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RADIATION BLISTERING OF STRUCTURAL MATERIALS FOR FUSION DEVICES AND REACTORS

by

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RADIATION BLISTERING OF STRUCTURAL MATERIALS FOR FUSION DEVICES AND REACTORS[†]

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Abstract

Radiation blistering can be an important erosion process when the
structural components of controlled thermonuclear fusion devices or reactors are
exposed to the impact of energetic particles leaving the plasma region. A brief review
of some of the important parameters governing the radiation blistering process is given.
Erosion rates associated with helium blistering in V, Nb and Type 304 stainless steel
are reported for different irradiation temperatures and different projectile energies.

Introduction

During the operation of thermonuclear devices and reactors, the surfaces
of such major components as container walls, beam limiters, diverter walls, and
beam-dump walls of the injector region will be exposed both to the primary plasma
radiations and to secondary radiations from (n, γ), (n, p), (n, α), and other nuclear
reactions and from various secondary-particle and photon-emission phenomena induced
by the primary radiation. For example, during the operation of a large steady-state
D-T fusion reactor, the vacuum walls are expected to reach 400—1000°C and
simultaneously will be bombarded with high fluxes of energetic neutral particles (e.g.,
MeV neutrons and also neutral atoms formed by such processes as charge exchange near
the plasma boundary), energetic ions leaking out of the confining fields, and energetic
photons (e.g., γ rays, bremsstrahlung, soft x-rays, and synchrotron radiation). Such
energetic radiations striking the surfaces of reactor components can cause plasma
contamination and wall erosion by a variety of surface phenomena as discussed
earlier (1—3).

One of the more important erosion processes has been identified to be
radiation blistering (1—5). In this process the gaseous atoms implanted in near surface

regions by energetic projectiles form gas bubbles. Such bubbles near the surface region can grow, and when the pressure is high enough, they can plastically deform the surface skin to form visible blisters that may eventually rupture. The bursts of gas released by blister rupture can contaminate the plasma and peeling of the blister skin can result in wall erosion. A brief review of the experimental results obtained by the authors on the importance of various parameters influencing the blistering process will be given in this paper.

Experimental Procedures

The target materials used for irradiations were of high purity (Marz grade) and were obtained from Materials Research Corporation, except for the Type 304 stainless steel samples which were of commercial grade. All targets were first mechanically polished and then electropolished by techniques described earlier (4—9). The targets were annealed at appropriate temperatures in high vacuum whenever required (4—9). All irradiations were done with a mass analyzed beam of $^4\text{He}^+$ or D^+ ions from a 2-MeV Van de Graaff accelerator and the vacuum in the target chamber was $\sim 1-2 \times 10^{-8}$ Torr during the irradiation. Other details of target irradiation and examination of the irradiated surfaces can be found elsewhere (4—9).

