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Three-dimensional simulation of threshold porosity for fission gas release in the rim region of LWR UO_2 fuel

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Abstract

The threshold porosity above which fission gas release channels would be formed extensively in the rim region of high burnup UO_2 fuel was estimated by the Monte Carlo method and Hoshen–Kopelman algorithm. With the assumption that both rim pore and rim grain can be represented by cubes, the pore distribution in the rim was simulated three-dimensionally by the Monte Carlo method according to rim porosity and pore size distribution. Using the Hoshen–Kopelman algorithm, the fraction of open rim pores interlinked to the outer surface of a fuel pellet was derived as a function of the rim porosity. The simulation revealed that it is the rim porosity rather than the pore size distribution or rim thickness that determines the fraction of open pores connected to the pellet surface. The analysis also indicated that the threshold porosity is around 24%, above which the number of rim pores forming release channels increases very rapidly.

1. Introduction

As the discharge burnup of LWR UO_2 fuel increases to improve the economics of fuel, the formation of the rim structure has been observed at the periphery of high burnup UO_2 fuel pellets. The concern is that it would contribute to fission gas release and hence to the buildup of internal rod pressure, which is understandable because a large fraction of fission gas accumulated in the rim pores [1–3] could be eventually released to the free volume of the fuel rod. The release might then induce a thermal feedback effect and lead to fuel rod overpressurization and clad lift-off [4]. Hence, it becomes very important to reveal the condition under which fission gas in the rim pores would be released significantly to the free volume of the fuel rod.

The formation of gas release channels in the rim region is a very complex phenomenon that involves pores whose size distribution depends on the as-fabricated microstructure, power history and operating parameters such as temperature and burnup. In addition, the interlinkage of rim pores would be a statistical process due to their random position and size distribution. Therefore, to estimate the connectivity of rim pores to the outer surfaces of a fuel pellet and hence to analyze the fission gas release, a quantitative estimation of pore connectivity to free surfaces is required considering the microstructural characteristics of the rim region.

Several theoretical studies have been made to analyze the gas release in the rim. Baron et al. [5] applied Ronchi's bond percolation method [6] to calculate the fission gas escape probability in the rim. By introducing an average number of bonds per pore, Ronchi [6] calculated the pore interlinkage probability as a function of fractional porosity and grain to pore size ratio. Although this study uses elaborated mathematical models and equations for the percolation theory, it is difficult to directly introduce the results into a fuel performance code to study the effect of various parameters on the fission gas release in the rim region.

Bernard et al. [7] proposed an empirical additional surface that serves as a release path which develops in

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the rim due to subdivision of grains and increase in porosity. The model parameters for the surface were adjusted to fit experimental data for irradiated PWR fuel rods. Lösönen's empirical fission gas release model for the rim [8] assumes that gas release starts in the rim if its burnup is higher than 55 MWd/kg U. It also presumes that the release increases linearly up to a rim burnup of 100 MWd/kg U, where all the fission gas in the grain boundaries of the rim is assumed to be released to the free volume of the fuel rod.

On the other hand, by using the Monte Carlo method, Koo et al. [9,10] and Kim and Cho [11] developed a simulation method that calculates the interlinked fraction of pores to open spaces. However, they applied the method to the pores located in the grain boundaries of the as-fabricated microstructure rather than the pores in the rim. In addition, to simplify the problem, they treated pores two-dimensionally instead of three-dimensionally.

In this paper, the interlinkage of rim pores to the outer surfaces of a fuel pellet is simulated three-dimensionally for the rim region where a large number of pores of around 1 μm exist among small subdivided grains of 0.2–0.3 μm produced from the restructuring of as-fabricated grains. With use of the Monte Carlo method, pores are distributed three-dimensionally in the rim region according to porosity and their size distribution. Then the number of pores interlinked to the free surfaces of the pellet is obtained as a function of the rim porosity. In addition, the feasibility of this simulation is discussed by comparing the calculated results with experimental data.

