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EXPOSURE AND TEMPERATURE DEPENDENCE OF ELONGATED BLISTER FORMATION IN COMPLEX RADIATION ENVIRONMENTS*

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Blistering of platinum alloy surfaces by ^{252}Cf alpha particle and fission fragment radiations occurred at relatively low concentrations of implanted helium during exposures at room temperature as well as at 1000°C . Distinctive configurations of the blisters resulting from transport of atoms displaced by the fission fragments persisted during the high temperature exposures. Post-exposure heating of specimens exposed at room-temperature produced no additional blistering until temperatures of 1300°C were reached. Post-exposure heating of 1000°C blistered specimens produced only thermal etching effects.

The low helium concentrations required for blistering and the distinctive blister configurations produced by ^{252}Cf exposures suggest a unique mode of surface distortion resulting from the large number and highly localized distributions of atom displacements generated by ^{252}Cf fission fragments. Such conditions may not be duplicated in the He-ion and fast neutron radiation environments of fusion reactors.

1. INTRODUCTION

Conditions under which surface blistering occurs in complex radiation environments are being characterized by the exposure of materials to ^{252}Cf alpha particle and fission fragment radiations. The alpha particles simulate helium ions and the fission fragments generate displaced atoms analogous to fast neutrons in fusion reactor environments. Previous investigations have demonstrated that blister formation during helium implantation by ^{252}Cf alpha particles is enhanced at low temperatures by the atom displacements which result from concurrent fission fragment exposure.^[1-3] The blisters assume vertically elongated and other distinctive configurations due to the transport of interstitials resulting from the atom displacements. The purposes of the present study were to determine the lower limits of exposure for the displacement-enhanced helium blistering and to establish the temperature range for formation of distinctive blister configurations due to interstitial transport.

2. EXPOSURE CONDITIONS

Blistering produced by ^{252}Cf radiations was characterized by examination of inner surfaces of ^{252}Cf - containing Pt-10 wt % Rh capsules that were exposed for various times at high and low temperatures in air. Exposure conditions of the specimens are summarized in Table 1. Resultant concentrations of implanted helium and numbers of displaced atoms decrease from maximum values at the exposed specimen surfaces to zero at depths of penetration corresponding to the 6 to $12\text{ }\mu\text{m}$ ranges of the incident particles.^[4]

Prior to examination, the capsule specimens were sectioned transversely at each end, and the ^{252}Cf contained as Cf-oxide was extracted by elution with dilute nitric acid. The capsules were then sectioned longitudinally and cleaned by soaking in concentrated nitric acid. Some specimens were heated in air after ^{252}Cf removal to establish effects of post-exposure heating on surface features. Surfaces of the various specimens were examined in a scanning electron microscope. Cross sections were characterized by optical microscopy, using equipment in alpha-containment boxes.

3. LOW TEMPERATURE BLISTERING

Incipient blistering produced near room temperature was observed after ^{252}Cf exposures producing maximum surface concentrations of 1.2 atom % implanted helium and 40 displacements per atom (dpa), as shown in Figure 1A. The blisters exhibited small circular cross sections not exceeding $1\text{ }\mu\text{m}$ in size and sometimes outlined grain boundaries in the specimen surface, but did not have the distinctive vertical or lateral elongations characteristic of higher exposures at room temperature (Figure 1B). A lower exposure specimen (11A), with maximum 0.03 atom % helium and 0.8 dpa, exhibited no surface blistering at room temperature. Cross section examinations using optical metallography revealed no evidence of surface blistering or subsurface gas bubbles; the low-temperature blisters were evidently confined to a very thin surface layer.

