



Original Article

Modeling of Pore Coarsening in the Rim Region of High Burn-up UO_2 Fuel

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ABSTRACT

An understanding of the coarsening process of the large fission gas pores in the high burn-up structure (HBS) of irradiated UO_2 fuel is very necessary for analyzing the safety and reliability of fuel rods in a reactor. A numerical model for the description of pore coarsening in the HBS based on the Ostwald ripening mechanism, which has successfully explained the coarsening process of precipitates in solids is developed. In this model, the fission gas atoms are treated as the special precipitates in the irradiated UO_2 fuel matrix. The calculated results indicate that the significant pore coarsening and mean pore density decrease in the HBS occur upon surpassing a local burn-up of 100 GWd/tM. The capability of this model is successfully validated against irradiation experiments of UO_2 fuel, in which the average pore radius, pore density, and porosity are directly measured as functions of local burn-up. Comparisons with experimental data show that, when the local burn-up exceeds 100 GWd/tM, the calculated results agree well with the measured data.

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1. Introduction

Sintered UO_2 pellets in rods are the standard nuclear fuel in light water reactors [1–4]. The discharge burn-up of fuel assemblies has been increasing throughout the past few decades in order to improve the economics of power plants. The local burn-up in the rim region of UO_2 pellets will exceed 100 GWd/tU when the average pellet burn-up is about 40 GWd/tU due to the configuration of the neutron energy spectrum in a light water reactor [5,6]. During in-pile irradiation, starting at a local burn-up around 50 GWd/tU and a temperature below 1,100°C, the UO_2 fuel matrix will undergo a restructuring

process which results in the appearance of a new fuel structure with the typical characterization of small submicron grains and coarsened fission gas pores. This new structure observed in the rim region of the UO_2 pellets is called high burn-up structure (HBS) or rim structure. Some other researchers [7,8] named this morphology cauliflower structure due to its similar appearance to the biological morphology of the cauliflower.

The occurrence of the HBS results in the fission gas pores coarsening and it will eventually accelerate the fuel swelling; these issues have raised some concern over the reliability and safety of extended fuel operation life in the reactor. Since this

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HBS was first observed in the late 1950s [6], numerous experimental as well as the theoretical studies have been performed in order to investigate this new structure in detail and understand the HBS formation mechanisms. However, a full scientific description of the HBS formation mechanisms is still missing and there are still very few mechanistic models that can be used in the fuel performance analysis code, although several international collaboration programs, such as High Burn-up Effects Program [9] and High Burn-up Rim Project [10], have been established since the early 1980s and several theoretical models have been presented [11–23].

Fortunately, the rim structure of PWR-fuels with local burn-ups of 240 GWd/tM has been studied in recent years and confirms that the coarsening process of the average pore size is close to Ostwald ripening (OR) [24] and coincides with the other studies in the literature [25–27]. Hence, in this paper, a theoretical analysis is presented to investigate the evolution of fission gas pores in the rim region of high burn-up UO_2 fuel pellets based on the OR mechanism, which was originally developed to explain the coarsening of precipitates in solids [28–31]. To describe the evolution of fission gas pores in the HBS of UO_2 fuel pellets, a numerical model of fission gas pore coarsening is developed, which includes the coarsening process of pores, and calculations of the pore radius and density in the fuel matrix during irradiation of UO_2 fuel pellets. The comparisons of theoretical predictions of the mean pore radius, mean pore density, and the porosity with the experimentally measured results is also discussed.

This paper is organized as follows: the next section provides a mechanistic and engineering model for the evolution of fission gas pores in the rim region of high burn-up UO_2 fuel pellets. Then, the calculated results of the mean pore radius, mean pore density, and porosity in the fuel as a function of burn-up are presented and compared to the available literature data. Finally, the main findings of this study are summarized in the conclusions.

