

HELIUM MIGRATION ALONG STRESS GRADIENTS IN FUSION REACTOR STRUCTURES

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ABSTRACT

Compared with fast breeder reactors (FBR), fusion reactor environments result in much higher helium production rates. Helium has significant impact on the evolution of microstructure of irradiated materials. Bubble migration along stress gradients in FBR fuel pins was estimated to be negligible (nearly 1% of the force due to temperature gradient). The large stress levels experienced by the first wall of a typical fusion design drives us to examine the importance of stress gradients in driving bubble motions in the fusion structures. Boundary integral method formulated for time dependent plastic deformation in one-dimension was used. Major changes in the forces affecting helium bubbles due to stress gradients were noticed in the first few millimeters of a 5 cm 316 SS structure placed in a fusion environment (dpa rate of $1 \cdot 10^{-7}$ dpa/sec for 10 years). Calculated force were nearly two orders of magnitude higher than that experienced in fast reactors. High dpa rates increase the force affecting gas bubbles especially at the first few millimeters on the structure surface due to the increase in swelling level with its impact on the stress level there. Forces differed by four orders of magnitudes as the bubble radius vary from 20 to 500 Å (i.e., 2 to 50 nm). velocity of a 100 Å gas bubble was estimated to be 0.002 Å/sec. Such a bubble would move 6.3 μm in a year. The impact of stress gradient on driving helium bubble migration is an important phenomena in analyzing the behavior of fusion reactor first wall structures.

Keywords: Fusion reactors, Irradiation behavior, Helium migration, Stress gradients.

1. INTRODUCTION

When materials are irradiated by high energy neutrons, the displacement damage is accompanied by the production of impurity atoms due to nuclear transmutation reactions. Attention has been focused on transmutant helium since mid-sixties [1] due to the significant impact that helium is known to have on the microstructure of irradiated materials [2] and the fact that the differences between the neutron spectra obtained in a fast fission reactor and in a deuterium-fusion reactor result in much higher helium production rates in the latter. For example, the ratio of transmutant helium to displacement production (He/dpa ratio) will be on the order of 10 to 20 appm He/dpa in the first wall of a fusion reactor compared with 0.3 to 0.5 appm He/dpa in a typical fast fission test reactor. Typical helium production rates in Fast Reactors are 5-15 ppm/yr. while they are of the order of 50-300 ppm/yr. in fusion devices [1]. This larger quantities of helium

will alter the temperature and fluence dependence of microstructural evolution, swelling, and mechanical property changes [3]. It is of interest to note that typical dimensions of helium gas bubbles in metals are of the order of 1-100 nm with typical number of helium atoms of $50 \cdot 10^6$ atoms [1].

The observation by Barnes and Mazey [4] in 1963 that helium filled bubbles within solid copper foils could, under the appropriate conditions, move bodily through that material has led to many subsequent studies of such phenomena, both experimental and theoretical. There are different types of potential gradients which can cause inclusions (e.g., gas bubbles) to migrate in solids. Table (1) lists these potentials as well as the different mechanisms by which the transport of atoms of the solid from one part of the surface or interface of the inclusion to another part takes place [5].

Table 1. Potential gradients and the atomic mechanisms for inclusion migration in solids [5].

Potential gradient in the host solid	Atomic mechanism of solid atom transport
(1) Temperature	(A) Vacancy diffusion in the host solid
(2) Stress	(B) Surface diffusion along the interphase boundary
(3) Electric field	(C) Volume diffusion in the interior of the inclusion
(4) Accelerational field (e.g. gravity)	
(5) Vacancy concentration in the solid	

For ceramic fuels used in Liquid Metal Fast Breeder Reactors (LMFBR), owing to their relatively poor thermal conductivity, steep temperature gradients of the order of 1 to $3 \cdot 10^3$ °C/cm may develop in such materials during operation [6]. Thus migration of fission-gas bubbles (typically 0.5 to 5 μ) up the thermal gradient in the columnar-grain region of the cylindrical fuel elements is a very likely mechanism for gas release in this region [7] which gives the phenomena of bubble migration a technological importance.

