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EFFECT OF He^{+} and D^{+} ION BEAM FLUX ON BLISTER FORMATION IN NIOBIUM AND VANADIUM

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ABSTRACT

The effect of incident ion beam flux on the blister formation in annealed polycrystalline niobium and vanadium has been investigated for 0.5-MeV He^+ and 0.25-MeV D^+ projectiles. For the He^+ ions the flux was varied from 1×10^{13} ions/(cm^2-sec) to 1×10^{15} ions/(cm^2-sec), the targets were held at 900°C, and the total dose was varied from 0.1 to 1.0 C/cm^2. For the D^+ ion irradiation the flux was varied from 1×10^{14} to 1×10^{15} ions/(cm^2-sec), the niobium targets were held at 700°C, and the total dose was 2.0 C/cm^2. For both the helium implanted vanadium and niobium the blister density shows a stronger flux dependence than the average blister diameter. For deuteron implanted niobium the blister size increased as the flux was increased from 1×10^{14} to 1×10^{15} ions/(cm^2-sec).

INTRODUCTION

Irradiation of metal surfaces such as niobium and vanadium with He^+ and D^+ projectiles are known [1-9] to form blisters on surfaces. Earlier blistering studies in other metals can be found in Refs. 1-9 and the references cited therein. The results of these studies [1-9] reveal that the shape, size, density and the degree of blister formation (defined here as the fraction of total irradiated area that is occupied by blisters) depend on such parameters as projectile energy [3,6], target temperature [2-8], channeling condition of the projectile [2,3], orientation of the irradiated surface planes [2,3,6,9-12], initial defect concentration in the target [1,4], and total dose [1,3,4,6]. Another important irradiation parameter is the projectile flux. To our knowledge the effect of flux on blister formation has received little attention. At a particular irradiation temperature the rate of gas build-up near the implant depth and the subsequent blister formation will depend on 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 the projectile flux on blister formation. In the present paper this effect of projectile flux on blister formation in annealed polycrystalline niobium and vanadium was investigated for two gases - namely helium and deuterium; the former having extremely low permeability and the latter having extremely high permeability.

Such studies of blistering in niobium and vanadium by implantation with helium ions with energies ranging from 0.1 to 3.5-MeV are of interest to the controlled thermonuclear fusion reactor program. For example, blistering processes can play an important role in the operation of D-T fusion reactors [12-14] in that they lead to severe erosion of the wall surfaces exposed to impact by energetic projectiles such as helium.

EXPERIMENTAL TECHNIQUES

The polycrystalline niobium and vanadium foils were obtained from Materials Research Corporation (Marz grade). The foils were first given a fine metallographic polish and then annealed at 1200°C for 2 hours in a vacuum of $3\text{-}5 \times 10^{-7}$ Torr and finally electropolished by techniques described earlier [1,4]. The polished areas had an average surface roughness of less than 0.02 μm as determined by a scanning electron microscope with a resolution limit of about 0.02 μm. A mass analyzed beam of $^4He^+$ ions with energy of 0.5-MeV, or of D^+ ions with energy of 0.25-MeV from a 2-MeV Van de Graaff accelerator was highly collimated (half angle of divergence = 0.01°) and was incident parallel (within ∿0.1°) to the surface normal of the target. For high temperature irradiations thin targets (∿25 μm thick) were used and were heated by ohmic heating. The target temperature was measured with an optical pyrometer; a correction was applied for absorption in the window of the target chamber. For He^+ ion irradiations the ion flux was varied from 1×10^{13} ions/(cm^2-sec) to 1×10^{15} ions/(cm^2-sec) and for D^+ ion irradiation the ion flux was varied from 1×10^{14} to 1×10^{15} ions/(cm^2-sec). In each case the irradiated area on each target was a circular spot of 2 mm in diameter. During the irradiation the vacuum in the target chamber was maintained at about $1\text{-}2 \times 10^{-8}$ Torr by ion pumping. The irradiated target surfaces were examined with a scanning electron microscope, Cambridge Stereoscan model Mark IIA. The size distribution of the blisters were measured with a Zeiss particle-size analyzer from enlarged micrographs. For blisters with irregular shapes the average diameter of the blister was defined as the diameter of a circle having the same area as the blister. The mean values of the average blister diameters presented in this paper are the weighted average values determined from the size distribution of the blisters. The results on blister size, density and fraction of total irradiated area occupied by blisters given in this paper represent average values from at least two irradiation runs. In cases where considerable scatter was observed the runs were repeated three or four times.

