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with Helium Ions of Different Energies (3-500 keV)

By

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Radiation Blistering of Nb Implanted Sequentially
with Helium Ions of Different Energies (3-500 keV)

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Abstract

Cold rolled, polycrystalline niobium samples were irradiated at room temperature with $^4\text{He}^+$ ions sequentially at 14 different energies over an energy range from 3 keV - 500 keV. The dose for each energy was chosen to give an approximately uniform concentration of helium between the implant depths corresponding to 3 keV and 500 keV. In one set of experiments the irradiations were started at the Kurchatov Institute with 3 keV $^4\text{He}^+$ ions and extended up to 80 keV in several steps. Subsequently, the same target area was irradiated with $^4\text{He}^+$ ions at Argonne National Laboratory (ANL) starting at 100 keV and increased to 500 keV in steps of 50 keV. Another set of irradiations were started at ANL with 500 keV $^4\text{He}^+$ ions and continued with decreasing ion energies to 100 keV. Subsequently, the same area was irradiated at the Kurchatov Institute starting at 80 keV and continued with decreasing ion energies to 3 keV. Both sets of irradiations were compared for two different total doses, 0.5 C cm^{-2} and 1.0 C cm^{-2} .

Introduction

The structural components in a fusion reactor will be exposed to irradiation by helium projectiles with a broad energy spectrum. The impact of helium ions on surfaces can cause surface erosion due to helium blistering. For $^4\text{He}^+$ ion energies larger than 10 keV most blistering studies reported to date have used monoenergetic helium beams. The present experiments were designed to study the effect of helium ion implantation over a broad energy range on the surface erosion by blistering. To achieve such implants over a broad energy range cold rolled Nb targets were irradiated sequentially with $^4\text{He}^+$ ion energies ranging from 3 keV to 500 keV at room temperature. The dose for each energy was chosen so that an approximately uniform helium concentration (within $\sim 10\%$) was obtained between the implant depth corresponding to 3 keV and 500 keV. These experiments are considered to represent a link between blistering studies done with monoenergetic ion beams and those which need to be performed under simultaneous impact with particles having a broad energy range to simulate the flux and energy spectrum of helium projectiles expected to reach components in a fusion reactor exposed to plasma radiations. Estimates of the fluxes and energy spectrum of helium projectiles striking for example the first wall give widely different values for different fusion reactor concepts [see Table 1 in Reference 1]. Recently, blistering studies of niobium implanted sequentially with low energy $^4\text{He}^+$ ions over a relatively small energy range from 0.5 keV to 9 keV have been reported [2].

In one set of the present experiments the irradiations were started at the Kurchatov Institute with 3 keV $^4\text{He}^+$ ions and extended up to 80 keV in several steps (3, 12, 25, 45, 80 keV). Subsequently, the same target area was irradiated with $^4\text{He}^+$ ions at Argonne National Laboratory (ANL) starting at 100 keV and increased to 500 keV in steps of 50 keV. In another set of

experiments the irradiations were started at ANL with 500 keV $^4\text{He}^+$ ions and continued with decreasing ion energies (in 50 keV steps) to 100 keV. Subsequently, the same area was irradiated at the Kurchatov Institute starting at 80 keV and continued with decreasing ion energies to 3 keV. Both sets of irradiations were completed for two different total doses, 0.5 C cm^{-2} and 1.0 C cm^{-2} , to study the effect of total dose on blister appearance, blister exfoliation, and the helium implant profile. Measurements of the depth profile of the implanted helium were conducted by Rutherford backscattering at ANL, using 1.5 MeV protons. The measured values were compared with calculated implant profiles.

