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DEUTERON AND HELIUM ION IRRADIATION OF CERAMIC COATINGS ON Nb-1% Zr[†]

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Abstract

The surface damage to insulating barium alumino-silicate glass coatings due to irradiation by 100 and 250 keV deuterons and helium ions at room temperature and at 300°C has been studied. Blisters are observed after irradiation at room temperature with both deuterons and helium ions with energies of 100 keV and 250 keV. For deuteron irradiation a large fraction of the blisters have diameters which are approximately 3 to 5 times larger than the diameter observed with helium ions for identical irradiation conditions, but the density of blisters is nearly an order of magnitude lower. For irradiation at 300°C, no blisters are observed with either type of particle. The sharp rise in permeation rate with temperature is thought to be responsible for this behavior. The blister skin thicknesses have been measured and correlated with calculated projected-range values.

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Introduction

Several fusion reactor concepts require that the entire first wall or parts of it consist of electrically-insulating material. For example, for the first wall of the reference theta-pinch reactor (RTPR) [1] a structure with an insulator on a metal backing has been suggested in order to electrically insulate the first wall metal from the emf which develops during the implosion heating. The insulating layer serves to prevent electrical breakdown between the plasma and blanket segments during the short implosion heating stage. For the pulsed mode operation of such a reactor it is necessary that the dielectric and mechanical properties of this composite structure of the first wall be maintained for a reasonable lifetime. Different types of ceramic insulating coatings are being developed by Atomics International for use in such fusion reactors.

A variety of surface effects have been observed when insulators are bombarded with energetic ions [2, 3]. The surface damage of two insulating ceramic coatings on Hastelloy-X and Nb-1% Zr at room temperature and at 300°C caused by 100 keV and 250 keV helium ion irradiation for doses ranging from 3.7×10^{18} to 1×10^{19} ions cm^{-2} has been studied recently [4]. Blisters were observed after room temperature irradiation with both 100 keV and 250 keV helium ions. However, for irradiation at 300°C no blisters could be observed. The sharp rise in the helium permeation with temperature, observed by others for some glasses and ceramics, was suggested to be responsible for this behavior.

We have extended these studies to include irradiations with D^+ ions under conditions identical to those used previously for helium ion irradiation in order to compare the effects of the two types of particles on the same type of ceramic surface. Measurements have been made to determine the distribution in blister diameters, the blister density, and the blister skin thickness.

Experimental Techniques

The ceramic coatings, consisting of a mixture of 52.3% SiO_2, 40.5% BaO and 7.2% Al_2O_3 on Nb-1% Zr, were furnished by D. W. Keefer, Atomics International. (The concentrations are given in wt %.) The slips for the coatings were prepared by ball milling the frits with the above composition and the slips were applied to the substrate surface by spraying. The coatings were then fired at high temperature (∿ 1200°C) in air or helium. Further details of preparation of these coatings can be found elsewhere [5]. The thickness of the coatings used in these studies ranged from 62 to 99 μm, which is much larger than the projected ranges of the ions used for irradiation.

The coatings were irradiated with 100 keV or 250 keV He^+ or D^+ ions from a 2-MeV Van de Graaff accelerator in high vacuum at a total pressure of ∿ 5 x 10^{-8} Torr. The flux of the incident ions was held at approximately 1 x 10^{14} ions cm^{-2} sec^{-1}. For the determination of the actual irradiation dose of these surfaces, corrections for changes in the electrical resistivity and for the secondary electron emission had to be applied. Such corrections were determined from measurements of ion-beam currents (at constant flux) on both metal and ceramic targets under appropriate conditions. Due to changes in the current signal of the ceramic-coated target with increasing irradiation time (resulting from changes in electrical resistivity and secondary electron emission yields), a maximum uncertainty of 20% in the total dose is expected. Other details of the irradiations are similar to those of the irradiations of metal targets described earlier [4]. No optical or chemical polishing of the targets was considered necessary, but all the targets were cleaned in ultrasonic baths of trichloroethylene, acetone, distilled water and methanol prior to irradiation. For irradiation at 300°C the targets were resistance heated, and the target temperature was measured by an infrared-sensitive pyrometer. The target surfaces were examined before and after irradiation in a scanning electron microscope (Cambridge Stereoscan S4-10). The ceramic surfaces were coated with ~ 200 Å of gold after irradiation to prevent charging during observation in the scanning electron microscope.