RESULTS

The blister formation in annealed polycrystalline niobium and vanadium samples will be described separately for the two types of projectiles used, $^4He^+$ and D^+ ions.

$^4He^+$-Ion Irradiation of Niobium

The scanning electron micrographs in Figs. 1(a), 1(b) and 1(c) show the blisters formed after annealed polycrystalline niobium surfaces at 900°C were irradiated with 0.5-MeV $^4He^+$ ions to a total dose of 0.1 C/cm^2 for fluxes of 1×10^{13}, 1×10^{14} and 1×10^{15} ions/(cm^2-sec), respectively. In each micrograph two types of blisters can be distinguished. The smaller size (type I) blisters have diameters ranging from approximately 0.3 to 1.0 μm (see insets in Figs. 1a–c) and are found to be nearly independent of the fluxes used. The other type (type II) of blisters have larger diameters, ranging from 3 to 15 μm. The value for the average blister density of the smaller size blisters is found to be approximately $(5 \pm 3) \times 10^6$ blisters/cm^2, and appears to be independent of the three dose rates studied within the quoted experimental error. For the larger blisters the blister density increases from $(3 \pm 2) \times 10^4$ blisters/cm^2 to $(6 \pm 3) \times 10^4$ blisters/cm^2 as the flux is increased from 1×10^{13} to 1×10^{14} ions/(cm^2-sec), but a further increase in flux to 1×10^{15} ions/(cm^2-sec) does not seem to change this value within the quoted error limits. The fraction of total irradiated area that is occupied by the blisters of both types shows a slight increase with increasing flux. The values increase from approximately (1.5 ± 0.5)% to (3.5 ± 1.0)% as the flux is increased from 1×10^{13} to 1×10^{15} ions/(cm^2-sec).

The scanning electron micrographs in Figs. 2(a) and 2(b) show blisters formed on polycrystalline niobium surfaces after irradiation under similar conditions as described above, except for a higher total dose of 1.0 C/cm^2 and for fluxes of 1×10^{14} ions/(cm^2-sec) and 1×10^{15} ions/(cm^2-sec), respectively. Again two types of blisters can be distinguished. For the flux range studied the average diameters of the smaller size blisters range from 0.3 to 1.8 μm while those for the large size blisters range from 3-15 μm. A comparison of the blister diameters for a given flux but for the two total doses of 0.1 and 1.0 C/cm^2 shows that the range of blister

