ELSEVIER

Journal of Nuclear Materials 279 (2000) 207–215

journal of nuclear materials

www.elsevier.nl/locate/jnucmat

An attempt to explain the high burnup structure formation mechanism in UO_2 fuel

Chan Bock Lee *, Youn Ho Jung

Korea Atomic Energy Research Institute, Yusung, P.O. Box 105, Taejon, South Korea

Received 23 June 1999; accepted 24 January 2000

Abstract

A formation mechanism of high burnup structure (HBS) or rim effect in UO_2 fuel was proposed. The fission gas bubbles in the HBS region may be nucleated and stabilized at the threshold atomic concentration of gas atoms inside the grain of UO_2 fuel. HBS formation in HBS region may be driven not by the conventional thermal diffusion process, which should increase with the temperature, but by the cascade of atomic movements by the fission fragments. Based upon the above mechanism, the threshold local burnup for HBS initiation was derived as a function of temperature, fission rate and grain size. The threshold burnup for HBS initiation was predicted to increase with temperature, and at high temperature, HBS would never form due to the fission gas release. Variations of HBS widths among the different irradiation tests could be explained by the proposed mechanism.

PACS: 28.41.Ak; 28.41.Bm

1. Introduction

Due to the characteristic neutron energy spectrum in light water thermal reactors, fissile ^{239}Pu is produced by resonance neutron absorption of ^{238}U in the narrow periphery region of the UO_2 pellet. Therefore, local burnup near the pellet edge becomes 2–3 times higher than the pellet average burnup. It was found that for high burnup fuel, small bubbles were nucleated and grains were finely divided at the periphery of the UO_2 pellet, which is called rim effect or high burnup structure (HBS). Since the temperature at the pellet periphery is low, the bubble formation in that region has received primary attention. Research on the rim effect or HBS has been extensively performed over 10 years. However, since HBS is formed at the outer narrow region of the high burnup UO_2 pellet, there have been difficulties and uncertainties in analyzing its microstructure and measuring the local burnup and fission gas inventories.

Through the cumulative efforts of many researchers [1–9], the characteristics of HBS were identified as follows. In the region where HBS developed, grains are finely divided into very small grains with a size of 0.1–0.5 μm, and fission gas bubbles with an average diameter of 1–2 μm are distributed uniformly between the subgrains. HBS starts to form at the local burnup of around 60–80 MWd/kgU at the periphery of the pellet. Spino et al. [4] showed that the porosity of HBS was 15–17% and fission gases in the HBS region would not be released up to the porosity of ~20%, and that during HBS formation, bubbles may form first, and then be followed by grain subdivision due to the overpressure of the bubbles.

Along with the microstructural examination of HBS, an explanation of its causes and mechanisms has been proposed [10–13], mainly through burnup and/or radiation damage, and therefore, primary variables in HBS formation seem to have been identified. However, further work is still needed for the completeness in explaining HBS formation and its progress. This work

* Corresponding author. Tel.: +82-42 868 2257; fax: +82-42 864 1089.

E-mail address: cblee@kaeri.re.kr (C.B. Lee).

0022-3115/00/$ - see front matter
PII: S0022-3115(00)00021-0

extends the study of the HBS formation mechanism. An attempt is made to explain the causes and mechanisms of HBS formation in relation with such parameters as the temperature, burnup, grain size, fission density, rod internal pressure and ^{235}U enrichment, etc.

2. Fission gas behavior

Fission gas atoms generated by fission inside a UO_2 grain tend to diffuse into the grain boundary to form bubbles. Bubbles formed at grain boundary surfaces have a lenticular shape, and they grow and interconnect with each other. Then the grain face bubbles move to the grain edge to form more stable bubbles. The bubbles in the grain edges can be interconnected with each other and ultimately become open to the external surface. When the bubbles in the grain edge become open to the external surface, the fission gases in the open bubbles are released instantaneously. Then, the open bubbles act as an open channel for the release of the fission gases which will diffuse into them later. Therefore, gas concentrations at the grain boundary depend upon the diffusion flux of the gas atoms from inside the grain, so that it depends upon temperature, irradiation time, and the grain size.