2. Physical processes and model

It is well known that the noble gas Xe is originally generated in the UO_2 fuel matrix in atomic form. All atoms formed by fission processes have a strong tendency to diffuse and precipitate into bubbles due to their low solubility. It is assumed that some processes of the pore coarsening in the UO_2 fuel matrix are the same as or similar to the coarsening of precipitates in solids, which are described as follows [28–31]:

- (1) The fission gas Xe atoms formed by the fission of uranium isotopes have a strong tendency to diffuse into bubbles due to their very low solubility and this process can decrease the total free energy of the system, which is the same as the second phase particles in solids.
- (2) The decreasing of the interfacial energy per unit volume is the driving force for the coarsening of both the fission gas pores in the UO_2 fuel matrix and the second phase particles in solids.
- (3) According to the Gibbs–Thomson effect, the growth of the larger second phase particles in solids is at the

expense of the smaller ones. Re-resolution of fission gas atoms from bubbles and/or pores due to the fission fragment will occur in UO_2 fuel during irradiation, and will induce the shrinkage of the small bubbles but has no obvious effect on the larger bubbles.

The basic premise of the theory presented in this work is that the pore coarsening is driven by the diffusion of fission gas atoms. Based on the similar processes and mechanisms of the pore coarsening and precipitates in solids described above, there is no difficulty in applying OR to develop a mechanistic model for pore coarsening in the rim region of high burn-up UO_2 fuel.

The schematic diagram of the fuel system which contains α phase (Xe pore) and β phase (UO_2 fuel matrix) is shown in Fig. 1. The α phase is composed of Xe atoms (A component). The β phase is composed of Xe atoms and pure UO_2 (B component). The diphasic equilibrium condition under a constant temperature and constant pressure requires:

$$dG = 0 \quad (1)$$

More specifically:

$$dG_g + dG_s + dE_{\text{solid}} + \sum_i \mu_i dn_i = 0, \quad (2)$$

where the first term on the left hand side of Eq. (2) represents the change of the Gibbs free energy of fission gas in the pore; the second term represents the change of the Gibbs free energy of the fuel system resulting from the change of the interfacial area between the α phase and β phase; and the third term represents the change of the strain energy of the UO_2 fuel matrix during the process of pore coarsening.

The internal pressure in the coarsened rim bubbles of UO_2 pellets is assumed to be much higher than the equilibrium pressure of $P_{\text{eq}} = 2\gamma/r$ (γ : surface energy), from the fact that a very high density of dislocations has been observed close to the surface of coarsened rim bubbles with radii between 100 nm and 300 nm in the boiling water reactor pellet peripheral region [32]. These dislocations are generated by a dislocation punching mechanism, and the internal pressure in bubbles is given by $P = \mu b/r + P_{\text{eq}}$ (μ is the shear modulus and b

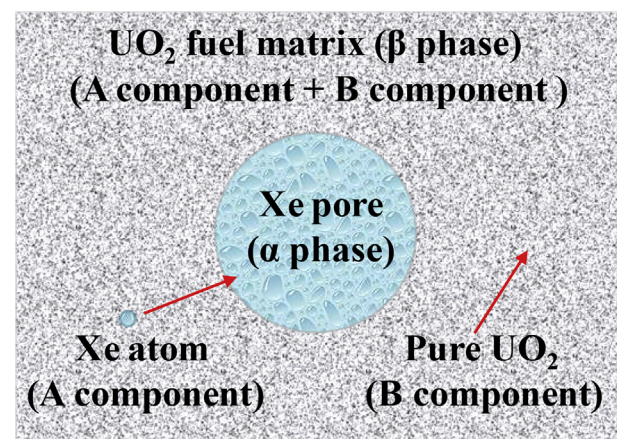


Fig. 1 – Schematic diagram to illustrate the UO_2 fuel system.

the Burgers vector) [33]. Hence, the change of the fission gas free energy in the pore during the process of pore coarsening can be expressed as:

$$dG_g = VdP = -\frac{4}{3}\pi(\mu b + 2\gamma)rdr, \quad (3)$$

where $r(m)$ is the radius of the pore.