2. Pore interlinkage in the grain boundaries of as-fabricated microstructure

2.1. Representation of a grain and grain boundary pores

The simulation of the interlinkage of rim pores to free surfaces is done by two steps: first, to distribute the pores in the rim according to rim porosity and their size distribution, and then to calculate the total number of pores connected to free surfaces. The Hoshen–Kopelman algorithm [12] is adopted here to study the interlinkage of the grain boundary pores in the as-fabricated microstructure and also to find the open paths to the free surfaces. This algorithm greatly reduces calculation time because the checking for interlinkage is limited only to neighbors.

To check whether the simulation method employed in this paper would yield reasonable results, we first applied it to the pore interlinkage in the grain boundaries of the as-fabricated microstructure. Fig. 1 shows how a basic simulation unit, a grain and its boundary, is represented. If a grain is represented by a large cube composed of $5\times5\times5$ small cubes and its grain boundary covered with pores is modeled as the layer with the thickness of one cube that surrounds the grain, a basic simulation unit is a $6\times6\times6$ small cube. However, the size of a basic unit can be changed according to the ratio of the grain-to-pore size. For example, if the ratio of the grain-to-pore were 9, a basic unit would consist of $10\times10\times10$ small cubes.

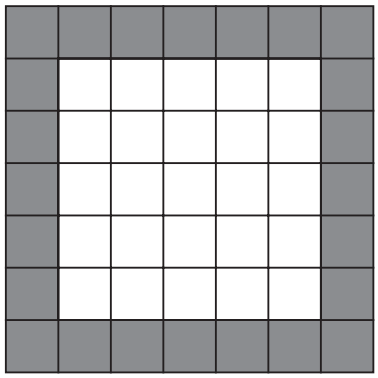

Fig. 1. Cross-sectional view of a three-dimensional simulation unit representing both a grain and its grain boundary region by $6\times6\times6$ cubes in the as-fabricated microstructure (white: grain, gray: grain boundary region).

2.2. Simulation of pore interlinkage

To locate pores in the grain boundary by the Monte Carlo method, we need to calculate the probability that a pore would be placed at a small cube in the layer surrounding the grain. With the assumption that pores exist only in the grain boundaries, the probability can be calculated by Eq. (1) using the concept that the total pore volume in a simulation unit is the same as the product of the probability and the volume of the grain boundary layer that belongs to a grain:

$$p_{\mathrm{gb}} = \frac{P_{\mathrm{pellet}} V_{\mathrm{unit}}}{V_{\mathrm{gb}}/2}, \tag{1}$$

where p_{gb} = the probability that a pore exists in a small cube of the grain boundary layer, P_{pellet} = the porosity of a fuel pellet, V_{unit} = the volume of a simulation unit, $V_{\mathrm{gb}}/2$ = the volume of the grain boundary layer that belongs to a grain.

It is to be noted that, since the pores exist both in the grain boundaries and within the grains, Eq. (1) would overestimate the probability that a pore exists in the grain boundary layer, and hence could lead to higher threshold porosity at which interconnection of the grain boundary pores would occur. However, the pores within the grains were ignored because the purpose of this section is to assess whether the present method would

provide reasonable result in terms of gas release in the as-fabricated region of a fuel pellet.

If a simulation unit is represented by $6\times6\times6$ small cubes, the ratio of V_{unit} to $V_{gb}/2$ is $6^3/(6^3-5^3)/2$. Hence, for a pellet porosity of 5%, the probability that a pore exists at a small cube in the grain boundary layer is 0.24 (24%). Then we locate the pores in the three-dimensional system representing the as-fabricated microstructure and calculate the fraction of open pores connected to the open surface of a fuel pellet.

Scanning electron micrographs of the fractured surfaces of irradiated UO_2 fuel show that lenticular pores with the size of mostly between 0.1 and 1.0 μm cover grain boundaries [13]. For a typical UO_2 grain size of 10 μm, the size ratio of a grain to a grain boundary pore ranges from 10 to 100. If the average size of grain boundary pores were taken to be 0.5 μm, the most probable size ratio would be 20 and therefore we can assume that the size ratio would be usually around 10–30 in typical LWR UO_2 fuels.

