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Simulation of Interlinkage of Grain Boundary Gas Bubbles to Free Surfaces by the Monte Carlo Technique

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몬테 카를로 기법을 이용한 결정립계 기포의 자유 공간으로의 연결 모사

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Abstract

A method to simulate the extent of interlinkage of grain boundary gas bubbles to the free surfaces of fuel pellet was developed. With the shape of UO_2 grain treated as tetrakaidecahedron (TKD), the interlinked fraction of fission gas bubbles to free surfaces at grain corners was calculated as a function of the radius of grain corner bubbles by the Monte Carlo technique. In spite of two dimensional analysis, the present method showed reasonable agreement between predicted and measured fuel swelling at the moment that complete bubble interlinkage was achieved. However, for more realistic simulation of interlinkage, grain corner bubbles should be treated three dimensionally.

요 약

결정립계에 존재하는 핵분열기체의 기포가 소결체의 외부와 연결되는 정도를 모사할 수 있는 방법을 개발하였다. UO_2 결정립의 형상을 TKD로 취급할 때, 결정립 Corner에서 자유 공간과 연결되는 핵분열기체의 기포 비율을 결정립 Corner에 형성된 기포 반경의 함수로서 몬테 카를로 방법을 이용하여 계산하였다. 2차원적인 분석에도 불구하고, 본 방법은 모든 기포가 자유 공간과 완전히 연결된 순간에서 예측된 핵연료 팽윤과 측정된 핵연료 팽윤이 비교적 잘 일치함을 보였다. 그러나 핵분열기체 기포가 외부와 상호 연결된 정도를 좀 더 사실적으로 모사하려면 결정립 Corner의 기포를 3차원적으로 취급해야 한다.

1. Introduction

Many workers have revealed that release of fission gases from fuel matrix to fuel exterior during irradiation depends on the extent of interlinkage of grain boundary gas bubbles to free surfaces. Beere and Reynolds [1] predicted that a stable tunnel network forms along grain edges when fuel swelling is larger than 8%. Turnbull [2] has shown experimentally that tunnels form when the swelling is larger than 7%. Villalobos et al. [3] assumed that bubble interlinkage is achieved when the amount of the swelling in grain edges is 7%. Dollins and Nichols [4] also assumed that no fission gas escapes from the fuel until gas bubble swelling on grain edges and corners reaches 5%. Tucker and White [5] who approximated UO_2 grain as tetrakaidecahedron (TKD) [6] (see Fig. 1) considered that long range interlinkage of grain corner bubbles connected by four grain edges (see Fig. 2) would be established, provided that the probability of finding a tunnel open at any time is greater than or equal to 0.4.

In the real situation, however, tunnel formation is a probabilistic phenomenon involving complex geometry and various size of grains. All the models described above, however, do not take into account the fact that the interlinkage of grain corner bubbles is statistical in nature due to arbitrary distribution of grain corners.

In the present work, with the shape of UO_2 grain treated as TKD, the fraction of fission gas bubbles interlinked to free surfaces at grain corners is determined in terms of the radius of curvature of grain corner bubbles r_c by the Monte Carlo technique. However, the method of calculating r_c, which is described by White and Tucker [7], is beyond the scope of this paper.

