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Simulation of Pore Interlinkage in the Rim Region of High Burnup UO_2 Fuel

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Abstract

Threshold porosity above which fission gas release channels would be formed 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 cube, pore distribution in the rim was simulated 3-dimensionally by the Monte Carlo method according to porosity and pore size distribution. Then, 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 rim porosity. The simulation showed that porosity of 24-25% is the threshold above which the number of rim pores forming release channels increases very rapidly. On the other hand, channels would not be formed if the porosity is less than about 23.5%. This is consistent with the observation that, for porosity less than 23.5%, almost no fission gas is released in the rim. However, once the rim porosity reaches beyond 25%, extensive open paths would be developed and considerable fission gas release would start in the rim.

Key Words : rim region, high burnup, UO_2 fuel, fission gas release, threshold porosity, monte carlo method

1. Introduction

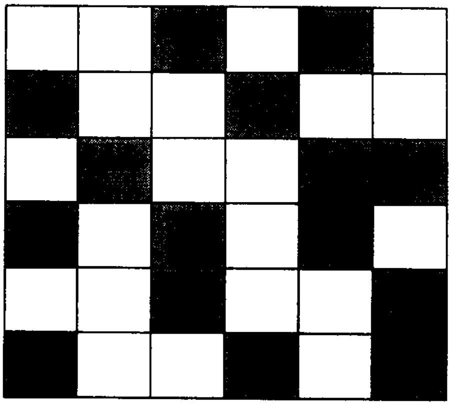
As discharge burnup of LWR UO_2 fuel increases, rim structure developed at the periphery of high burnup UO_2 fuel raises the concern that it could contribute to fission gas release and hence to the buildup of rod internal pressure. This concern is understandable because large fraction of fission gas accumulated in the rim pores created by restructuring [1-3] could eventually be released to the free volume of the fuel rod resulting in thermal feedback effect and possibly fuel rod over-pressurization and clad lift-off [4]. All safety authorities require that, to extend the discharge burnup of current UO_2 fuel, the thermal and mechanical behavior of high burnup fuel should be analyzed properly from the viewpoint of overall in-reactor performance and safety. Hence it becomes more and more important to reveal that, at which rim burnup or rim porosity, considerable fission gas release would start in the rim.

In a real situation, the formation of gas release channels in the rim region is a very complex phenomenon that involves pores whose size distribution depends on as-fabricated microstructure, power history and operating parameters such as temperature and burnup. In addition, the interlinkage of pores in the rim 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, microstructural characteristics of the rim region should be considered. Most of the models developed so far did not consider the statistical nature of the interlinkage of gas bubbles. On the other hand, some attempts were made by using the Monte Carlo method to simulate the connectivity of grain boundary gas bubbles to free surfaces [5-7]. However, the attempts used 2-dimensional methods rather than 3-dimensional ones to simulate the distribution and interlinkage of grain boundary bubbles.

In this paper, interlinkage of rim pores created by restructuring to the outer surfaces of a fuel pellet is simulated 3-dimensionally by using the Monte Carlo method and percolation theory for the rim where a large number of pores of around 1 μm or so exist among small grains of 0.2-0.3 μm produced by restructuring of as-fabricated grains [1]. In addition, feasibility of this simulation is discussed by comparing the present results with related experimental data.

2. Percolation Theory

To simulate the interlinkage of rim pores to free surfaces, two works need to be done; first, to



(a) Locating pores in some squares
(white: pore, gray: grain)

10	8		11		9
	8	7		9	9
6		7	4		
	1		4		5
3	1		4	2	
	1	1		2	

(b) Assigning a label to occupied squares

2	2		11		9
	2	2		9	9
6		2	2		
	1		2		5
1	1		2	2	
	1	1		2	

(c) Updating labels of occupied squares

Fig. 1. Application of Hoshen-Kopelman Algorithm to a 2-dimensional System

distribute the rim pores in the rim according to porosity and their size distribution, and then to calculate the total number of pores connected to the free surfaces. In this paper, the Hoshen-Kopelman algorithm [8] is adopted to check pore interlinkage and also to find out the open paths to the free surfaces. Here is an example of a procedure where the algorithm is used to check the pore interlinkage [8] in a 2-dimensional system for porosity of 58% together with the Monte Carlo method. It is assumed for simplicity that both rim pore and rim grain are of the same size and both of them can be represented by a square as shown in Fig. 1:

(1) Calculate the probability that a rim pore would be placed at a specific square. Here the probability, which is the same as the porosity, is assumed to be 0.58. Using the Monte Carlo method, locate rim pores in some number of squares according to the probability. First, for each square, generate a random number between 0 (zero) and 1 (one). If the random number is equal or less than the probability of 0.58, the square is taken as a pore. Otherwise, the square is assumed to be a grain. This procedure is repeated for all 36 squares and finally we get the pore distribution of Fig. 1(a). Since there are 21 white squares (pores) from the total of 36, the porosity in Fig. 1(a) is found to be 58% as was assumed before simulation. If we try another run, we could have a different porosity from 58%. However, for a large number of squares, almost the same porosity would be obtained even though the pore distribution is different.

(2) Starting at the origin (0,0), which is the leftmost square at the bottom row, assign a cluster label only to squares occupied by a pore. If a square at (x,y) is occupied, check only its neighboring squares with lower coordinates (x-1,y) and (x,y-1); that is, left and lower squares. If one of the two neighbors are occupied, the label of (x,y) is assigned to the same one as its occupied neighbor. In case that both neighbors are occupied and have different cluster labels, the label of (x,y) should be the lower one. On the other hand, if both of the two neighbors are unoccupied, the square gets the next cluster label available. Let's see an example at the uppermost row in Fig. 1(b). The first label is 10 because it's the next label available after the label 9 in the row below and, at the same time, it does not have any adjacent occupied squares. The label for the second occupied square is 8 because it is the lower one of the two neighbors' 8 and 10. For the third label, it is 11 since its two neighbors are unoccupied. The label for the last square is 9 due to its lower square whose label is 9.

(3) Update each cluster label in case that there are different labels for the interconnected same cluster. If two adjacent occupied squares have different labels, the higher label should be replaced by the lower one. We can see an example of updating by comparing the second rows from the bottom of Fig. 1(b) and Fig. 1(c), respectively. The label 3 in Fig. 1(b) is replaced by the label 1 because of its adjacent square. On the other hand, the label 4 is changed into 2 due to its right square whose label is 2. This procedure is repeated until all labels are changed into numbers that are the lowest possible in the same cluster. With this process, each square can be easily checked to see whether they are interlinked or not. If two squares have the same label, they are interlinked even if they are physically separated. Therefore, the total number of pores interlinked to a pellet surfaces can be obtained by counting the total number of pores whose labels are extended to the surface of the system.

(4) Calculate the number of pores connected to free surfaces. If the uppermost row is assumed to be a free surface, the pores whose cluster labels appear at the uppermost row are open pores. Since the pores with the label of 2, 9 and 11 are open ones, the total number of open pores is 14; 10 with the label 2, 3 with the label 9 and 1 with the label 11. Therefore, the fraction of open pores is 0.67 because 14 pores from the total of 21 are connected to the surface.

The above algorithm greatly reduces calculation time because checking for interlinkage is limited only to neighbors. In this paper, the algorithm has been extended to a 3-dimensional system so that we can apply it to the rim region and hence can calculate the fraction of rim pores connected to a pellet's outer surfaces.

3. Thickness of the Rim Region

Many investigators measured rim thickness by both EPMA and optical microscopy. The measured thickness is shown as a function of rim burnup in Fig. 2. The filled symbols in Fig. 2 represent data measured by EPMA and the open ones by optical microscopy. While there exists some controversy over whether rim thickness increases linearly or exponentially with fuel burnup, Fig. 2 shows that threshold local burnup required for rim formation is about 52 GWd/tU [9]. Recent data [10] obtained under typical PWR conditions suggest that rim thickness increases linearly with rim burnup at least up to 70 GWd/tU. Fig. 2 also indicates that rim thickness is about 300-350 μm for a rim burnup of 120 GWd/tU, which could be the maximum burnup observed in typical commercial LWR fuels.