The system to simulate pore interlinkage in the as-fabricated microstructure is assumed to be composed of $300\times300\times300$ small cubes. Since the system consists of 27 million small cubes, it contains 1000–27 000 simulation units depending on whether the size ratio is 10–30. Our simulation shows that even 1000 simulation units are large enough to produce nearly the same open pore fraction for many trials.

2.3. Results and discussion

Fig. 2 shows the fraction of open pores as a function of the ratio of grain to pore size. It is found that there is a threshold porosity above which the open pore fraction starts to increase abruptly for all five cases. The threshold porosity decreases as the ratio increases, showing agreement with Ronchi's calculation [6]. For the size ratio of 10–30, the threshold porosity for gas release in the as-fabricated microstructure has been calculated to be around 5–12%.

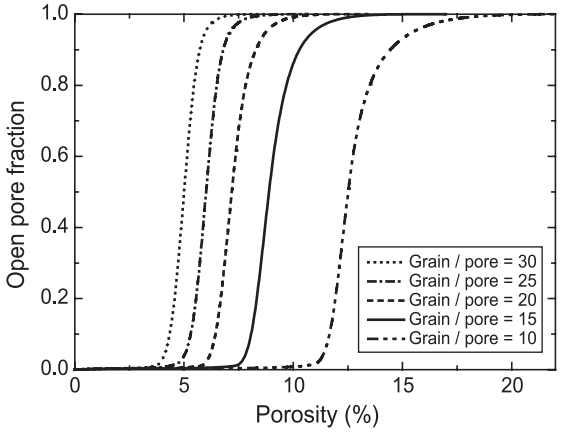

Fig. 2. Open pore fraction in the as-fabricated microstructure as a function of porosity.

Release pathways at grain boundaries are generally considered to be achieved at the moment when the gaseous swelling due to grain boundary pores reaches 5–8%. Both Koo et al. [14] and Dollins and Nichols [15] assumed that no fission gas escapes until pore swelling in the grain boundaries reaches 5%. While Turnbull [16] showed experimentally that release tunnels form when the swelling is larger than 7%, Villalobos et al. [17] assumed that pore interlinkage is achieved when the gaseous swelling in the gain boundaries is 7%. On the other hand, Beere and Reynolds [18] predicted that a stable tunnel network forms along the grain boundaries when fuel swelling is larger than 8%.

Therefore, the result of the present simulation, which indicates that release paths are formed suddenly when the grain boundary porosity reaches 5–12%, gives a slightly higher porosity than the experimental finding [16]. And it also yields a porosity that is a little bit higher than the one used by some gas release models. They assume that the gaseous swelling in the grain boundaries required for the completion of release channels in the grain boundaries, is in the range of 5–8% [14,15,17,18]. These two comparisons indicate that the present simulation method provides slightly higher threshold porosity than the previous works, as was expected from the assumption used in this paper that pores exist only in the grain boundaries. Therefore, it can be concluded that the present simulation method can be applied to the pore interlinkage in the periphery of high burnup UO_2 fuel.

3. Pore interlinkage in the rim region

3.1. Representation of the rim region

Many investigators measured the rim thickness by both EPMA and optical microscopy. Although there exists some controversy over whether the rim thickness increases linearly or exponentially with rim burnup, the threshold local burnup required for the rim formation is thought to be 52 GWd/t U [19]. Lassmann et al. [20] also claimed that UO_2 grains begin to recrystallize when the local burnup reaches about 60 GWd/t U. Recent data [21] obtained under typical PWR conditions suggest that the rim thickness increases linearly with the rim burnup at least up to 70 GWd/t U.

It is assumed that the rim is composed of many small cubes as shown in Fig. 3. In addition, we assume that a part of the rim, which is a cube system consisting of a very large number of small cubes, can represent the whole rim in terms of gas release. This would be a reasonable assumption because there would be no special

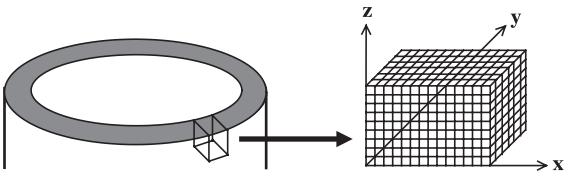

Fig. 3. Representation of rim region for the simulation of interlinkage.