2. Problem Setup

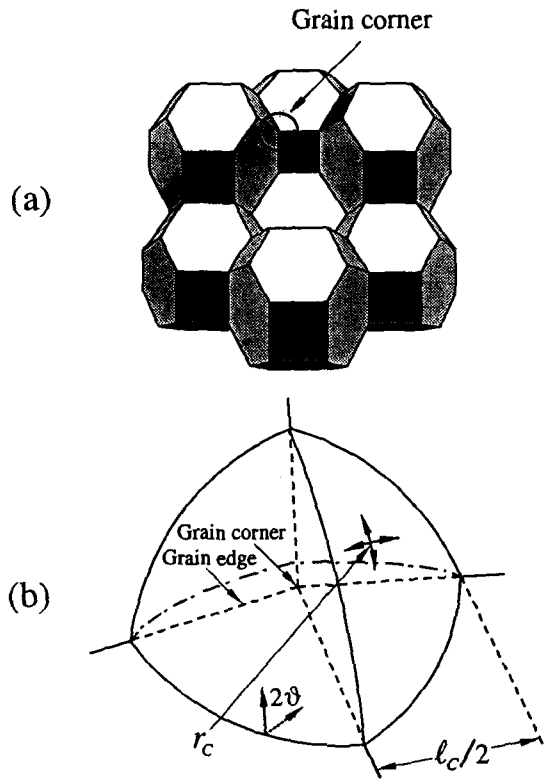
Before irradiation, UO_2 grains can be considered to be tightly sintered together with only a small amount of initial porosity produced during manufacturing process. It is assumed that the geometrical shape of this fuel and the initial porosities define an effective Booth's [8] surface area for release such that the surface-to-volume ratio may be approximated as a large sphere of radius A, which is called as the poly-granular aggregate (PGA). This radius will obviously depend on the manufacturing method, initial density and open porosity present in the fuel as well as the basic UO_2 grain size. In addition, the radius A will clearly depend on the presence of cracks and thus could decrease with increasing burnup. In the present work, it is assumed that A is constant and therefore does not change during irradiation. According to White and Tucker [7], the radius of PGA is 2×10^{-4}m in case that UO_2 grain radius is 5 μm.

Based on this argument, it is assumed that the basic building block of fuel pellet is a PGA whose radius is A. This means that once the fission gas atoms generated within the matrix migrate to the surface of the PGA, they are released to the outside of fuel pellet.

Under the assumption that the shape of UO_2 grain is that of TKD, the prerequisite for the formation of grain boundary tunnels through which fission products can be released to the outside of fuel pellet is the establishment of fission gas bubbles at grain corners (see Fig. 1). Once the grain corner bubbles are established, they grow as irradiation proceeds and the radius of bubble would also increase. Therefore, the problem to be solved in this paper is to determine the fraction of grain corner bubbles interlinked to the surface of PGA as a function of the radius of curvature of grain corner bubble r_c after grain corner bubbles are established.

3. Simulation of Problem by the Monte Carlo Technique

The shape of fission gas bubbles that form at grain corners is that of a tetrahedron constructed of four



$r_c = 1.226\,\ell_c$

ℓ_c : Bubble length along grain edge
r_c : Radius of grain corner bubble
ϑ : Dihedral angle (= 50°)

Fig. 1. Geometry of (a) TKD Grains [6] and (b) Grain Corner Bubbles [7].

