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A GENERAL MECHANISM FOR HELIUM BLISTERING INVOLVING DISPLACED ATOM TRANSPORT*

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A mechanism developed to account for formation of vertically elongated blisters in high displacement environments produced by ^{252}Cf alpha particles and fission fragments has been extended to formation of dome-shaped blisters in the low displacement environments produced by simple helium ion beams. In this mechanism, transport of displaced atoms to relieve compressive stresses in the helium-implanted layer allows interconnections of small, subsurface bubbles to form the blister cavity. The same transport may cause thickening of the blister caps at low implantation energies. The transition from dome-shaped to vertically elongated blistering occurs between the 300 and 3000 displacements per helium atom produced by simple helium ions and ^{252}Cf radiations, respectively.

1. INTRODUCTION

Blistering and related surface distortions of materials implanted with high concentrations of helium represents a major problem in the development of structural components for thermonuclear reactors [1]. Extended exposures of the materials to accelerator He-ion beams, to simulate the thermonuclear reactor radiations, produces characteristic dome-shaped blisters (Fig. 1A), whose interconnection and exfoliation can cause a significant surface erosion. Formation of such blisters is generally attributed to the agglomeration of the implanted helium in subsurface regions of the exposed material, but the details of the agglomeration mechanism, especially at low temperatures where thermal diffusion is limited, are not yet completely defined.

Recent observations of a unique form of blistering of surfaces exposed at low temperatures to ^{252}Cf alpha particles and fission fragments have demonstrated the potentially important role of displaced atom transport in the blistering mechanism [2,3]. In contrast to the relatively low number of atoms displaced per helium atom implanted during exposures in simple helium ion beams, the implantation of helium atoms by ^{252}Cf alpha-particles is accompanied by a large number of atoms displaced by the fission fragments. The blisters formed by ^{252}Cf exposures exhibit vertically elongated configurations (Fig. 1B), among others, best accounted for by a transport of the displaced atoms that relieves lateral compressive stresses in the helium implanted layer. The purpose of this paper is to extend the mechanism developed for elongated blister formation in such high displacement environments to the formation of conventional dome-shaped blisters in the low-displacement environment produced by simple helium-ion beams.

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A. Dome-Shaped Blisters Produced by Accelerator He-ion Beams

B. Vertically-elongated Blisters Produced by ^{252}Cf Alpha Particles and Fission Fragments

Figure 1. Types of Blisters Produced by Helium Implantations in Low and High Displacement Environments

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2. BACKGROUND

Formation of dome-shaped blisters has been observed on surfaces of many materials implanted with helium by exposure to monoenergetic helium ion beams in particle accelerators [1]. The blister formation occurs characteristically at a threshold helium concentration, which is dependent on temperature and radiation characteristics. At low temperatures, the many small (5 μm) blisters formed on palladium metal irradiated with 300 keV He^+ -ions, for example, require exposures $>10^{18}$ particles/ cm^2 , which produce helium concentrations exceeding 30 atom % at depths of penetration corresponding to the most probable range ($\sim 0.6 \mu\text{m}$) of the helium ions [4]. Increasing temperature in general decreases the threshold helium concentration for blistering, and the implantation of relatively small helium concentrations (<3 atom %) produces characteristically porous surfaces at high temperatures [5]. Decreasing energy of the incident beam also decreases the helium concentration threshold for blistering [1].

Two general models have been proposed to account for the blistering phenomena. In the Gas Pressure Model [1,6,7], blistering occurs when the size and distribution of subsurface bubbles allows them to interconnect, releasing gas whose pressure deforms the nearby surface. Several mechanisms for formation, growth and interconnection of the bubbles have been suggested. An alternative Lateral Stress Model attributes blistering to the buckling of the helium-implanted surface layer under lateral compressive stresses resulting from restraint of its volume expansion by underlying material [7,8,9]. However, observations of blistering on the unexposed side of a thin specimen, when the peak concentration of implanted helium is closer to the unexposed side than to the exposed side [10], effectively eliminates this model as originally proposed.

2.1 Gas Pressure Model for low temperature blistering

Two additional observations have special bearing on the Gas Pressure Model for helium blistering.

a. The thickness of the blister caps are greater by factors up to two than the most probable range of the incident ions at energies less than about 50 keV [11,12]. This discrepancy indicates a significant swelling, attributed to formation of helium bubbles, in the blister caps.

b. Multiple energy implantations which produce uniform concentrations of implanted helium exceeding 30 atom % from the surface inward avoid blister formation [13]. The implanted material has a microstructure of small interconnected

pores which allows helium release to the specimen surface. Similar structures in subsurface layers may permit helium agglomeration in monoenergetically implanted specimens [14].