As Olander [5] pointed out, there have been no measurements of migration of inclusions (i.e., helium bubbles in our case) in a stress gradient. On the other hand, there is no shortage of theoretical speculation concerning the magnitude and direction of this phenomenon. Bubble migration along stress fields was investigated for fast reactor fuel pins [8]. Due to the low stress levels experienced (as shown in Figure 1), it was showed that the stress gradient exerts a force on the bubble that is only about 1% of that due to the temperature gradient. Thus, at least in fast reactor fuels, stress gradient migration does not appear to be an important means of gas bubble motion within bulk fuel material, but in regions of locally high stress gradients, this force can be of significance.

The large difference between stress levels in a typical high power LMFBR fuel pin (as was shown

in Figure 1) and that experienced by the first wall of a typical fusion design (Figure 2) points to reexamine the effect of stress gradients in driving bubble motions in the fusion structures. This is specially true if we take into consideration that the temperature gradients in these structures are lower than those experienced in fast reactor fuel pins (nearly two orders of magnitude less) which may reverse the order of significance between the stress and temperature gradients. Taking into consideration that fusion reactor structures will be subjected to complicated stress histories makes the investigation of each structure with its own stress history a point that deserves careful attention. As would be explained later, since bubbles move down the stress gradient, it is worth investigation whether migration along stress gradients will be significant enough to cause release of certain gas bubbles (helium and/or tritium for example) if they move to free surfaces.

The above argument gains more support in case of the bonded structures in the fusion reactors. It is known that the bonding technique leads to complicated stress fields which must be understood in order to evaluate the probability of failure for the device [1]. According to stress distribution in a Bonded structure of a typical DEMO fusion design [10], and since bubbles move down the stress gradient, this would mean that bubbles would migrate to the interfacial region with the

consequences on coating adhesion. This behavior should be evaluated as a function of component life time and in the relevant stress environment. The important point to stress is that these calculations have to be performed in the relevant stress environment for each structure, not just because of the varying stress level as a function of time but also because of the change in the stress gradient with time. What needs to be stressed is that while radiation-effects data on (as well as modeling for) individual fundamental phenomena can be correlated to structural effects, the uncertainty in synergistic effects (heat, high dpa, and stress) could jeopardize the success of the final design [2].

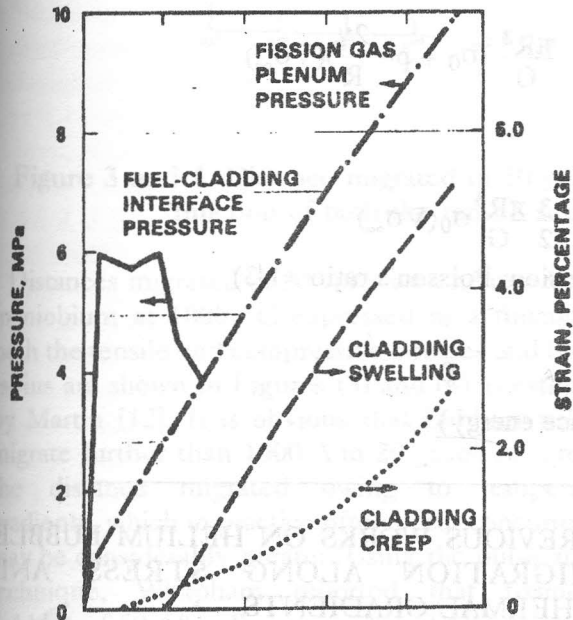


Figure 1. Calculated pressures of a high power LMFBF fuel pint to design burnup [8].

2. MECHANISMS OF HELIUM MIGRATION ALONG STRESS GRADIENTS

Calculation of gas bubble migration in stress and temperature gradients can be divided into two classes, which can be labeled "global" and "local" [5]. The first one deals with the change in the system (bubble and medium) free energy while the second one deals with the distribution of the potential (temperature or stress) on the bubble surface. Both the global and local methods have been applied to temperature-gradient induced bubble migration [5]

and the migration velocities computed by the two techniques are the same. In the case of a stress gradient, only the global method has been examined in the western literature with widely discordant results. In the Russian literature, the local method was used. Table (2) lists several expressions for the force on a bubble due to a stress gradient.