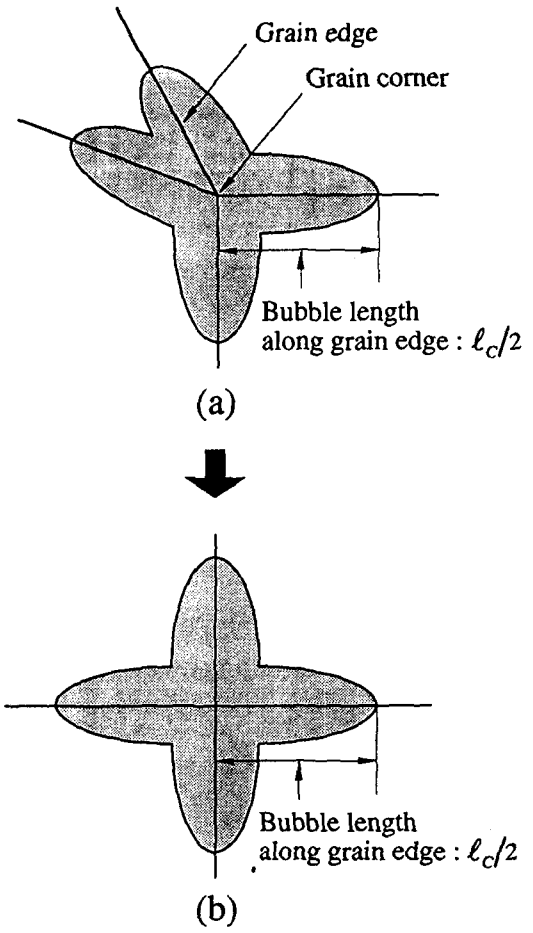


Fig. 2. Two Dimensional View of (a) Grain Corner Bubble and (b) Its Simplification for the Present Analysis.

spherical segments of radius r_c with four vertices extending the grain edges (see Fig. 2). As grain corner bubbles grow, they eventually join to form chains at grain corners and give rise to a network of channels.

Two grain corner bubbles belong to the same cluster if there is an unbroken sequence of bonds from the first bubble, through other bubbles, to the second bubble. Consequently, of all the bubbles within a PGA, it is required not only to determine all clusters connected to the surface of PGA but also to calculate the total number of bubbles within these clusters.

Since this is a three dimensional problem which is complex and takes too much computing time to simulate, it is reduced to a two dimensional problem as follows. Let's consider the cross-sectional circle of a PGA which has the same radius as that of the PGA. Then it is assumed that the fraction of grain corner bubbles interlinked to the circumference of the cross-sectional circle through clusters (see Fig. 3) is equal to the fraction of grain corner bubbles connected to the surface of the PGA. That is, this means that following relation holds:

$$f_{2D} = f_{3D}, \quad (1)$$

where

$f_{2D} = C_{2D}/N_{2D}$,

$f_{3D} = C_{3D}/N_{3D}$,

C_{2D} = number of grain corner bubbles connected to the circumference of a cross-sectional circle,

N_{2D} = total number of grain corner bubbles in a crosssectional circle,

C_{3D} = number of grain corner bubbles connected to the surface of a PGA,

N_{3D} = total number of grain corner bubbles in a PGA.

Furthermore, for two dimensional simulation, the geometry of grain corner bubble is assumed to have the two dimensional crux with one side length l_c (see Fig. 2) rather than three dimensional shape as shown in Fig. 1.

For a given number of grains within the cross-sectional circle, it is attempted to determine the total number of grain corner bubbles within the clusters that are interlinked to the circumference as a function of the radius of grain corner bubble. To determine how many grain corner bubbles randomly positioned within the circle are interlinked to the circumference, following procedure is adopted:

(1) Generate a given number of grain corner bubble with the shape of two dimensional crux within the cross-sectional circle with its position and orientation being determined by the Monte Carlo technique. That is, the position and the orientation of crux are statistical variables generated from the random number. Additional fixed variables needed for the Monte Carlo simulation are the number of grain corner bubbles within the circle N_{2D} and the length of grain corner bubbles along grain edge l_c. Mathematical description of these fixed variables and the values of these variables together with other related parameters are described in section 4.

(2) Determine all clusters where at least one of its bubbles are interlinked to the circumference of the circle.

(3) Obtain the fraction f_{2D}, which will be denoted as f hereafter, by dividing the total number of bubbles within these clusters by a given number of bubbles within the circle. As may be easily expected, the fraction of bubbles interlinked to the circumference would increase with bubble length.

4. Statistical and Other Related Variables

The number of grains within a PGA, N, is

$$N = \frac{4\pi A^3/3}{4\pi a^3/3} = \left(\frac{A}{a}\right)^3,$$

where

a = grain radius.