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Table 1. Exposure Conditions for ^{252}Cf Capsules

Capsule Spec. No.	Contained ^{252}Cf (μg) ^(a)	Temperature ($^{\circ}\text{C}$)	Time at Temp. (days) ^(b)	Alpha Exposure ^(c)		Fission Frag Exp. ^(c)	
				α/cm^2	He (at %) $\bar{Z}$	part/cm	dpa ^(d)
243Z	17.8×10^3	20	1050	1.2×10^{18}	3	4×10^{16}	100
Post-exposure heating		1100	2 hr				
Post-exposure heating		1300	2 hr				
11A	220 (500 Tb)	20	9	1×10^{16}	.03	3×10^{14}	0.8
13A	219 (500 Tb)	20	500	5×10^{17}	1.2	1.6×10^{16}	40
15A	126 (500 Tb)	1000	(43) 4.4 (2)	3×10^{15}	.01	1.2×10^{14}	0.3
7A	108 (870 Gd)	1000	(109) 250 (172)	1.5×10^{17}	0.4	5×10^{15}	12
8B	171 (810 Pr)	1000	(104) 250 (86)	1.5×10^{17}	0.4	5×10^{15}	12
Post-exposure heating		1100	2 hr				
6A	134 (850 Eu)	1000	(110) 250 (179)	1.5×10^{17}	0.4	5×10^{15}	12
Post-exposure heating		1300	2 hr				
8A	171 (810 Pr)	1000	(104) 250 (86)	1.5×10^{17}	0.4	5×10^{15}	12
Post-exposure heating		1500	2.5 hr				

a. Micrograms carrier given in parentheses.

b. Times at room temperature before and after high temperature exposure given in parentheses.

c. Alpha and fission fragment exposures given for time at test temperature only. Maximum exposures calculated assuming ^{252}Cf -oxide concentrated near one end of capsule.

d. Displacements per atom.

A. Low Exposure Spec. 13A
(1.2 at % He, 40 dpa)

B. High Exposure Spec. 243Z
(3 at % He, 100 dpa)

Figure 1. Surface Blistering Produced by ^{252}Cf Exposure at Low Temperatures (20°C)

The low exposure thresholds for blistering of surfaces exposed to ^{252}Cf radiations (~ 1 atom % helium) have previously been attributed to the generation of high concentrations of displaced atoms by the ^{252}Cf fission fragments concurrent with helium implantation by the ^{252}Cf alpha particles. In contrast, surfaces exposed to ^{242}Cm alpha particles producing up to 5 atom % concentrations of implanted helium without concurrent fission fragment exposure were not blistered,^[5] and exposures greater than 20 atom % were required to produce low-temperature blistering of specimens exposed to He-ion accelerator beams.^[6]

4. HIGH TEMPERATURE BLISTERING

Incipient blistering at high temperature (1000°C) was observed after exposures that produced a maximum 0.01 atom % helium and 0.3 dpa (Figure 2A). The circular high-temperature blisters, somewhat more pronounced than those near threshold exposures at low temperature, formed predominantly along grain boundaries, leaving the grain interiors relatively blister free.

Higher exposures generating 0.4 atom % helium and 12 dpa at 1000°C produced a markedly distorted surface structure comprised of pronounced globular protrusions interspersed with large holes (Figure 2B). Metallographic examination of cross sections of the high-temperature exposed specimens showed the protrusions to contain many large and small gas bubbles. Comparison with specimens exposed at low temperatures and subsequently heated indicated the globular protrusions resulted from atom displacements produced by the ^{252}Cf fission fragments at 1000°C as well as near room temperature.

5. BLISTERING DURING POST-EXPOSURE HEATING

Room-temperature ^{252}Cf -exposed specimens, which exhibited characteristic low-temperature surface structures, suffered no additional blistering due to implanted helium during post-exposure heating at temperatures below 1300°C . Heating a low-temperature-exposed specimen at 1100°C collapsed the elongated blisters, leaving only remnant surface features (Figure 3A). Heating at 1300°C , however, produced a new and differently blistered surface that was characterized by irregularly contoured areas containing many large holes (Figure 3B). The pattern of the contoured areas corresponded to the grain structure of the specimen surface. The contoured surface produced by 1300°C heating after low temperature exposure did not exhibit the pronounced globular protrusions characteristic of the high temperature (1000°C) exposure, a difference attributed to the absence of distortions due to atom displacements by fission fragments. Cross sectional examinations of the 1300°C -heated specimen revealed many large and small gas bubbles

A. Low Exposure Spec. 15A
(0.01 at % He, 0.3 dpa)

B. High Exposure Spec. 7A
(0.4 at % He, 12 dpa)

Figure 2. Surface Blistering Produced by ^{252}Cf Exposure at High Temperatures (1000°C)

within the distorted surface layer.

