Conf-780431--8

MASTER

DP-MS-77-119

WHISKER GROWTH — A NEW MECHANISM FOR HELIUM BLISTERING
OF SURFACES IN COMPLEX RADIATION ENVIRONMENTS

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Extended abstract for poster presentation at the
3rd International Conference on Plasma Surface Interactions
in Controlled Fusion Devices
Culham Laboratory, Abingdon, Oxfordshire, UK
April 3-7, 1978

and for publication in the Proceedings

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WHISKER GROWTH — A NEW MECHANISM FOR HELIUM BLISTERING OF SURFACES IN COMPLEX RADIATION ENVIRONMENTS*

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Implantation of helium concurrent with the generation of large numbers of displaced atoms in surface layers of materials exposed to ^{252}Cf α -particles and fission fragments produces a unique form of low temperature surface blistering [1]. Observations of vertically elongated blisters, among other configurations, formed under such conditions suggest that the blisters result from a whisker-growth mechanism. In this mechanism, the blisters, like filamentary whiskers on certain electroplated metals [2,3], represent a surface deformation that relieves the lateral compressive stresses developed in the material during helium implantation [4]. The implanted helium combines with vacancies resulting from atom displacements by the fission fragments to produce gas bubbles, leaving residual interstitial atoms around the bubbles to form whisker-like blister walls. Such a mechanism may under some conditions contribute to surface blistering in other complex radiation environments, such as the thermonuclear reactor. The purpose of this paper is to formulate a basis for the whisker-growth mechanism for helium blistering as an aid to the specification of conditions under which the mechanism might apply.

* Work done under USDOE Contract AT(07-2)-1.

Blistering of ^{252}Cf -Exposed Surfaces

Surface blistering by ^{252}Cf radiations was first observed at the Savannah River Laboratory in May 1976 on a Pt-10% Rh capsule which had contained an initial 17.8 mg ^{252}Cf as an oxide deposit for about 2-1/2 yr near room temperature. Decay of the ^{252}Cf (half-life 2.65 yr) had exposed adjacent surfaces to 1.2×10^{18} α -particle/cm² and implanted helium in concentrations up to 3 atom %. The capsule surfaces were concurrently exposed to 4×10^{16} fission fragments/cm², which produced lattice damage up to about 100 displacements per atom [5]. The concentrations of implanted helium were much less than the 20 atom % or more required to produce blistering at low temperatures by α -particles (or accelerator helium ions) alone [6,7].

The blisters occurred in a variety of configurations. Near the Cf-oxide deposit, they exhibited circular cross sections, 1-10 μm diameter, but farther away they appeared in both vertically and laterally elongated configurations preferentially aligned parallel to the capsule length. The vertically elongated blisters (Fig. 1), with diameters about 1 μm and lengths up to 10 μm , formed farthest from the radiation source; they were connected to the capsule surface at one end only, with their free ends directed away from the radiation source, and they sometimes overlapped in tangled arrays. The laterally elongated blisters, formed at intermediate distances, were connected to the capsule surface over most of their length; they were generally shorter and wider

than the vertically elongated blisters, but in some areas were found in parallel columns extending lengthwise 50 to 100 μm or more.

Whisker Growth

The vertically elongated blisters observed on ^{252}Cf -exposed surfaces exhibited many of the characteristics of filamentary whiskers formed at low temperatures on electroplated zinc, cadmium, and tin surfaces [2,3]. Electroplates of these metals, in contrast to most others, crystallize with a lateral compressive stress. The whiskers, typically 0.5 to 2 μm in diameter, grow as single crystals by addition of metal atoms at the whisker base. No pits or holes of comparable volume are found within the electroplate. Whisker-growth rates depend most importantly on the magnitude of the lateral compressive stress in the electroplate, with rapid growth occurring only above a critical threshold, typically 40 MN/m^2 for zinc electroplates. Certain crystal textures of the electroplate also promote whisker growth.

