Behavior of Helium Gas Bubbles in Neutron-Irradiated Beryllium

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Transmission electron microscopic observations were made on hot-rolled Be sheet prepared from cast ingots irradiated to about $5 \times 10^{19}$ neutrons (fast) in JRR-2. The aim of the study was to elucidate the effect of irradiation on changes in microstructure and the effect of pre-irradiation heat treatment on the formation of He gas bubbles during post-irradiation annealing.

The formation and growth of He bubbles during post-irradiation annealing was distinctly less inhibited in the Be irradiated as hot-rolled than in the metal irradiated after recrystallization treatment. In the former case, He bubbles could be observed upon post-irradiation treatment of only 3 hr at 750°C, while in the latter case, a similar formation of bubbles could only be seen after a final treatment at 900°C for 50 hr.

The difference thus observed in the tendency of bubble formation depends upon the grain boundary mobility during post-irradiation heat treatment.

The nucleation and growth of bubbles occurred preferentially at the grain boundaries, inclusions and dislocation sites. The recrystallization of irradiated Be was retarded by the interaction between dislocations and He atoms.

The inclusion which has the largest affinity to the bubbles in Be is Be$_3$Fe.

I. INTRODUCTION

It is commonly known that He atoms are produced by the fast neutron reactions $^9$Be$(n, 2n)^7$He and $^9$Be$(n, \alpha)^6$Li, in addition to provoking the normal displacement damage. Studies have been made by several workers(1)-(4) on the effect of neutron irradiation on the mechanical properties of Be. As a result of these studies, it has been shown(5) that the He atoms thus produced have an influence on the loss of ductility of Be.

Because of the inherently insufficient circumferential ductility of Be tubing, it is important to minimize the loss of ductility due to He gas bubbles.

It is known that in order to minimize the swelling and the loss of ductility in a metal due to inert gas bubbles produced by nuclear fission, it is effective to reduce the diameter of the gas bubbles and to disperse them in the matrix. Barnes et al.(6) have studied swelling in metallic U, and reported that finely dispersed precipitates produce significantly less swelling of this metal. Hickman & Chute(7) reported that the distribution of He gas bubbles in neutron-irradiated Be was influenced by pre-irradiation heat treatment. They suspected this phenomenon to be due to modification of the distribution of inclusions in the Be, which distribution was in turn influenced by heat treatment.

It is considered that the behavior of He gas bubbles in Be is the most important factor in the loss of ductility and the swelling of the metal, but the details of this behavior are still unknown. No method has yet been established to prevent the swelling and the loss of ductility in the metal. Of recent, Be tubing with good circumferential ductility has become available from cast ingots. It should be of interest to elucidate the behavior of He gas bubbles in irradiated Be specimens made from such cast ingots.

The purpose of the present study is to observe the effect of pre-irradiation heat treatment on the formation of He gas bubbles in neutron-irradiated Be and the effect of...
irradiation on the change in microstructure during post-irradiation annealing. Transmission electron microscopy was employed for the studies.

II. EXPERIMENTAL PROCEDURE AND MATERIALS

Cast ingot made from electrolytic Be flakes was rolled first at 900°C into sheet 1 mm thick, and then at 700°C to 0.4 mm thickness. By chemical polishing and subsequent mechanical cutting, the specimen was shaped into 15 x 20 x 0.25 mm sheets. The impurities in the specimen are given in Table 1. Some of the specimen pieces were recrystallized before irradiation by annealing at 900°C for 3 hr. The specimens, either as hot-rolled or recrystallized, were both irradiated in JRR-2 at 232°C for 180 hr. The total neutron fluence was determined from the induced γ-activity of Al-0.6%Co alloy monitor wires to be $1.1 \times 10^{20} \text{n/cm}^2$. The fast neutron dose was obtained from the measured total dose and from the ratio of total to fast neutron fluxes measured previously in the same irradiation hole. In the present study, the fast neutron fluence was estimated to be about $5 \times 10^{19} \text{n/cm}^2$.