Results

Figure 1 shows scanning electron micrographs of blisters on the ceramic coatings formed during irradiation with 100 keV and 250 keV He^+ and D^+ ions to total doses of 3.7×10^{18} ions cm^{-2} at room temperature. The micrographs reveal a striking difference in the blister diameters for deuteron and helium ion irradiation for both energies. For 100 keV irradiation the blister diameters range from 3.0 to 25 μm for the D^+ case and only from 0.6 to 4.0 μm for the He^+ case. For 250 keV irradiation the values for the blister diameters range from 3 to 60 μm for the D^+ case, and from 0.6 to 6 μm for the He^+ case. A distribution of the blister diameters is shown in the histograms in Figure 2. For both energies a large fraction of the blisters observed for D^+ irradiation have diameters which are approximately 3 to 5 times larger than for He^+ ion irradiation. However, for both energies the blister density for D^+ ion irradiation is $\sim 2.2 \pm 0.5 \times 10^5$ blisters cm^{-2}, which is nearly an order of magnitude lower than the value of $1.8 \pm 0.3 \times 10^6$ blisters cm^{-2}, observed for He^+ ion irradiation.

Exfoliation of blisters is observed for both 100 keV and 250 keV D^+ ion irradiation in some areas (e.g. see Figure 1b), but no exfoliation is observed for the He^+ ion irradiation at either energy. From the ruptured blisters,measurements of the blister-skin thickness have been made. The skin thickness values for D^+ ion irradiation at 100 keV and 250 keV are 0.8 ± 0.2 μm and 1.5 ± 0.2 μm, respectively (see Figure 3). For He^+ ion irradiation the skin thickness values for 100 keV and 250 keV had been estimated earlier [4] as 0.25 ± 0.05 μm and 0.5 ± 0.1 μm, respectively.

The projected ranges of helium ions in such a ceramic coating have been calculated using a program by Brice [6] and an estimated [7] density of 3.2 gr. cm^{-3} for this coating. This program utilizes Thomas-Fermi nuclear stopping cross-sections [8]. The calculated projected ranges for 100 and 250 keV He^+ ions are 0.34 μm, and 0.67 μm, repsectively. These values are slightly higher than the upper limit values for the observed skin thicknesses. If the value of the estimated density of the coating were larger, the projected range values would be reduced.

The projected ranges of deuterons in the ceramic coating used have been calculated using Thomas-Fermi nuclear stopping cross sections together with electronic stopping cross sections of Lindhard et al. [9], which are proportional to projectile velocities. The calculated projected ranges for 100 and 250 keV D^+ ions are 0.76 μm and 1.48 μm, respectively. These values are in reasonable agreement with the observed skin thicknesses.

When the temperature during irradiation was raised to 300°C no blisters were observed for either He^+ or D^+ irradiation at the two energies. In fact, no significant difference is noted between the irradiated and unirradiated areas of the coating held at 300°C (see Figures 4a and 4b), and an unirradiated area held at room temperature (Figure 4c).

Discussion

The appearance of blisters as a result of irradiation at room temperature with either deuterons or helium ions indicates that these glassy coatings can be deformed plastically. This is in contrast to the pits observed in other ceramic materials such as SiC after helium-ion irradiation [3]. The skins of blisters from 250 keV ions of both kinds are about twice as thick as those from

100 keV ions of the same type. Deuterium blister skins are approximately 3 times thicker than helium blisters at each energy. Blister skin thicknesses correlate favorably with calculated projected ranges of the ions at each energy.

The fact that deuterium blisters are 3 - 5 times larger in diameter at each energy is due in part to the greater depth of penetration of deuterons but a more important factor is the lower permeation rate for deuterium in glassy materials. This is illustrated by the fact that blisters due to 100 keV deuterons are substantially larger, on the average, than those due to 250 keV helium ions, although the projected ranges are within 15% of each other. For the same reason, no rupturing of helium blisters was observed, whereas a few deuterium blisters were ruptured at both energies.