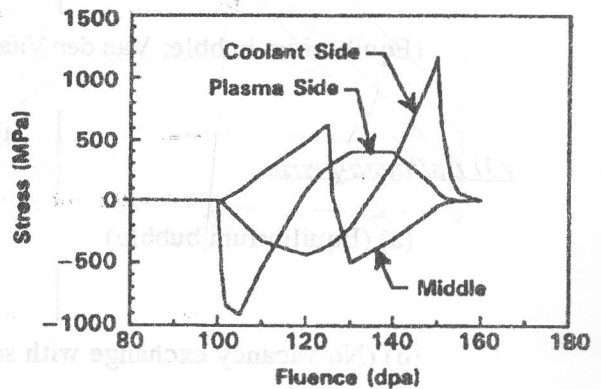


Figure 2. Stress due to swelling in a completely unconstrained plate of primary candidate alloy (PCA) with a through thickness temperature gradient from 400 to 550°C [9].

In Table (2), p is the pressure of gas in bubble, γ is the surface tension of solid, G is the shear modulus, K is the bulk modulus, R is the bubble radius, σ is the stress (positive in tension), and $\nabla\sigma_\infty$ is the stress gradient far from bubble.

It is worth mentioning that random migration of very small bubbles is theoretically much more important than thermal-gradient migration [7]. However, it should be pointed out that random migration has a dominant effect at lower temperatures whereas at higher temperatures biased motion is a dominating influence [7].

A discussion on bubble migration can not be completed without considering the restraining forces on the bubble motion and their coalescence as well. It is well known that structural heterogeneities will apply restraining forces to the bubbles and act as potential trapping centers. Whether or not bubble migration occurs depends on the number of traps present compared to the number of bubbles and the relative magnitudes of the driving forces. Among the restraining forces are the following [6]:

Table 2. Force on a gas bubble in a solid with a stress gradient [5].

<p>(1) <i>Eshelby</i> (Poisson's ratio = 1/3)</p>	$\frac{3}{2} \frac{\pi R^3}{G} \sigma_0 (\nabla \sigma_{\infty})$
<p>(2) <i>Martin</i> (Equilibrium bubble; Van der Waals correction neglected)</p>	$-\frac{4\pi R^4 \sigma_0 (\nabla \sigma_{\infty})}{4\gamma - 3\sigma_0 R} + \frac{2\pi R^3}{3K} \left(\frac{8\gamma - 3\sigma_0 R}{4\gamma - 3\sigma_0 R} \right) \sigma_0 (\nabla \sigma_{\infty})$
<p>(3) <i>Bullough-perrin:</i> (a) (Equilibrium bubble)</p>	$-\frac{4\pi R^4 \sigma_0 (\nabla \sigma_{\infty})}{4\gamma - 3\sigma_0 R} - \frac{2\pi R^3}{3K} \left(\frac{8\gamma - 3\sigma_0 R}{4\gamma - 3\sigma_0 R} \right) \sigma_0 (\nabla \sigma_{\infty})$
<p>(b) (No vacancy exchange with solid)</p>	$\frac{\pi R^3}{G} \left(\sigma_0 + p - \frac{2\gamma}{R} \right) (\nabla \sigma_{\infty})$
<p>(4) <i>Nichols-Leiden</i> (Equilibrium bubble; gradient of uniaxial tension; Poisson's ratio = 1/3)</p>	$\frac{3}{2} \frac{\pi R^3}{G} \sigma_0 (\nabla \sigma_{\infty})$
<p>(5) <i>Speight</i> (Equilibrium bubble; surface tension = surface energy)</p>	0

- (1) Forces when bubbles intersect a dislocation or grain boundary. In the case of dislocation, account should also be taken of the dislocation stress field which results in an induced interaction with bubbles.
- (2) Restraining forces due to the existence of precipitates. If there is a stress field around the precipitates there should also be an induced interaction as discussed in connection with the dislocation.
- (3) Point defects generated during irradiation will also induce restraining forces.

Two comments are worth mentioning [6]: (1) Driving forces are considerable more dependent on bubble size than the restraining forces, and (2) Traps have different strengths as measured by the restraining forces they exert on the bubbles.

3. PREVIOUS WORKS ON HELIUM BUBBLE MIGRATION ALONG STRESS AND THERMAL GRADIENTS

The force on a bubble in a stress gradient has been determined by Bullough and Perrin [11], Martin [12], Eyre and Bullough [6], and Leiden and Nichols [13]. None of these works considered the fusion reactor environment with the evolution of stress and stress gradients with time. Following is only representative examples of the forces and velocities expected in certain nuclear application environments as determined both theoretically and experimentally in order to give an appreciation of the magnitude of the forces involved and the corresponding acquired bubble velocities.