Since six grain corners are present per grain, the number of grain corner bubbles within a PGA, N_{3D}, is

$$N_{3D} = 6N = 6\left(\frac{A}{a}\right)^3.$$

Then the number of grain corner bubbles in a cross-sectional circle of radius A is roughly equal to two-thirds power of the number of grain corner bubbles in a PGA. Therefore, the two dimensional number of grain corners, N_{2D}, is

$$N_{2D} = N_{3D}^{2/3} = 6^{2/3}\left(\frac{A}{a}\right)^2. \quad (2)$$

Since all grains are nearly the same size, grain corner bubbles can be considered to be uniformly distributed not only within the cracked body but also in the circle. For the purpose of uniformly placing the grain corner bubbles within the circle, the circle is divided into n_r sections both radially and circumferentially; that is, the circle is divided into $n_r \times n_r$ area-units which have equal area of $\pi A^2/n_r^2$. Then

the position and the orientation of each grain corner bubble with the shape of crux as shown in Fig. 2 is generated within each area-unit by the Monte Carlo technique. Hence the number n_r is obtained as follows:

$$n_r = N_{2D}^{1/2} = 6^{1/3}\left(\frac{A}{a}\right). \quad (3)$$

Since the value of A is very small as can be seen in Table 1, simulation is performed in a circle of radius A_f greater than A. Let

A = radius of a PGA circle,
A_f = radius of the circle in Fig. 3,
a = grain radius,
a_f = grain radius in the circle of radius A_f,
l_c = length of grain corner bubble along grain edge,
$l_c^f = l_c$ in the circle of radius A_f.

Then a_f and l_c^f in simulating the present physical situation are determined in such a way that the ratios of these two values to A_f become equal to those of a and l_c to A. Then the following relations are obtained:

$$a_f = \left(\frac{a}{A}\right) A_f,$$

$$l_c^f = \left(\frac{l_c}{A}\right) A_f.$$

According to White and Tucker's model [7], grain corner bubbles begin to interlink each other at the radius of 0.4965 a and they interlink completely at the radius of 0.88a. In addition, from the relation of $r_c = 1.226 l_c$ (see Fig. 1), the range of l_c^f in the circle of radius A_f is between 0.4050 a_f and 0.72 a_f, that is,

$$0.4050\left(\frac{a}{A}\right)A_f \le l_c^f \le 0.72\left(\frac{a}{A}\right)A_f. \quad (4)$$

In conclusion, additional fixed variables used in the Monte Carlo technique are as follows:

- radius of the cross-sectional circle, A_f
- number of grain corner bubbles in the circle, N_{2D}
- number of division, n_r
- length of grain corner bubble along grain edge, l_c.

The values of fixed variables and the other related parameters needed for the present simulation are given in Table 1.

Table 1. Values of Fixed Variables and Other Related Parameters

a	A (cm)	a/A	A_f (cm)	N_{2D}	n_r	$l_c^{f,\max}$ (cm)
5×10^{-4}	2×10^{-2}	1/40	3	73^2	73	0.0539

5. Results and Discussion

Fig. 3 presents one example showing how interlinkage of grain corner bubbles to free surface develops with bubble radius when A_f is 3cm. At the moment that grain corner bubbles are established at the bubble radius of 0.47 a [7], only bubbles located very near the surface are interlinked to the circumference of the circle. As bubbles grow with temperature and burnup, bubble networks are more and more extended to the interior region. At the geometrical maximum bubble radius of 0.88a, almost all bubbles are interlinked to the circumference through clusters. In the real situation, however, the bubble can keep growing and hence the bubble radius can become larger than 0.88a. All bubbles are interlinked completely to the circumference when the bubble radius is greater than around 0.94a.