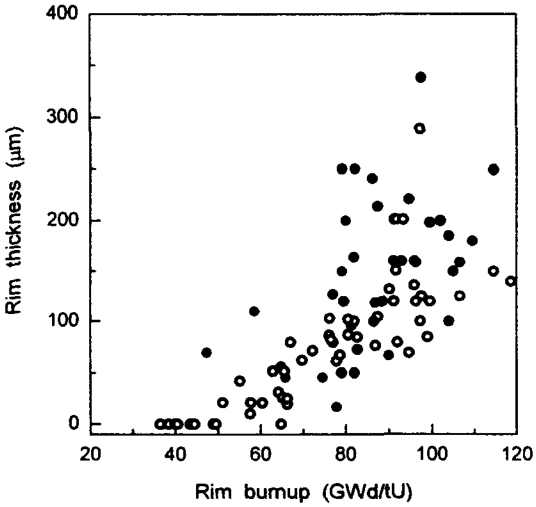


Fig. 2. Measured Rim Thickness by EPMA(filled symbols) and Optical Microscopy(open symbols)[9]

4. Representation of the Rim Region

It is assumed that the rim region 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 fission gas release. This would be a reasonable assumption because there would be no special reason or evidence to believe that the gas release behavior in the rim would be different along circumferential direction as long as burnup distribution in the rim which determines microstructural evolution - porosity and pore size distribution - would be also uniform.

There are two faces in the cube system through which fission gas release can occur; upper and right face. In this case, rim pores that are retained in the cluster labels appearing at one of the two faces are open ones. For a pellet whose radius, height, and rim thickness is R, H and t, respectively, the ratio of upper to right face area is approximately $2\pi Rt/2\pi RH$ which is reduced to t/H. If the rim thickness and pellet height are assumed to be 350 μm and 10 mm, the ratio is 0.035. Since there are two release faces in a fuel

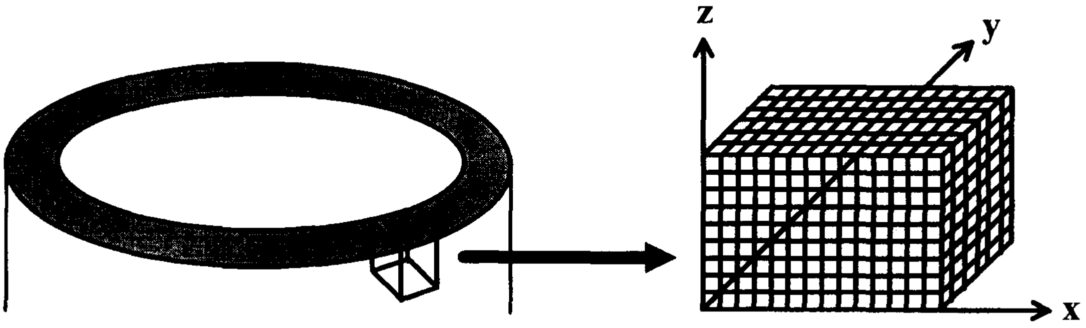


Fig. 3. Representation of Rim Region for the Simulation of Interlinkage

pellet, that is upper and lower face, the ratio is 0.070. Therefore, we can neglect the upper face in Fig. 3 and can consider only the right face, which is actually a circular face of a pellet, to be a major release path.

Rim region in a high burnup UO_2 fuel is represented by a system of 300(radial) × 300(circumferential) × 300(axial) small cubes, where the length of a small cube corresponds to 0.5 μm. Therefore, the system is a cube of 150 μm. The rim thickness of 150 μm along the radial direction has been chosen because it corresponds to about half the thickness of the rim observed in high burnup fuels. And the other two lengths have been taken to be the same as the rim thickness. The size of the system, which consists of 27 million small cubes, is 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 CPU of Pentium IV 2.53 GHz. 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 trials.

Since the mean size of rim pores is around 1 μm [1], a rim pore is represented by $2 \times 2 \times 2$ small cubes, a cube of 1 μm. However, rim pores observed by Spino et al. [11] in high burnup fuel have a different size distribution depending on burnup and microstructural characteristics. To investigate the effect of pore size on open pore fraction, 5 cases of pore size were analyzed; four cases of uniform pore size and one case with size distribution as shown in Fig. 4. The uniform pore sizes were $2 \times 2 \times 2$, $3 \times 3 \times 3$, $4 \times 4 \times 4$, and $10 \times 10 \times 10$ small cubes, respectively.

