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Surface Roughness Effects on Blister Formation in Polycrystalline Molybdenum

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Polycrystalline molybdenum targets with electropolished and roughened surfaces were bombarded with 100 keV He^+ and 200 keV H_2^+ ions at room temperature. It has been demonstrated that the blister formation is largely or completely suppressed by roughening the electropolished surface with emery paper of #1200, #400 and #100. Up to a He^+ fluence of 1.0 x 10^{19} particles /cm^2, no blisters are observed in the targets with the two roughest surfaces, while on the smooth surface blisters begin to occur at a fluence of 7.5 x 10^{17} particles/cm^2. The surface roughness effect on blister suppression is discussed in relation to the projected range of incident particles.

Keywords: Surface Roughness Effects, Blistering, Molybdenum,
Blister Suppression, 100 keV Helium Ion, 200 keV H_2^+ Ion,
Surface Erosion, Surface Topography, Ion Bombardment

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多結晶モリブデンにおけるブリスター生成におよぼす表面粗さ効果

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電解研磨した表面および粗した表面を持つ多結晶モリブデンに，100keVのHe^+イオンおよび200keVのH_2^+イオンを室温で衝撃した。電解研磨した表面を，#1200,#400および#100のエメリー紙で粗すことによって，ブリスターの生成は大いに抑制されるか完全に抑制されることが明らかになった。電解研磨した滑らかな表面では7.5×10^{17} particles/cm^{2}のヘリウム衝撃によってブリスターが発生し始めるが，一方，2つの最も粗した表面では1.0×10^{19} particles/cm^{2}のフルエンスまでヘリウム衝撃をおこなったが，ブリスターは観察されなかった。ブリスターの発生を抑制する表面粗さの効果については，入射粒子の入射方向の飛程と関係づけて考察した。

目次なし

1. INTRODUCTION

Radiation blistering of materials due to high energy helium, proton and deuteron bombardment has been extensively investigated within the last years since it has been regarded as one of the most dangerous sources of first wall erosion in fusion reactors$^{1-7)}$. Assuming that the critical fluence for the appearance of blistering is 5 x 10^{17} particles/cm^2 and that the flux of the helium ions impinging on the first wall is 1 x 10^{13} particles/cm^2·sec, blisters can appear on the first wall after the reactor has been running for only a day or so. Recently, investigations on the reduction of surface erosion caused by blistering have been reported by several authors. Das et al.$^{8)}$ have demonstrated that the flaking due to blister rupture is crucially reduced in SAP with a suitable microstructure. Ziegler et al.$^{9)}$ have also shown that the blister formation is drastically suppressed by covering a tungsten surface with a lot of tungsten needles. On the other hand, the blister once formed are all sputtered away further blister formation is suppressed. Roth et al.$^{10)}$ have explained this result by using the model that the implanted gas can diffuse out through pores and cracks which are made in the surface layer bombarded to high fluences. Evans$^{11)}$ has also suggested that the surface roughness plays an important role for the blister suppression. Therefore, a systematic examination of the surface roughness effects on blister formation should be required.

The purpose of this paper is to clarify the effects of the surface roughness on the blister formation. In this experiment polycrystalline molybdenum which is a candidate material for the first wall was used as a target material. A part of this work has been reported elsewhere$^{12)}$.

2. EXPERIMENTAL PROCEDURE

Molybdenum targets of 10 mm x 10 mm x 1 mm cut from a cross rolled sheet with 99.96 % purity were purchaced from Plansee. They were annealed at 1000 °C for 10 minutes. Prior to bombardment they were electropolished in a solution consisting of 50 cc sulphuric acid and 350 cc ethyl alcohol under the conditions of 10 °C and 0.5 A/cm^2 for 5 minutes. Roughened areas of three different grades were made by abrading an electropolished molybdenum surface with emery paper of #1200, #400 and #100, a quarter of which was kept as polished (see Fig. 1).