The irradiation temperature has a strong effect on blister formation. No deuterium blisters were observed at 300°C at the dose studied; this is similar to the result reported for helium irradiation [4]. The absence of blisters at high temperatures is thought to be due to the sharp increase in permeation rate with temperature observed in for both helium and deuterium most glasses. For example, the permeation rate through borosilicate g... [10] increases by more than two orders of magnitude from room temperature to 300°C. The pits appearing in all three photographs in Figure 4 are apparently not due to irradiation or heating of the sample.

The total blistered area, obtained by multiplying the blister density times the area of an average blister, is substantially higher after deuteron irradiation compared to helium-ion irradiation under the same conditions at room temperature. In either case, however, surface erosion due to blistering is easily eliminated by a sufficient increase in temperature during irradiation.

Acknowledgements

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References

[1] R. A. Krakowski, F. W. Clinard, Jr., F. L. Ribe and T. A. Coultas, Surface Effects in Controlled Fusion, eds. H. Wiedersich, M. S. Kaminsky, K. M. Zwilsky, (North-Holland Publishing Company, Amsterdam, 1974) p. 54; also in J. Nucl. Mat. 53 (1974) 54.

[2] W. Primak, J. Nucl. Mat. 53 (1974) 238.

[3] M. Kaminsky and S. K. Das, "Erosion of Silicon Carbide Surfaces Under Helium Ion and 14 MeV Neutron Irradiations", in proceedings of the First Topical Meeting on the Technology on Contrclled Nuclear Fusion, p. 508, Vol. II, CONF-740402-P2, U.S.A.E.C. Office of Information Services, Technical Information Center.

[4] M. Kaminsky, S. K. Das, and R. Ekern, Nuclear Technology (in press).

[5] T. G. Parker, Jr., (ed.) Atomic International Report No. AI-73-45 (1973).

[6] D. K. Brice, Sandia (USA) Report, SLA-73-0416 (1973).

[7] D. W. Keefer, (private communication).

[8] D. K. Brice, Phys. Rev. A6 (1972) 1791.

[9] J. Linhard, M. Scharff, and H. E. Schiott, Matt. Fys. Medd. Dan. Vid. Selsk. 35 (1966) No. 9.

[10] L. C. Walters, J. Am. Ceramic Soc. 53 (1970) 288.

Figure Captions

Figure 1. Scanning electron micrographs of ceramic coating after irradiation at room temperature to a total dose of 0.5 C cm^{-2}: a) 100 keV D^+ ions; b) 250 keV D^+ ions; c) 100 keV He^+ ions; d) 250 keV He^+ ions.

Figure 2. Distributions of blister diameters in ceramic coatings after irradiation with 100 keV and 250 keV D^+ and He^+ ions.

Figure 3. Enlarged view showing blister skins after irradiation: a) 100 keV D^+ ions; b) 250 keV D^+ ions; c) 100 keV He^+ ions; d) 250 keV He^+ ions.

Figure 4. Scanning electron micrograph of ceramic coating: a) after irradiation with 100 keV D^+ ions at 300°C to a total dose of 0.5 C cm^{-2}; b) unirradiated (heated to 300°C); c) unirradiated and unheated.