Experimental Techniques

Cold-worked foils of electron-beam zone refined polycrystalline niobium (Marz grade) were obtained from Materials Research Corporation. The surface preparation of all targets was done at ANL by mechanical polishing and lapping and degreased in ultrasonic baths of trichlorethylene, acetone, distilled water and 2 rinses in methanol. Subsequently, the targets were electropolished in an electrolyte containing 92.5% methanol, 5% H_2SO_4 , and 2.5% HF at -70°C at a current density of $\sim 0.05 \text{ A cm}^{-2}$. Finally, the samples were rinsed in ultrasonic baths of methanol. After this treatment the surfaces had an average surface roughness of less than $0.02 \mu\text{m}$ over 90% of the polished area as determined by scanning electron microscopy. Niobium foils with an average thickness in the range from 50 to $70 \mu\text{m}$ and from 2 to $3 \mu\text{m}$ were used. The thin foils were used for the Rutherford backscattering studies. For the implantations with $^4\text{He}^+$ ions for the energy range from 3 keV to 80 keV mass analyzed beams from an isotopic separator at the Kurchatov Institute was used [3]. During these irradiations the $^4\text{He}^+$ ion flux on target was held at 1.2×10^{17} ions

$\text{sec}^{-1} \text{ cm}^{-2}$ and the vacuum was $\sim 5 \times 10^{-7}$ Torr. At such a high flux a heating of the foil is to be expected. For the implantations with $^4\text{He}^+$ ions for the energy range from 100 to 500 keV mass analyzed beams from a 2-MeV Van de Graaff accelerator at A&L was used. The beam was highly collimated (half angle of divergence $\sim 3 - 5 \times 10^{-8}$ Terr. To implant to a total dose of $\sim 1.0 \text{ C cm}^{-2}$ the following dose values were used (listed in parenthesis in units of C cm^{-2}) for a given energy in keV: 3 (0.02), 12 (0.02), 25 (0.05), 45 (0.10), 80 (0.04), 100 (0.06), 150 (0.12), 200 (0.08), 250 (0.06), 300 (0.10), 350 (0.06), 400 (0.04), 450 (0.06), and 500 (0.2). To implant to a total dose of only 0.5 cm^{-2} , half the dose values listed above for each energy were used. The irradiations from 3 keV to 80 keV in the sequence 3, 12, 25, 45 and 80 keV will be referred to as type A irradiations. The irradiations with a reversed sequence will be referred to as type B irradiations. The irradiations starting at 100 keV and increased to 500 keV in steps of 50 keV will be called type C irradiations. The irradiations with the reversed sequence will be referred to as type D irradiations.

The niobium surfaces were examined before and after the irradiation with a Cambridge Stereoscan S4-10 scanning electron microscope. The Rutherford - backscattering measurements were made with a 1.5-MeV H^+ beam. The backscattered ions were energy-analyzed using solid-state surface-barrier detectors, the scattering angle being 174° .

Results and Discussion

Figures 1 (a) and (b) show at two different magnifications surfaces of cold rolled, polycrystalline niobium foil after the sequential implantation with $^4\text{He}^+$ ions with increasing energies from 3 - 80 keV to a total dose of 0.11 C cm^{-2} (at the five discrete energies with the corresponding dose mentioned above -

type A irradiations). Figure 1 (c) shows the same area as in Figure 1 (a) after 100 keV to 500 keV $^4\text{He}^+$ implantations (type C irradiations) to a dose of 0.39 C cm^{-2} . The total $^4\text{He}^+$ ion dose for all energies is 0.5 C cm^{-2} . No detectable changes of the type A irradiated surfaces can be observed after the subsequent type C irradiations [cf. figures 1 (a) and 1 (c)]. Figure 1 (d) shows an enlarged view of the surface after type A and C irradiations. One can readily notice blisters whose diameters fall into four major class sizes. The larger size blisters (class 1) have diameters ranging from 1.5 to 2.0 μm , the next smaller size blisters (class 2) have diameters in the 1 to 1.5 μm range, the next size blister (class 3) have diameters in the 0.4 - 0.6 μm range, and the smallest size blisters (class 4, - see for example small blisters on top of exfoliated skin) have diameters of 0.1 to 0.2 μm . These four major size classes of blister diameters are thought to be due to the $^4\text{He}^+$ ion implantations at 45 keV (for class 1), at 25 keV (for class 2), at 12 keV (for class 3), and at 3 keV (for class 4), based on measurements of blister diameters with monoenergetic $^4\text{He}^+$ ions in single irradiations [4, 5]. Measurements of the blister skin thicknesses for the class 1 size blisters [figure 1 (b)] give a value of $0.24 \pm 0.03 \mu\text{m}$, which is close to the expected blister skin thickness for 45 keV $^4\text{He}^+$ ion implantation in niobium [5]. This further indicates that the blisters of class 1 are caused by the 45 keV $^4\text{He}^+$ ion component. The measured blister density for class 1 and 2 blisters is $\sim 2 \times 10^7 \text{ blisters cm}^{-2}$. The appearance of blisters of these four size classes indicates that the critical dose for blister appearance must have been exceeded.