Type of Projectile

In a D-T fusion reactor the energetic projectiles such as deuterium, tritium and helium will strike structural components. Since the permeability of the hydrogen isotopes in most metals is quite different from that of the inert gas atoms like helium, it is important to study the effect these different projectiles have on the blistering process. Figures 1(a) and 1(b) compare the blisters formed in niobium irradiated at 700°C with $^4\text{He}^+$ and D^+ ions, respectively. The blisters formed during helium-ion irradiation (Fig. 1a) after a total dose of 1.0 C/cm^2 are of two types. The larger size blisters have an average diameter of 5—8 μm, while the smaller size blisters have an average diameter of ~0.5 μm. The blisters formed during deuteron irradiation after even a higher dose of 2.0 C/cm^2 are also of two types, but are much smaller in size (Fig. 1b) than the helium blisters. The deuterium blisters appear in different shapes depending upon the orientation of the grains [e.g., dome shaped (10), elongated, crow-foot shaped (6)]. Most of the deuterium blisters are unruptured (Fig. 1b) whereas some of the large helium blisters have ruptured. The observation
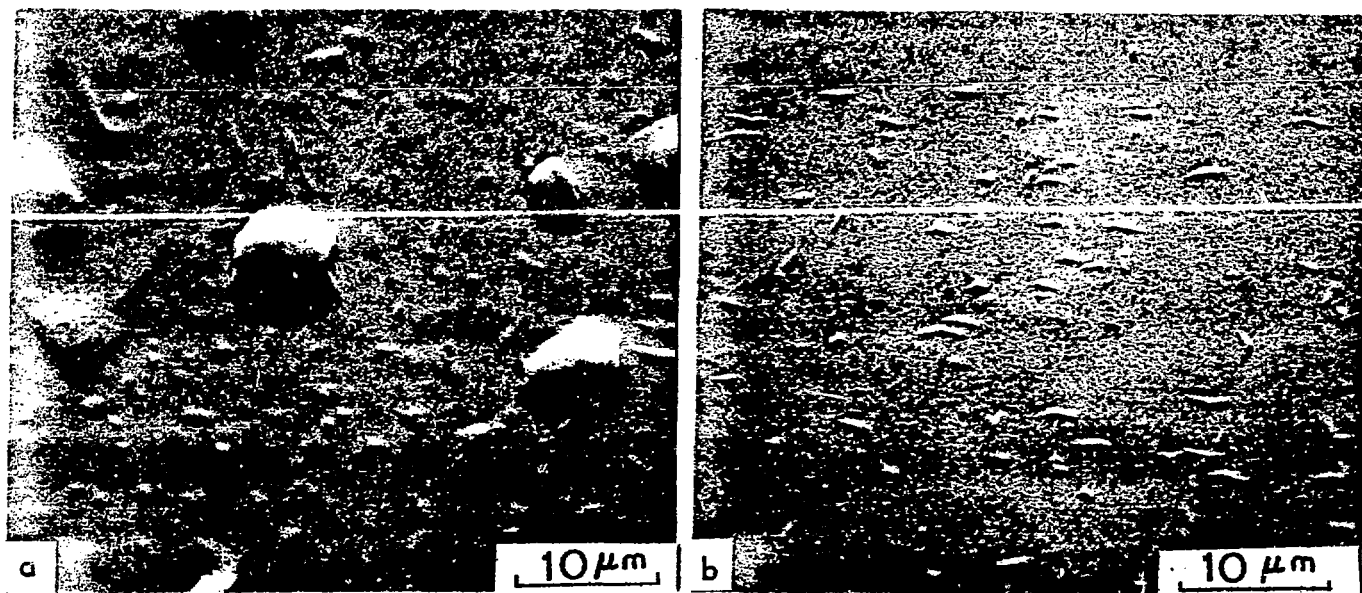
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FIG. 1

Scanning electron micrographs (SEMs) of annealed polycrystalline niobium surfaces irradiated at 700°C (a) with 0.5-MeV ^{4}He$^+$ ions to a total dose of 1.0 C/cm^2 (b) with 0.25-MeV D$^+$ ions to a total dose of 2.0 C/cm^2.

that blister size for deuteron irradiation is smaller than for helium ion irradiation under nearly comparable conditions (note that the dose for deuteron irradiation was higher) can be related to the fact that the gas buildup is greatly reduced for deuterium in niobium since the deuterium permeability (determined by the solubility and diffusivity) is many orders of magnitude larger than that of helium. For example, the diffusion coefficient of deuterium in niobium (11) is $D_D = 1.3 \times 10^{-4}$ cm^2/sec at 800°C, while that for helium in niobium (12) ranges from 10^{-19} to 10^{-14} cm^2/sec between the temperatures of 600°C and 1200°C.