Table 1 Chemical analysis of impurities in Be used

<table>
<thead>
<tr>
<th></th>
<th>BeO</th>
<th>Fe</th>
<th>Al</th>
<th>Si</th>
<th>Cu</th>
<th>Ni</th>
<th>Be</th>
</tr>
</thead>
<tbody>
<tr>
<td>ppm</td>
<td>270</td>
<td>145</td>
<td>195</td>
<td>265</td>
<td>65</td>
<td>140</td>
<td>Bal</td>
</tr>
</tbody>
</table>

Both irradiated and nonirradiated specimens were heat-treated at 750°C, 800°C and 1,000°C for a variety of periods in Ar, and then thinned down electrolytically by window technique for electron microscopy. An electron microscope, JEM-6A (100 kV), with goniometer attached, was used for direct observation of the Be foils.

III. RESULTS OF THE EXPERIMENTS

1. Effect of Pre-irradiation Heat Treatment on the Formation of He Gas Bubbles in Be

No bubble formation was observed under as-irradiated conditions in the specimens, either annealed or unannealed before irradiation. Upon post-irradiation heat treatment at 900°C for 50 hr, many He gas bubbles from 1 to $3 \times 10^{-4} \text{cm}$ in diameter were observed on the grain boundaries and at the inclusions in the specimens irradiated as hot-rolled. Those subjected to annealing treatment before the irradiation were found, on the other hand, to contain extremely small bubbles. Photographs of both specimens are shown in Photos. 1 and 2. Upon post-irradiation annealing at 1,000°C, the specimens annealed before irradiation had He bubbles located on the grain boundaries, inclusions and dislocation lines.

With lengthening annealing period the bubbles grew in size, but remained at the same sites as the above. After 43 hr of post-irradiation annealing at 1,000°C, the specimens annealed before irradiation presented the condition revealed by the transmission electron micrographs shown in Photo. 3. The He gas bubbles are seen to have nucleated preferentially along the dislocation lines.

2. Effect of Neutron Irradiation on the Recrystallization of Be

Photomicrographs of specimens irradiated as hot-rolled and annealed at 750°C for 3 hr are shown in Photo. 4(a). Photo. 4(b) shows the microstructure of an unirradiated control specimen. As seen in Photo. 4(a), the structure of the irradiated Be is composed of subgrains with highly concentrated dislocations,
He gas bubbles preferentially nucleated on a dislocation line in Be irradiated after recrystallization and subsequently annealed at 1,000°C for 43 hr

Photo. 3

Microstructures of (a) Be irradiated as hot-rolled and subsequently annealed at 750°C for 3 hr and (b) unirradiated Be of the same heat treatment as (a)

Photo. 4

while in the unirradiated Be (Photo. 4(b)), many recrystallized grains are found and the dislocations have disappeared. In Photo. 5(a), a similar comparison is possible for the material similarly treated after irradiation but for the same 3 hr at 900°C. In Photo. 5(a), for the irradiated specimen, recrystallization is not complete and the structure is a mixture of recrystallized and unrecrystallized grains; in contrast recrystallization is complete and even grain coarsening has commenced in the unirradiated Be shown in Photo. 5(b). From the above results, it is confirmed that the recovery and recrystallization in the hot rolled markedly are retarded by neutron irradiation.

3. Interaction between He Bubbles and Inclusions

As seen in Photo. 6, the He gas bubbles have nucleated preferentially by the heat-treatment at 1,000°C. When the specimens are post-irradiation heat-treated at 900°C for 50 hr, the He gas bubbles at the inclusion are observed only in the material after neutron irradiation as rolled. In the specimen irradiated after recrystallization, no He bubbles are observed at the inclusions.

Photo. 5

Microstructures of (a) Be irradiated as hot-rolled and subsequently annealed at 900°C for 3 hr and (b) unirradiated Be of the same heat treatment as (a)

Photo. 6

He gas bubbles nucleated on an inclusion in Be irradiated after recrystallization and subsequently annealed at 1,000°C for 43 hr

Photo. 6

From the above facts, it is deduced that the He bubbles observed on the inclusions in the Be irradiated as rolled and annealed at 900°C for 50 hr are not produced by the pro-
cess of nucleation and growth, but are formed on the grain boundaries and then carried away from the moving grain boundaries by the inclusions.

Selected-area electron diffraction has revealed that the inclusions having the highest affinity to He bubbles are Be11Fe.