Fig. 1 shows the variation of the gas atom concentration at the grain boundary as a function of temperature and grain size. Calculations were performed numerically by solving the following differential equation, based upon Speight's derivation [14] under the conditions of intragranular bubble formation and resolution

$$\frac{\partial}{\partial t}C(r,t) = D\left(\frac{b}{b+g}\right)\left(\frac{\partial^2}{\partial r^2}C(r,t) + \frac{2}{r}\frac{\partial}{\partial r}C(r,t)\right) + YF(t), \quad (1)$$

where $C(r,t)$ is the gas atom concentration as atoms and bubbles (atoms/m^3), D the diffusion coefficient of gas atoms (m^2/s), b the resolution rate of gas atoms from the in-grain bubbles (s^{-1}), g the capture rate of gas atoms by the in-grain bubbles (s^{-1}), and $YF(t)$ is the gas atom production rate by fission (atoms/m^3 s).

Gas atom concentration at the grain boundary increases with temperature and grain size. It is notable that gas concentration per unit area increases with grain size. Therefore, bubble interconnection at the grain boundary surface may occur earlier for a larger grain than for a smaller grain. After the bubble interconnection has occurred, the fractional release of the fission gas will be smaller for larger grains according to Booth's derivation [15], since the diffusion distance of the larger grain is longer, as shown in Fig. 2. Fig. 2 shows the fractional fission gas release of the fuels with different grain sizes when all of the fission gases in the grain boundary are assumed to be released after reaching the saturation concentration for bubble interconnection, which was assumed as 8×10^{19} atoms/m^2 [16], and the resolution of gas bubbles at the grain boundary back to the grain inside was not taken into account. Irradiation tests of different grain sizes by Hirai et al. [17] showed this behavior. At a burnup of less than 30 MWd/kgU, the fractional fission gas release of larger grains was higher than or comparable to that of smaller grains, while at high burnup after enough interconnection of the bubbles at the grain boundary, the fractional fission gas release of the larger grain is smaller. In the Halden fuel

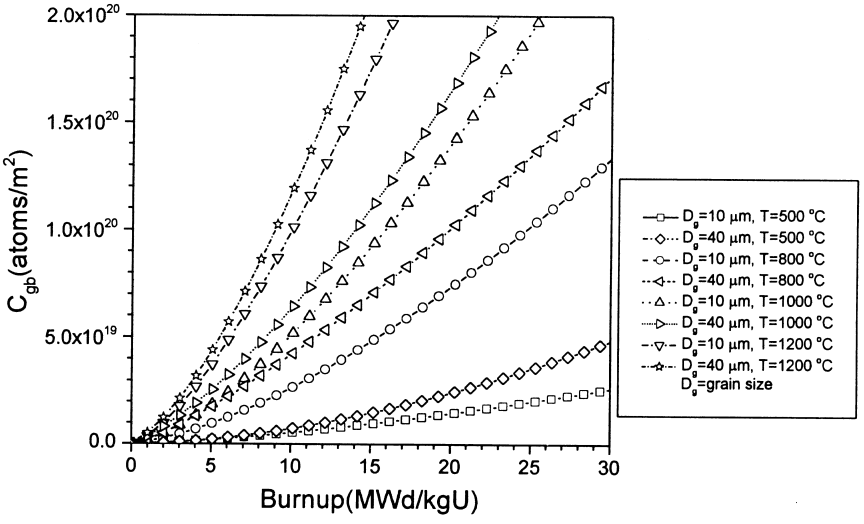

Fig. 1. Variation of the gas atom concentration at the grain boundary.

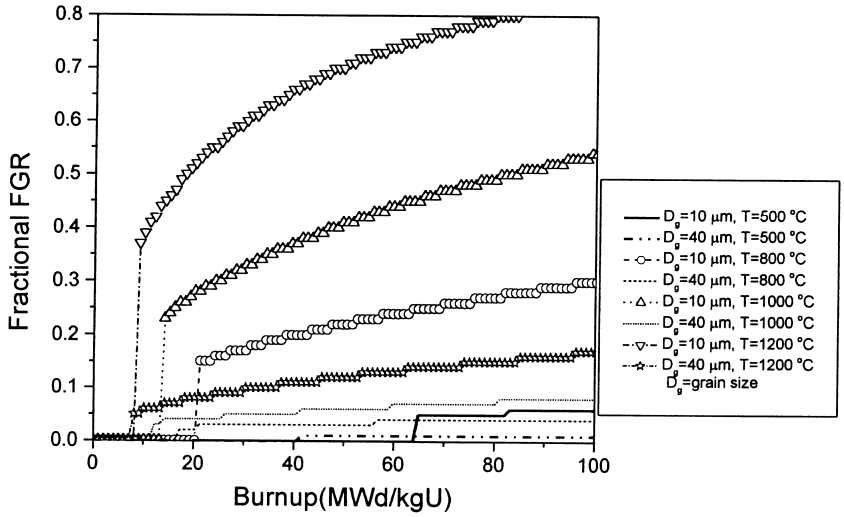

Fig. 2. Fractional release of fission gases.

irradiation tests, larger grain fuel showed a higher fission gas release at low burnup and the reverse at high burnup [18,19].