Energy of Projectile

Figures 2(a) to 2(f) illustrate the effect of projectile energy on helium blister formation in niobium irradiated at room temperature. Irradiation to a dose of 0.5 C/cm^2 with 100-keV ^{4}He$^+$ ions results in a blister density of ~2×10^6 blisters/cm^2, and the average blister diameter ranges from 1 to 30 μm (Fig. 2a). Many of the large blisters show exfoliated skins and in some cases (marked by arrows) a second blister skin has ruptured and fallen off. Figure 2(b) shows an enlarged view of a blister skin from which the skin thickness can be estimated to be ~0.4 μm. Irradiation with 250-keV ^{4}He$^+$ ions to the same dose of 0.5 C/cm^2 showed blisters with average
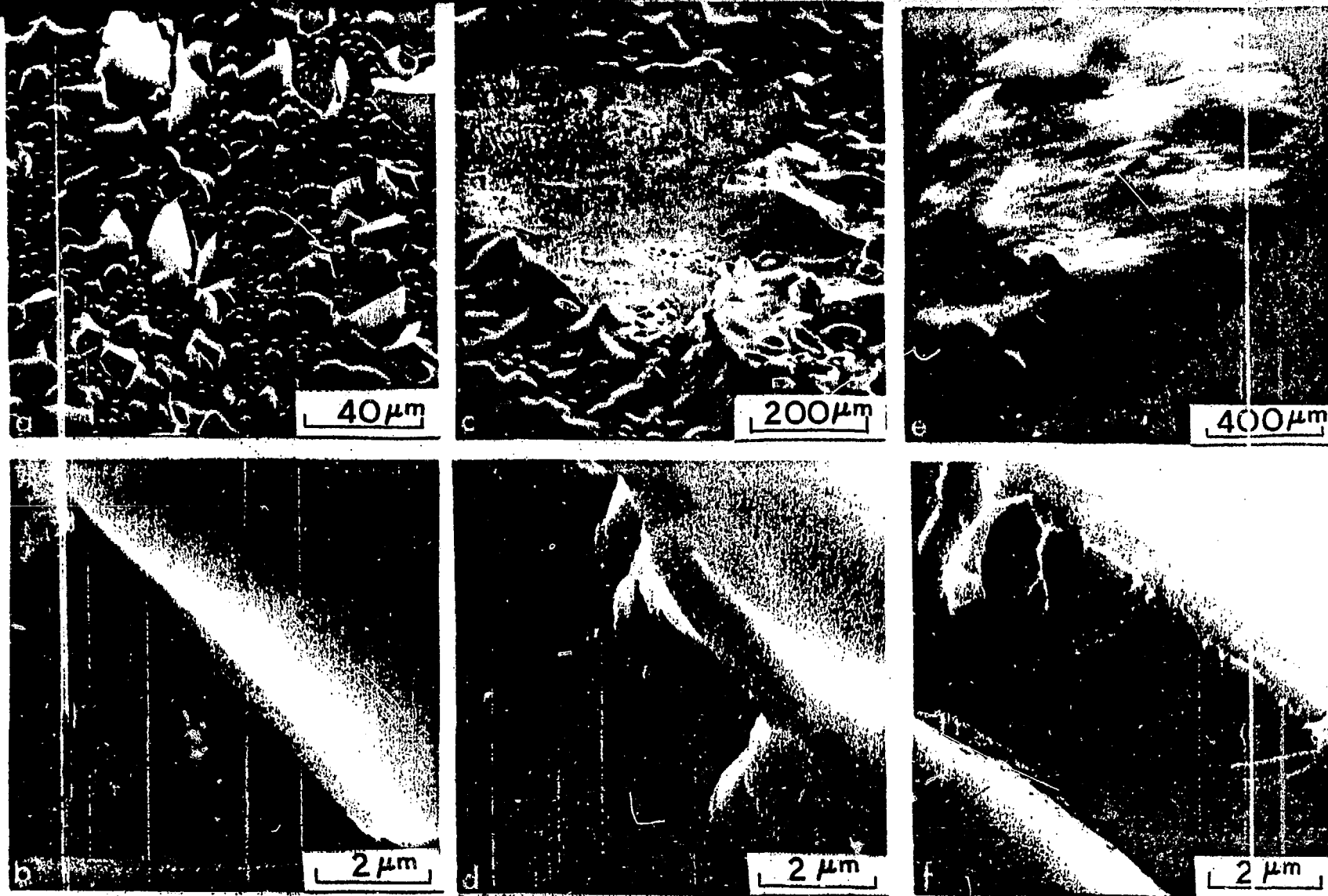
FIG 2.

SEMs of polycrystalline niobium surfaces irradiated at room temperature (a) with 100-keV ^{4}He$^+$ ions to a total dose of 0.5 C/cm^2, (b) an enlarged view of the blister skin in (a), (c) with 0.5-MeV ^{4}He$^+$ ions to a dose of 1.0 C/cm^2, (d) an enlarged view of blister skin in (c), (e) with 1.5-MeV ^{4}He$^+$ ions to a dose of 1.0 C/cm^2 and (f) an enlarged view of the blister skin in (e).