Table 1
Typical pore size distribution in the rim [22]

Pore size (μm)	Fraction of pores (%)
0.5	1.9
1.0	42.3
1.5	34.6
2.0	17.3
2.5	3.9

reason or evidence to believe that the gas release behavior in the rim would be different along the circumferential direction, as long as the burnup distribution in the rim which determines microstructural evolution (porosity and pore size distribution) were uniform along the circumferential direction.

There are two, upper and right, faces in the cube system through which fission gas release can occur. For a pellet whose radius, height, and rim thickness are R, H and t, respectively, the ratio of upper-to-right face area is approximately $2\pi Rt/2\pi RH$. However, since the upper and lower face of a fuel pellet are the same in terms of gas release, the above ratio should be multiplied by 2 and hence the final ratio is $4\pi Rt/2\pi RH$. If the rim thickness and pellet height are assumed to be 300 μm and 10 mm, respectively, the ratio is 0.06. Therefore, we can neglect the gas release through the upper face in Fig. 3 and then can consider only the right face, a circular face of a pellet, to be a major release path.

The rim region is represented by a system of 300 (radial) × 300 (circumferential) × 300 (axial) small cubes. Since the length of a small cube is taken to be 0.5 μm, the system corresponds to a cube of 150 μm. The rim thickness of 150 μm along the radial direction has been chosen because it is about half the thickness of the rim observed in the high burnup fuels [19]. The other two lengths along the circumferential and axial directions have been taken to be the same as the rim thickness. The size of the system, which consists of 27 million small cubes, is sufficiently large enough to produce nearly the same open pore fraction in many trials for the same porosity. It takes less than 6 minutes to complete one case of simulation in a PC with the Pentium IV 2.53 GHz CPU. If the system representing the rim were small, simulation results would be different for each trial even if the same porosity were used for all the trials.

Since the mean size of rim pores is around 1 μm [1], a rim pore is represented by 2 × 2 × 2 small cubes, a cube of 1 μm. However, the rim pores observed by Spino et al. [22] in the high burnup UO_2 fuel have a size distribution that is dependent on burnup and microstructural characteristics. Consequently, to investigate the effect of pore size on the open pore fraction, five cases of pore size were analyzed; four cases of uniform pore size and one case with the size distribution as given in Table 1. The uniform pore sizes were simulated by 2 × 2 × 2, 3 × 3 × 3, 4 × 4 × 4 and 10 × 10 × 10 small cubes, respectively.

3.2. Simulation of pore interlinkage

To locate a rim pore by the Monte Carlo method in the cubic system of Fig. 3, we need to calculate the probability that a pore would be placed at a small cube. This probability can be calculated by Eq. (2) using the concept that the total pore volume in a simulation unit is the same as the product of the probability and the volume of a simulation unit representing a rim pore:

$$p_{\text{pore}} = \frac{P_{\text{rim}} V_{\text{cube}}}{V_{\text{pore}}}, \tag{2}$$

where p_{pore} = the probability that a pore exists at a small cube, P_{rim} = the porosity of the rim region, V_{cube} = the volume of a small cube, V_{pore} = the volume of a simulation unit representing a rim pore.

For example, let us assume that the rim porosity is 20% and a rim pore is represented by a small cube. Then the volume of a rim pore and that of a cube is the same and, according to Eq. (2), the probability that a pore exists at a small cube is 0.2 (20%). However, the situation is different when a rim pore is larger than a rim grain, as in the case of the high burnup UO_2 fuel. If we simulate a rim pore by a 2 × 2 × 2 cube, the probability that a pore exists at a small cube size is calculated to be 2.5% because V_{pore} is eight times larger than V_{cube}. Due to the overlapping of rim pores during the simulation process, the resulting simulated porosity is always smaller than that used to derive the probability by Eq. (2).

Our simulation shows that the resulting porosity obtained after locating pores in the system is usually 1–2% lower than the given porosity used to derive the probability. However, when the open pore fraction is plotted as a function of rim porosity, we use the simulated porosity rather than the one for deriving the probability.