Post-exposure heating of specimens previously exposed at 1000°C produced little change in the as-irradiated surface structure of the specimens. Heating at 1100°C in air left the globular protrusions of the 1000°C -exposed specimens unchanged (Figure 4A). Heating at 1300°C in air produced a pronounced faceting of the protrusions and uncovered many subsurface holes, probably by oxidative evaporation of the metal. Heating at 1500°C in air produced a thermal contouring but less-pronounced faceting of the globular protrusions, revealing many subsurface holes within the globules (Figure 4B).

A. Heated 1100°C (Spec. 243Z)

A. Heated 1100°C (Spec. 8B)

B. Heated 1300°C (Spec. 243Z)

B. Heated 1500°C (Spec. 8A)

Figure 3. Effect of Post-Exposure Heating on Low Temperature Blistered Specimen

Figure 4. Effect of Post-Exposure Heating on High Temperature Blistered Specimens

6. INTERPRETATION

Blistering of ^{252}Cf alpha-exposed specimens at low temperatures is caused by displacement-enhanced agglomeration of helium bubbles in near-surface layers of the specimen. [1-3] Distinctive vertically elongated configurations of the blisters in some areas result from the transport of interstitial atoms produced by the fission fragments into the blisters walls to relieve lateral compressive stress in the helium implanted layer. In this mechanism, reciprocally critical values of both lateral stress and bubble size are necessary to initiate blistering. Since lateral stress depends on the concentration of implanted helium and bubble size depends (at low temperature) on the number of atoms displaced by the fission fragments, then the low helium concentration required for the formation of blisters during ^{252}Cf exposure results from the high number of atoms displaced by the fission fragments.

Enhancement of helium blistering at low temperatures should therefore be expected, to some degree, in any complex radiation environment that produces atom displacements concurrent with implantation of helium in an exposed material. The helium ion and fast neutron radiations to which first wall materials are exposed in a fusion reactor produce such an environment, and the displacement-enhanced blistering should persist to fusion reactor temperatures. The magnitude of the enhanced blistering will depend, however, on the displacement-to-helium atom ratio, which is generally much higher for ^{252}Cf radiations (~3000 displ/He) than for the fusion reactor radiations dominated at first wall surfaces by alpha particles (300 displ/He). The dependence of enhanced blistering on the distribution of displacements is not characterized, but it seems likely, moreover, that the highly localized displacement distributions produced by fission fragments would also produce blistering at lower helium concentrations than the more uniform displacement distributions generated by the fast neutron exposures. The displacement-enhanced helium blistering characteristic of ^{252}Cf -exposed surfaces may, therefore, not be observed in fusion reactor environments, except in devices with uniquely high displacement to helium atom ratios.

The very low helium concentration thresholds for blistering of ^{252}Cf exposed surfaces and the persistence of distinctive blister configurations at 1000°C indicate that the fission-fragment displacement and transport of interstitial atoms enhances blister formation up to temperatures of about 0.6 of the absolute melting point of the capsule material. This temperature approximates the upper limit at

which displacement-induced distortions resulting from fission fragment exposures are observed in other metals, such as uranium. [7] The somewhat higher temperatures at which the displacement-induced distortions are observed in fission-fragment exposed materials compared to fast-neutron exposed materials are attributed to a higher rate of generation of displacements relative to their disappearance at sinks. [8] If displacement enhanced surface distortions of materials do occur in the complex alpha particle and fast neutron radiation environments of fusion reactors, then they should be found only at the somewhat lower temperatures (<0.5 absolute melting point) at which the fast neutron effects are observed.

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