During the process of pore coarsening, the change of the free energy of the UO_2 fuel system due to the change of the interfacial area $A = 4\pi r^2$ between the α phase and β phase is given by:

$$dG_s = \gamma dA = 8\pi\gamma r dr \quad (4)$$

The contribution to dG of the change of the strain energy of the fuel during the pore coarsening process is calculated as follows. The density of elastic energy in the UO_2 fuel matrix under the condition of externally applied hydrostatic pressure is given by [34]:

$$E_{el} = \frac{\sigma^2}{2k}, \quad (5)$$

where σ is the hydrostatic pressure in Pa and k is the bulk modulus of the UO_2 fuel in Pa. The total strain energy stored in the fuel is the integral of E_{el} over the entire volume. In this work, it is assumed that the equilibrium Xe pore is produced by removing the atoms of fuel matrix from the region to be occupied by the Xe pore to the outer surface of the fuel. So, the elastic energy of the fuel matrix containing a pore with a radius of r can be expressed as:

$$E_{solid} = E_{solid}^0 - \frac{\sigma^2}{2k} \cdot \frac{4}{3}\pi r^3, \quad (6)$$

where E_{solid}^0 is the elastic energy of the fuel before the pore is introduced. Hence, during the process of pore coarsening, the change of the total strain energy in the fuel is calculated by taking the differential of Eq. (6), which is:

$$dE_{solid} = -\frac{2\pi\sigma^2}{k} \cdot r^2 dr \quad (7)$$

Therefore, substituting Eqs. (3), (4), and (7) into Eq. (2), one can obtain another expression of the diphasic equilibrium condition under constant temperature and constant pressure as follows:

$$2\pi r \left(\frac{8}{3}\gamma - \frac{2}{3}\mu b - \frac{\sigma^2}{k} r \right) dr + \sum_i \mu_i dn_i = 0 \quad (8)$$

During the process of pore coarsening, according to the OR mechanism, it is assumed that there is diffusion dn_A moles A component (Xe atoms) into the α phase from the β phase. During this process, the molar of B component (n_B) is constant, which means $dn_B = 0$. Therefore, dn_A moles A component will add to the α phase and dn_A moles A component will subtract from the β phase. So, Eq. (8) can be transferred to Eq. (9) which can be expressed as:

$$2\pi r \left(\frac{8}{3}\gamma - \frac{2}{3}\mu b - \frac{\sigma^2}{k} r \right) dr + \mu_A^\alpha dn_A - \mu_A^\beta dn_A = 0 \quad (9)$$

and solving for $\mu_A^\beta - \mu_A^\alpha$

$$\mu_A^\beta - \mu_A^\alpha = \frac{2\pi r \left(\frac{8}{3}\gamma - \frac{2}{3}\mu b - \frac{\sigma^2}{k} r \right) dr}{dn_A} = 2\pi r \left(\frac{8}{3}\gamma - \frac{2}{3}\mu b - \frac{\sigma^2}{k} r \right) \frac{dr}{dn_A} \quad (10)$$

By contrast, proceeding from strict thermodynamic equations, a three-parameter equation of state (EOS) for Xe was established by Kaplun and Meshalik [35]. More recently, a preferable modified EOS for Xe was proposed based on the MD simulation and Kaplun's work [36]. The comparison with the MD simulation data show that the proposed EOS can be applied up to 550 MPa at 900 K and 3 GPa at 1373 K. The EOS for Xe with a consideration of the Van der Waals force is:

$$P = \frac{RT}{V} \left(1 + \frac{c_0}{V - b_0} \right) - \frac{a_0}{V^2} \quad (11)$$

where a_0 , b_0 , and c_0 are constants.