Figure 2 (a) shows the surface of a niobium foil after 500 keV to 100 keV $^4\text{He}^+$ irradiation (type D) to a dose of 0.39 C cm^{-2} . Over most of the irradiated area no blisters could be detected. The irradiated area shown in figure 2 (a) is similar to the unirradiated area shown in figure 2 (b). Figures 2 (c) - (e) show

surfaces after the subsequent irradiations with 80 keV to 3 keV $^4\text{He}^+$ ions (type B) to a dose of 0.11 C cm^{-2} of the type D irradiated surface. The total dose is 0.5 C cm^{-2} . Again, blisters of the four class sizes can be seen. For the blisters of class 1, however, the blister diameters range to higher values of $\sim 4 \mu\text{m}$. Many of the exfoliated blisters of class 1 have skin thicknesses of about $0.24 \pm 0.03 \mu\text{m}$, a value identical to the observations made in Figure 1. The blister density value for the class 1 and 2 blisters is $\sim 2 \times 10^7 \text{ blisters cm}^{-2}$. A comparison of Figure 2 (e) and Figure 1 (b) shows that after the combination of type B and D irradiations [500 keV to 3 keV, see Figure 2 (e)] the blisters of size class 4 form at the bottom of the blister craters, while after the combination of type A and C irradiations (3 keV to 500 keV) the blisters of the same size class appear on the top of the blister skins. This observation is plausible in view of the irradiation sequences chosen. For both irradiation sequences the ion components in the 100 keV to 500 keV range do not seem to contribute to blister appearance or rupture even though the total dose was 0.39 C cm^{-2} . It had been previously reported [6] that for 500 keV $^4\text{He}^+$ ion irradiation to a dose of 0.1 C cm^{-2} (which is the same dose used for the 500 keV component in the present irradiations) blisters appeared on a cold rolled niobium surface but not on an annealed niobium surface. Work is in progress to determine the differences in the microstructure of the samples used in the present investigations and those used earlier.

Figures 3 (a) and (b) show at two different magnifications surfaces of a niobium foil after 500 keV to 100 keV $^4\text{He}^+$ irradiation (type D) to a dose of 0.78 C cm^{-2} , which is twice the dose given to the foil shown in Figure 2 (a). A few blisters can be observed. An unirradiated area is shown for comparison in Figure 3 (c). Figures 3 (d) - (f) show at different magnifications surfaces after the subsequent irradiations with 80 keV to 3 keV $^4\text{He}^+$ ions (type B) to a

dose of 0.22 C cm^{-2} of the type D irradiated surface. Several large exfoliated areas were observed and one is shown in Figure 2 (d). It appears that these exfoliated areas are due to the rupture of the large blisters (formed during type D irradiations) caused by the subsequent type B irradiations. Blisters of five size classes can be seen. Four size classes are identical to those observed before for a dose of 0.5 C cm^{-2} , and a fifth size class includes the larger blisters ($\sim 7 - 25 \text{ }\mu\text{m}$ diameter) during type D irradiations. Many of the exfoliated blisters of class 1 have formed craters with a depth of $\sim 0.28 \pm 0.04 \text{ }\mu\text{m}$, a value which is close to the expected blister skin thickness for 45 keV He^+ ion irradiations. The blister density value for the class 1 and 2 blisters is approximately $4 \times 10^7 \text{ blisters cm}^{-2}$, a value which is a factor of 2 larger than the one observed for the dose (0.5 C cm^{-2}) which is a factor of two lower.