The diffusion coefficient of the fission gases is one of the key parameters in the fission gas behavior. There exist some variations among the several correlations for the diffusion coefficient of xenon in UO_2 under the irradiation environment. In this study, the diffusion coefficient [20] determined by using the classifications of Turnbull et al. [21], such as temperature, fission rate and a mixed contribution from both, was used. Fig. 3 shows the dependence of the diffusion coefficient upon the temperature and fission rate. At temperatures less than 700°C, diffusion coefficient of the gas atoms is controlled by the fission rate rather than the temperature. There is a difference of one-order of magnitude between the diffusion coefficients at the temperatures of 500 and 1000°C.

Fig. 4 shows the predicted fission gas distribution inside the grain after 1200 days of irradiation. At temperatures below ~700°C, it can be seen that few are diffused to the grain boundary, while at temperatures higher than 1200°C, the diffusion process is stabilized to show a parabolic radial distribution of the gas atom concentration. Accordingly, since the temperature near

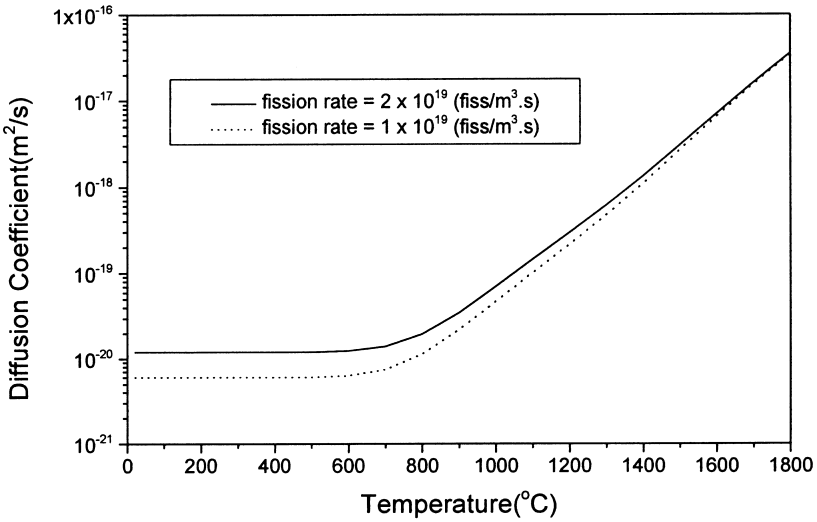

Fig. 3. Diffusion coefficient of fission gas atoms in UO_2 fuel [20].

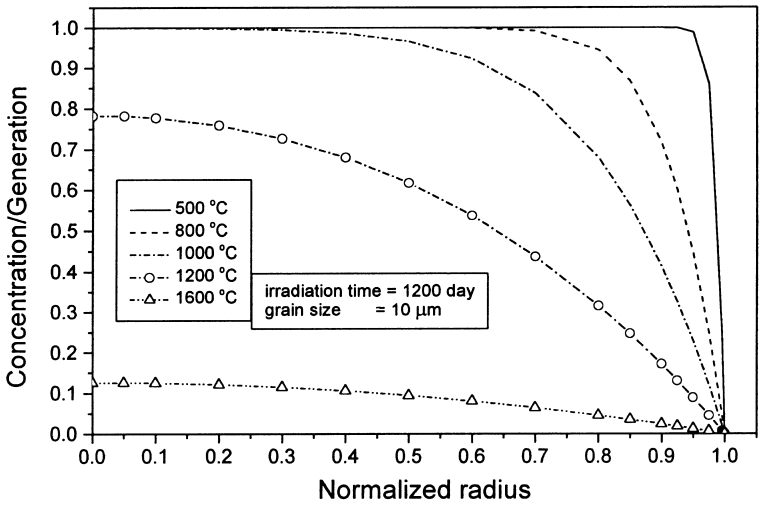

Fig. 4. Fission gas atom distribution inside the spherical grain.