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diameters ranging from 3 to 30 μm and the blister density was $\sim 1.2 \times 10^6$ blisters/cm^2.
For 0.5-MeV ^{4}He$^+$ ion irradiation to a total dose of 1.0 C/cm^2 a very large blister
(Fig. 2c) occupied about two-thirds of the bombarded area. From an enlarged view of
a portion of the ruptured skin shown in Fig. 2(d) the skin thickness is determined to be
~ 1.1 μm. On further increase of the projectile energy to 1.5 MeV only one large
blister occupied the entire bombarded area (Fig. 2e). A portion of the ruptured skin
can be seen in Fig. 2(f) and the skin thickness is found to be ~ 2.8 μm. From a
comparison of the blister skin thicknesses for different helium ion energies (Figs. 1b,
1d, and 1f) it is clear that the blister skin thickness is increasing with increasing
projectile energy. The measured skin thicknesses for different projectile energies fall
within 30% of the corresponding projected ranges calculated according to Schiøtt (13).

The increase in blister size with increase in projectile energy can be
understood if one considers the expressions for the gass pressure "p" inside a blister
as discussed earlier (4). The pressure "p" is directly proportional to the skin thickness
"t" and inversely proportional to the square of the radius "a" of the blister. Hence for
a given blister height "h" and pressure "p" (which is roughly porportional to the total
dose), a higher projectile energy (and correspondingly higher skin thickness "t") will
require a larger blister radius "a". At high temperature (e. g., niobium at 900°C) the
effect of projectile energy on blister size is less pronounced. It should be pointed out
that the erosion rates S estimated by techniques described earlier (4, 5) for 100-keV
and 0.5-MeV helium ion irradiations (see Figs. 1a and 1c, respectively) yield values
of $S \sim 1.0 \pm 0.4$ and $S \sim 0.20 \pm 0.05$ niobium atoms per incident helium ion,
respectively. Such an increase in erosion rate with decreasing projectile energy has
also been observed for Type 304 stainless steel (9, 14).

Total Dose and Dose Rate

It has been observed (6—8) that for helium ion irradiation of niobium the
blister size increases with increasing total dose. Figures 3(a), 3(b) and 3(c) show
blisters formed in Type 304 stainless steel irradiated at 450°C with 0.5-MeV ^{4}He$^+$ ions
to total doses of 0.1, 0.5 and 1.0 C/cm^2, respectively. For a dose of 0.1 C/cm^2 the
average blister diameter is ~ 700 μm (Fig. 3a) and a large portion of the irradiated area
is occupied by this single blister. At a higher dose of 0.5 C/cm^2 three exfoliated skin
layers are observed (Fig. 3b) as compared with one for a dose of 0.1 C/cm^2 (Fig. 3a).
For an even higher dose of 1.0 C/cm^2 the number of exfoliated layers increases to five

200 μm

200 μm

200 μm

SEMs of surfaces of annealed Type 304 stainless steel after irradiation at 450°C with 0.5-MeV ^{4}He$^+$ ions for dotal doses of (a) 0.1 C/cm^2 (b) 0.5 C/cm^2 and (c) 1.0 C/cm^2.
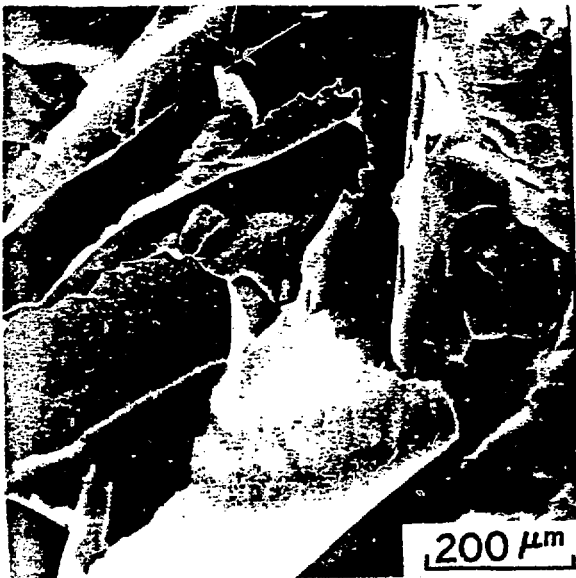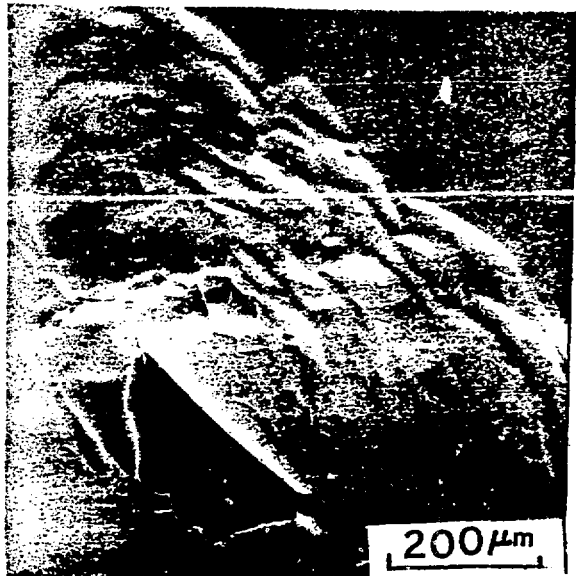
in some regions (Fig. 3c). The estimated erosion rates for the three doses 0.1, 0.5 and 1.0 C/cm^2 are 0.1 ± 0.05, 0.45 ± 0.1 and 0.8 ± 0.2 atoms per incident ion, respectively. One should note that within the experimental error the erosion rates depend nearly linearly on the dose. One should keep in mind that these rates are determined only from that part of the blister skin which has been totally lost. Therefore the observed increase in the erosion rates with increasing dose (for the dose range studied here) is related to the increase in the exfoliation of blisters and to the subsequent loss of blister skin with increasing dose.

