 3 gives the new evolution of D_i with fission rate:

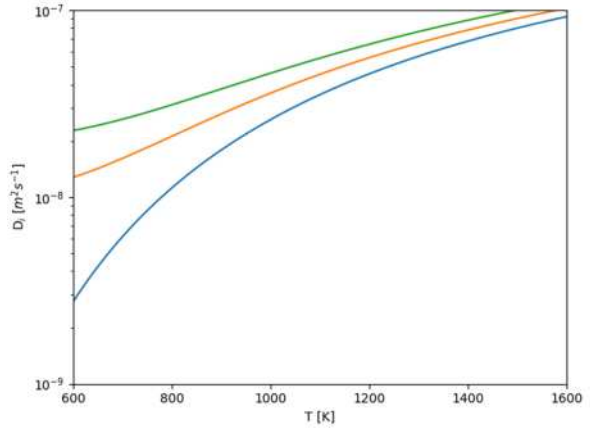

Figure 3: Self interstitial diffusion coefficient (m^2s^{-1}) versus temperature (K) with inclusion of the a-thermal term for fission rate = 0 (blue), $1.10^{19}m^{-3}s^{-1}$ (orange), $2.10^{19}m^{-3}s^{-1}$ (green).

2.3 Test of the model of formation-growth of the HBS zone

With the corrections described in section 2.2, the model is compared to the experimental data reported in [13] in which the HBS zone size evolution was given as function of the fuel pellet mean burn up. The following figure 4 shows a good agreement between the MFPR-F model and the experimental data, using a HBS formation dislocation density threshold adjusted to $2.10^{15}m^{-2}$.

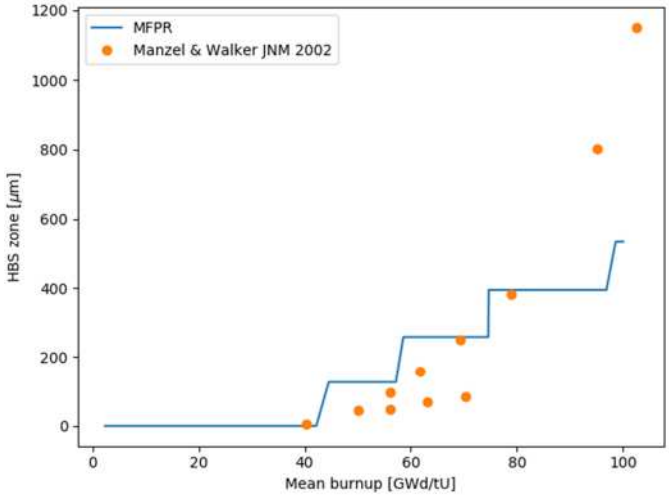

Figure 4: Evolution of the HBS size simulated by the MFPR-F code with density threshold equal to $2.10^{15}m^{-2}$, compared to the experimental data given in [13].

The model is also compared to the more analytical results of the HBRP program [15]. In these tests UO_2 samples were irradiated in controlled conditions of fission rate and temperature. Post exams of these irradiated samples revealed the restructuring of the samples. In [15] the authors defined a zone in the temperature-burnup diagram in which the restructuration was completely obtained and a zone in which only a beginning of restructuration was found. As shown in figure 5, the MFPR-F model reproduces correctly these results. An underestimation of the HBS formation above 1200K is noted (as also revealed by the underestimation of the HBS zone size above 100GWd/t in the figure 4).

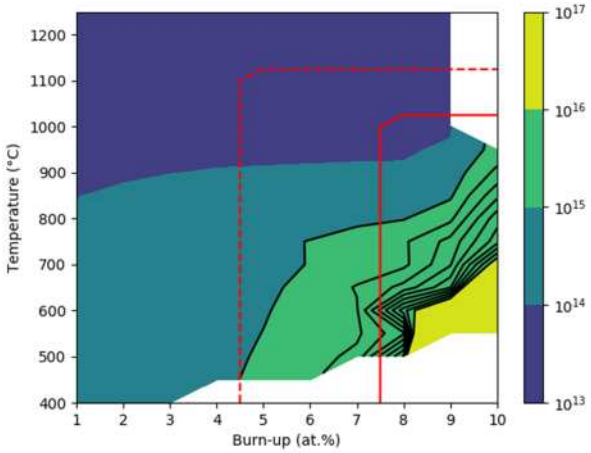

Figure 5: Simulation of the HBRP samples behaviour with the MFPR-F HBS model. The dislocation density obtained for each sample is reported in m^{-2} (see colour code). The areas corresponding to HBS formation are given by the red (solid) line for complete restructuration or dashed line for beginning of restructuration. A critical dislocation density ~$10^{15}m^{-2}$ would allow a correct reproduction of experimental results.