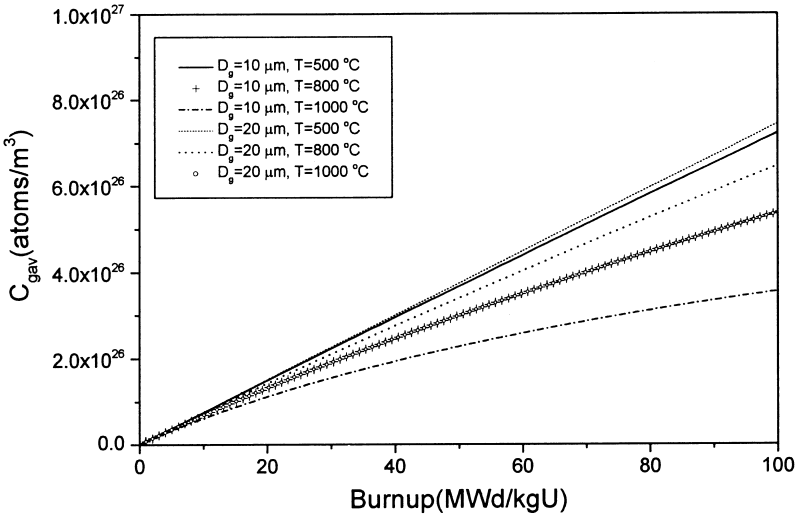

Fig. 5. Average concentration of the gas atoms inside the grain.

the pellet edge is in the range of 400–700°C during irradiation in LWRs, fission gas atoms generated by fission stay inside of the grains. Fig. 5 shows the average concentration of the gas atoms inside the grain as a function of temperature, fission rate and grain size. The concentration of the fission gas atoms at a local burnup of 70 MWd/kgU, which is considered as the typical threshold burnup for HBS formation [22], is about 5.1×10^{26} atoms/m^3.

3. HBS formation mechanism and discussion

Microstructural examination of the HBS region in a UO_2 pellet showed that the diameter of the bubbles would be in the range of 1–2 μm [4]. During the irradiation of the UO_2 fuel, it is known that very small intragranular bubbles with diameters of $\sim 10^{-9}$ m can nucleate along the tracks of fission fragments [23,24] and are in equilibrium due to the continuous nucleation and resolution by irradiation.

Fig. 6 shows the atomic concentration in the bubbles in equilibrium as a function of bubble radius, temperature and the external pressure on the bubble, assuming the ideal gas law. It can be seen that the external pressure on the bubbles caused by such parameters as the rod internal pressure and the contact pressure between the pellet and the cladding is one of the key parameters. Under the typical operating conditions of PWR fuels at high burnup, the rod internal pressure is in the range of

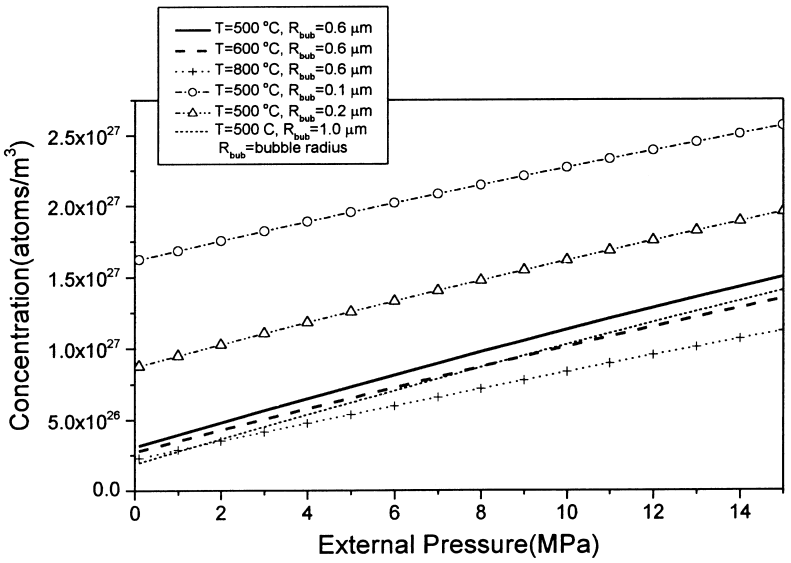

Fig. 6. Gas atom concentrations in the bubbles in equilibrium.

8–11 MPa. At the external pressure of 9 MPa, the atomic concentration in the bubbles with a diameter of 1.2 μm is about 1×10^{27} atoms/m^3, which is just about two times the gas atom concentration inside the UO_2 grain at the local burnup of 70 MWd/kgU.