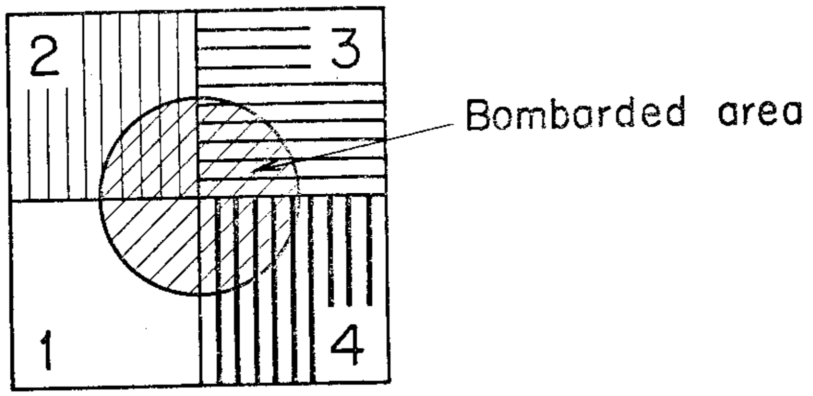


Fig. 1 Target surface with different roughnesses.

1. Electropolished.
2. Abraded with emery paper of #1200.
3. Abraded with emery paper of #400.
4. Abraded with emery paper of #100.

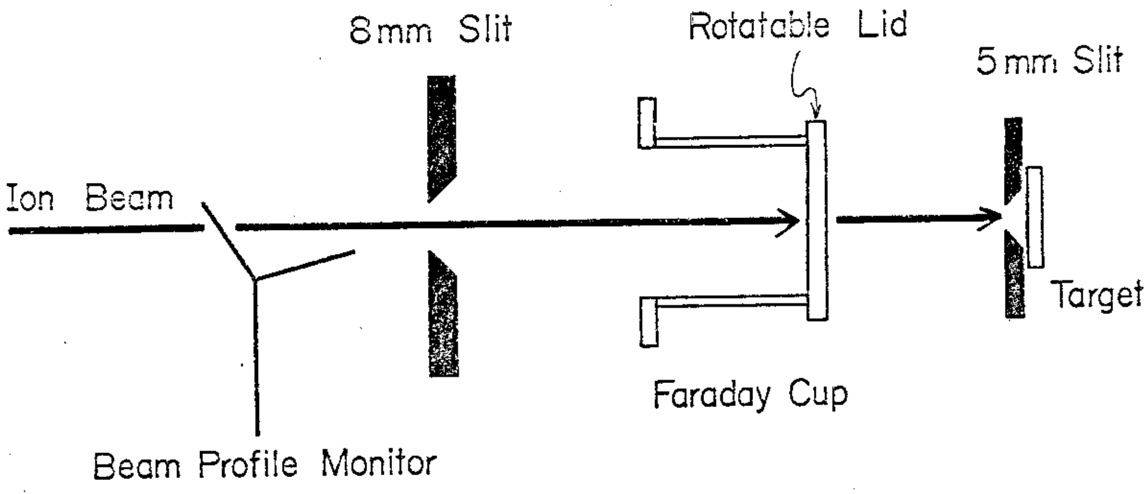


Fig. 2 Schematic of experimental set up.

Bombardments were carried out with 100 keV helium and 200 keV H_2^+ ions from a 400 kV Cockcroft-Walton accelerator in JAERI, where a 200 keV H_2^+ ion is equivalent to two 100 keV H^+ ions. Figure 2 shows the experimental set up schematically. The beam was defined by a slit of 8 mm in diameter and hit the target at normal incidence. Target mounted on a holder with a stainless steel cap and a slit of 5 mm in diameter. A Faraday cup consisting of a cylindrical tube and a rotatable lid was placed between the two slits. Ion current could be monitored alternatively by rotating the lid. The Faraday cup was also used for determining a suitable voltage on the target to suppress the secondary electrons caused by ion bombardment. The beam current was measured at the target with the suppressing voltage of +360 V. The current densities were 30 - 50 $\mu A/cm^2$ and 20 - 120 $\mu A/cm^2$ for helium and H_2^+ ion beams, respectively. A beam profile monitor was used to sense the shape of the beam in both horizontal and vertical modes. Usually, the beam was scanned two-dimensionally by applying potential with a frequency of 1000 Hz on two sets of deflection plates. The scanning area was approximately 10 mm x 10 mm on the slit of 8 mm in diameter. For hydrogen bombardment to fluences of 1.0×10^{19} and 2.5×10^{19} particles/cm^2, however, the beam was not scanned in order to achieve bombardment to the high fluences. One of four targets on a turntable could be positioned in the beam during each bombardment. The target chamber was evacuated with an ion pump and the pressure was around 1×10^{-7} Torr during bombardment. Temperature of the targets was ambient room temperature. The surface structure of the targets was observed in a scanning electron microscope (SEM).