3. The HBS zone and fission product behaviour modelling

When the criterion on dislocation density for HBS formation is reached, the fuel microstructure is modified in the corresponding pellet radial zone according to the main features reported for the HBS in [1, 2, 7, 8, 9].

The grain size is immediately decreased to a sub-micrometric value (0,9 µm was chosen and it is foreseen in an improved form of the model to decrease this size up to 0,5µm).

The fission gas intergranular porosities (face and edge) as well as as-fabricated porosities are replaced by large, non-interconnected porosities. The behaviour of these new HBS porosities might be a complex issue. Recently, an analytical approach was proposed in [21] to model the coalescence of randomly distributed immobile pores owing to their growth and triple impingement in order to reproduce the porosity behaviour as a function of burn up in the HBS zone. This model seems in fact very promising. In the present work we followed a more simplified approach by fitting the porosities size evolution with burn up on the experimental data given by Cappia et al [12], while the total porosity is fixed. The results, for a total porosity of 15%, are illustrated in figure 6 and compared to the corresponding experimental data for porosity size and pores concentration in the HBS zone given by Spino et al [11]. The model reproduces correctly the beginning of pores coalescence and growth when a local burn up of around 10at% is obtained as well as the global evolution of the porosities for higher burn up. Note that, the total porosity being fixed, the pore concentration is proportional to the inverse of the pore size. This explains the overestimate of the pore concentration for burnup lower than 10at%.

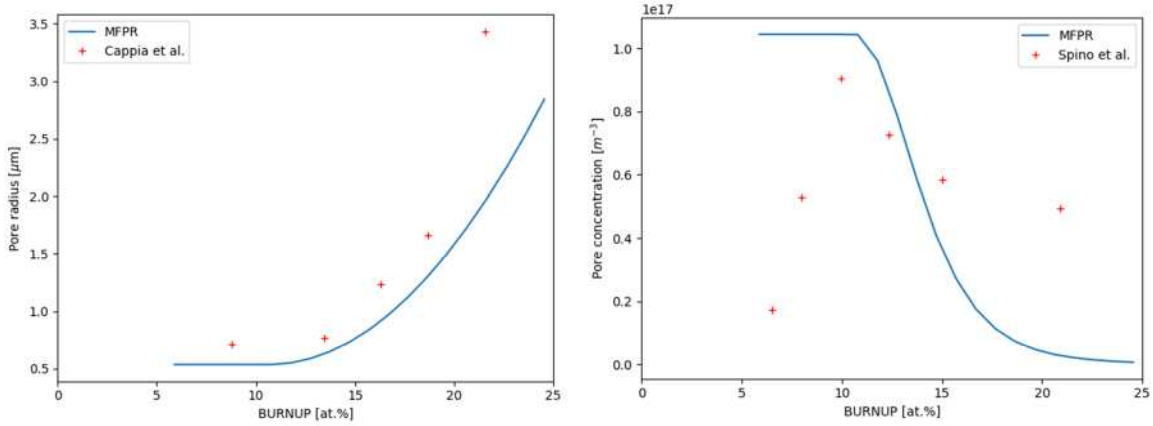

Figure 6: Simulation of the porosity evolution versus burn up with the MFPR-F HBS model. Comparison to the experimental data from [12] (pores radius in µm) and [11] pores concentration (m^{-3})

3.1 The fission product behaviour modelling

When the HBS is formed, the volatile fission products (fission gases, Cs, I, Te, Sb) are totally removed from the grains and transferred into the pores. The diffusion problem for the other fission products is reformulated, taking into account the reduced grain size. For these fission products, the normal chemistry model is still acting and additional gaseous species might be produced and transferred in pores with the volatile species (see [22] for a description of the chemistry model).
The pores pressure is then evaluated by the ideal gas law taking into account all the gaseous species transferred in the pores.
As shown for example in [1] the HBS pores are highly pressurized. In annealing conditions [3], the interpretation of the fission product release is based on the assumption of an over-pressurization of the pores during the transient. This is the basis of the model implemented in

the MFPR-F code.
The pores pressure P_{pores} is calculated from the ideal gas law. The pore of radius R_{pore} is over-pressurised if the following condition is fulfilled: $P_{pore} > (2\gamma/R_{pore}) + P_h$ (P_h the hydrostatic pressure and γ the pore surface tension). In these conditions, when the over-pressurization $P_{pore} - (2\gamma/R_{pore}) - P_h$ exceeds a critical value ΔP_{crit}, a fraction of species is released so that the over-pressurization remains equal to ΔP_{crit} . The critical amount of fission product atoms in the pore is related to the critical over-pressure via the ideal gas law:

$$Y_{crit}^{pore} = \frac{P_{crit}\alpha V_{fuel}}{kT}$$

In which α is the total porosity, k the Boltzmann constant, and $P_{crit} = \Delta P_{crit} + (2\gamma/R_{pore}) + P_h$.