Examination of tracks of fission fragments with high kinetic energy due to fission showed that fission fragment moved about 6.5 μm, and that the width of the track was about 15 nm [23,24]. Bubbles of very small size ($\sim 10^{-9}$ m) are formed along the tracks of fission fragments. In the displacement spikes formed along the tracks of the fission fragments, cascade of atomic displacement would occur [24]. During the recombination stage of the displaced atoms, bubbles could be nucleated from the super-saturated gas atoms inside the grain. As the gas atom concentration inside the grain increases, number density and the size of the nucleated bubbles would increase. It would be probable that the nucleated bubbles of high number density near the displacement spikes could be interconnected with each other, and gas atoms in a highly super-saturated concentration inside the grain could be captured by the adjacent bubbles by enhanced mobility due to irradiation. Enhanced mobility includes both the short range movement during the recombination stage of the displaced atoms in the displacement spikes as well as the enhanced thermal diffusion of gas atoms through the point defects such as vacancy generated by irradiation [23]. Therefore, nucleated bubbles could be stabilized by growth inside the grain where gas atoms are highly super-saturated.

When the bubbles are nucleated and stabilized by the localized short range movement of the gas atoms in the displacement spikes at low temperature, bubbles would be rather uniformly distributed and their growth would be limited to a certain extent, which can be seen in the microstructure of the HBS region. Bubble nucleation by fission fragments in the HBS region has also been proposed by Lemekhov [10] and Kinoshita [12]. However, Lemekhov indicated that as the temperature increases, HBS would form at lower burnup, which as will be explained later, is in opposition to this work.

If there is not enough supply of vacancy compared with that of gas atoms into the bubbles at low temperature, the bubbles may be over-pressurized to cause stress to the matrix. It was suggested [4] that over-pressurization itself of the bubble in HBS region would not be enough to cause the grain subdivision. However, considering the fact that grain subdivision is shown quite localized around bubbles during HBS development, surface of the bubbles could be the starting point of the grain subdivision, and grain subdivision could be initiated and enhanced by both the radiation damage such as dislocation network [13] and the stress caused by the bubble over-pressure. Once the grain is subdivided, diffusion of the gas atoms inside the grain into the bubbles may be enhanced through the newly formed subgrain boundary and therefore, the bubble over-pressurization and grain subdivision might be accelerated by the feedback. Then, the gas atom concentrations inside the very small subgrain with a 0.1–0.5 μm diameter could be depleted, as suggested by Spino et al. [4,5].

There exist currently two opposite approaches to explain the HBS development. One is that grain subdivision occurs first by the radiation damage such as dislocation network and bubble formation follows the grain subdivision [8,9]. The other is that the bubble

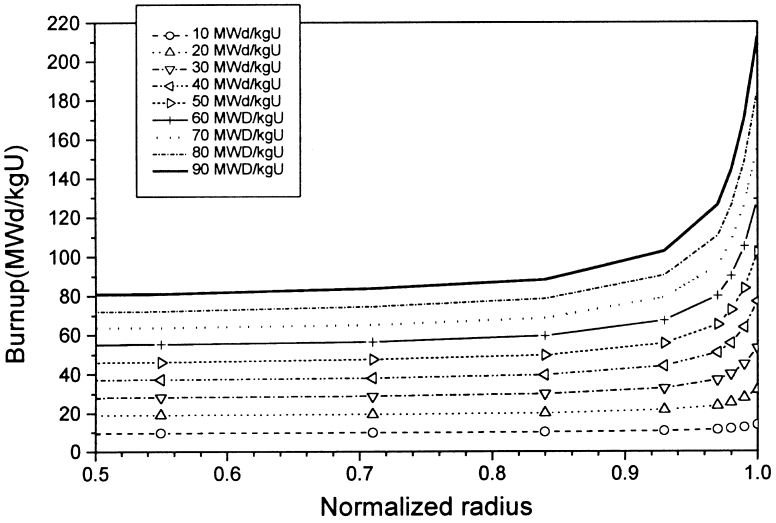

Fig. 7. Radial burnup distribution calculated by RAPID program.

formation comes first and then grain subdivision follows it [4,5]. The model proposed in this work belongs to the latter. Many parameters involved in HBS development have been found, such as radiation damage, bubble formation and grain subdivision which could be interdependent. However, it would be still quite necessary to find the primary driving parameter of HBS development.

Based upon the above proposed mechanism that the bubbles in the HBS are formed at the critical concentration of gas atoms inside the grain, HBS initiation, that is, bubble nucleation and stabilization depends upon such parameters as temperature, grain size and fission rate, etc. by which the diffusion coefficient of gas atoms is affected. As the temperature increases, the burnup at which the critical concentration is reached is increased due to the diffusional release of gas atoms to the grain boundary, as shown in Fig. 5. When the temperature is high enough, the critical concentration inside the grain would be never reached due to the diffusion release of gas atoms into the grain boundary.