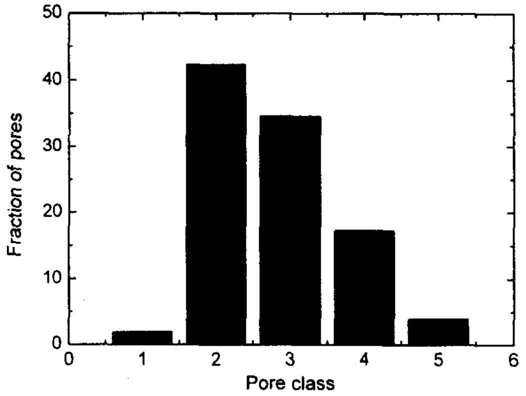


Fig. 4. Typical Pore Size Distribution in the Rim[11](Pore class 1 corresponds to 0.5μm in pore size. as the pore class increases, the pore size increases by 0.5μm)

To locate a rim pore in the large cubic system, we need to calculate the probability that a pore would be placed at a small cube. This probability can be calculated by Eq. (1) using the concept that the fractional volume occupied by a pore in a small cube is the same as the product of the probability and the pore volume:

$$P_{Pore} = \frac{P_{rim} V_{cube}}{V_{Pore}} \tag{1}$$

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 rim pore.

For example, let's assume that rim porosity is 23.5% and a rim pore is represented by a small cube. Then the volume of a rim pore and a cube would be the same and, according to Eq. (1), the probability that a pore exists at a small cube would be 23.5%. The procedure of locating pores and calculating the fraction of open pores is the same as explained in Sec. 2 except that the 2-dimensional system has been extended to 3-dimensional one.

However, situation is different when a rim pore is larger than a rim grain, which is the case for the high burnup UO_2 fuel. If we simulate a rim pore by a 2x2x2 cube, the probability that a pore exists at a small cube is calculated to be 2.94% because V_{Pore} is eight times larger than V_{cube}. In reality, pore distribution in the rim could not be random. Since the region around the pre-existing rim pores would be fission gas depleted, new pore creation here would be difficult compared to that in other region where large fission gas inventory is available. However, this effect was not taken into account in this paper due to difficulty in quantifying how new pores are created in the fission gas depleted zone.

Then the procedure of applying the Hoshen-Kopelman algorithm is slightly changed. First, starting at (0,0,0) position, generate a random number between 0 (zero) and 1 (one). If the random number for the first cube is equal or less than 0.0294 (2.94%), eight small cubes contained in a 2x2x2 cube are taken as a pore. The lowest (left, front, bottom) coordinates for eight cubes are (0,0,0), (1,0,0), (0,1,0), (0,0,1), (1,1,0), (1,0,1), (0,1,1) and (1,1,1), respectively. If a random number for the second cube of (1,0,0) is also equal or less than 0.0294 (2.94%), 2x2x2 cubes starting with the second cube having the lowest coordinate of (1,0,0) is taken as a rim pore.

If random numbers for two adjacent cubes indicate that both of them are pores, the size of the two pores is 3x3x3 small cubes rather than 4x4x4. Therefore, if a rim pore is simulated by a cube that is equal or larger than 2x2x2 small cubes, the resulting simulated porosity is usually 1-2 % smaller than the one used to derive the probability that a pore exists at a specific small cube. This is because some cubes are taken to be occupied simultaneously by the two adjacent pores. 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.

5. Results and Discussion

Fig. 5 shows the effect of pore size distribution on open pore fraction as a function of rim porosity for 5 cases. We can see that, for the

