Pressure Calculation of Intergranular Bubbles in UO_2

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1. Introduction

The nuclear fuel of the light water reactor consists of zirconium cladding tubes and polycrystalline UO_2 fuel pellets. About 15% of fission products generated during operation are inert gas atoms such as krypton and xenon. This inert gas atom has very low solubility in UO_2 and diffuses to the grain boundary from inside the matrix. [1] The fission gas diffused into the grain boundary forms an intergranular bubble, which has a lenticular shape due to the difference between the grain boundary energy of UO_2 and the free surface energy. The grain boundary bubbles grow and connect to each other, and fission gas is released to the outside of the pellet through this tunnel. [2,3] The formation of fission gas bubbles is the main cause of nuclear fuel swelling, and fuel pulverization due to rupture of grain boundaries caused by over-pressurization of intergranular bubbles during the transient accident is also being actively studied. [4–10]. In order to accurately model these phenomena, it is necessary to obtain pressure information inside the intergranular fission gas bubble. Since the size of the intergranular bubbles generated in the UO_2 under normal operation is very small and the resulting pressure is high, direct bubble pressure measurement is impossible. Therefore, in most studies, the amount of fission gas in a bubble is measured through energy dispersive X-ray spectroscopy (EDX), secondary ion mass spectrometry (SIMS), and electron probe microanalysis (EPMA) or it is calculated using a burnup and diffusion model. Nogita et al. measured the pressure of the intragranular bubble using TEM-EDX. [11] Cagna et al. measured the under surface bubble pressure using SEM-SIMS-EPMA. [12] Noirot et al. and Walker et al. also measured the amount of fission gas and the size of the fission gas bubble inside the fission gas bubble and calculated the pressure. [13,14] The pressure value derived from this experiment has a much higher value than the capillary pressure expressed as below equation.

$$P = \frac{2\gamma}{r} \tag{1}$$

Gao et al. calculated the pressure of the intergranular bubble in a high burnup structure without experiments based on the equations of state of xenon, the empirical formulae for the burnup distribution along the radial direction and, the fission gas behavior according to the burnup. [15] This study showed that the pressure of the fission gas bubble existing in the high burnup structure was about 40 times higher when compared with the capillary pressure of the bubble. As the experiments and calculation results show, the fission gas bubble is over-pressured, with pressure much higher than the capillary pressure. However, there is a lack of research on how over-pressurized intergranular fission gas bubbles in areas where recrystallization has not occurred. If over-pressurization of these intergranular fission gas bubbles is well understood according to the local burnup of nuclear fuel or the bubble size, it can be very helpful in fuel swelling or pulverization modeling research. In this study, a study was conducted on how the pressure changes according to the size of local burnup and intergranular bubbles in nuclear fuel without recrystallization and investigated how over-pressurized these bubbles are compared to the capillary pressure.

2. Method

In order to calculate the intergranular bubble pressure, it is necessary to know the amount, volume, and temperature of fission gas. Gao et al. calculated the pressure of bubbles by assuming that the bubble in the high burnup structure has a size of 0.2 to 2 μm, and the temperature is 800 K, regardless of the radial direction and burnup. However, in the area where recrystallization has not occurred, the temperature gradient is very large in the normal operation state along the radial direction, and the bubble size accordingly is also greatly different, so the bubble size and temperature cannot be assumed. Therefore, it is necessary to obtain temperature and bubble size information based on microstructure analysis of actual nuclear fuel grain boundary.

White et al. conducted microstructure analysis on intergranular bubbles present in the grains of nuclear fuel pellets of about 21 GWd/tU. [16] A total of 9 pellets were divided into 4 to 6 in each radial direction for microstructure analysis, and the bubble size and density analysis and the output history of each nuclear fuel pellet were contained in the NEA report. [17] The temperature information of each part where microstructure analysis took place was also calculated using the ENIGMA code.

The amount of fission gas in the grain boundary was calculated using temperature information and power history information. The modified Forsberg-Massih model was used for calculation following the gas diffusion equation. [18]

$$\frac{dC}{dt} = D(t)\Delta_r C(r,t) + \beta(t) \qquad (2)$$

C = gas concentration
β = gas production
$\Delta_r = \frac{d^2}{dr^2} + \frac{2}{r}\left(\frac{d}{dr}\right)$
D = diffusion constant
t = time

The modified Forsberg-Massih model also considers diffusion from the grain boundary to the grain, so the following boundary conditions are used.