As the fission rate increases, the critical concentration is reached earlier due to the less time available for diffusion. It is interesting that as the grain size increases, the critical concentration for HBS initiation is reached at lower burnup due to less fractional diffusion of the gas atoms to the grain boundary. It may also be probable that grain subdivision is more easily initiated from the

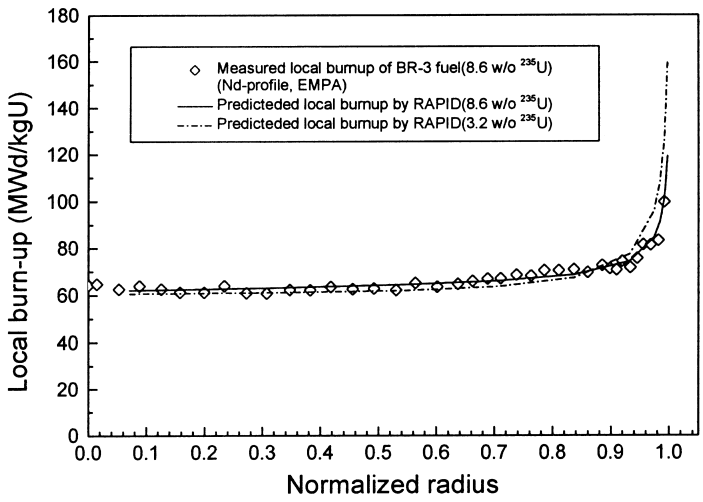

Fig. 8. Radial burnup distribution for different ^{235}U enrichments [5].

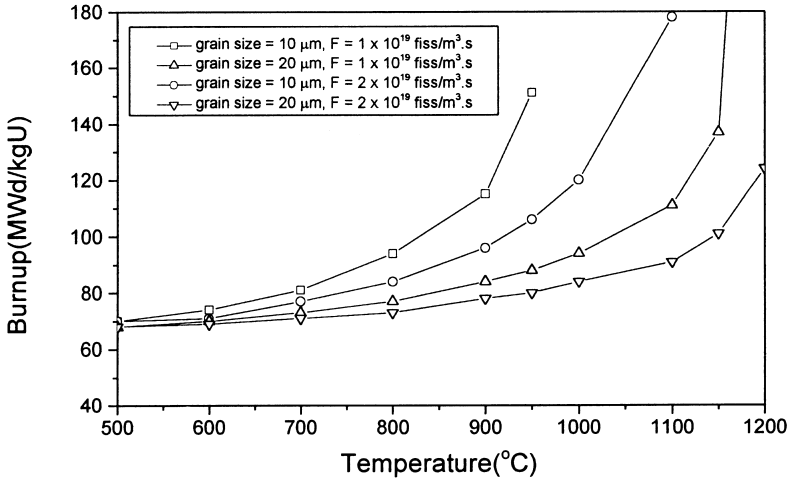

Fig. 9. HBS initiation local burnup as a function of temperature, grain size and fission rate.

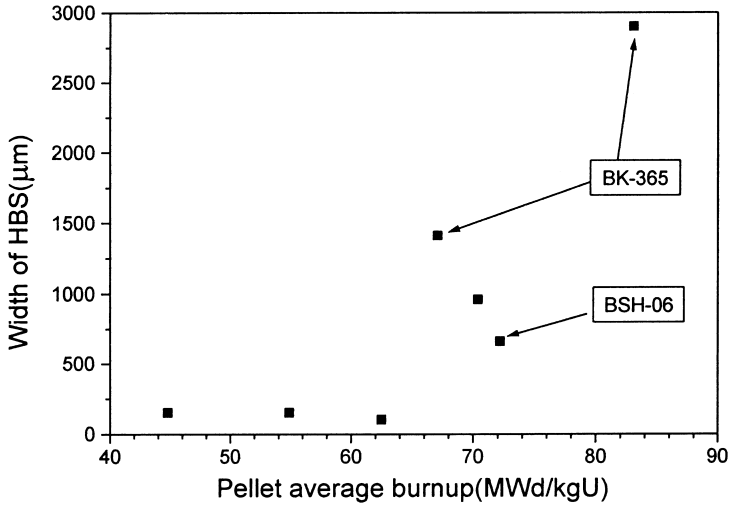

Fig. 10. Measured data of HBS width in the HBEP irradiation tests [6].

external surfaces and the grain boundary, so that grain subdivision may proceed more slowly for the larger grain, as investigated by Nogita et al. [9]. Therefore, the final effect of the grain size on the HBS development seems to need further investigation.