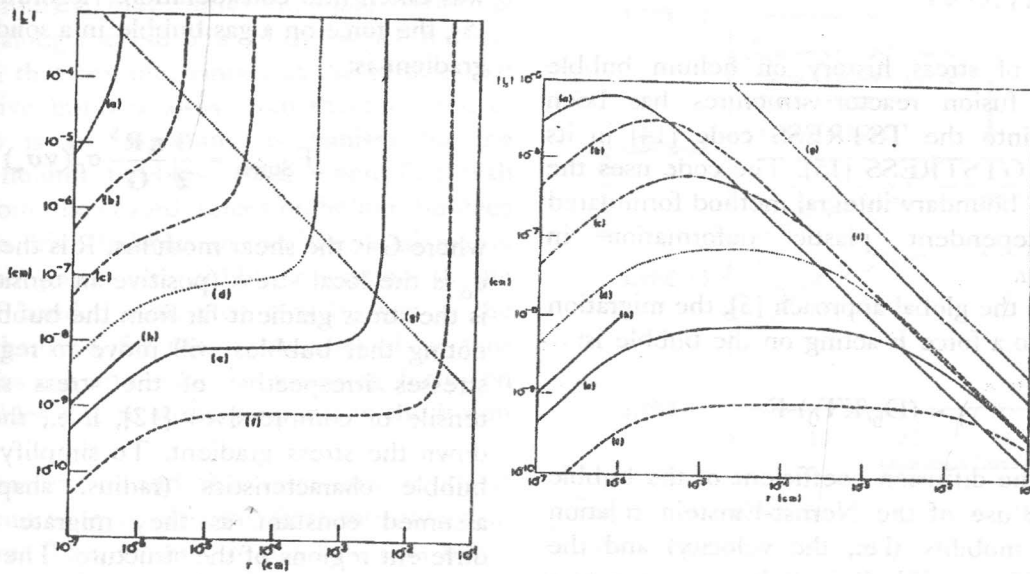


Figure 3 and 4. Distance migrated in 20 years by a helium bubble in niobium at 100°C expressed as a function of both the tensile and compressive stresses and bubble radius [12].

Distances migrated in 20 years by a helium bubble in niobium at 1000 °C expressed as a function of both the tensile and compressive stresses and bubble radius are shown in Figures (3) and (4) as estimated by Martin [12]. It is obvious that bubbles will not migrate further than 1000 Å in 20 years. As a result, the distance migrated owing to temperature gradients, which in practice often occur concurrently, may be considerably greater. Using the pulse-anneal technique, Whapham reported that fission-gas bubbles of 50 Å radius in irradiated UO₂ moved at a rate of about 10³ Å/sec in a temperature gradient estimated at 10⁶ °C/cm and an estimated temperature of 1600 °C [4]. Leiden and Nichols [13] estimated the force due to stress gradient of 2*10⁹ dynes/cm³ in a material where $\nu=0.3$ and $E=2.6*10^7$ psi (~1.5*10¹² dyne/cm²). The bubble radius was assumed to be 100 Å and the bubble was assumed to move a distance of roughly one micron under the influence of the stress gradient. The force is estimated to be 1.6*10⁻⁸ dyne. Velocities of fission gas bubbles in columnar grains of a typical fast reactor fuel pin are shown in Figure (5) [7].

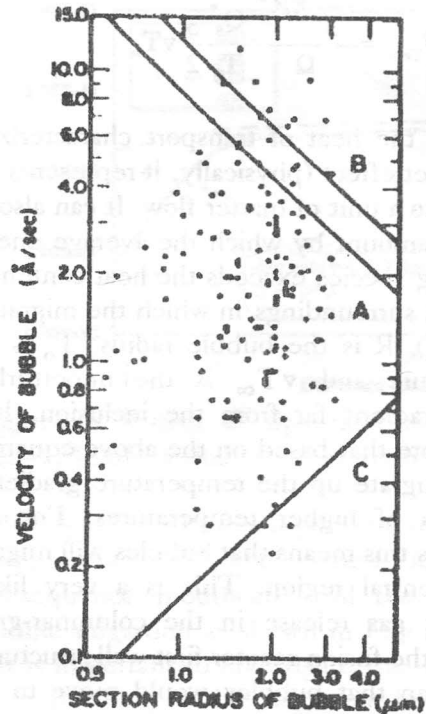


Figure 5. Velocities of fission gas bubbles in columnar grains of a typical fast reactor fuel pin [7].