IV. DISCUSSION

1. Effect of Pre-irradiation Heat Treatment on the Behavior of He Gas Bubbles in Be

As seen in Photos. 1 and 2, He gas bubbles are formed during post-irradiation heat treatment more easily in Be irradiated as hot-rolled than after recrystallization treatment. Photograph 7 shows the distribution of bubbles in a specimen irradiated as rolled and annealed for 50 hr at 900°C. It is evident that the He bubbles are located mainly on the grain boundaries and within the grains in the recrystallized areas, and they grow during the post-irradiation annealing; but they are not observed on the sub-boundaries and within the sub-grains in the unrecrystallized areas. These phenomena indicate that during the post-irradiation recrystallization moving grain boundaries sweep the He atoms in the matrix, and as a consequence, the He is enriched at the grain boundaries. When the sweeping grain boundaries raise the concentration of He to a level high enough to nucleate the bubbles, preferential nucleation and growth of the bubbles will occur on the grain boundaries. The formation of large He bubbles on the grain boundaries by such sweeping mechanism has been reported by Barnes for Cu, and by Loomis et al. for F.P. gas bubbles in metallic U.

If the formation of He bubbles on the grain boundaries depends on the sweeping-up of He atoms in the matrix by moving grain boundaries, even at such low concentrations of He as in the present study (1×10⁻³%), the He bubble should be found mainly on the moving grain boundaries, except in the case of post-irradiation annealing at considerably high temperatures. The lowest temperature for the formation of bubbles, therefore, may approximate the recrystallization temperature of the specimen. In the case of 3 hr post-irradiation annealing at 750°C of the specimen irradiated as rolled, small He bubbles are observed only on the boundaries of recrystallized grains (Photo. 8). It is thus apparent that the migration of grain boundaries play an important role in the formation of He bubbles.

The distribution of He gas bubbles Be irradiated as hot-rolled and subsequently annealed at 900°C for 50 hr

Photo. 7

He gas bubbles located on grain boundaries in Be irradiated as hot-rolled and subsequently annealed at 750°C for 3 hr

Photo. 8

2. Effect of Neutron Irradiation on the Recrystallization of Be

Lillie and Murray et al. have report-
ed independently that recrystallization and grain growth of neutron-irradiated Al-Li alloy are retarded by He formation. In α-ray bombarded Al, Ells\(^{13}\) also observed a similar phenomenon. The recrystallization of an un-irradiated Be sheet prepared from cast ingot will be complete at about 750°C\(^{14}\).

In the present study, as shown in Photo. 5(a), the recrystallization was not complete, and many sub-grains with dense dislocations remained even after post-irradiation annealing at 900°C.

When He bubbles are present at a grain boundary, the movement of the grain boundary during recrystallization and grain growth is impeded by the stabilizing effect due to the decrease in surface energy between grain boundaries and the bubble\(^{15}\).

Ells studied the recrystallization behavior of Al bombarded with α-particles\(^{13}\). He showed that α-bombardment had a large influence in inhibition the start of recrystallization. He concluded that such inhibition of recrystallization was not caused by the presence of He filled bubbles \(>10^{-3}\) cm in radius, but by individual He atoms in solution or by small clusters of He atoms.

In the present study, observation by transmission electron microscopy revealed that bubbles in the specimens annealed below 900°C after irradiation were aligned along the boundaries of grains that had recrystallized and migrated during the post-irradiation annealing. As in Photos. 9 and 5(b), no bubbles are observed on grain boundaries, dislocations and dislocation networks in the unrecrystallized areas.

It is thus considered that the inhibition of grain growth during recrystallization in the neutron irradiated Be is also caused by the interaction with dislocations or grain boundaries brought about by He atoms in solution or by small clusters of He atoms.

Weir, in his study on the effect of high-temperature irradiation on the mechanical and physical properties of Be\(^{14}\), suggested that He atoms has a tendency to be attracted around a dislocation. Such a collection of He atoms would stabilize the dislocation, and its motion would be impeded.

The inhibition of recrystallization in neutron-irradiated Be should therefore be due to the formation of He atoms.

3. Interaction between He Bubbles and Inclusions
A number of studies\(^{63)-(7)\) have shown that second phase particles in a metal provide appropriate nucleation sites for F.P. gas bubbles. In the present study, preferential nucleation of He bubbles at the inclusions was observed, as shown in Photo. 6.