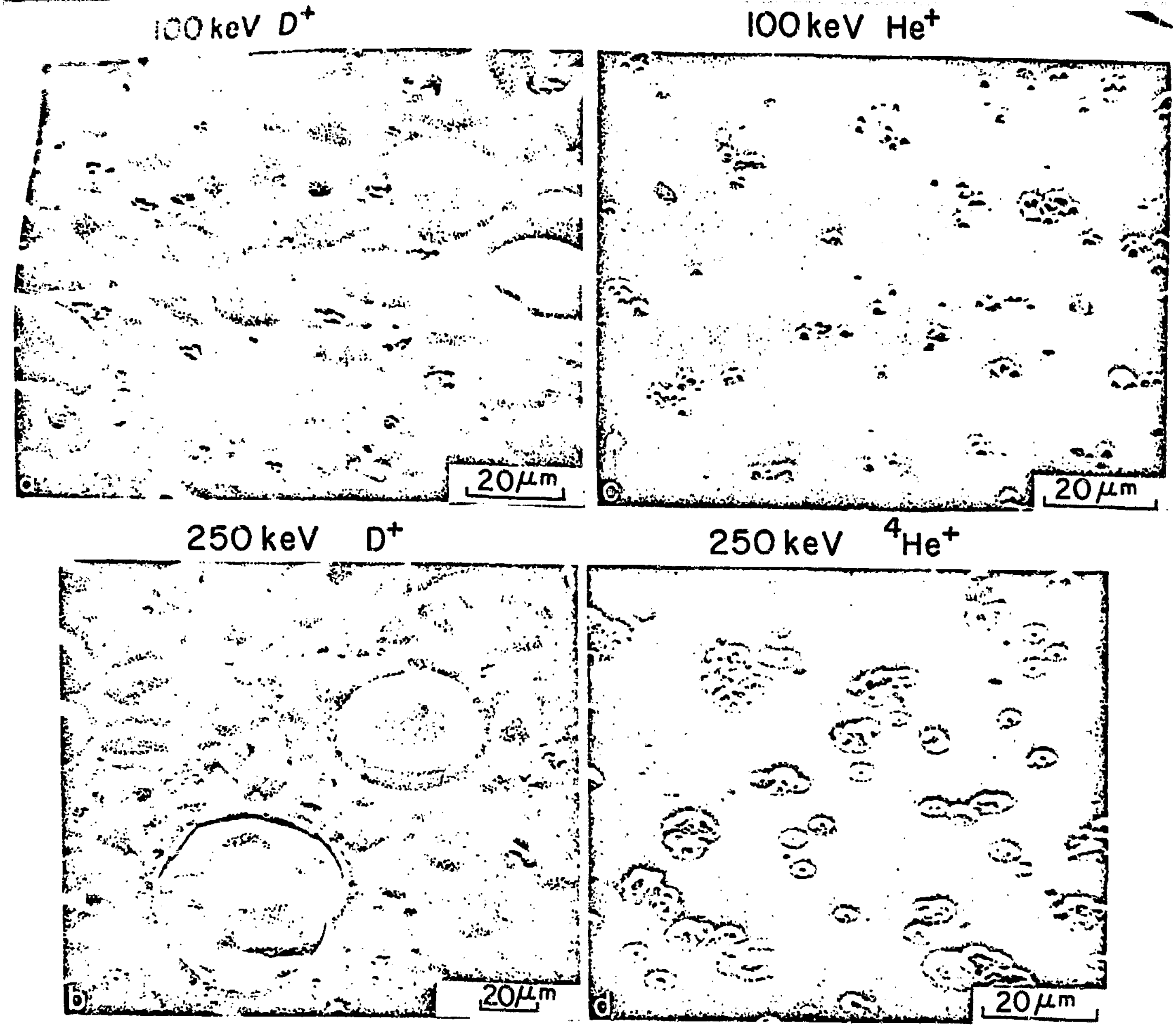

Fig. 1

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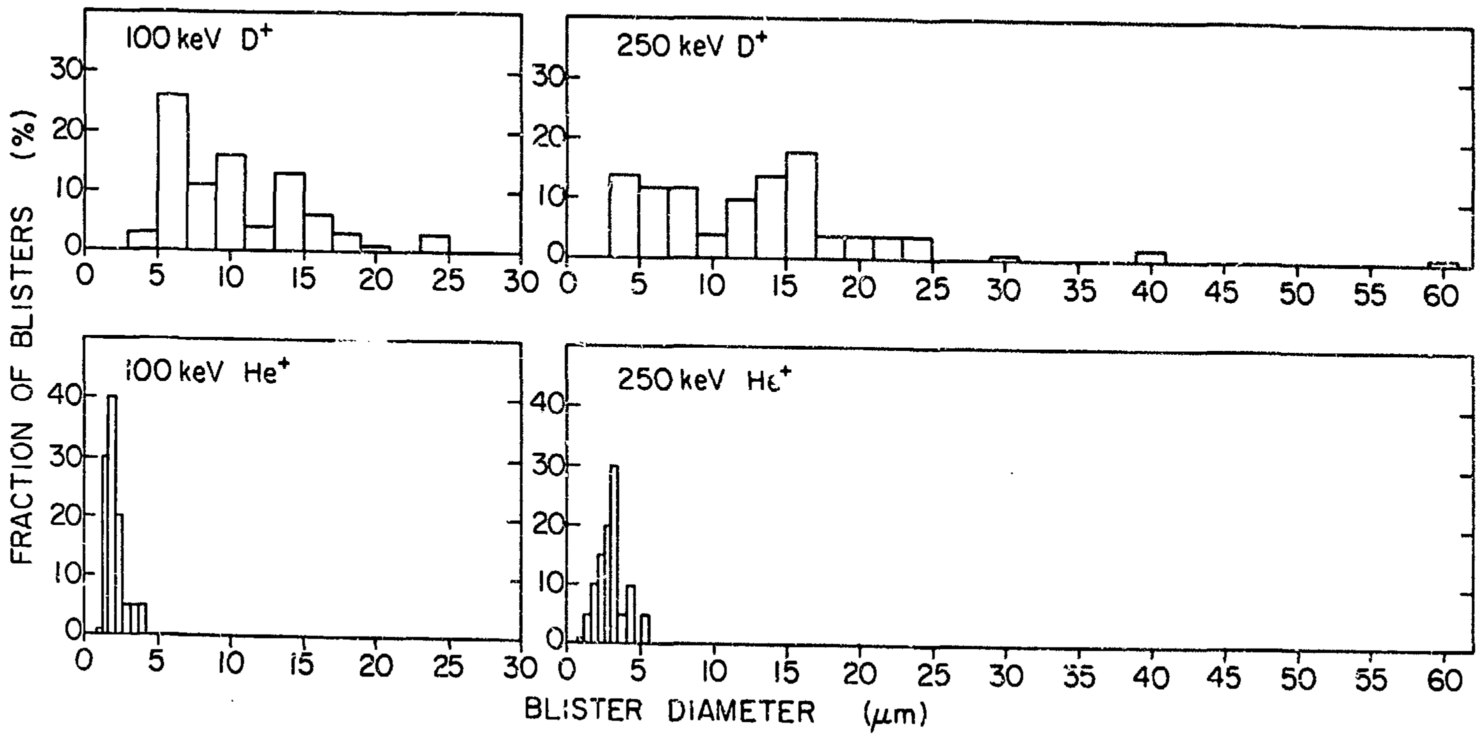

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INSULATOR CERAMIC COATING (52.3% SiO_2, 40.5% BaO and 7.2% Al_2O_3) IRRADIATED AT ROOM TEMPERATURE TO A DOSE OF 0.5 C/cm^2 WITH