4. CALCULATIONS

The effect of stress history on helium bubble migration in fusion reactor structures has been incorporated into the TSTRESS code [14] in its latest version GTSTRESS [15]. The code uses the method of the boundary integral method formulated for time dependent plastic deformation in one-dimension.

According to the global approach [5], the migration velocity due to a force F acting on the bubble is:

$$v_1 = (D_b/KT_0) F$$

where D_b is the diffusion coefficient of the bubble and we made use of the Nernst-Einstein relation between the mobility (i.e., the velocity) and the diffusion coefficient [7]. K is Boltzmann constant ($1.3805 \cdot 10^{-23}$ J/k) and T_0 is the local temperature.

For a nonconduction inclusion (which is the case for gas bubbles) in a temperature gradient, the force is [5]:

$$F_{\text{Temp}} = \frac{P_4}{\Omega} \frac{\pi R^3}{T_0} \frac{Q_s^*}{2} \nabla T_\infty$$

where Q_s^* is the heat of transport characterizing the surface Soret effect (physically, it represents the heat flow due to a unit of matter flow. It can also be viewed as the amount by which the average energy of the migrating species exceeds the heat content or enthalpy of the surroundings in which the migration takes place [7]), R is the bubble radius, T_0 is the local temperature and ∇T_∞ is the unperturbed temperature gradient far from the inclusion. It is important to note that based on the above equation, bubbles will migrate up the temperature gradients, i.e., to regions of higher temperatures. For fast reactor fuel pins this means that bubbles will migrate towards the central region. This is a very likely mechanism for gas release in the columnar-grain region [7]. For the fusion reactor first wall structures, this would mean that bubbles would move to the plasma side away from the coolant side.

To take helium bubble migration due to stress gradient into consideration, the simple expression for the force on the bubble as expressed by Eshelby

was taken into consideration. According to Eshelby [5], the force on a gas bubble in a solid with a stress gradient is:

$$F_{\text{Stress}} = \frac{3 \pi R^3}{2 G} \sigma_0 (\nabla \sigma_\infty)$$

where G is the shear modulus, R is the cavity radius, σ_0 is the local stress (positive in tension), and $\nabla \sigma_\infty$ is the stress gradient far from the bubble. It is worth noting that bubbles will move to regions of lower stresses irrespective of the stress sign (whether tensile or compressive) [12], i. e., they will move down the stress gradient. To simplify the analysis, bubble characteristics (radius, shape, etc.) are assumed constant as they migrate between the different regions of the structure. The stress history that is calculated is that due to both thermal as well radiation induced stress (due to swelling).

5. PARAMETRIC STUDY OF FORCE AFFECTING HELIUM BUBBLE MIGRATION DUE TO STRESS GRADIENTS

The structure considered is 20% cold-worked 316 stainless steel plate free to expand but not to bend (an ideal simulation for first wall fusion structures). It was subjected to different irradiation environment. Surface temperature was taken to be 525 °C, Back side temperature was taken to be 400 °C. Plate thickness was taken to be 5 cm. Membrane stress was considered to be zero. dpa rates were taken as: $5 \cdot 10^{-8}$, $1 \cdot 10^{-7}$, $5 \cdot 10^{-7}$, and $1 \cdot 10^{-6}$ dpa/sec respectively. Calculations were performed for 31.5 dpa (corresponding to 10 years of operation with a dpa rate of $1 \cdot 10^{-7}$ dpa/sec). It is important to note that helium production rate in SS 316 is 140-240 appm/year. The shear modulus of iron was calculated as a function of temperature. Bubble radii in the range 20-500 Å were considered.

The stress history as a function of time is depicted in Figure (6). The corresponding force evolution with time is shown in Figure (7). It is to be noted that forces would be directed towards the surface of the 316 SS structure (i. e., down the stress gradient). It is obvious that major changes in the forces affecting helium bubbles due to stress gradients are noticeable in the first few millimeters of the

structure. Forces may differ by one order of magnitude along the life time of the structure. It is well known that any mechanism at the surface that tends to drive bubbles away from the bulk and to the surface is an important mechanism for the release of helium bubbles. Also, when first wall structure would be coated, forces on helium bubbles would be to drive bubbles to the interfacial region stressing the point that bubble migration to the interface of bonded structures may be one of the failure mechanisms for these structures. It is thus quite obvious the importance of assessing forces on helium bubbles due to stress gradients in the fusion structures.

