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THE EFFECT OF DISPLACEMENT CASCADES ON SMALL HELIUM BUBBLES IN ALUMINUM AND GOLD*

S E. DONNELLY¹, R.C. BIRTCHER², C. TEMPLIER³, R. VALIZADEH¹ and V. VISHNYAKOV¹

¹Joule Laboratory, Science Research Institute, University of Salford, M5 4WT, UK

²Materials Science Division, Argonne National Laboratory, Argonne, IL 60439, USA

³Laboratoire de Métallurgie Physique, University of Poitiers, 86022 Poitiers, France

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¹Joule Laboratory, Science Research Institute, University of Salford, M5 4WT, UK

²Materials Science Division, Argonne National Laboratory, Argonne, IL 60439, USA

³Laboratoire de Métallurgie Physique, University of Poitiers, 86022 Poitiers, France

ABSTRACT

The evolution of individual helium bubbles in thin foils of gold and aluminum irradiated with 400 keV Ar⁺ and 200 keV Xe⁺ has been followed with *in-situ* transmission electron microscopy for a comparison between the effects of dilute (Al) and dense (Au) collision cascades. Bubble shrinkage in Al has been attributed to direct displacement of the gas out of the bubbles. Effects in Au, include the disappearance and Brownian motion of bubbles under irradiation, and are consistent with thermal spike processes seen in molecular dynamics simulations.

INTRODUCTION

Interest in the interaction of inert gases with solids has arisen over the last few decades primarily because of potential materials problems associated with the operation of nuclear fission and fusion reactors. Despite a considerable worldwide interest¹ and the relative simplicity of a system consisting only of an inert gas and an elemental metal, a number of fundamental questions remain unanswered about the inert gas/metal system. An interesting issue from a technological point of view is the behavior of gas bubbles in a reactor environment where both high temperatures and damaging irradiation control bubble kinetics. Although the effect of temperature on bubble behavior has been extensively studied^{1,2}, scant information is available on bubbles under damaging irradiation. The present paper contrasts helium bubble behavior in aluminum and gold subjected to a heavy ion bombardment. The host metals were chosen to compare dilute (Al) and dense (Au) collision cascades on bubble behavior.

EXPERIMENTAL

Jet-polished polycrystalline Au and Al (99.9999% pure) TEM discs were implanted at temperatures of 300 and 500 K respectively ($T_m = 0.31$ and 0.37) with 3 keV He⁺ ions at a flux of 6×10^{13} ions/cm²/sec. Ar or Xe irradiations and *in-situ* observations were performed at the HVEM-Accelerator Facility located at Argonne National Laboratory³. Specimens were irradiated with the ion beam 10° away from the surface normal. 200 keV Xe irradiation of Al and 400 keV Au irradiation of Au produced displacement cascades without trapping significant numbers of heavy ions in the foil. The conditions of the helium implantations and subsequent damaging irradiations are given in Table 1.

The effect of the irradiation on specific areas of specimens was monitored using photographic film and—for bubbles of diameter 10 nm or greater—videotape. Higher resolution TEM was carried out using a JEOL 3010 TEM at the University of Salford.

Table 1 Implantation and irradiation conditions

	He Implantation			Heavy Ion Irradiation			
	Energy	Dose	Temperature	Species	Energy	Temp.	Dpa/ 10^{15} ions/cm ²
Al	3 keV	1.10^{17} cm ⁻²	0.32 T _m (R.T.)	Xe	200 keV	0.31 T _m	2.7
Au	3 keV	5.10^{15} cm ⁻²	0.37 T _m (500K)	Ar	400 keV	0.37 T _m	5.3

RESULTS AND DISCUSSION

The micrographs in Figure 1 show helium bubbles in aluminum and gold following irradiation to two doses, as given in the figure caption. The increment in dose is 2.7 displacements per atom (dpa) in both. Note that all observed changes occurred as a consequence of heavy ion irradiation and no changes were observed when irradiation was discontinued and the specimens maintained at the experimental temperature. For Al, three types of changes in bubbles were observed: reduction in size, disappearance and coalescence. In Au, bubble motion was also observed.

Bubble shrinkage and disappearance

Figure 2a) shows the radii of individual bubbles, as observed by TEM in aluminum as a function of Xe dose. There are two distinct processes operating here (A and B on figure 2). The Xe dose for bubbles to disappear by the more rapid process is commensurate with the dose to sputter a thickness of aluminum similar to the bubble diameter. After sputtering has proceeded to the point where the bubble contacts the surface, the TEM image of the bubble will begin to diminish in size. If it is assumed that a thickness equivalent to half the bubble diameter must be eroded for the bubble TEM image to disappear, then a sputtering yield of between 7 and 15 is required consistent with the expected value of 10 for 200 keV Xe on Al⁴.

Figure 2. Bubble shrinkage and disappearance in a) Al and b) Au. Images in b) are still frames from videotape. Numbers indicate time in seconds.