As described already in Sec. III-3, after post-irradiation annealing of the Be irradiated in warm rolled state, many bubbles remain on the inclusions as a result of the interaction between moving grain-boundaries and inclusions. The interaction between moving grain-boundaries and inclusions is shown in Photo. 10. It is evident from this figure that the grain boundaries eventually come to be parallel to the foil surface, because of the difference in grain boundary mobility between sites near inclusions and other sites. As shown in Photo. 10, it is possible to determine the direction of movement of the grain boundaries. The direction is indicated by an arrow in Photo. 10. It is evident that the He bubbles observed have remained on the inclusions as a result of this interaction.

It is possible to evaluate the retaining...
Transmission electron micrograph of Be irradiated as hot-rolled and subsequently annealed at 900°C for 50 hr.

(Note interaction between grain boundaries and inclusion.)

**Photo. 10**

force $F_p$ exerted by each inclusion on a He bubble. For example, the contact angle of the He bubbles in reference to the inclusion amounts to $91^\circ$ in the case shown in Photo. 6. The binding energy $E_b$ of He bubbles on the inclusion can be obtained by the measured values and the formula derived by Nelson\(^{18}\).

On the other hand, the grain boundary driving force $F$, exerted upon a spherical bubble of radius $r$, has been given by Barnes & Nelson\(^{19}\):

$$F = \gamma_b \sin \theta,$$

where $\gamma_b$: Tension of the grain boundary

$\theta$: Angle of the cone

Since the retaining force $F_p$ can be expressed by the formula $F_p = E_b/R_1$, it is possible to discuss the stability of a He bubble located on a moving grain boundary by comparison with these forces.

In this formula, $R_1$ denotes the radius of bubble before attachment, and $E_b$ is the binding energy between bubble and inclusion. The values calculated by the formula on presented in **Table 2**, where a value of 1,000 dyne/cm has been adopted for the surface tension of Be $\gamma$, and 500 dyne/cm for the grain boundary tension $\gamma_b$, pertaining to bubbles of the same radius. From the results obtained therefrom, it is evident that the retaining force of the inclusion on the He bubble is about twice as large as the maximum grain boundary driving force. It shows that a He bubble attached to an inclusion would not be swept away by a moving grain boundary, while a He bubble moving along with a grain boundary would, upon encountering an inclusion, be left on the inclusion as a result of interaction between grain boundary and inclusion. The occurrence of such phenomenon is shown in Photo. 10.

**Table 2** Comparison between retaining force of Be$_{11}$Fe against He gas bubble and maximum grain boundary driving force

<table>
<thead>
<tr>
<th>Contact angle ($^\circ$)</th>
<th>Radius of bubble (cm)</th>
<th>Binding energy $-E_b$ (erg)</th>
<th>Retaining force (dyne)</th>
<th>Maximum grain boundary driving force (dyne)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$R_1$</td>
<td>$R_2$</td>
<td>$R_3$</td>
<td>$R_4$</td>
</tr>
<tr>
<td>91.0</td>
<td>$9.42 \times 10^{-6}$</td>
<td>$1.35 \times 10^{-3}$</td>
<td>$2.67 \times 10^{-7}$</td>
<td>$2.83 \times 10^{-4}$</td>
</tr>
</tbody>
</table>

$R_1$: Equilibrium radius before contact, $R_2$: Equilibrium radius after contact

**V. CONCLUSIONS**

A comprehensive study by transmission electron microscopy was carried out on neutron irradiated Be to elucidate the effect of irradiation on changes in microstructure, and the effect of pre-irradiation treatment on the formation of He gas bubbles generated by post-irradiation annealing.

(i) As compared with specimens irradiated after recrystallization, those irradiated as hot rolled were found to be less inhibited in the formation and growth of the bubbles during post-irradiation annealing of the Be.

The formation of fine gas bubbles along grain boundaries and dislocation lines was observed in the specimen irradiated in annealed state only after 50 hr of post-irradiation treatment at 900°C, while in
the specimen irradiated as hot-rolled, a similar formation was observed after only 3 hr of treatment at 750°C.

The observed difference in the tendency of bubble formation depends upon the grain boundary mobility during post-irradiation annealing.

(2) In Be of relatively low He concentration as in the present case, the nucleation and growth of the bubbles occurred preferentially at grain boundaries, inclusions and dislocation sites.

(3) The recrystallization of irradiated Be was impeded by the interaction of dislocations with the He atoms.

(4) The inclusion that has the largest affinity to the bubbles in Be has been found to be Be$_{11}$Fe.

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References