It is of interest to add that force due to stress gradients tend to level off away from the surface of the structure. This is due to the fact that major changes in stress levels (as well as stress gradients) with time occur at the surface due to high swelling there, while stress gradients and stress levels are slowly varying with time away from the surface.

It is important to compare the magnitude of the force due to stress in fusion and fast reactor fuel pin. According to Olander [8], force on gas bubbles in a typical LMFBR fuel pins due to stress gradient is around 2.04×10^{-15} N. It is apparent that force due to stress gradient in fusion structures is nearly two orders of magnitude higher than that experienced in fast reactors.

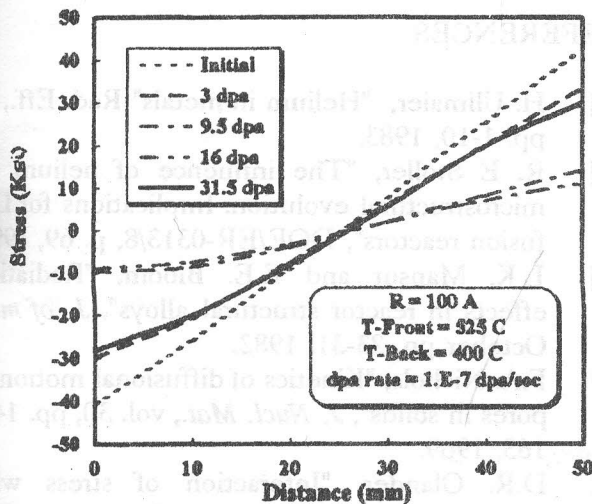


Figure 6. Stress evolution with time for the 316 SS structure.

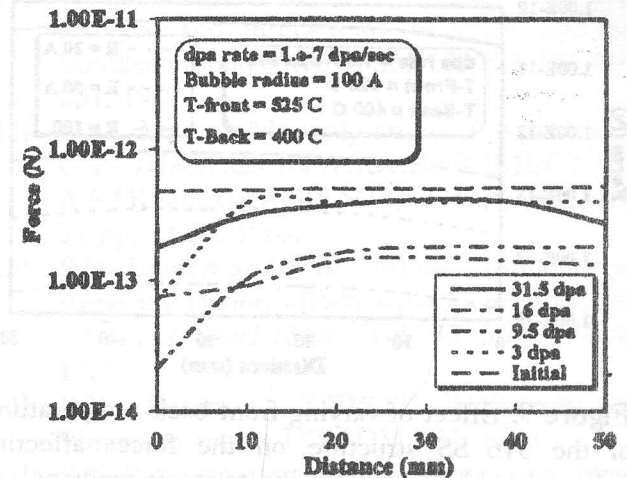


Figure 7. Evolution of force affecting helium bubble due to stress gradient.

As shown in Figure (8), for higher dpa rates, force affecting helium bubbles tend to be increased especially at the first few millimeters on the structure surface due to the increase in swelling level with its impact on the stress level there.

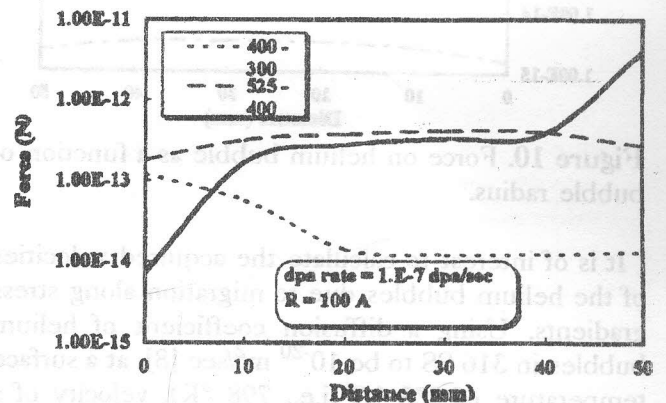


Figure 8. Effect of dpa rate on force affecting helium bubbles due to stress gradients.