The slower bubble shrinkage process in Al (B on figure 2a) occurred for deeper-lying bubbles unaffected by surface erosion. The shrinkage of the bubble radius was typically about a tenth of the rate observed for shrinkage by surface removal (A on figure 2a). This process is believed to be due to a combination of He resolution by Al recoil atoms coupled with preferential absorption of Al interstitials to maintain equilibrium bubble pressure. (Athermal bubble equilibration processes will be discussed below). Irradiation-induced resolution of gas from bubbles has previously been reported by Evans for Kr in Zr⁷. Simulation of cascade effects on a layer of helium sandwiched between two aluminum layers using the Monte Carlo code TRIM91⁸ indicates a tendency to homogenize the system at a resolution rate in reasonable agreement with the observations⁹.

For gold, bubble disappearance occurs much more rapidly. Figure 2b illustrates the dynamics of one event recorded on videotape as the event took place over a very short dose range. Many bubble disappearances in Au occurred after a typical dose step of $\approx 0.5 \times 10^{15}$ Ar/cm². Unfortunately, when bubbles had grown (by coalescence) to a size where they could be recorded on videotape very few were then observed to disappear. However, figure 2b shows one disappearance event captured on videotape in which the bubble contrast is observed to vanish over a period of approximately 1.5 seconds. Although, as will be discussed below, we believe that many disappearances of smaller bubbles in gold take place as a result of direct interactions with individual cascades, the timescale of the event shown in figure 2b) is clearly too long for this to be an explanation. (For annihilation by a single cascade, bubble contrast should vanish in significantly less than 1 ns). However, the small Ar dose delivered in 1.5 s ($\approx 2.5 \times 10^{12}/\text{cm}^2$) implies that the bubble disappearance must have been triggered by the passage of only one or two Ar ions. A possible explanation of this event is the type of process recently modeled using molecular dynamics (MD) by Ghaly and Averbach⁵ in which a single cascade at or near the surface of gold gave rise to viscous flow processes having effects as deep as 7 nm. Such a process would convert a near surface bubble to a crater. Surface diffusion has been observed to cause the modification of small features on gold at room temperature⁶. We believe that the observed contrast change in TEM is due to the conversion of the bubble to a surface crater followed by filling in of the crater by surface diffusion on the otherwise flat surface. Such processes clearly do not occur, however, at the curved or faceted bubble surfaces as this would give rise to thermally-driven bubble diffusion and this is not observed.

Surface cratering and radiation-driven diffusion of bubbles (see below) is not sufficient to account for all bubble disappearances in Au. A possible explanation is that direct annihilation (resolution) occurs as a result of interaction with a single cascade. Helium and vacancies incorporated into the molten spike region of a cascade would subsequently be redistributed to submicroscopic defects or to several smaller bubbles. This mechanism is only expected to operate in materials that support dense cascades where the cascades are larger than the bubbles. This may result in subsequent He loss by diffusion of submicroscopic agglomerates to the specimen surfaces.

Fig. 3a) Experimentally measured and b) calculated bubble motion in gold (mean bubble diameter 5.5 nm) following post-irradiation with a dose increment of 8.55×10^{14} Ar/cm². Scatter plots show distance and direction of motion of individual bubbles.

Bubble Motion

No bubble motion, other than that due to coalescence of very close bubbles was observed in aluminum. In contrast, considerable bubble motion was observed in gold. The scatter plot in figure 3a) shows the distance and direction of motion of a sample of bubbles (mean diameter 5.5 nm) measured for a dose step of 8.55×10^{14} Ar/cm². There is a stochastic bubble motion with a root mean square displacement per bubble of ≈ 4.5 nm in the plane of the micrograph.

Although Brownian motion of bubbles due to thermal diffusion has been previously observed at higher temperatures¹⁰, to our knowledge, this is the first direct observation of bubble motion induced by irradiation. The presence of a possible oxide film on the inside surface of bubbles in Al may hinder displacement-driven motion—however, we believe that the principal difference between Al and Au is the nature of cascade processes in the two materials. In contrast to the large dilute displacement cascades occurring in Al, TRIM91⁸ calculations reveal that each Ar ion incident on the ≈ 50 nm thick Au TEM foil will produce an average of ≈ 6 discrete subcascades with a mean volume equivalent to a sphere with a diameter typically between 2 and 3 nm. This size is in reasonable agreement with the size of both the initial molten spike zone and the resulting vacancy rich region (≈ 4 nm) observed in MD simulations^{5,11}. We suppose that a cascade initiated adjacent to a bubble will form a melt zone that will disrupt the bubble shape. Although we do not propose a precise mechanism, viscous flow processes may allow a bubble to deform into the molten region. We have calculated bubble motion resulting from such a process using a simple model with the following assumptions: (i) bubbles and cascades are spherical in shape, (ii) the spatial distribution of cascades through the gold foil is random and unaffected by the bubble distribution, (iii) bubble-cascade interaction only occurs when a cascade volume touches or overlaps with a bubble, and (iv) the cascade thermal spike and localized melting results in the bubble moving to the center of the combined volume of the bubble and the cascade. For a given bubble, there is no preferential direction in which cascades will be created, and its motion will appear to be Brownian in nature.