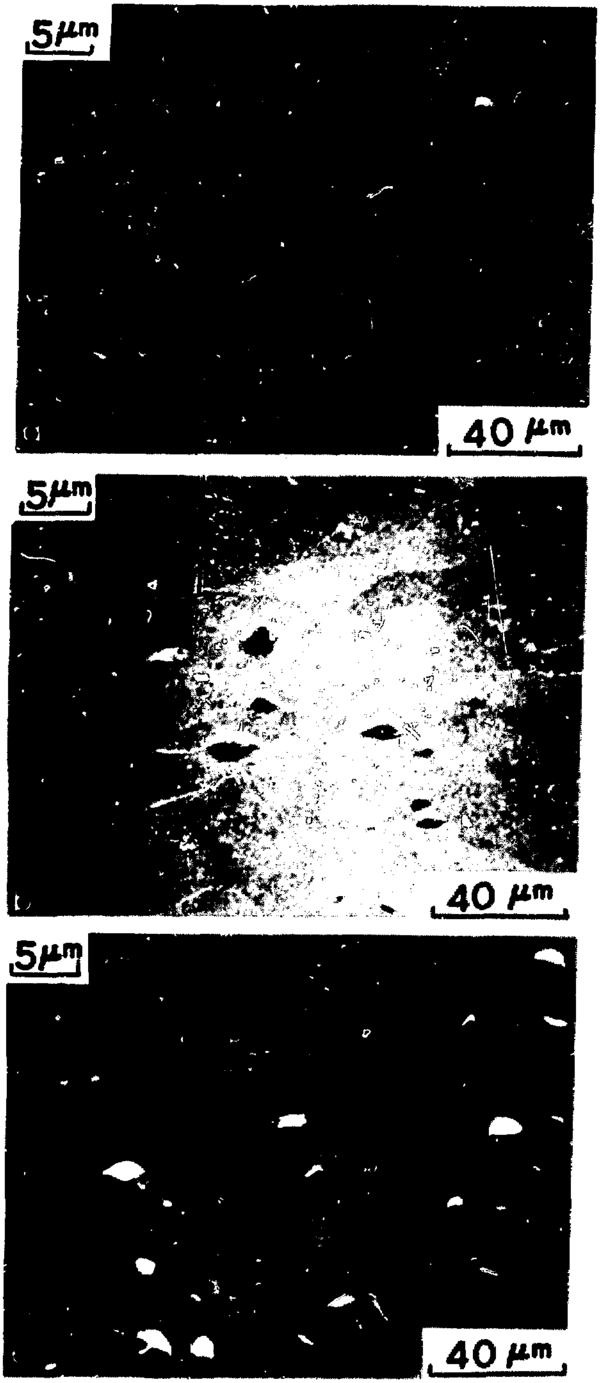


Fig. 1. Scanning electron micrographs (SEM) of surfaces of annealed polycrystalline Nb after irradiation at $900^{0}C$ with 0.5-MeV $^{4}He^{+}$ for a total dose of 0.1 C/cm^{2} at fluxes of:

(a) 1×10^{13} ions/(cm^{2}-sec)
(b) 1×10^{14} ions/(cm^{2}-sec)
(c) 1×10^{15} ions/(cm^{2}-sec)

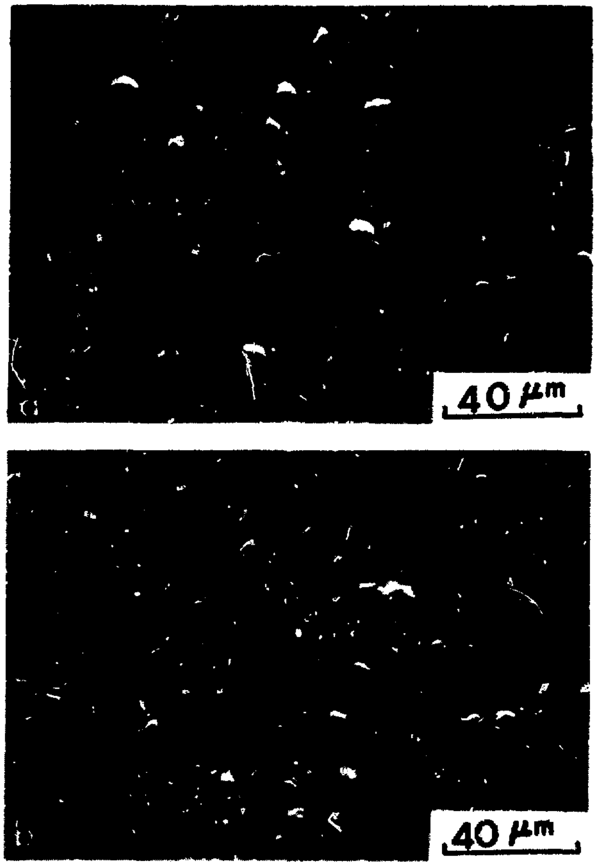


Fig. 2. SEM's of surfaces of polycrystalline Nb after irradiation at 900°C with 0.5-MeV $^4He^+$ for a total dose of 1.0 C/cm^2 at fluxes of (a) 1×10^{14} ions/(cm^2-sec) and (b) 1×10^{15} ions/(cm^2-sec).

diameters has practically the same value for the two doses studied. The values for the blister densities for both the smaller and the larger size blisters are approximately $(8 \pm 2) \times 10^6$ blisters/cm^2 and $(1.5 \pm 0.5) \times 10^5$ blisters/cm^2, respectively, and show (within the quoted error limits) no significant flux dependence. The average value for the fraction of total irradiated area that is occupied by blisters of both size classes is (9 ± 2)% and is, within the quoted error limits, the same for the flux range studied.