Accounting for these observations requires detailed consideration of the growth and interconnection of helium bubbles.

Evans has described the microstructure of the helium-implanted region of a specimen exposed at low temperature to monoenergetic helium ions as a layer of small (2 to 3 nm) highly overpressurized bubbles resulting from helium agglomeration under conditions of no vacancy diffusion [6]. Such overpressurized bubbles impose complex stresses on the surrounding material, resolved compressively in directions parallel to the bubble layer. The bubbles form and grow, according to Evans' mechanism, either by capture of vacancies produced by atom displacements at the bubble surface, or by stress-induced creation of interstitial dislocation loops in the surrounding material (loop-punching). Interconnection of the bubbles occurs at the threshold helium concentrations for blistering by interbubble cracking. The interbubble cracking mechanism has some difficulty in accounting for a sufficient quantity of helium in the blister cavity to cause surface deformation.

An alternative mode of gas agglomeration, suggested by Baskes and Wilson to account for bubble formation, occurs by a spontaneous rearrangement of closely-spaced helium-vacancy clusters produced during helium ion exposures [15]. The growth rate of the largest bubbles rapidly accelerates above a critical cluster concentration, generating the pressurized blister cavity. Neither the Evans nor Baskes-Wilson mechanisms for helium agglomeration considers the disposition of interstitial atoms produced concurrent with growth of gas bubbles during helium ion exposures.

2.2 Elongated blister formation in high displacement environments

The importance of interstitial transport processes to the blistering mechanism is illustrated by the formation of vertically elongated blisters on surfaces exposed to ^{252}Cf alpha particles and fission fragments [2]. Inner surfaces of ^{252}Cf -containing Pt-10% Rh capsules exhibit such blisters after exposures of 1.2×10^{18} alpha particles/ cm^2 , producing concentrations of implanted helium up to a maximum of 3 atom % at the capsule surface; concurrent fission fragment exposures of 4×10^{16} particles/ cm^2 produce displacement damage in excess of 100 displacements per atom. The vertically elongated blisters, about 1 μm in diameter and up to 10 μm long, form at locations along the capsule wall no farther than about 0.5 cm from the ^{252}Cf deposit.

The unique features of the blistering of ^{252}Cf -exposed surfaces are accounted for by interaction of the implanted helium with the lattice defects produced by the fission fragments [3]. The implanted helium combines with vacancies resulting from atom displacements by the fission fragments to form relatively large gas bubbles, leaving residual interstitials around the bubbles to agglomerate into specially oriented dislocation loops. Glide of the loops to the surface, to relieve lateral compressive stresses produced by the implanted helium, displaces successive layers of lattice atoms into the elongated blister wall.

3. GENERAL MECHANISM FOR HELIUM BLISTERING INVOLVING DISPLACED ATOM TRANSPORT

An interstitial transport mechanism analogous to that for formation of vertically elongated blisters in the high displacement environment produced by ^{252}Cf radiations can be used to explain the growth and interconnection of small over-pressurized bubbles formed during helium implantation in the low displacement environment produced by simple helium-ion beams. In the low as in the high displacement environment, helium atoms with enhanced mobility due to atom displacements agglomerate, along with vacancies resulting from displacements, to form the array of small over-pressurized bubbles observed. Growth of the bubbles can occur by either the vacancy capture or the loop punching mechanism envisioned by Evans [6], or by the random coalescence of helium-vacancy complexes described by Baskes and Wilson [15].

Each of these bubble growth processes leaves residual interstitial atoms to form dislocation loops in the surrounding material. Transport of the residual interstitials, by glide of the dislocation loops in suitable orientations to relieve lateral compressive stresses in the implanted layer, produces important effects in the formation of conventional dome-shaped blisters. In particular, such transport allows the small helium bubbles to interconnect and form the helium-filled cavity required for blistering by the Gas Pressure Model. The same interstitial transport, in the macroscopic stress gradients resulting from restraint on expansion of the implanted layer by underlying material, contributes to the swelling of the blister caps on specimens implanted in low energy helium ion beams. These effects of interstitial transport on blistering in low displacement environments are illustrated schematically in Fig. 2.

Detailed comparisons of the blistering processes in high and low displacement environments requires consideration of the differing helium distributions produced by ^{252}Cf radiations and accelerator He-ion beams. Exposure to ^{252}Cf radiations produces concentrations of implanted helium (and numbers of displaced atoms) which

decrease from maximum values at the exposed specimen surfaces to zero at depths of penetration corresponding to the range of the incident particles [2]. Bubble distributions in ^{252}Cf -exposed material are therefore somewhat like those resulting from multiple-energy helium ion exposures, for which no conventional dome-shaped blistering is observed. It is thought, however, that in the high displacement environments of the ^{252}Cf exposures, the fission-fragment-induced mobility of implanted helium depletes the surface layers to depths of a few tenths micron, the approximate thickness of the elongated blister wall. This distance is comparable to the most probable range of monoenergetic He-ions at 100 keV and less. The distribution of implanted helium near the surfaces of ^{252}Cf -exposed specimens may therefore more closely resemble that produced by implantations using monoenergetic He-ion beams than that produced using multiple energy He-ion beams.