3.3. Results and discussion

Fig. 4 shows the effect of pore size distribution on the open pore fraction as a function of rim porosity for the

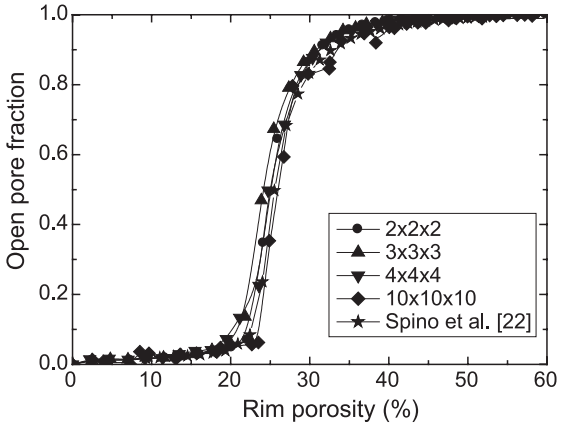

Fig. 4. Effect of pore size distribution on open pore fraction.

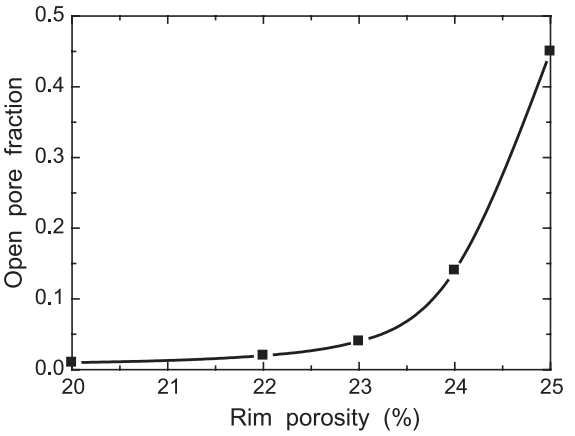

Fig. 5. Relationship between rim porosity and open pore fraction.

five cases. We can see that, for the same porosity, the effect of pore size is negligible and all curves for the five cases show the similar threshold porosity above which the open pore fraction increases very rapidly. Another simulation was also made to reveal the effect of rim thickness on the open pore fraction with a system of 600 (radial) × 300 (circumferential) × 300 (axial) small cubes, where the rim thickness is 300 μm in contrast to 150 μm used for the previous simulation. We chose 300 μm because it is close to the maximum rim thickness observed in the very high burnup fuel. The simulation shows that, for the same rim porosity, the open pore fraction was almost identical irrespective of the rim thickness. Consequently, it can be concluded that it is the rim porosity which determines the open pore fraction in the rim rather than the pore size distribution or the rim thickness. Based on this result, the rim porosity rather than the pore size or its distribution was chosen as a main simulation parameter.

The maximum rim porosity has been reported to be up to 24%; 14.5% [22], 23.5% [23] and 24% [21] for a respective pellet average burnup of 66.6, 56.3 and 102 GWd/t U. Therefore, to check whether considerable release paths would be formed for the rim porosity observed in LWR high-burnup UO_2 fuel, three-dimensional simulations were performed for the rim porosity ranging from 20% to 25%. Fig. 5 shows that the open pore fraction for the porosity of 24% is only about 14%. However, if the porosity is larger than around 24%, the open pore fraction increases abruptly. This implies that, for the rim porosity less than 24%, open channels in the rim through which considerable fission gas release could take place would not be formed. Manzel and Walker [21] performed a variety of destructive examinations for commercial PWR fuel pellets whose average burnup reached up to 102 GWd/t U. They found that the porosity in the pellet outer region, whose relative radius is greater than 0.9, increases from 7% at a pellet average burnup of 50 GWd/t U to almost 15% at 102 GWd/t U. In addition, they observed that the porosity at the outermost pellet rim, whose local burnup could be higher than 150 GWd/t U, is as high as 24%. However, they concluded that the outer region of the fuel, where the rim structure has been observed at fuel rod burnup above 50 GWd/t U, evidently does not make an important contribution to the level of fission gas release even at burnup as high as 100 GWd/t U. This observation is consistent with our analysis that almost no fission gas is released in the rim of a high burnup fuel pellet as long as its rim porosity is lower than 24%.