Hence, the r of a pore is related to n_A moles gas atoms in one pore through the EOS of Xe with the internal pressure in rim bubbles ($P = (\mu b + 2\gamma)/r$) [33] of Eq. (11).

$$\frac{\mu b + 2\gamma}{r} = \frac{n_A RT}{4\pi r^3/3} \left(1 + \frac{n_A c_0}{4\pi r^3/3 - n_A b_0} \right) - \frac{n_A^2 a_0}{(4\pi r^3/3)^2} \quad (12)$$

where μ is the shear modulus of the UO_2 fuel in Pa; b is the Burgers vector in m, and γ is the surface energy of the UO_2 fuel in J/m^2 .

According to Eq. (12) above, we can express dr/n_A as follows:

$$\frac{dr}{dn_A} = f_1(r) \quad (13)$$

It is assumed that the μ_A^α will be invariable with r .

When $r \rightarrow \infty$, substituting Eq. (13) into Eq. (10), then:

$$\mu_A^\beta(\infty) = \mu_A^\alpha + f_2(r) \quad (14)$$

When $r = r$, substituting Eq. (13) into Eq. (10), then:

$$\mu_A^\beta(r) = \mu_A^\alpha + f_3(r) \quad (15)$$

Using Eqs. (14) and (15),

$$\mu_A^\beta(r) - \mu_A^\beta(\infty) = f_3(r) - f_2(r) \quad (16)$$

According to the Gibbs–Thomson relationship [28], the chemical potential of the A component in the β phase is $\mu_A = \mu_A^\theta(T) + RT \ln P_A$, then:

$$\mu_A^\beta(r) - \mu_A^\beta(\infty) = RT \ln \frac{P(r)}{P(\infty)} \quad (17)$$

Using the concentration $C(r)$, substitute the pressure $P(r)$ into Eq. (17) since the β phase is in solids [25], then:

$$\mu_A^\beta(r) - \mu_A^\beta(\infty) = RT \ln \frac{C(r)}{C(\infty)} \quad (18)$$

When $(C(r) - C(\infty))/C(\infty) \ll 1$, then:

$$\ln \frac{C(r)}{C(\infty)} \cong \frac{C(r)}{C(\infty)} - 1 \quad (19)$$

Using Eqs. (18) and (19),

$$C(r) = C(\infty) \cdot \left(1 + \frac{\mu_A^\beta(r) - \mu_A^\beta(\infty)}{RT}\right) \quad (20)$$

Substituting Eq. (16) into Eq. (20) results in the following expression:

$$C(r) = C(\infty) \cdot \left(1 + \frac{f_3(r) - f_2(r)}{RT}\right) \quad (21)$$

Imposing gas-atom conservation, i.e., requiring that the sum of the gas atoms diffusion into the pores with the radius of r is equal to the total flux of the A component through the spherical surface with the radius of r , will result in the following expression:

$$\frac{dn_A}{dt} = \frac{4\pi Dr}{N_A} \cdot [C(\bar{r}) - C(r)] \quad (22)$$

where D is the diffusion coefficient of single gas atoms in m^2/s ; N_A is Avogadro's number; and \bar{r} is the average radius of Xe pores.

Hence,

$$\frac{dr}{dt} = \frac{dr}{dn_A} \cdot \frac{dn_A}{dt} = f_1(r) \cdot \frac{4\pi Dr}{N_A} \cdot [C(\bar{r}) - C(r)] \quad (23)$$

In this work, since it is assumed that $d\bar{r}/dt = (dr/dt)_{\max}$, substituting Eq. (21) into Eq. (23), one can obtain the following expression:

$$r = 2\bar{r} + \frac{2\mu b + 4\gamma}{3\sigma} \quad (24)$$

Substituting Eq. (24) into Eq. (23) and solving for \bar{r} :

$$\frac{d\bar{r}}{dt} = \frac{4\pi D}{N_A} \cdot \left. \frac{r dr}{dn_A} \right|_{r=2\bar{r}+\frac{2\mu b+4\gamma}{3\sigma}} \cdot [C(\bar{r}) - C\left(2\bar{r} + \frac{2\mu b + 4\gamma}{3\sigma}\right)] \quad (25)$$