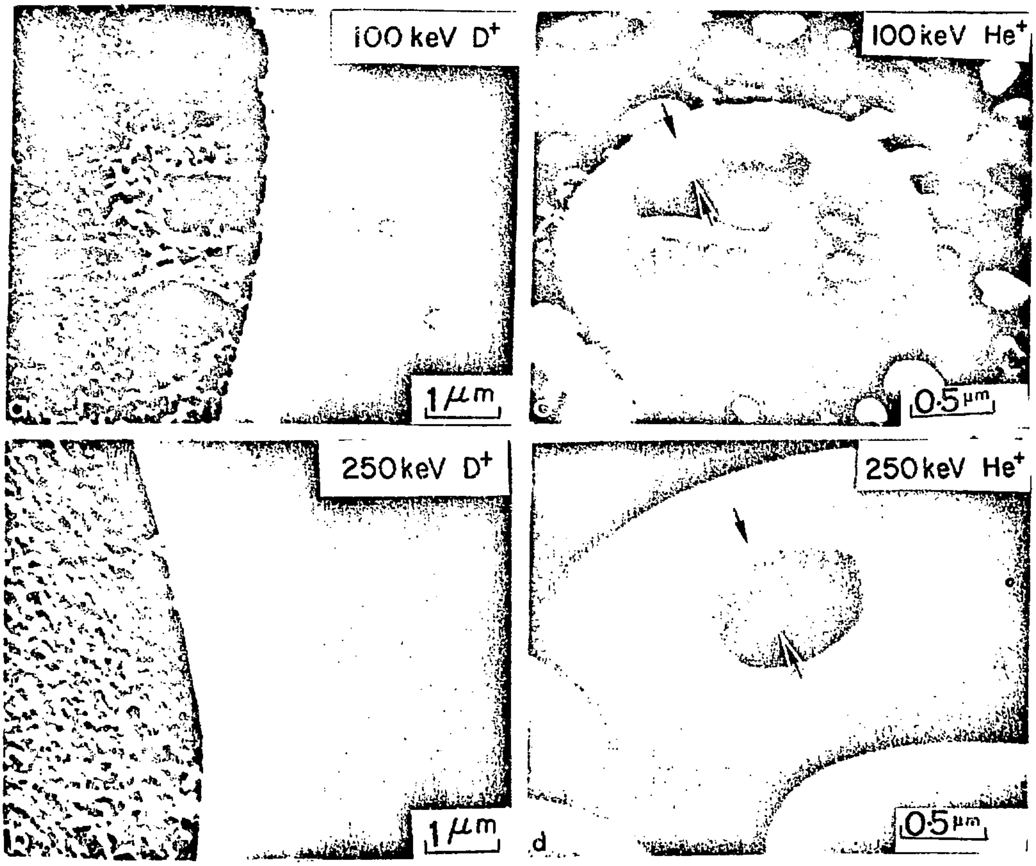

Fig. 3

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INSULATOR CERAMIC COATING (52.3% SiO_2, 40.5% BaO, and 7.2% Al_2O_3) ON Nb-1% Zr IRRADIATED AT 300°C WITH 100keV D^+ IONS TO A DOSE OF 0.5 C/cm²

IRRADIATED AREA — UNIRRADIATED AREA — UNIRRADIATED AREA (UNHEATED SAMPLE)

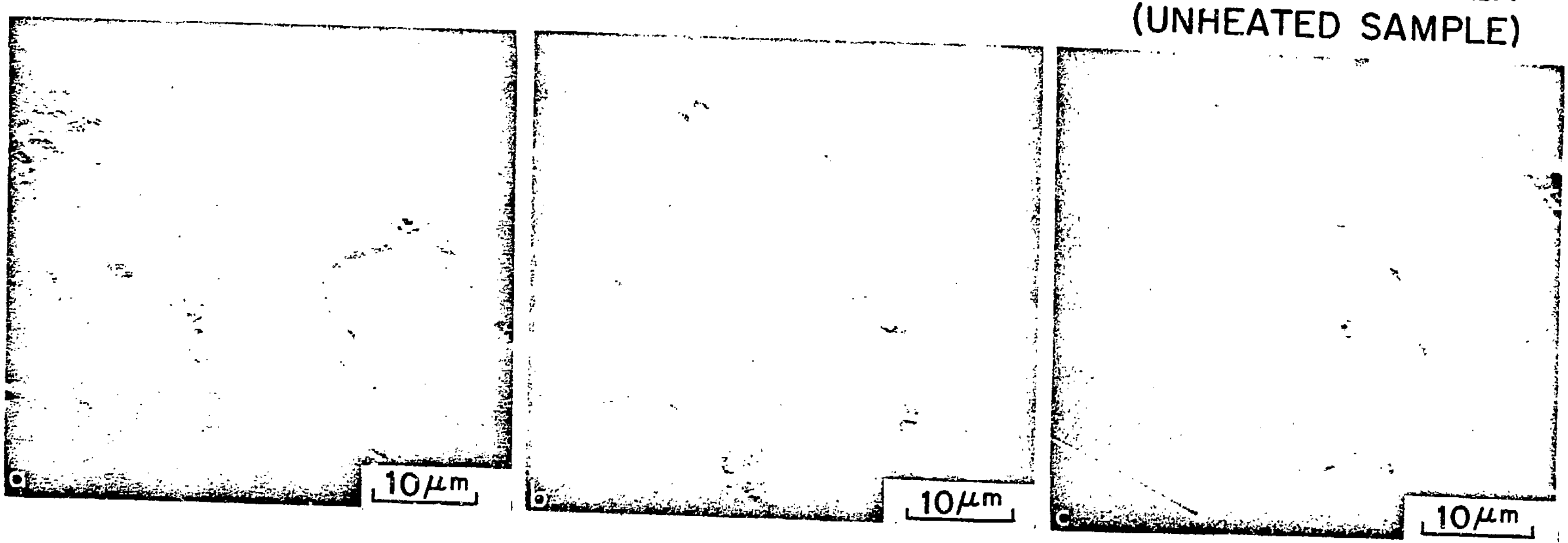

Fig. 4

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