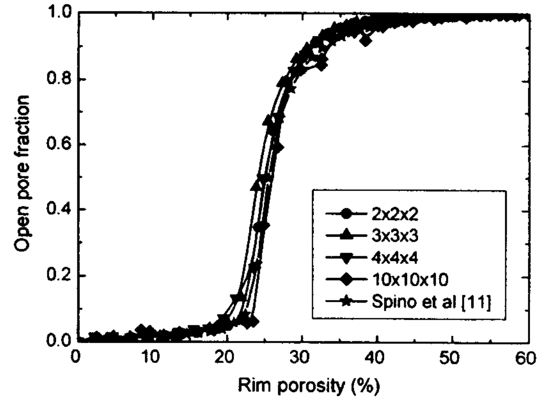


Fig. 5. Effect of Pore Size Distribution on Open Pore Fraction

same porosity, the effect of pore size is negligible. In addition, all curves for the 5 cases show the similar threshold porosity above which open pore fraction increases very rapidly. This simulation indicates that it is not the pore size distribution but the rim porosity which determines the open pore fraction in the rim. Based on this result, the rim porosity was chosen as a main simulation variable instead of pore size or its distribution.

Rim porosity has been reported to be 10~23.5% for a pellet average burnup up to 100 GWd/tU [11-12]. Therefore, to check whether considerable release paths would be formed for typical rim porosity observed in LWR UO_2 fuel, 3-dimensional simulations were performed as a function of rim porosity ranging from 20 to 25%. Fig. 6 shows that open pore fraction at 23.5% is only about 7%. However, if porosity is larger than around 23.5%, open pore fraction increases abruptly. This implies that, for a rim burnup of 100 GWd/tU whose corresponding rim porosity could be up to 23.5%, open channels in the rim through which considerable fission gas could be released would not be formed. This result is consistent with the observation that almost no fission gas is released in the rim up to 100 GWd/tU [2,13,14]. 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 23.5%.

At a rim burnup of 98 GWd/tU, though restricted to the first several few tens of μm from the pellet outer surface, a trend to pore coalescence and incipient pore channel formation has been found [15]. To confirm this observation, open pore fraction near the pellet surface was also simulated as a function of distance from the pellet surface for rim porosity of 23.5% with the same system and pore size of Fig. 6. Fig. 7 shows that the fraction of open pores decreases with the distance from the surface and becomes negligible above 20 μm, which supports 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 some threshold value of around 24-25%, 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.

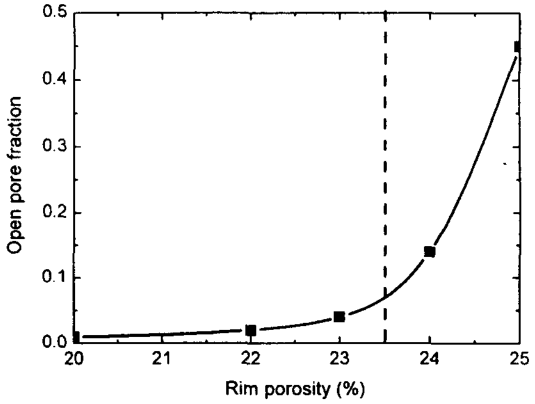


Fig. 6. Relationship Between Rim Porosity and Open Pore Fraction

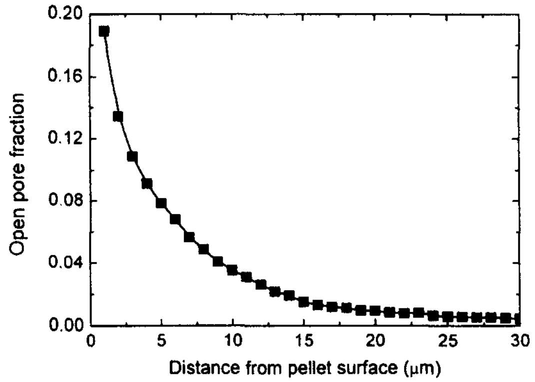


Fig. 7. Open Pore Fraction as a Function of Distance from Pellet Surface for Rim Porosity of 23.5%

6. Conclusions

Interlinkage of rim pores and formation of release paths in the outer region of high burnup UO_2 fuel was 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 that determines the fraction of open pores connected to the pellet surface.
2) The number of rim pores that connects with each other and form release channels in the rim increases very rapidly above the threshold porosity of 24-25%.
3) If the rim porosity is less than about 23.5%, only a small number of rim pores located within 20 μm from the pellet surface could contribute to gas release resulting in very low gas release.
4) Once the rim porosity reaches some threshold value of around 24-25%, extensive release channels would be developed and considerable gas release would occur in the rim resulting in fuel rod over-pressurization and clad lift-off.

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