Fig. 7 shows the radial burnup distribution for 4 w/o ^{235}U fuel calculated by the RAPID program [25]. At the pellet average burnup of 60 MWd/kgU, it can be seen that HBS is formed at the width of ~100 μm when the critical concentration of the gas atoms is assumed to be 5.1×10^{26} atoms/m^3 or a burnup equivalent of 70 MWd/kgU [22], and HBS starts to form at the pellet average burnup of 45 MWd/kgU. Fig. 8 shows both the measured and predicted radial burnup distributions for fuels of different ^{235}U enrichments of 8.6 w/o and 3.2 w/o. It can be seen that local peaking of the burnup at the pellet periphery decreases with the increase of ^{235}U enrichment. Therefore, HBS formation in the highly enriched fuel will occur at a higher pellet average burnup than the lowly enriched fuel.

Fig. 9 shows the local burnup for HBS initiation as a function of temperature, grain size and fission rate for the 4 w/o ^{235}U fuel under the assumption that HBS forms at the critical gas concentration of 5.1×10^{26} atoms/m^3. At temperatures less than 700°C, HBS forms at a local burnup of less than 80 MWd/kgU, while at

temperatures higher than 1000°C, HBS would never develop due to the diffusional release of the gas atoms. It can be seen that as the grain size or the fission rate increases, HBS develops at a lower local burnup.

In summary, according to the proposed mechanism of HBS formation, as the fuel temperature increases, as the fission rate decreases, as the ^{235}U enrichment increases, and as the grain size decreases, initiation of HBS could be discouraged.

In the high burnup experiment program (HBEP) fuel irradiation tests [1,6], there were two fuel rods, where one (BSH-06) was irradiated at high power and the other (BK-365) was irradiated at low power. The BK-365 rod was irradiated at a power less than 220 W/cm during its whole lifetime, while the BSH-06 rod was irradiated at a power higher than 300 W/cm for over 400 days. The burnup and fractional fission gas release of the BK-365 rod were 69.4 MWd/kgU-rod avg. and 3.8%, while those of the BSH-06 rod were 59.8 MWd/kgU-rod avg. and 8.4%, so that fission gas release of the BSH-06 rod is much higher than the BK-365 rod. Fig. 10 shows the measured width of the HBEP test rods [6]. Linear dependence of the HBS width upon the local burnup cannot be found. The effect of temperature upon HBS formation, according to the proposed mechanism may explain the difference in the width of the HBS region, such as 1410 and 2900 μm at the pellet average burnups of 67.1 and 83.1 MWd/kgU, respectively, for the BK-365 rod and 660 μm at the pellet average burnup of 72.2 MWd/kgU for the BSH-06 rod. Fuel temperature of the BK-365 rod with annular pellet was below 800°C during most of the irradiation, while the BSH-06 rod was irradiated at a relatively high temperature [6]. Given above is a quite qualitative interpretation so that well-characterized irradiation tests, specially in terms of fission gas release and temperature would be needed to evaluate the proposed model.

Then, what will the effects of HBS formation on fuel performance be? It has been considered that due to the high porosity, the thermal conductivity in the HBS region would be reduced to increase the fuel temperature and, subsequently, the fission gas release. An irradiation test showed enhanced fission gas release at high burnup [26]. However, enhancement of the fission gas release at high burnup may be the inherent characteristic of the fission gas release, considering its mechanism. As shown in Fig. 2, the concentration of the gas atoms at the grain boundary would increase with the burnup, and so would the fraction of open bubbles. Therefore, the release probability of the fission gas would increase with the burnup under the same conditions. In addition, in-pile fuel temperature measurement in the Halden Reactor Project did not show any meaningful temperature increase by HBS formation up to the fuel burnup of 70 MWd/kgU, which should be observed if the thermal conductivity is decreased in the HBS region [27]. In addition, it was found [28] that the thermal conductivity of the irradiated fuel was increased after annealing at a high temperature above 1500°C which resulted in the formation of fission gas bubbles at the annealing of radiation defects on the thermal conductivity. This indicates that bubble formation and gas atom depletion in the matrix, which is similar to the microstructural characteristics of the HBS region might decrease the thermal conductivity due to the increased incoherence of the matrix at the grain boundaries. Therefore, the combination of bubble formation and grain subdivision would determine the thermal conductivity of the HBS region. Anyhow, it seems that the effects of HBS formation on fuel performance may not be so harmful as previously thought.