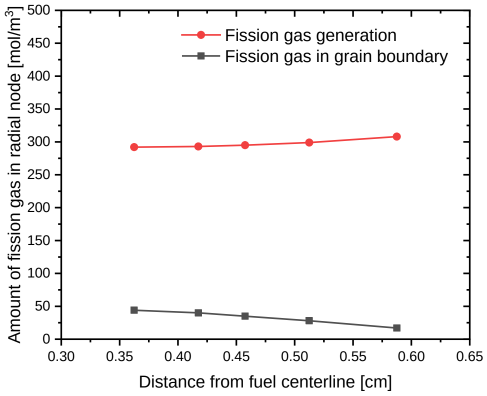
$$C(r,0) = 0$$
$$C(a,t) = \frac{b(t)\lambda N(t)}{2D} \qquad (3)$$

N = surface gas concentration
λ = resolution layer depth
a = hypothetical grain radius
b = resolution rate

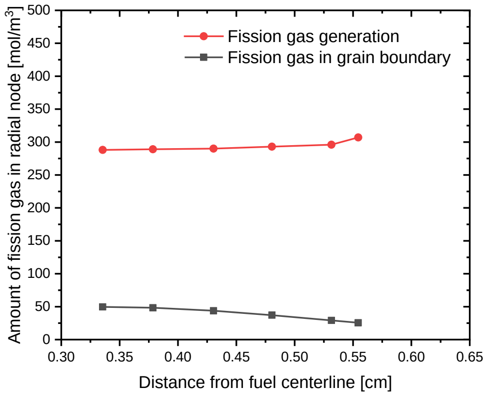
It is necessary to use an appropriate equation of state for fission gas for pressure calculation. Nogita et al. used Ronchi's equation of state to calculate the intragranular fission gas bubble pressure. [11,19] Cagna et al. used Soave's equation of state for pressure calculation. [12,20] Noirot et al. used the Van der Waals equation, and Walker et al. used the equation of state of Brearley et al. [13,21,22] Gao et al. used Ronchi's equation of state to calculate the intergranular bubble pressure of a high burnup structure. [15,19] In this study, the pressure was calculated and compared using the Ideal gas law and Van der Waals equation, as well as Ronchi's equation of state and Xiao's equation of state. [19,23] The calculated pressure was compared with the capillary pressure according to local burnup and local temperature.

3. Result

Figure. 1 is a graph showing the amount of fission gas generated and accumulated in the grains of each nuclear fuel calculated by the modified Forsberg-Massih model. Because of the annular fuel design, there is a hole from the center to a radius of about 0.3 cm, so the data is empty in that part. Although the burnup is higher in outside part of the nuclear fuel and the fission gas production is high, the diffusion from the grain inside to the boundary is smaller than the center part of the nuclear fuel due to the relatively low temperature, so the amount of fission gas in grain boundary tends to decrease overall toward the outside of the nuclear fuel.



(a)



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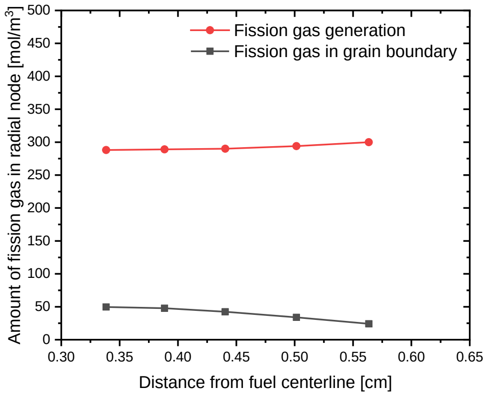
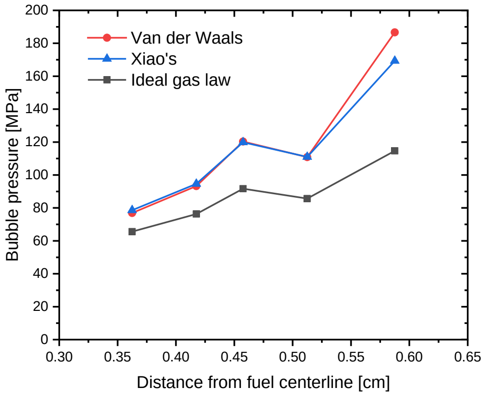


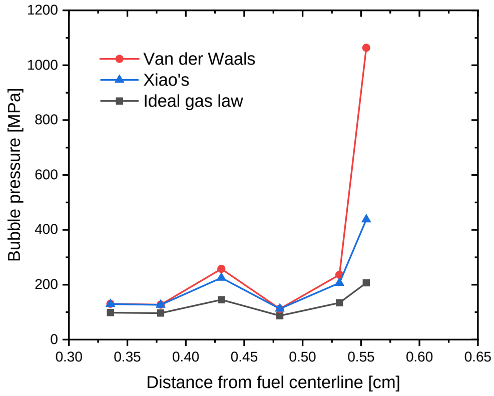
(c)

Figure. 1 Calculated amount of generated fission gas and accumulated in grain boundary. Pellet 4000 (a), pellet 4004 (b), pellet 4005 (c).