The number of i-th fission product atoms released from this process is simply expressed by:

$$Y_i^{released} = \left(1 - Y_{crit}^{pore}/Y_{Nf}^{pore}\right)Y_i^{pore}$$

In which $Y_{N_f}^{pore}$ is the current atoms number of the i-th gas species in pores and $Y_{Nf}^{pore} = \sum_i Y_i^{pore}$. The total amount of atoms released is thus given by:

$$Y_{N_f}^{released} = \sum_i Y_i^{released} = Y_{Nf}^{pore} - Y_{crit}^{pore}$$

It means that only the amount of atoms above the critical amount is released, likely to lead to an underestimation of the released fraction.
This simplified approach must be considered as a first step toward a more comprehensive description of the complex phenomena involved in the FP release process from the HBS zone. In [3], the authors noted a correlation between peak pressures occurring during the transient and the fission gas release, suggesting a process driven by pressure evolution in the pores. In addition, the microstructure of the samples was highly disturbed as the samples were totally pulverized at 1500K. In [23], the processes of fuel fragmentation were investigated in detail by experimental approaches. The possibility of cracking by pores overpressure was notably investigated and was finally considered as not relevant for pores pressure up to 160MPa. The addition of a stress field from the pellet clad mechanical interaction did not allow the HBS cracking either. Lastly, the study revealed the existence in the HBS zone of two crystalline phases. The second phase is close to the classical F3-3m UO_2 symmetry but with a different lattice parameter and different thermal expansion properties. The amount of this second phase increases with burn up and its existence is likely linked to the gap of miscibility in the U-Pu-O phase diagram (or other U-lanthanides-O diagram). It was shown in [23] that in annealing conditions, when the grain deformation is not free, the differential extension of the two phases induces a significant increase of the stresses at grain boundaries of which the most fragile are then ruptured.
Despite the simplified character of our fission product behaviour model in the HBS, it is interesting to compare the results obtained with the experimental data for LOCA conditions [4] or high temperature annealing [3].

3.2 Gas release from HBS in LOCA conditions

In [4], a 103 GWd/t UO_2 sample fully restructured was submitted to a LOCA-like temperature transient (0,2°C/s as temperature ramp, 1200°C as final temperature, held during 15min followed by a quick cool down).
The results expressed in cumulated gas release fraction obtained with the MFPR-F HBS model are compared to the experimental data in figure 7.

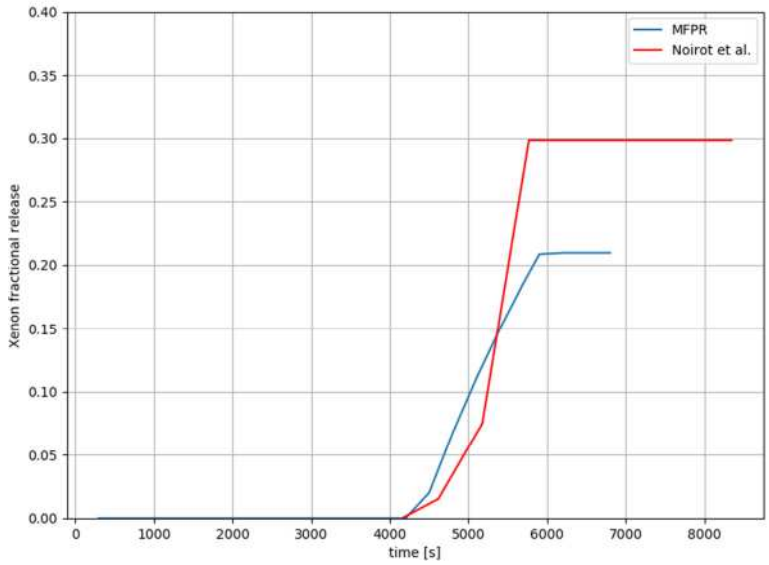

Figure 7: Simulation of the gas release in LOCA-type conditions with the MFPR-F HBS model for a 103GWd/t sample, obtained with a critical overpressure of 130 MPa, and comparison to the experimental data from [4].