Eq. (25) is the major result of this section. Using Eqs. (12) and (21), an approximate solution for the average radius of Xe pores $\bar{r}(t)$ is given by:

$$\bar{r}(t) = \left[\frac{D \cdot (\mu b + 2\gamma) \cdot (\mu b - 4\gamma) \cdot RT}{96\pi \cdot N_A \cdot \sigma^3} \right]^{1/4} \cdot [C(\infty)]^{1/4} \cdot t^{1/4}, \quad (26)$$

where D is the diffusion coefficient of single gas atoms in m^2/s ; μ is the shear modulus of the UO_2 fuel in Pa; b is the Burgers vector in m; γ is the surface energy of the UO_2 fuel in J/m^2 ; σ is the external hydrostatic pressure in Pa; R is the constant of gas in $J/mol/K$; T is the absolute temperature in K; N_A is Avogadro's number; t is the time in s; and $C(\infty)$ is the concentration of the A component in the β phase at infinite distance, which is approximately equal to the average concentration of gas atoms $\bar{c}(t)$ and can be obtained by solving the basic diffusion equation of fission gas with a collection of accompanying equations accounting for the irradiation effects and the bubble nucleation induced by the low solubility of the fission gases in the UO_2 fuel matrix [37–40].

Gas atom conservation is imposed, i.e., the sum of the fission gas in dynamics solution and in pores is equal to the amount of fission gas generated. So, the concentration of gas in bubbles, $N_b(t)$, is given by:

$$N_b(t) = \beta f t - \bar{c}(t), \quad (27)$$

where f is the fission rate in fission/ m^3s/β ; and β is the number of gas atoms produced per fission event.

The average number of gas atoms in a single pore, $n_b(t)$, can be obtained by the EOS of Xe with the average radius of pores from Eq. (12). The density of pores in the fuel matrix, $C_b(t)$, is given by:

$$C_b(t) = \frac{N_b(t)}{n_b(t)} \quad (28)$$

3. Results and discussion

In order to obtain the data of the radius and density of bubbles and the porosity in the fuel matrix, and eventually validate the pore coarsening model, Eqs. (24) and (26) in the previous section are solved simultaneously. The inputs to this model are time increment, fission rate, burn-up, fuel temperature, and hydrostatic pressure in the fuel pellets. The data of the key parameters used in this model are summarized in Table 1. In addition, it is reasonable to use an approximate temperature of $380^\circ C$ to calculate the mean pore radius and the mean pore density in this work in order to correspond to the experimental data [24] under the same irradiation temperature.

Fig. 2 shows the comparison between the calculated values and the experimental results [24] of the mean pore radius of as-irradiated UO_2 as a function of burn-up. The experimental results of Fig. 2 with maximum local burn-ups of 240 GWd/tM show the critical local burn-up point associated with the pore coarsening process for the rim region of high burn-up UO_2 fuel is about 100 GWd/tM, which has been discussed in detail by Spino et al [24]. This can be better appreciated in Figs. 3 and 4 where both the pore density and the porosity are plotted as a function of the local burn-up. As clearly seen in Fig. 2, at local burn-up below 100 GWd/tM the mean pore radius decreases appreciably with increasing local burn-up and for local burn-up above about 100 GWd/tM a sustained increase with increasing local burn-up is verified. The trendline with the local burn-up above 100 GWd/tM fitted by experimental data suggests with reasonable precision that the mean pore radius increases proportionally to $BU^{0.4}$, which is close to the average cavity size increasing proportionally to $t^{0.5}$ for the Ostwald-ripening mechanism [25–27] based on the assumption that time is equal to burn-up. Additionally, the mean pore size increases proportionally to the $[C(\infty)]^{1/4}$ in Eq. (26) and the

Table 1 – Values of parameters used in the model.