$^4He^+$-Ion Irradiation of Vanadium

The micrographs in Figs. 3(a), 3(b) and 3(c) show typical blisters formed after irradiating annealed polycrystalline

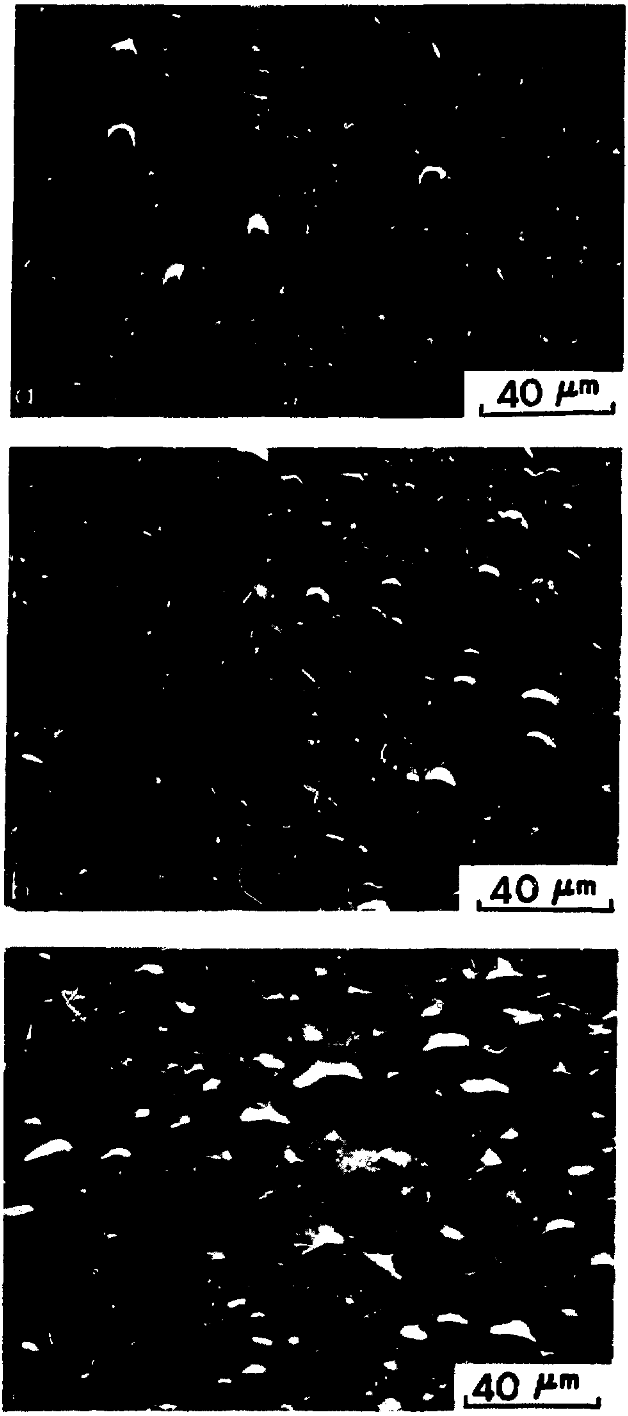


Fig. 3. SEM's of surfaces of annealed polycrystalline V after irradiation at 900°C with 0.5-MeV $^{4}He^{+}$ for a total dose of 0.1 C/cm^2 at fluxes of:

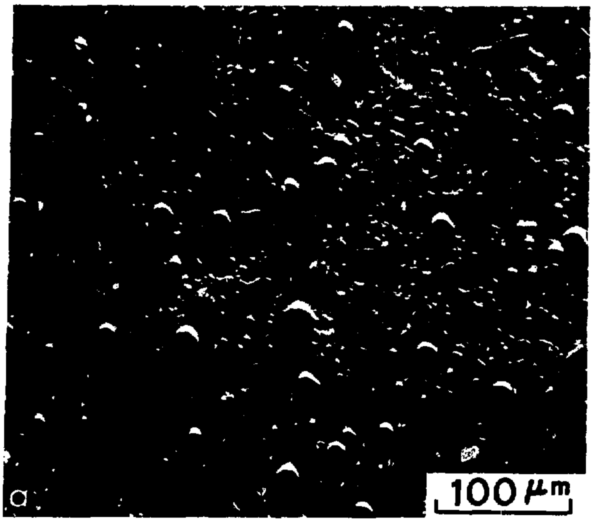
(a) 1×10^{13} ions/(cm^2-sec)
(b) 1×10^{14} ions/(cm^2-sec)
(c) 1×10^{15} ions/(cm^2-sec)