4. Conclusion

A formation mechanism of HBS or rim effect in UO_2 fuel was proposed. The bubbles in the HBS region may be nucleated and stabilized by fission fragments at the threshold atomic concentration of the gas atoms inside the grain. HBS initiation was driven not by the conventional thermal diffusion process, which should increase with the temperature, but by the cascade of atomic movements in the displacement spikes along the tracks of the fission fragments. Based upon the above mechanism, the threshold local burnup for HBS formation was derived as a function of temperature, fission rate and grain size, which showed that the local burnup for HBS initiation increased with temperature and at a high temperature, HBS would never occur due to the fission gas release. The proposed mechanism could explain variation of the HBS widths among different irradiation tests through the effect of the temperature and ^{235}U enrichment on HBS formation, and it is consistent with the finding [5] that bubble formation is followed by grain subdivision in the HBS region.

Acknowledgements

This work has been carried out under the Nuclear R&D Program supported by Ministry of Science and Technology in Korea. The authors would like to thank J. Spino and J. Rest for the critical reading and comments of the manuscript.

References

[1] M.E. Cunningham, M.D. Freshley, D.D. Lanning, J. Nucl. Mater. 188 (1992) 19.
[2] K. Une, K. Nogita, S. Kashibe, M. Imamura, J. Nucl. Mater. 188 (1992) 65.

[3] Hj. Matzke, M. Kinoshita, J. Nucl. Mater. 247 (1997) 108.
[4] J. Spino, K. Vennix, M. Coquerelle, J. Nucl. Mater. 231 (1996) 179.
[5] J. Spino, D. Baron, M. Coquerelle, A.D. Stalios, J. Nucl. Mater. 256 (1998) 189.
[6] M. Mogensen, J.H. Pearce, C.T. Walker, J. Nucl. Mater. 264 (1999) 99.
[7] L.E. Thomas, C.E. Beyer, L.A. Charlot, J. Nucl. Mater. 188 (1992) 80.
[8] K. Nogita, K. Une, J. Nucl. Mater. 226 (1995) 302.
[9] K. Nogita, et al., J. Nucl. Mater. 248 (1997) 196.
[10] S.E. Lemekhov, Proc. ANS Top. Meeting on LWR Fuel Performance, Florida, 1994.
[11] Hj. Matzke, M. Kinoshita, J. Nucl. Mater. 247 (1997) 247.
[12] M. Kinoshita, J. Nucl. Mater. 248 (1997) 185.
[13] J. Rest, J. Nucl. Mater. 210 (1994) 187.
[14] M.V. Speight, Nucl. Sci. Eng. 37 (1969) 180.
[15] A.H. Booth, AECL-496, AECL, 1957.
[16] L.C. Bernard, P. Blanpain, E. Bonnaud, E. van Schel, in: Proceedings of Enlarged Halden Program Group Meeting, HPR-349, Norway, 1998.
[17] M. Hirai, et al., Proc. ANS Top Meeting on LWR Fuel Performance, Oregon, 1997.
[18] I. Matsson, H. Teshima, in: Proceedings of Enlarged Halden Program Group Meeting, HPR-349, Norway, 1998.
[19] I. Matsson, in: Proceedings of Enlarged Halden Program Group Meeting, HPR-349, Norway, 1998.
[20] D. Baron, B. Hermitte, J.P. Piron, in: Proceedings of the IAEA Technical Committee Meeting on Advances in Fuel Pellet Technology for Improved Performance at High Burnup, Tokyo, 1996, p. 185.
[21] J.A. Turnbull, C.A. Friskney, J.R. Findlay, F.A. Johnson, A.J. Walter, J. Nucl. Mater. 107 (1982) 168.
[22] K. Lassmann, C.T. Walker, J. Van der Laar, F. Lindstrom, J. Nucl. Mater. 226 (1995) 1.
[23] J.A. Turnbull, J. Nucl. Mater. 38 (1971) 203.
[24] D.R. Olander, Fundamental aspects of nuclear reactor fuel elements, ERDA, 1976.
[25] C.B. Lee, et al., KAERI/TR-1217/99, KAERI, 1999.
[26] R. Manzel, M. Coquerelle, Proc. ANS Top Meeting on LWR Fuel Performance, Oregon, 1997.
[27] H. Devold, in: Proceedings of Enlarged Halden Program Group Meeting, HPR-349, Norway, 1998.
[28] M. Lippens, L. Mertens, EPRI TR-106501, 1996.