On the other hand, using X-ray fluorescence analysis (XRF) and electron probe microanalysis (EPMA), Mogensen et al. [24] measured the concentration of retained xenon along radial direction of the many fuel sections obtained from the high burnup effects program (HBEP). The measurement showed that, for the two cases whose rim burnups were higher than 75 GWd/t U, fission gas had been released from the rim to the rod free volume. Unfortunately, since the rim porosities of the two fuel sections where the measurement was made were not available, it is impossible to decide whether the gas release in the rim was related to its high porosity.

In case that the measured rim porosity is overestimated due to grain pullout, there would be a possibility that fission gas release in the rim would not be observed even for the rim porosity higher than the threshold value of 24% and hence it could be argued that the threshold rim porosity should be higher than 24%. However, since the highest rim porosity observed so far seems to be 24% [21], we can say at least that the threshold rim porosity of this paper could be used for interpreting almost no gas release in the rim with porosity less than 24%.

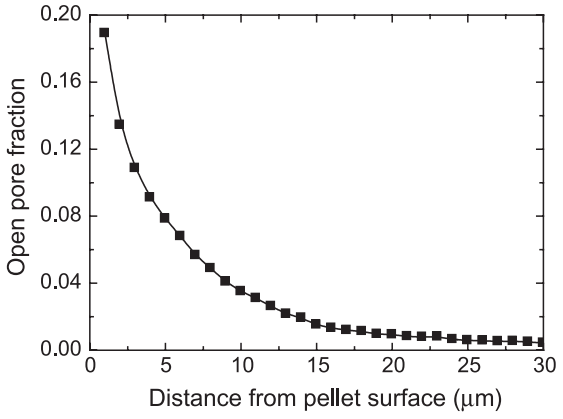

Fig. 6. Open pore fraction as a function of distance from a pellet surface for the rim porosity of 24%.

Therefore, the present study suggests that, if the rim temperature were low enough to maintain its microstructure, there would be no significant gas release in the rim as long as its porosity is lower than 24%. It is interesting to note that the threshold porosity and the maximum porosity observed in LWR fuel are almost in the same range.

Although restricted to a few tens of μm from the pellet outer surface, a trend to pore coalescence and incipient pore channel formation has been found at a rim burnup of 98 GWd/t U [5]. To confirm this observation, the open pore fraction near the pellet surface was also simulated as a function of distance from the pellet surface for the rim porosity of 24%. Fig. 6 shows that the fraction of open pores decreases with the distance from the surface and becomes negligible above 20 μm, supporting the observation that release channels are extended to only a few tens of μm from the pellet edge. In addition, the present study suggests that, once the rim porosity reaches a threshold value of 24%, extensive release channels would be developed and gas release would occur considerably in the rim resulting in fuel rod over-pressurization and clad lift-off.

If a model is available which predicts the average rim porosity as a function of rim burnup, the present method can be linked with a fuel performance code such as COSMOS [25] to calculate gas release in the rim region of high-burnup UO_2 fuel and also to analyze its effect on rod internal pressure and clad lift-off.

4. Conclusions

The interlinkage of rim pores and the formation of release paths in the outer region of high-burnup UO_2 fuel were simulated by the Monte Carlo method and the Hoshen–Kopelman algorithm. The following results have been obtained in the present study:

(1) It is the rim porosity rather than pore size distribution or rim thickness that determines the fraction of open pores connected to the pellet surface.
(2) The number of rim pores that connect with each other and form release channels in the rim increases very rapidly above the threshold porosity of around 24%. If the rim porosity is less than 24%, only a small number of rim pores located within 20 μm from the pellet surface could contribute to gas release resulting in a very low gas release.
(3) Once the rim porosity reaches a threshold value, extensive release channels would be developed and considerable gas release would occur in the rim leading to fuel rod over-pressurization and clad lift-off.

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