vanadium surfaces held at 900°C with 0.5 MeV $^4He^+$ ions to a total dose of 0.1 C/cm^2 for fluxes of 1 x 10^{13}, 1 x 10^{14}, 1 x 10^{15} ions/(cm^2-sec), respectively. The average diameters of blisters of both types do not seem to depend significantly on beam flux. For the small size blisters the value for the average diameter ranges from approximately 0.1 to 0.3 μm and for the large size blisters the average diameter ranges from about 3 to 15 μm for the range of fluxes studied. It is observed that the value for the blister density for the small size blisters is (5 ± 2) x 10^6 blisters/cm^2 and that within the error limits quoted this value is the same for the three fluxes. For the larger size blisters, however, a dependence of the blister density on the flux is observed. The values increase from (1.0 ± 0.5) x 10^4, to (3 ± 1) x 10^4 and to (7 ± 2) x 10^4 blisters/cm^2 as the flux increases from 1 x 10^{13}, to 1 x 10^{14} and to 1 x 10^{15} ions/(cm^2-sec), respectively.

Figure 4 shows blisters formed after irradiating annealed polycrystalline vanadium surfaces under the same irradiation condition as those described above (Fig. 3), but for a higher total dose of 1.0 C/cm^2. The micrographs in Fig. 4(a) and 4(b) show typical blisters observed for fluxes of 1 x 10^{14} and 1 x 10^{15} ions/(cm^2-sec), respectively. It is found that the blister diameters for both types of blisters do not show an observable flux dependence. For the smaller size blisters the value for the diameter ranges from 0.5 to 1.0 μm and for the larger size blisters it ranges from 3 to 15 μm for the two flux values studied. The blister density for the small size blisters is about (8 ± 3) x 10^6 blisters/cm^2 for the two fluxes studied. The value for the large size blisters, however, increases from (2 ± 1) x 10^4 to (7 ± 2) x 10^4 blisters/cm^2 as the flux is increased from 1 x 10^{14} to 1 x 10^{15} ions/(cm^2-sec). As the dose is increased from 0.1 to 1.0 C/cm^2 the range of the values of the average blister diameters does not show an appreciable change but the mean value of the average blister diameters shows a slight increase from 6.2 to 7.4 μm. It is of interest to note that the values for the average blister diameters for both types of blisters are similar to those observed for niobium surfaces (Fig. 2) within the ranges quoted. The fraction of total irradiated area that is occupied by both types of blisters is found to increase from approximately 5% to 15% as the flux is increased from 1 x 10^{14} to 1 x 10^{15} ions/(cm^2-sec).

D^+-Ion Irradiation of Niobium

The micrographs in Figs. 5(a), 5(b) and 5(c) show blisters formed on the surfaces of annealed polycrystalline niobium at 700°C after irradiation with 250-keV deuterons to a total dose of 2.0 C/cm^2 for the fluxes of 1 x 10^{14}, 4 x 10^{14} and 1 x 10^{15} ions/(cm^2-sec), respectively. It has been observed earlier [6,9] that the blister shape strongly depends on the orientation of the grains.