Parameter	Value	Ref.
β	0.26	[37]
γ	1 J/m ²	[41]
D	$1.2 \times 10^{-39} f$	[41]
b	0.39 nm	[33]
σ	1.5×10^7 Pa	[42]
μ	75 GPa	[33]
a_0	0.25978 J m ³ /mol ²	[36]
b_0	2.39276×10^{-5} m ³ /mol	[36]
c_0	5.56583×10^{-5} m ³ /mol	[36]

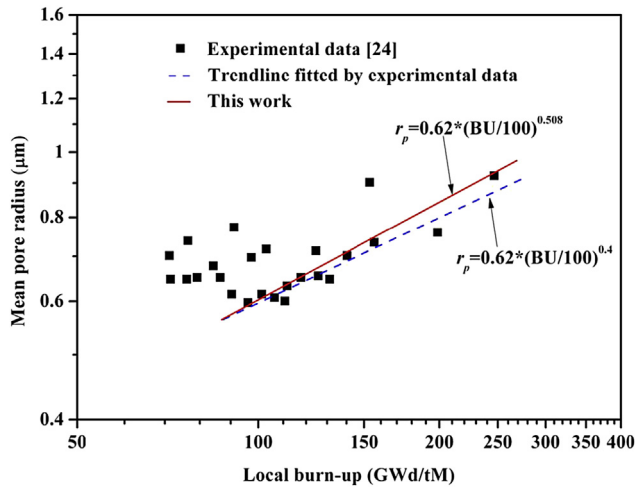


Fig. 2 – Comparison of measured and calculated mean pore radius data as a function of local burn-up.

concentration of the A component in the β phase at infinite distance $C(\infty)$ is approximately equal to the average concentration of gas atoms $\bar{c}(t)$. The average concentration of gas atoms $\bar{c}(t)$, which can be obtained by solving the basic diffusion equation of fission gas in the fuel matrix, increases proportionally to $t^{1.032}$. Ultimately, the mean pore size increases proportionally to $t^{0.508}$ and also to $BU^{0.508}$. It is clear that our calculated mean pore growth rate is in reasonable agreement with the experimental data, although the calculated growth rate is appreciably overestimated.

The comparison of measured and calculated mean pore density as a function of local burn-up is shown in Fig. 3. As can be seen, the maximum value of the pore density with measured data is achieved at about 100 GWd/tM and the pore density decreases constantly with increasing local burn-up above 100 GWd/tM. As clearly seen in the figure, the turning point of about 100 GWd/tM signals the onset of the pore coarsening process. The trendline with the local burn-up above 100 GWd/tM fitted by experimental data suggests

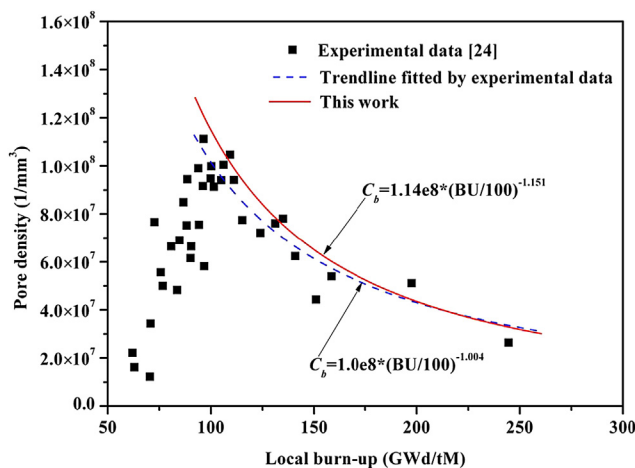


Fig. 3 – Comparison of measured and calculated mean pore density as a function of local burn-up.