The effect of total dose on the blister size is less pronounced at high target temperature (e.g., Nb or V at 900°C) than at room temperature (6, 15). For example, for annealed polycrystalline vanadium irradiated at 900°C with 0.5-MeV ^{4}He$^+$ ions one observes that as the dose is increased by one order of magnitude from 0.1 to 1.0 C/cm^2 the mean value of the average blister diameters shows (15) only a slight increase from 6.2 to 7.4 μm. This is in part due to a higher rate of helium

release at 900°C than at room temperature (16). This effect of temperature will be discussed later.

At a particular irradiation temperature the rate of gas buildup near the implant depth and the subsequent blister formation will depend on the dose rate (i. e., the incident ion flux) and the rate of gas release from the surface. Depending on the balance of gas trapping and gas release, there may or may not be an effect of dose rate on blister formation. Recent results on blistering in helium implanted vanadium show (15) that with increasing dose rate from 1×10^{15} ions/(cm^2-sec) the blister density increases from $(1.0 \pm 0.5) \times 10^4$ to $(7 \pm 2) \times 10^4$ blisters/cm^2. For deuterium blistering in niobium the effect of dose rate is found to be much more pronounced than for helium blistering for similar irradiation conditions. An increase in the dose rate has also been observed (17) to reduce the critical dose for blister formation in proton irradiated molybdenum.

Target Temperature

The target temperature is one of the most important parameters affecting blister formation. Figure 4A shows a plot of the erosion rates vs. target temperature for helium blistering of Type 304 stainless steel. It can be seen in Fig. 4A that the maximum erosion rate occurs at an irradiation temperature of ~450°C. A qualitatively similar temperature dependence has been observed for Type 316 stainless steel (18). Another striking result that is apparent from Fig. 4A is that as the projectile energy is reduced from 0.5 MeV to 100 keV the erosion rate increases by approximately a factor of 7 for the same dose of 0.5 C/cm^2 and a temperature of ~450°C. Figure 4B shows the effect of temperature on the blister formation in polycrystalline vanadium irradiated with 0.5-MeV ^{4}He$^+$ ions for the same dose of 1.0 C/cm^2. As the irradiation temperature is increased from room temperature to 300°C the number of exfoliated skin layers increases from one to two (c.f. Figs. 4a and 4b). Irradiation at 500°C shows (Fig. 4c) an even stronger increase in blister exfoliation since six exfoliated skin layers are observed in certain regions. On further increasing the irradiation temperature to above 800°C blisters (Fig. 4d) with average diameters of only 10-20 μm and with no large scale exfoliation are observed. The erosion rate appears to be at a maximum somewhere between 300°C and 500°C.