Recent theory has accounted for whisker growth by a two-stage model [3]. Whisker formation locally disturbs the lateral compressive stress and generates shear and tensile components of the stress normal to the surface. In the first stage of whisker growth, an interstitial dislocation loop forms and expands laterally by emission of vacancies to a remote sink. In the second stage, the interstitial loop glides to the surface to form an extra plane of atoms at the whisker base. The stress

required for the glide stage of whisker growth must exceed a threshold value representing the resistance afforded by various barriers to dislocation motion in the electroplate.

Whisker-Growth Mechanism for Helium Blistering

The whisker-growth mechanism accounts for blistering of ^{252}Cf -exposed surfaces in terms of the interactions of implanted helium with lattice defects produced by the fission fragments (Fig. 2). Impact of the fission fragments displaces both metal atoms and implanted helium atoms, forming vacancies and interstitials, and promoting agglomeration of the helium atoms along with the vacancies into gas bubbles [8]. The residual interstitial atoms around the bubbles gather into platelets and form dislocation loops preferentially aligned to relieve the lateral compressive stresses in the helium-implanted surfaces. Glide of the interstitial loops under these stresses displaces successive layers of lattice atoms into the blister walls.

This mechanism suggests that reciprocally critical values of lateral stress and bubble size are necessary for blister nucleation. If implanted helium concentrations are high enough to provide abundant vacancy sinks, the second-stage glide of the interstitial loops formed will control blister growth. As bubbles increase in size, the normal components of the disturbed lateral stress increase in magnitude, until the threshold required for glide of the interstitial loops to the surface is exceeded.

Larger bubble size allows blister formation at lower lateral stress. Because lateral stress depends principally on the concentration of implanted helium and bubble size depends (at low temperatures) on the number of atoms displaced by the fission fragments, blistering in a high displacement environment occurs at relatively low implanted helium concentrations.

Once nucleated, the blisters grow by lateral extension of the blister bases into surrounding areas in which blistering thresholds are exceeded, as well as by vertical extension of the blister walls. Lateral growth, like nucleation, is promoted by high numbers of displacements, and vertical growth, sensitive to lateral stress, is promoted by high helium concentration. The varying configurations of blisters on the ^{252}Cf -capsule surfaces result from radiation intensity gradients within the capsule. Helium implanted by α -particles and lattice atoms displaced by fission fragments decrease with distance from the radiation source, as does the displacement-to-helium ratio (because the fission fragments have a shorter range than the α -particles) [5]. The wide circular blisters near the ^{252}Cf deposit form in uniform radiation environments producing relatively high displacement-to-helium ratios. The laterally elongated blisters in intermediate regions form as the front demarking the blister threshold moves along the capsule surface. The narrow, vertically elongated blisters farthest from the ^{252}Cf deposit form in non-uniform radiation environments producing relatively low displacement-to-helium ratios.

Limits on Blistering Conditions

The primary limit on radiation conditions under which blistering by the whisker-growth mechanism should occur is the critical helium concentration required. This limit, typically a few atom percent for ^{252}Cf radiations at low temperatures, increases with decreasing displacement-to-helium atom ratio to the 20 atom % helium at which other blistering mechanisms, such as interbubble fracture [9], predominate. The apparent lack of whisker-type blisters on metal surfaces implanted in simple helium ion beams indicates that the displacement-to-helium atom ratio must exceed that produced by helium ions alone. First wall materials, such as stainless steel, in magnetically confined fusion reactors with surface displacement-to-helium atom ratios not much higher could thus escape whisker-type blistering. More susceptible materials, such as graphite [10], in such reactors, or materials in other devices with significantly higher displacement-to-helium atom ratios might exhibit the effect.

FIG. 1. Vertically Elongated Blisters on Pt-10% Rh Capsule Surface Exposed to ^{252}Cf α -Particles and Fission Fragments

FIG. 2. Conceptual Model for Helium Blister Formation on Surfaces Exposed to ^{252}Cf α -Particles and Fission Fragments.

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