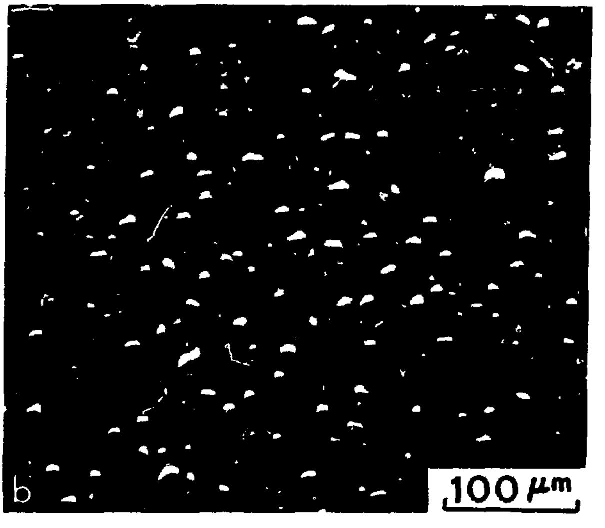


Fig. 4. SEM's of annealed polycrystalline V surfaces after irradiation at 900°C with 0.5-MeV $^4He^+$ for a total dose of 1.0 C/cm^2 at fluxes of (a) 1 x 10^{14} ions/(cm^2-sec) and (b) 1 x 10^{15} ions/(cm^2-sec).

In Fig. 5(a) most of the blisters in one of the grains exhibit three fold symmetry resembling the "crow-foot" shaped blisters described earlier [2,3,6]. Since the orientation of this particular grain is unknown, one can only conjecture that the prongs of the "crow-foot" shaped blisters are aligned along the [$\bar{1}2\bar{1}$], [$\bar{1}\bar{1}2$] and [$2\bar{1}\bar{1}$] directions of the niobium lattice was observed previously

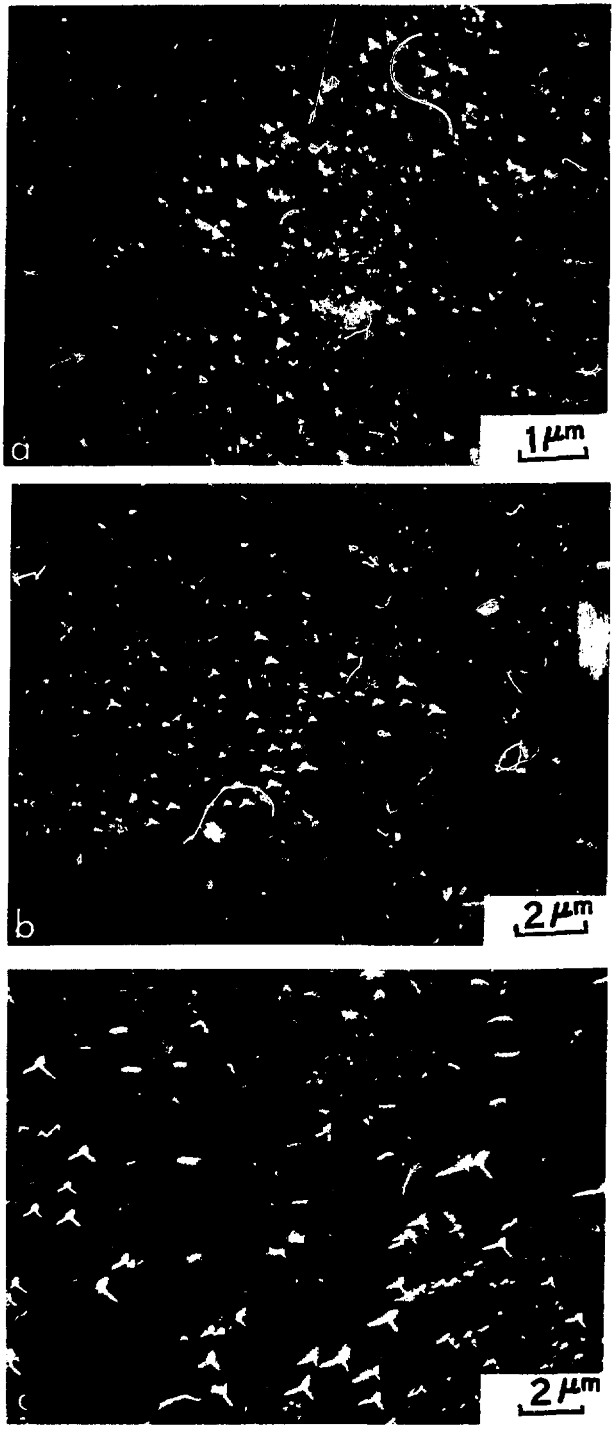


Fig. 5. SEM's of annealed polycrystalline Nb surfaces after irradiation at 700°C with 250-keV D^+ ions to a total dose of 2.0 C/cm^2 at fluxes of:
(a) 1×10^{14} ions/(cm^2-sec)
(b) 4×10^{14} ions/(cm^2-sec)
(c) 1×10^{15} ions/(cm^2-sec)

[2,3,6] for a (111) surface plane of a monocrystalline niobium target irradiated at 900°C with $^4He^+$ ions.