As expected, as the temperature increases, thermal creep tends to relax stress levels with the consequence modification of the forces affecting bubble migration as shown in Figure (9).

It is important to investigate the magnitude of the force affecting bubble migration for different values of bubble radius. As shown in Figure (10), these forces may differ by four orders of magnitudes as the bubble radius vary from 20 to 500 A (i.e., 2 to 50 nm).

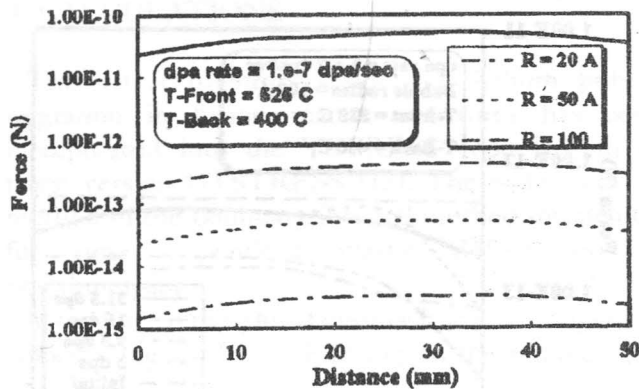


Figure 9. Effect of varying front-back temperatures of the 316 SS structure on the forces affecting helium bubble migration due to stress gradients.

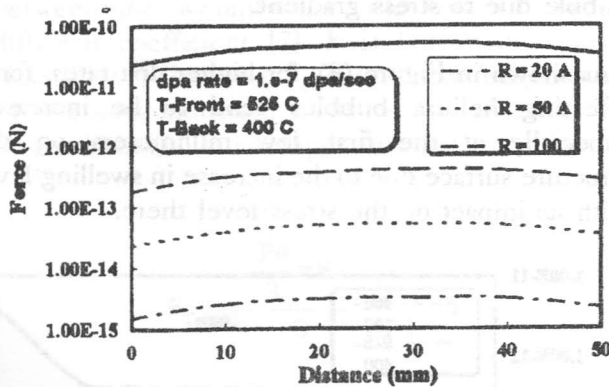


Figure 10. Force on helium bubble as a function of bubble radius.

It is of interest to calculate the acquired velocities of the helium bubbles due to migration along stress gradients. Using a diffusion coefficient of helium bubbles in 316 SS to be $10^{-20} \text{ m}^2/\text{sec}$ [8], at a surface temperature of $525 \text{ }^\circ\text{C}$ (i.e., $798 \text{ }^\circ\text{K}$), velocity of a 100 A gas bubble would be $0.002 \text{ A}/\text{sec}$. For a 500 A bubble, the velocity would be $0.2 \text{ A}/\text{sec}$. At these velocities, bubbles would move $6.3 \text{ }\mu\text{m}$ and $630 \text{ }\mu\text{m}$ in a year respectively. An important point to stress is that in the above calculations, trapping of bubbles was not considered. This is expected to slow them down as they would not migrate freely in the structure.

6. CONCLUSIONS

- (1) Major changes in the forces affecting helium bubbles due to stress gradients are noticeable in

the first few millimeters of a 5 cm 316 SS structure placed in a fusion environment. Forces may differ by one order of magnitude along the life time of the structure.

- (2) Force due to stress gradient in fusion structures is nearly two orders of magnitude higher than that experienced in fast reactors.
- (3) High due rates increase the force affecting gas bubbles especially at the first few millimeters on the structure surface due to the increase in swelling level with its impact on the stress level there.
- (4) As the temperature increases, thermal creep tends to relax stress levels with the consequence modification of the forces affecting bubble migration.
- (5) Forces may differ by four orders of magnitudes as the bubble radius vary from 20 to 500 A (i.e., 2 to 50 nm).
- (6) For a diffusion coefficient of helium bubbles in 316 SS of $10^{-20} \text{ m}^2/\text{sec}$ [8], at a surface temperature of $525 \text{ }^\circ\text{C}$, velocity of a 100 A gas bubble would be $0.002 \text{ A}/\text{sec}$. These bubbles would move 6.3 in a year. Trapping of bubbles is expected to slow them down as they would not migrate freely in the structure.
- (7) The impact of stress gradient on driving helium bubble migration is an important phenomena in analyzing the behavior of fusion reactor first wall structures.

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