Global kinetics and total gas release are both reasonably reproduced, although the released fraction is slightly underestimated.

3.3 Gas release from HBS in high temperature annealing conditions

In [3], a sample is extracted from the outer part of a highly irradiated UO_2 fuel (~102MWd/t mean pellet burn up). The sample corresponds to two different restructured zones: one with a local burn up of 160GWd/t and the other with 240GWd/t. This sample is submitted to a temperature transient (0,5°K/s, up to 2750 K) and the gas and fission product release is measured by Knudsen effusion mass spectrometry. The release is almost complete for fission gases and volatile fission product (Cs, Te, I) and appears in two steps; the first one around 1000K, likely corresponding to the 240GWd/t part of the sample, and the second one at 1500K for the part at 160GWd/t.
A simulation was performed with the MFPR-F model, reproducing the sample with restructured zones of local burn up of 160 and 240GWd/t. The results are given in the figure 8 for fission gases with a comparison to experimental data. The model results are presented both including the HBS model (solid blue line) and excluding it (dashed blue line).

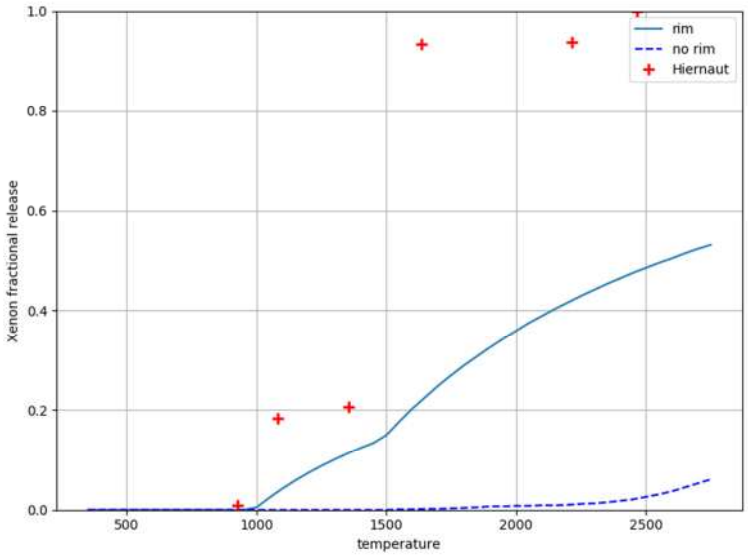

Fig 8: *Simulation of the gas release in high temperature annealing conditions with the MFPR-F HBS model for a sample with restructured zones of 160 and 240GWd/t (with and without activation of the HBS model), obtained with a critical overpressure of 250 MPa, and comparison to the experimental data from [3].*

The simulation underestimates the experimental data despite a correct reproduction of the global slope of the release curve and an improvement compared to the simulation without the HBS model. The underestimation is easy to justify mainly by considering that the release mode implement in the model does not allow the total release of gases contained in the pores.

4. Conclusions

A model for the HBS formation and behaviour in highly irradiated UO_2 fuel has been presented. The formation and growth modelling of the HBS zone is based on a mechanistic simulation of dislocation density evolution. It follows the detailed approach described in [16] which has been modified in the present work to take into account the specific effects at high fission rate occurring in the external part of the pellet. This model reproduces correctly the experimental data [13, 15] for the formation and growth of the HBS zone. It is noted that the HBS formation at temperature above 1000K reported in [15] or the width of HBS zone at very high mean bun up (> 100GWd/t) is not reproduced by the model. This will be a topic for improvements of the model. For example, on the basis of the present model a different approach for simulating the thermal self-interstitial diffusion in Uranium could be taken. Another, more complex possibility is a total re-formulation of the model not based, as currently on nucleation of dislocations by migrating interstitials but on the direct formation in the collision cascades volume by clustering of interstitials in the external part of the cascade as shown in [18].
On this basis, a simplified model for HBS description was proposed, the characteristics of which being mainly adjusted on data given in [11, 12] for pores size and concentration. For the fission product behaviour in the HBS, a simplified approach was proposed in a first stage. It is based on the pressure evolution in pores during temperature transient with the simple law of perfect gas or Van der Waals equations. A stress based criterion for release is chosen for the release process. This gives promising results in LOCA type conditions, but under-estimates the release at high temperature which suggests the importance of the significant structure alteration during the release process, not considered in the present model. This is one issue for further improvements, along with experimental validation.

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