Figure 2. Effect of Interstitial Transport on Helium Blistering in Low Displacement Environment Produced by Simple Helium Ion Beams (Schematic)

The critical difference between high and low displacement environments is the size and spacing of the helium bubbles formed. The bubbles produced in ^{252}Cf exposed specimens are presumed to be about the same diameter as the depth of the depleted layer, a few tenths μm and much larger than the 2 to 3 nm bubbles produced by the helium-ion beams. In the low displacement environments

produced by the helium ion beams, regions in which interstitial transport occurs around the small, closely spaced bubbles overlap, allowing interconnection of the bubbles at sufficiently high exposures. These interconnections may be crystallographically oriented, and the residual material between the interconnected bubbles may be massively recrystallized by such interstitial transport processes [14].

The direction of interstitial transport producing interconnection of small bubbles, and the ultimate disposition of atoms involved, will be determined by gradients in the lateral compressive stresses of the implanted layer. Superimposed on the gradient caused by different helium concentrations at different depths, which induces transport away from the regions of maximum helium concentration, are the gradients caused by restraint on expansion of the implanted layer by the underlying material. For specimens implanted with maximum helium concentration near the exposed surface, these stress gradients direct the interstitial transport from the helium implanted layer towards the exposed surface of the specimen. The interstitial transport processes can thus contribute to the thickening of the blister cap of specimens exposed in low energy ion beams. Such thickening of the blister cap decreases with increasing energy of the incident ion as increasing depth of penetration reduces stress gradients promoting transport toward the exposed surface. For specimens implanted with maximum helium concentrations near the unexposed surface, the stress gradients will direct interstitial transport from the helium implanted layer toward the unexposed surface, producing thickening of the blister caps formed on the unexposed side of such specimens.

Precise correlation of blister cap thickening with interstitial transport rates will require quantitative description of the effect of compressive stress gradients on the transport process. With 100 displacements per atom incurred even in low displacement environments by the time threshold helium concentrations for blistering are reached [16], it seems evident that interstitial transport processes can contribute significantly to the blister cap thickening.

Careful measurements by St. Jacques, Terreault and associates confirm previous reports of blister cap thickness about twice the most probable range of He^+ -ions at energies of 10 to 20 keV [17,18]. This discrepancy they attribute to helium bubble formation causing swelling in the material of the blister caps. The magnitude of swelling required, estimated at 30 to 60%, is, however, generally greater than the volume of bubbles (<15%) observed using transmission electron microscopy [19]. The extra swelling of the blister cap may be due to bubbles too small to

be observed. Alternatively it may be caused by deposition of interstitial atoms transported from helium-implanted regions at greater depths in the specimen.

4. HELIUM CONCENTRATION THRESHOLDS FOR BLISTERING

In the displacement transport mechanisms for blistering, the helium concentration threshold for blister formation, like the blister configuration, depends on the ratio of displacements to implanted helium atoms. This threshold increases from the 3 atom % or less helium characteristic of the high displacement environment produced by ^{252}Cf radiations to the >30 atom % helium required for blister formation in the low displacement environment produced by simple helium beams. The displacement/He atom ratio (and corresponding helium concentration) at which the transition from vertically elongated to dome-shaped blistering occurs is not precisely determined, but lies within the range of displacement/He atom ratios of the high and low displacement environments (Figure 3).

Figure 3. Dependence of Helium Concentration Thresholds for Blistering on Displacement/He ratio (Conceptual)

The lower limit is the ~ 300 displacement/He atom ratio produced by exposures to the simple helium-ion beams [16]. The upper limit is the ~ 3000 displacement/He atom ratio produced by the alpha particles and fission fragments from a ^{252}Cf source [20]. Termination of the vertically elongated blistering observed on ^{252}Cf capsule walls a relatively short distance (<0.5 cm) from the radiation source at one end

indicates the displacement/He atom ratio required for such blistering in high displacement environments is within 10% of the maximum displacement/He ratio produced by the ^{252}Cf radiations. The sharp decrease in helium threshold for elongated blister formation with increasing displacement/He atom ratio reflects recent preliminary observations of such blistering at very low concentrations of helium implanted during ^{252}Cf exposures.

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