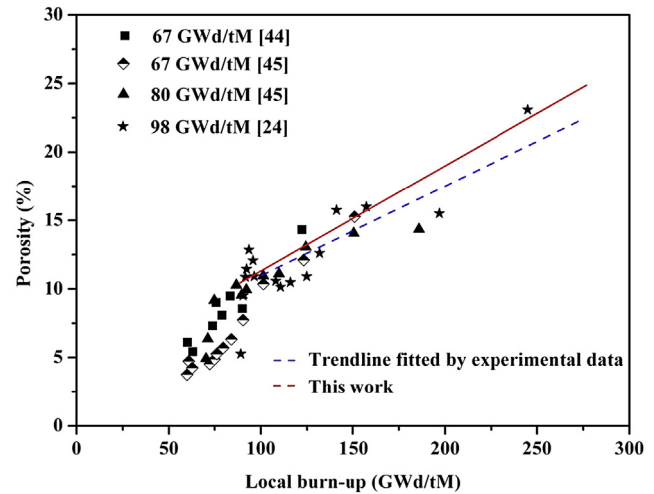


Fig. 4 – Comparison of measured porosity data for fuels with 67–98 GWd/tM average burn-ups and calculated porosity data as a function of local burn-up.

that the pore density decreases proportionally to $BU^{-1.004}$. This coincides fairly well with the t^{-1} dependence of the decrease of the number of second phase particles coarsening in supersaturated solutions [25] and of monatomic gas filled pores coarsening in solids [43]. Imposing gas atom conservation, the concentration of gas in bubbles as a function of time, $N_b(t)$, can be calculated from Eq. (27). The average number of gas atoms in a single pore, $n_b(t)$, can be obtained from solving the EOS of Xe [Eq. (12)] combined with the average radius of Xe pores [Eq. (26)]. Then, the density of pores in the fuel matrix, $[C_b(t)]$, can be obtained from Eq. (28). Ultimately, a density of pores proportional to $BU^{-1.151}$ ($BU = f \cdot t$, where f is the fission rate and t is the irradiation time), is expected. As can be seen, our predicted pore density is in reasonable agreement with the experimental value, although the predicted pore density-drop is appreciably faster than the experimental data.

The comparison of measured porosity data [24,44,45] for fuels with 67–98 GWd/tM average burn-ups and calculated porosity data as a function of local burn-up is shown in Fig. 4. As described above, the burn-up about 100 GWd/tM is the turning point of the porosity growth-rate. It should be noted that after exhibiting a relatively high value for porosity growth-rate at burn-ups below 100 GWd/tM, at the burn-ups above 100 GWd/tM the porosity growth-rate decreases and stabilizes at a much lower level near about 0.6%/10 GWd/tM. This is close to the matrix swelling rate due to the fission gas predicted in the literature [42] for high burn-up UO_2 fuels. It is clear that, at the burn-ups above 100 GWd/tM, the porosity growth-rate which was calculated in this work is in reasonable agreement with the available experimental data, although the calculated value is appreciably overestimated (about 0.63%/10 GWd/tM). The fact that the porosity growth-rate was overestimated by this numerical model can be explained by the difference between the calculated values and the measured data of the mean pore radius and pore density with the porosity data in UO_2 fuel using the relation $P = 4\pi r_p^3 \cdot C_p / 3$.

4. Conclusions

In this paper, a numerical model based on the Ostwald-ripening mechanism of pore coarsening in the rim region of high burn-up UO_2 fuel is presented. The model includes the coarsening process of pores and calculation of the pore radius and density in the fuel matrix during irradiation. The fission gas atoms are treated as the special precipitates in the irradiated UO_2 fuel matrix in this model. The capability of this model was validated by a comparison with the measured data of mean pore radius, pore density, and porosity in the HBS of UO_2 fuel. The calculated results suggest that pore coarsening kinetics proportional to 0.508 power of burn-up, and a density of pores proportional to -1.151 power of burn-up occur on surpassing a local burn-up of 100 GWd/tM. Such results reasonably agree with the measured results in the wide range of local burn-ups up to 240 GWd/tU. It is demonstrated that this numerical model is appropriate for describing the pore coarsening in the rim region of high burn-up UO_2 fuel.

Conflicts of interest

All authors have no conflicts of interest.

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