Interlinked fraction of grain corner bubbles is presented in Fig. 4 as a function of bubble radius. The result shown here is obtained by best-fitting of 40 calculations for the same radius of grain corner

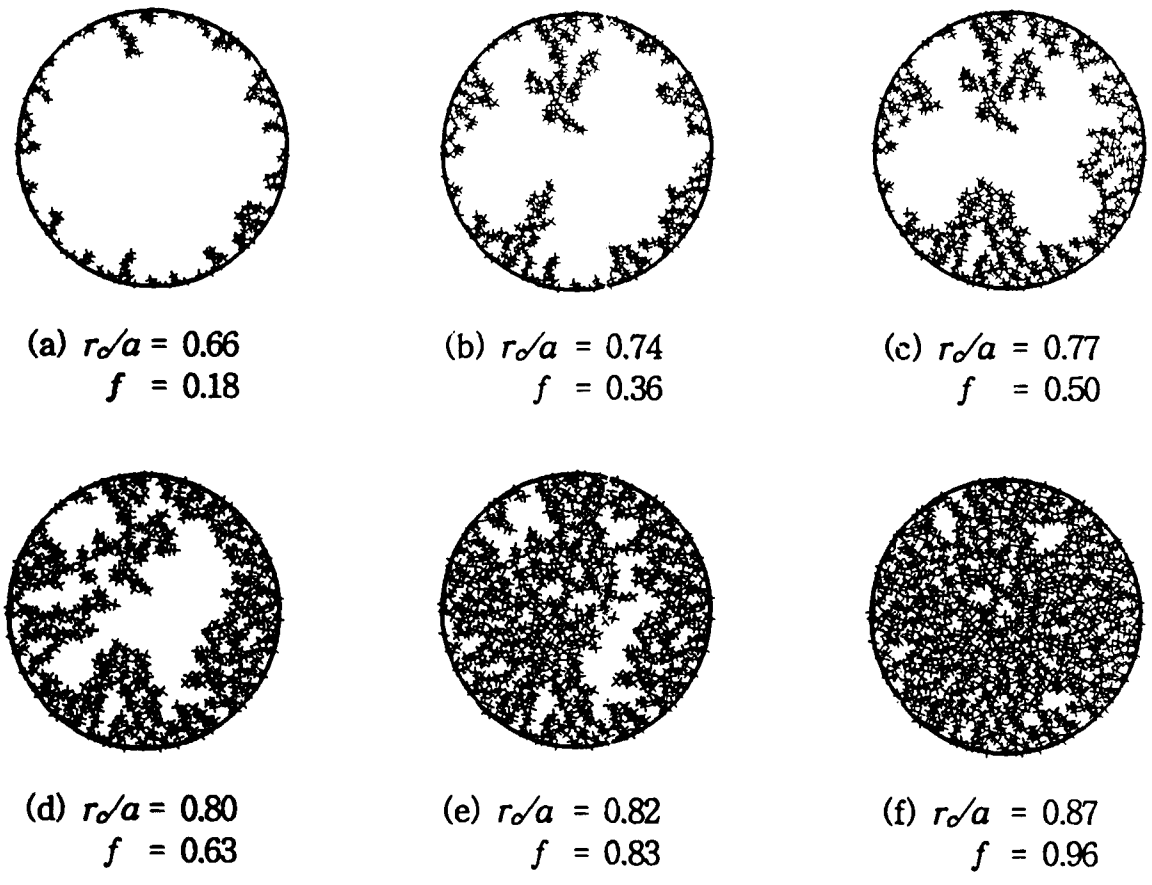


Fig. 3. Development of Bubble Interlinkage to Free Surface at Grain Corners and Interlinked Fraction as a Function of Relative Bubble Radius

bubble within the circle. Scattering of interlinked fraction for the same radius is due to the statistical nature. After grain corner bubbles are established at the bubble radius of 0.47a, about 5% of bubbles is interlinked to the free surface. Interlinked fraction increases with bubble radius almost linearly up to 30% and then abruptly up to about 95%. When bubble radius reaches its maximum value of 0.88a [7], interlinked fraction is close to unity. This result is understandable for the following physical background : for this value of bubble radius, grain corner bubbles in one grain would be connected to those in the next grain via interlinked bonds at each grain corner. This means that all grain corner bubbles would be interlinked among themselves and finally to the surface of the PGA.