The observed change in erosion rates with temperature is related to the strong temperature dependence of the yield strength of the material. For example, the
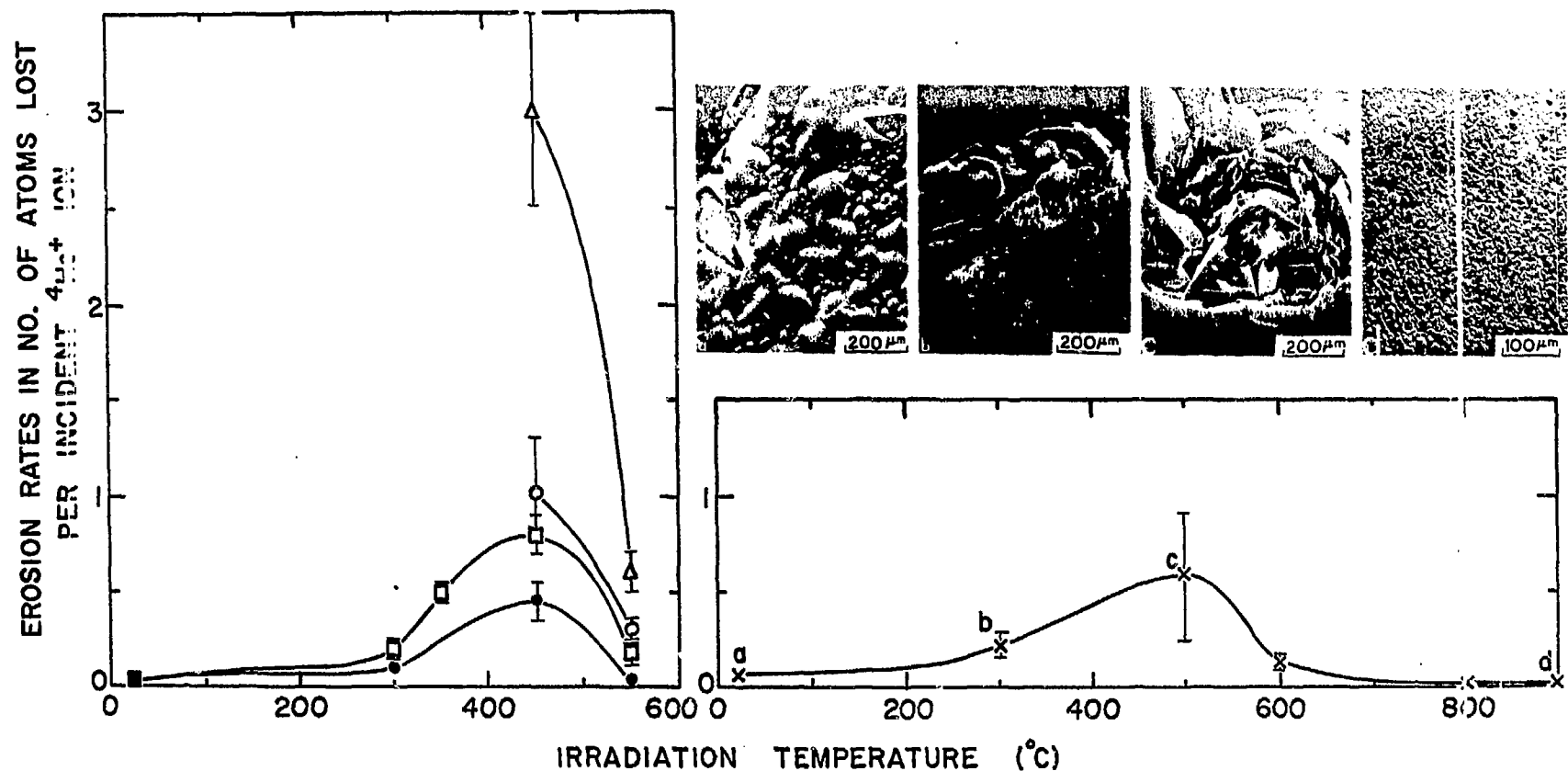
FIG. 4

(A) Erosion rates for annealed Type 304 stainless steel as a function of irradiation temperature for different projectile energies and total doses; : 0.5 C/cm^2 with 0.5-MeV ^{4}He$^+$, : 1.0 C/cm^2 with 0.5-MeV ^{4}He$^+$, : 0.1 C/cm^2 with 100-keV ^{4}He$^+$, : 0.5 C/cm^2 with 100-keV ^{4}He$^+$. (B) Erosion rates for annealed polycrystalline vanadium as a function of irradiation temperature for 0.5-MeV ^{4}He$^+$ ions and for a total dose of 1.0 C/cm^2. The figures (a)-(d) at the top of Fig. (B) show SEMs of irradiated surfaces for the temperatures indicated therein.