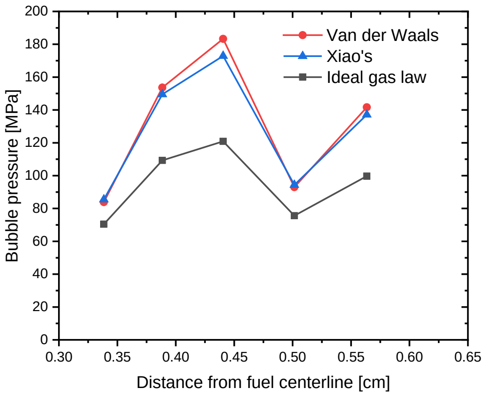
Based on the calculated amount of fission gas present in the grain boundary, the internal pressure of the intergraunlar bubble of each part was calculated using three different equations of state. Figure. 2 is a graph showing the pressure calculated by each equation of state.



(a)



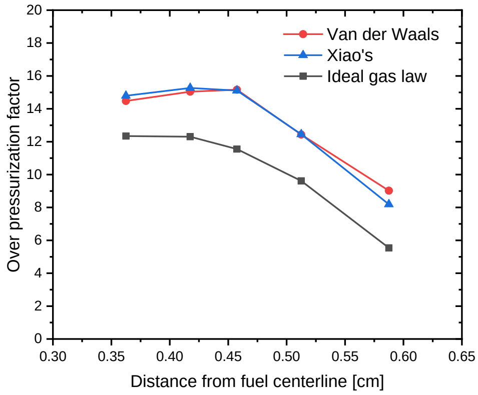
(b)



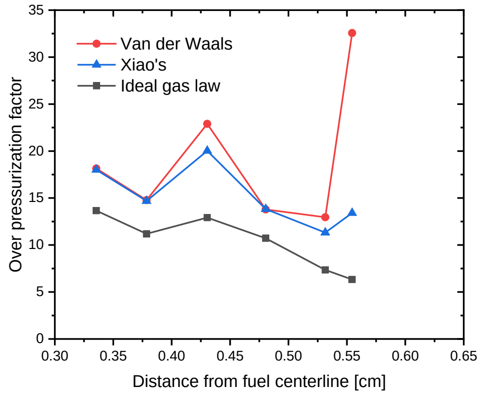
(c)

Figure. 2 Calculated pressure of intergranular bubble. Pellet 4000 (a), pellet 4004 (b), pellet 4005 (c).

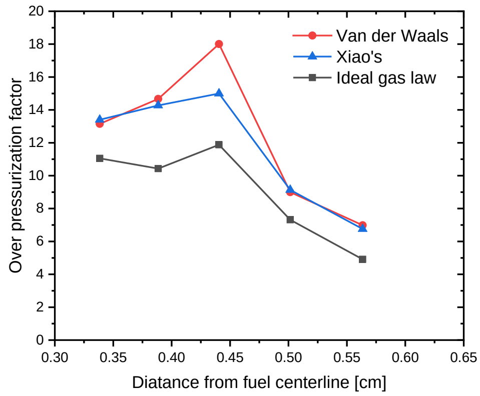
The Van der Waals equation tends to calculate the pressure very large when there are a large number of gas atoms in a small volume. This is why the pressure at the outermost radial node of the 4004 pellet was calculated to be over 1 GPa. Xiao's equation, a newly developed equation of state using molecular dynamics, is gaining the most credit. Based on the calculated pressures, it was determined how much higher pressures these pressures had when compared to the capillary pressure. It was calculated how many times the calculated pressure has a value compared to the capillary pressure.



(a)



(b)



(c)

Figure. 3 Over pressurization factor. Pellet 4000 (a), pellet 4004 (b), pellet 4005 (c).

Except for the outermost radial node of 4004 pellets calculated by Van der Waals, the over pressurization factor tends to decrease as it goes to the outside of nuclear fuel. Basically, the reason for over pressurization is that the rate at which the fission gas diffuses into the intergranular bubble is faster than the rate at which the bubble expands due to the pressure driven diffusion of UO_2 outward by the bubble pressure. This difference in diffusion rate will of course be larger as the temperature increases, and thus the over-pressurization will be greater in the hot area. Therefore, it has a larger over-pressurization factor in the center of the fuel.

4. Acknowledgement

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