This result is consistent with White and Tucker's assumption [7] that complete interlinkage of gas tunnel network occurs when the radius of grain corner bubbles is 0.88a. In this situation grain corner swelling is 8.7%, which is comparable to those presented by Beere and Reynolds [1] and Turnbull [2] but is a little higher than those suggested by Villalobos et al. [3] and Dollins and Nichols [4].

There would exist clusters, however, that are connected to the surface of the PGA even though they

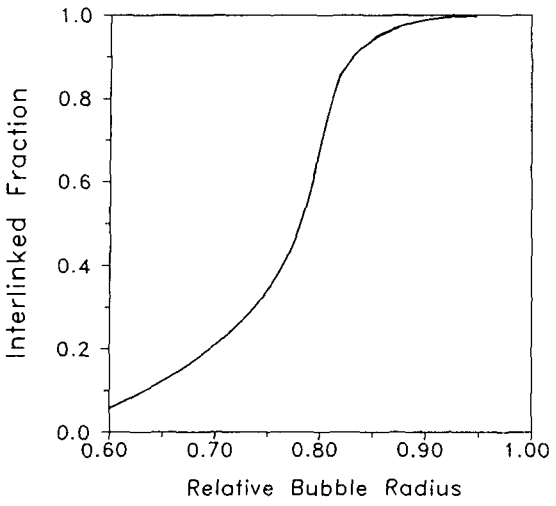


Fig. 4. Interlinked Fraction of Grain Corner Bubbles to Free Surface

are regarded as not being interlinked to the circumference of the circle. Therefore, two dimensional simulation would underpredict the fraction of gas bubbles interlinked to the free surfaces of fuel pellet.

Sensitivity study which was made to see the effect of A_f on f showed that f was almost insensitive to the change in A_f. This insensitivity can be understood by the following reasoning: when A_f becomes large, interlinkage among bubbles becomes easier due to the increased bubble radius. On the other hand, clusters formed inside the circle have more difficulty in touching the circumference of a circle because the cirlce radius is increased. These two opposite facts make f insensitive to the decrease in A_f. In a similar way, f is almost unchanged when A_f becomes small.

6. Conclusion

In spite of two dimensional analysis for the prediction of interlinkage of grain corner bubbles to free surfaces, the present method shows reasonable agreement between predicted and measured fuel swellings at the moment that complete bubble interlinkage is achieved. However, for more realistic simulation of interlinkage, grain corner bubbles should be treated three dimensionally.

References

[1] W. Beere and G.L. Reynolds, "The Morphology and Growth Rate of Interlinked Porosity in Irradiated UO_2," *J. Nucl. Mater.*, **47**, 51 (1973).

[2] J.A. Turnbull, "The Effect of Grain Size on the Swelling and Gas Release Properties of UO_2 During Irradiation," *J. Nucl. Mater.*, **50**, 62 (1974).

[3] A. Villalobos, A.R. Wazzan and D. Okrent, "Analysis of Fission Gas Disposition in Light Water Reactor Steady-State Operation," *Nucl. Tech.*, **58**, 492 (1982).

[4] C.C. Dollins and F.A. Nichols, "Swelling and Gas Release in UO_2 at Low and Intermediate Temperatures," *J. Nucl. Mater.*, **66**, 143 (1977).

[5] M.O. Tucker and R.J. White, "The Release of Unstable Fission Products from UO_2 During Irradiation," *J. Nucl. Mater.*, **87**, 1 (1979).

[6] H.V. Atkinson, "Theories of Normal Grain Growth in Pure Single Phase Systems," *Acta. Metall.*, **36**, 469 (1988).

[7] R.J. White and M.O. Tucker, "A New Fission Gas Release Model," *J. Nucl. Mater.*, **118**, 1 (1983).

[8] A.H. Booth, "A Suggested Method for Calculating the Diffusion of Radioactive Rare Gas Fission Products from UO_2 Fuel Elements and a Discussion of Proposed In-Reactor Experiments That May Be Used to Test Its Validity," AECL-700 (1957).