Preliminary results for a combination of type A and C irradiations of a cold rolled niobium foil for a total dose of 1 C cm^{-2} have been obtained. Unlike the case illustrated in Figure 1, the type C irradiations caused exfoliation of some of the blisters formed in type A irradiations. Rutherford backscattering studies were performed to measure the depth profile of implanted helium after type A and C irradiations of a thin ($2.8 \text{ }\mu\text{m}$) niobium foil to a dose of 0.5 C cm^{-2} .

Figure 4 (a) shows the depth profile of helium implanted, for example, in a combination type A and C irradiations of niobium, determined from the backscattering spectra of 1.5 MeV protons and using the stopping power values from Williamson et al. [7]. Figure 4 (b) shows a calculated implant profile for the implant conditions stated above. For the energy range from 100 keV to 500 keV the individual implant profiles were calculated assuming a Gaussian distribution, the first and second moments of which were obtained using Thomas-Fermi nuclear stopping cross sections together with Brice's [8] expression for the electronic

stopping cross sections which uses the semiempirical values of stopping power from Ziegler and Chu [9]. For the energy range from 3 keV to 80 keV the individual implant profiles were calculated by the authors from the Kurchatov Institute using the method suggested by Gibbons [10]. The shape of the calculated implant profile [Figure 4 (b)] resembles the experimentally determined profile [Figure 4 (a)] quite well.

References

- [1] M. Kaminsky, in Plasma Physics and Controlled Nuclear Fusion Research, Vol. II (International Atomic Energy Agency, Vienna, 1975) p. 287.
- [2] J. Roth, R. Behrisch, and B. M. V. Scherzer, J. Nucl. Mat. 57 (1975) 365.
- [3] V. M. Gusev et al. P T $\mathfrak{D}$, (Soviet Journal for Instruments and Methods) 4, 1969, 19.
- [4] J. Roth, R. Behrisch and B. M. V. Scherzer, J. Nucl. Mat. 53 (1974) 147.
- [5] R. G. St-Jacques, J. G. Martel, B. Terreault, G. Veilleux, S. K. Das, M. Kaminsky and G. Fenske, in these proceedings.
- [6] S. K. Das and M. Kaminsky, J. Appl. Phys. 44 (1973) 25.
- [7] C. F. Williamson, J. P. Boujot, and J. Picard, Tables of Range and Stopping Power of Chemical Elements for Charged Particles of Energy 0.05 to 500 MeV, CEA-R3042 (1966).
- [8] D. K. Brice, Phys. Rev. A6 (1972) 1791.
- [9] J. F. Ziegler and W. K. Chu, Thin Solid Films 19 (1973) 281.
- [10] James F. Gibbons, Proceedings of IEEE, Vol. 56, No. 3, p. 295, (March 1968).

COLD-ROLLED POLYCRYSTALLINE NIOBIUM IRRADIATED
AT ROOM TEMP. WITH

3-80 keV $^4\text{He}^+$ at KURCHATOV

3-80 keV $^4\text{He}^+$ at KURCHATOV,
DOSE: 0.11 C/cm^2

and

100-500 $^4\text{He}^+$ at A.N.L.,
DOSE: 0.39 C/cm^2

TOTAL DOSE: 0.11 C/cm^2

TOTAL DOSE: 0.5 C/cm^2

Fig. 1.

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COLD-ROLLED POLYCRYSTALLINE NIOBIUM IRRADIATED
AT ROOM TEMP. WITH

500-100keV $^4\text{He}^+$ at A.N.L.

500-100keV $^4\text{He}^+$ at A.N.L.
DOSE: 0.39 C/cm^2

80-3keV $^4\text{He}^+$ at KURCHATOV
DOSE: 0.11 C/cm^2

TOTAL DOSE: 0.39 C/cm^2

TOTAL DOSE: 0.5 C/cm^2

Fig. 2
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POLYCRYSTALLINE NIOBIUM IRRADIATED
AT ROOM TEMP. WITH

50-100keV $^4\text{He}^+$ at A.N.L.

500-100keV $^4\text{He}^+$ at A.N.L.

DOSE: 0.76 C/cm^2

80-3 keV at KURCHATOV

DOSE: 0.22 C/cm^2

TOTAL DOSE: 0.78 C/cm^2

TOTAL DOSE: 1.0 C/cm^2

Fig. 4
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