The scatter plot in figure 3b) shows the results for bubbles of diameter 5.5 nm in a 50 nm Au film subjected to 'cascade bumping' for cascades with a diameter of 2.7 nm (this value gives the best fit to the experimental bubble movement). The results are fairly sensitive to the value of cascade dimension so that averaging over the cascade size distribution would be more suitable but given the simplicity of the model it is encouraging that bubble movement in agreement with experiment is calculated for a mean cascade diameter consistent with the results of both Monte-Carlo and MD calculations.

Bubble coalescence

Although the mechanisms differ, coalescence occurs in both aluminum and gold under damaging irradiation. After coalescence of two approximately spherical bubbles, the resulting elongated single bubble becomes more spherical with increasing irradiation. In the case of the immobile bubbles in aluminum, coalescence takes place as a result of the net displacement of Al atoms out of the volume between the bubbles⁹ whereas in gold, coalescence is a result of cascade-induced mobility. An example of the coalescence of two bubbles on a grain boundary

in gold, recorded on videotape, is shown in figure 4a. The bubble changes towards a spherical shape. It is important to note that in both systems this process is driven by the heavy ion irradiation and is not thermally driven i.e. coalescence does not occur without irradiation over the timescale of the experiments (several hours).

The total bubble volume increases on coalescence whereas the total surface area of the bubbles appears to be conserved. This is illustrated for aluminum in figure 4b). This is an important observation as it indicates that the concept of an equilibrium bubble may be important even in circumstances where bubbles have no thermal mechanisms for regulating their pressure. Thermal equilibrium within a bubble of radius R occurs when the gas pressure balances the surface tension of the gas/metal interface—a condition that is met when the pressure, P , is given by: $P = 2\gamma/R$, where γ is the surface tension (or surface free energy) at the bubble surface. This condition necessitates that a bubble conserve surface area rather than volume after coalescence. A bubble is generally regarded as a neutral sink for both vacancies and self-interstitial atoms (SIAs) and, at high temperatures, maintains its pressure at equilibrium by means of a vacancy emission from the surface at a rate that is dependent on internal pressure¹². In the current experiments, the temperature is too low for equilibrium to be achieved thermally. Also the absence of bubble growth (except by coalescence) under high (equal) fluxes of vacancies and SIAs indicates that, in general, bubbles are neutral sinks for the two types of defect and that no biased traps exist for either type of defect. However, the increase in total volume on coalescence indicates that, when the bubble pressure increases during an initially constant volume coalescence event, the bubble must develop either slightly increased sink strength for vacancies or reduced sink strength for SIAs (presumably via strain fields from a now overpressurized bubble). In a similar way, strain effects could modify the interaction of vacancies and SIAs with different parts of an initially elongated bubble so that it tends to become more spherical.

Fig. 4 a) Coalescence of grain-boundary bubbles in Au (numbers indicate elapsed time in minutes—dose rate 1.7×10^{12} ions/cm²/sec) b) Bubble volume and surface area changes on coalescence of bubbles in Al.

The role of dislocations

Dislocations are believed to affect void and bubble growth through the biased attraction of self-interstitials. This can result in an excess of vacancies that may contribute to rapid growth. We have found no evidence for this process in the present work. In addition, we observed the formation and movement of dislocation loops under irradiation of Au, but when irradiation ceased motion also stopped. The driving force for motion of both dislocation loops and bubbles thus appears to be cascade processes. Finally, although dislocations might be expected to play an important role in thermally-driven bubble shrinkage, growth and coalescence these thermal effects have not been observed in our experiments. Dislocations, thus, do not seem to be of importance in interpreting our observations in the present experiments.

Bubble equilibration

The conservation of surface area on coalescence and the 'rounding out' of bubbles, reported above, indicate that a mechanism exists for bubbles to equilibrate athermally when fluxes of SIAs and vacancies are present. When inert gas bubbles are formed during implantation, defect fluxes may similarly permit equilibration to occur—except perhaps for the lowest energy He irradiations. This could provide an explanation of the fact that experimentally derived pressures (by diffraction from solid Ar, Kr and Xe within bubbles) has indicated that bubbles are generally close to equilibrium pressure. This is contrary to theoretical predictions of much higher pressures based on loop-punching models¹³.

CONCLUSIONS

A TEM study has been carried out on helium bubbles in aluminum and gold during *in-situ* heavy ion irradiation in a high voltage transmission electron microscope. This study enabled the fate of individual bubbles to be followed and a comparison to be made between the effects of dilute and dense cascades. Dense cascades give rise to a stochastic bubble motion that is believed to result from direct interaction between bubbles and the melt-zone of individual cascades ('cascade bumping'). In both systems, bubbles appear to equilibrate athermally under damaging irradiation perhaps by means of a sink strength for vacancies and interstitials that is dependent on pressure.

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