If one compares the size of such crow-foot shaped blisters for the different fluxes, it is seen that the length of each of the three prongs of a particular blister increases by approximately a factor of three as the flux is increased from 1×10^{14} to 1×10^{15} ions/(cm^2-sec) as can be seen in Figs. 5(a) and 5(c). The density of the "crow-foot" shaped blisters decreases from approximately $(3 \pm 1) \times 10^8$ to $(1.0 \pm 0.5) \times 10^8$ blisters/cm^2, as the flux increases from 1×10^{14} to 1×10^{15} ions/(cm^2-sec). It may be noted that the blister density for grains of other orientations can be different by many orders of magntidue [see also Fig. 3 in Ref. 6].

DISCUSSION

The results presented here show that the blister density for the large size blisters (type II) is flux dependent for the case of niobium and vanadium irradiated at 900°C with helium ions. Such a dependence can in part be related to the increase in displacement rate of lattice atoms with increasing flux. For the flux range studied the displacement rate can vary approximately from 10^{-4} to 10^{-2} dpa/sec. As the displacement rate increases the vacancy supersaturation increases and this in turn can cause a sharp increase in the void nucleation rate for the ranges of temperature and displacement rates considered here (see discussion by Wiedersich and Katz [15] for nickel at 600°C). The effect of higher displacement rates on the nucleation rates of helium bubbles may be similar. However, at this time it is not quite clear why for the helium ion irradiated niobium samples the effect of flux is less pronounced. It is possible that the differences in the interstitial impurity concentrations in niobium and vanadium may be responsible in part for the observed differences.

For helium ion irradiation at 900°C the blister size for both types of blisters does not change with flux significantly for vanadium, but for niobium the blister size changes slightly with flux. The blister size for 900°C irradiation is considerably smaller than for room temperature irradiation [16], which is in part due to the enhanced helium release [5,18] at 900°C. The simultaneous processes of gas implantation, diffusion and surface erosion have been discussed by Biersack [19]. For our irradiation conditions the loss of blister skin due to sputtering amounts to about 0.1-0.2 percent of the total skin thickness. Therefore, this sputtering erosion process plays an insignificant role in the blister formation and rupture in the present experiments. It appears that the blister size is much more strongly dependent on the irradiation temperature than on the incident beam flux (for the flux range studied), even though a higher beam flux can cause an increased helium release via an increased vacancy production.

The blisters formed in annealed polycrystalline niobium after deuteron irradiation show an increase in size as the flux is increased (Fig. 5(a)-5(c)). As has been pointed out earlier [6] the smaller amount of blistering by deuteron irradiation than by helium-ion irradiation even for doses which are much higher for deuteron than for helium, can be related to the fact that the solubility and diffusivity in niobium is many orders of magnitude larger for deuterium than for helium. The diffusion coefficient of deuterium in niobium [20] is $D_D = 1.3 \times 10^{-4}$ cm^2/sec at 800°C while that for helium in niobium [21] it can vary from 10^{-19} to 10^{-14} cm^2/sec as the temperatures are changed from 600°C to 1200°C.

Since the deuterium trapping rate in the lattice has to be larger than the deuterium release rate, in order to build up large enough bubbles for blister formation, it can be expected that the bubble nucleation and growth rate will be flux dependent. This is in accordance with our observation that the deuteron blister size increased as the flux was increased (for the flux range studied). In addition, the increase in blister size with increasing flux can in part be caused by the lowering of the threshold dose for blister formation with increasing flux. For example, Verbeek and Eckstein [22] observed for the case of 15-keV proton irradiation of molybdenum, that the threshold dose decreased with increasing flux. Qualitatively, a similar dependence can be expected to occur for our case of deuteron irradiated niobium.

The observation that the fraction of the irradiated area occupied by both types of helium blisters in vanadium formed at 900°C increases with increasing flux is related to the flux dependence of blister density and blister size, as described above.

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