yield strength of annealed 304 stainless steel at 450°C is half of its value at room temperature (19). Similarly the tensile strength of annealed vanadium at 900°C is less than a third of its value at room temperature (20). Thus, for a particular dose the collected gas in a bubble can deform the surface skin more readily at high temperature (e.g. stainless steel at 450°C) than at room temperature. In addition the kinetic pressure of the gas in the bubble will increase with temperature and may also enhance this process. However, at very high temperatures (e.g. at 900°C for vanadium) helium may be released through the surface either by atomic diffusion or by migration of small bubbles. Some recent work (21) shows that helium bubble mobility in niobium at 950°C is at least two orders of magnitude higher than that calculated earlier by Martin (22). Mass spectrometric evidence for helium release for niobium, vanadium and 316 stainless steel has been reported (16, 18). Furthermore, at high temperatures partial annealing of the lattice defects may occur and thereby reduce the nucleation and growth rate of the helium bubbles. Therefore, the amount of helium trapped in the lattice will be affected by the helium release rate and the bubble nucleation rate and will in turn determine the size and density of helium bubbles and the subsequent blister size. Thus the degree of blistering and exfoliation of blister skin is maximized if the temperature is high enough so that the surface can be deformed easily (lower yield stress) but low enough so that the helium release from the surface is still very small.

The critical dose for the appearance of blisters is strongly temperature dependent. For example, for helium bombardment of annealed polycrystalline niobium at room temperature for a total dose of 0.1 C/cm^2 no blisters could be detected (4); but blisters were readily detected (6) for 900°C irradiation even after a dose of only 0.01 C/cm^2. This temperature dependence of critical dose for blister appearance is also related to the temperature dependence of yield strength as discussed above.

Other Parameters

In addition to the parameters discussed above there are many other parameters that affect the degree of blistering and only a few will be mentioned here. Recent experiments (7) on helium blistering in cold worked and annealed niobium and vanadium show that for room temperature irradiation the blister size and the degree of blistering is reduced by cold working. Cold working has also been observed to reduce the void number density in neutron irradiated stainless steel (23, 24), but the void size does not change very much. However, for irradiation at 900°C, the degree of

blistering is similar for cold worked and annealed samples for both vanadium and niobium.

In bombarding monocrystalline metals with helium ions or deuterons, it has been observed (25) that the blister shape and the orientation of the blisters with respect to each other depend strongly on the crystallographic orientation of the irradiated surface plane of the target. The shapes of blisters formed on the (111), (110) and (001) surface planes of monocrystalline niobium samples irradiated at 900°C with axially channeled 0.5-MeV $^4He^+$ ions have been related to the intersection of certain active slip planes with the surface plane of the monocrystal during blister formation (25). It has also been observed that for the helium blisters formed on monocrystals the blister size is greater if the projectiles are axially channeled than if they were unchanneled (8, 17, 25). The blister density in helium implanted (111) niobium monocrystals irradiated at 900°C has been observed to decrease by approximately two orders of magnitude for the channeled helium projectiles as compared to the unchanneled ones (26). Such a behavior is not unexpected since the channeling of projectiles helps to reduce the radiation damage in near surface regions (corresponding to the initial part of the trajectory of the channeled projectiles) and thereby reduces the number of radiation induced nucleation sites for bubble formation.

Conclusion

The material presented in this paper suggests that helium blistering should be considered a serious erosion process for such fusion devices and reactor components as container walls, direct converter surfaces, diverter surfaces, and beam limiters. For example, from the erosion rates quoted above for 304 stainless steel for 100-keV He^+ ion irradiation to a total dose of 0.5 C/cm^2 at 450° C an annual thickness loss of about 0.09 mm per year can be estimated for a helium flux of 1×10^{13} projectiles $cm^{-2}sec^{-1}$. As has been pointed out earlier (5) such annual thickness loss values will be enhanced by the vaporization of the blister skin due to heating by energetic photon absorption (e.g. bremsstrahlung). Furthermore, it should be pointed out that our results on the temperature dependence of helium blistering of niobium and stainless steel indicate that the low wall temperatures considered by some authors for the operation of fusion reactors [e.g. 466° C of a niobium wall (27), and 300 — $\sim 500^{\circ}$ C of a stainless steel wall (28)] would lead to significantly more wall erosion than if such surfaces would be operated at a higher, more optimum wall temperature (e.g. $\sim 900^{\circ}$ C

for niobium).   Work is in progress to find ways to reduce the helium blistering process by choice of materials and their structures (e.g. porous materials).

Acknowledgments

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