3. EXPERIMENTAL RESULTS

Typical surface microtopographies of rough surfaces abraded with emery paper of #1200, #400 and #100 are sown in Fig. 3. Since abrading was made unidirectionally, numerous grooves were made on the surface parallel to the direction of abrading. Fine cracks were also introduced densely on the surface by abrading especially with emery paper of #100. The depth of the grooves and the distance between two neighbouring grooves were obtained from SEM micrographs, typical values of which are summarized in table 1. Hereafter we name four different surfaces surface A, surface B, surface C and surface D. They correspond to electropolished surface and rough surfaces

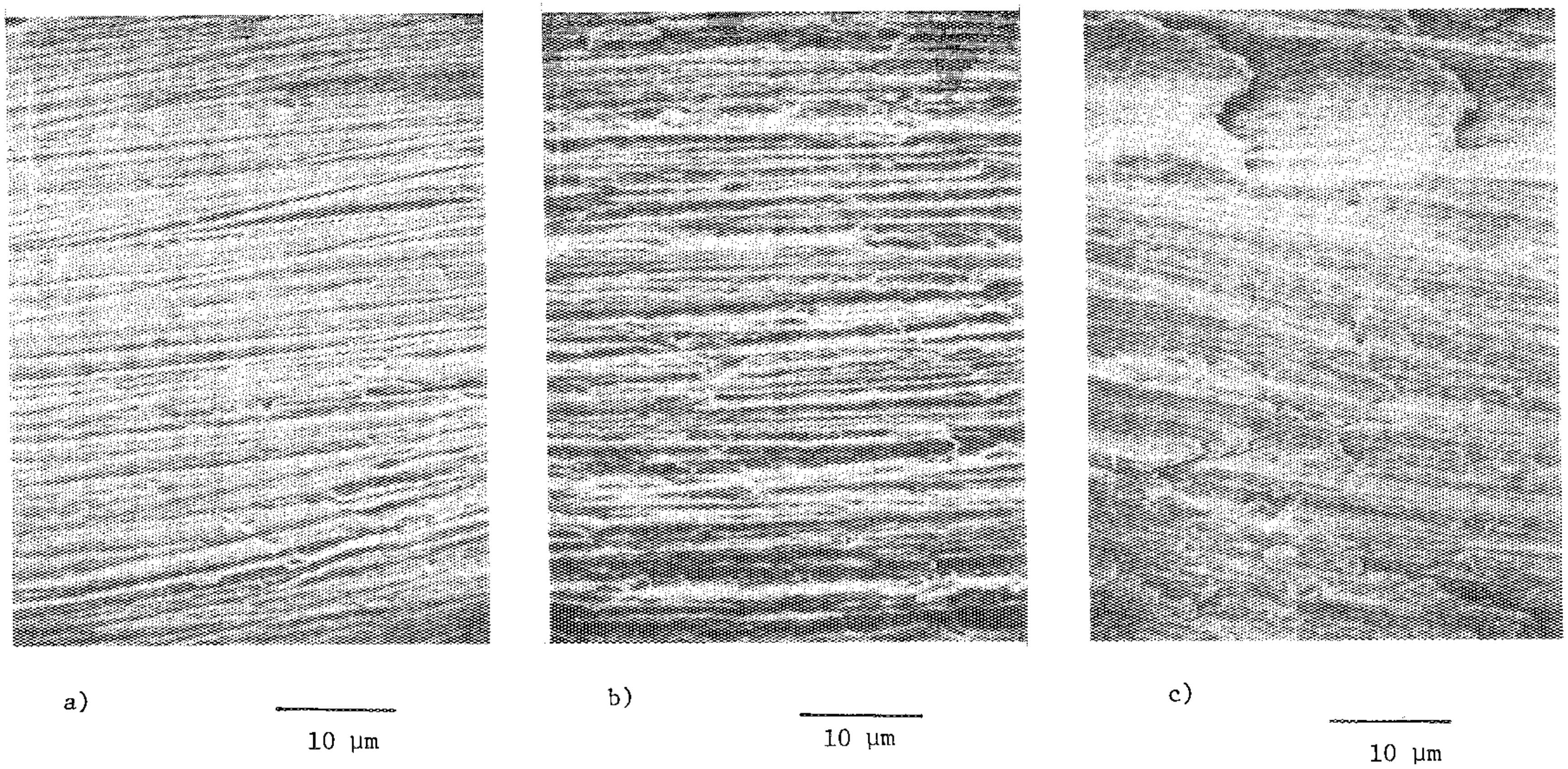
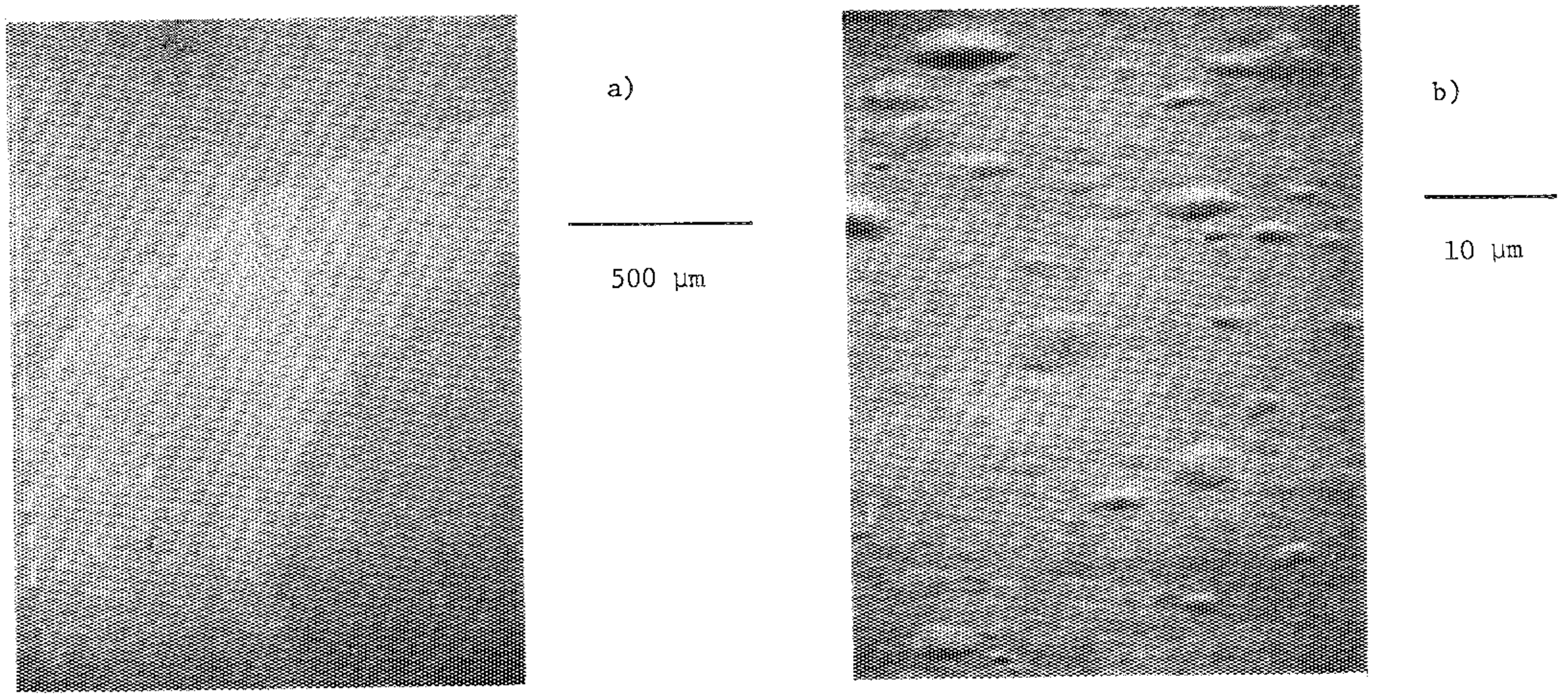


Fig. 3 Typical SEM micrographs of rough surfaces abraded with emery paper of a) #1200, b) #400 and c) #100. They show surfaces before bombardment.

abraded with emery paper of #1200, #400 and # 100, respectively.

Figure 4 shows the SEM micrographs of surface A after bombardment with 200 keV H_2^+ ions to a fluence of 2.5×10^{19} particles/cm^2. Two circular boundaries are seen on the bombarded target (Fig. 4a). The inner circular and outer annular regions correspond to the bombarded area 1 through the slit of 5 mm in diameter and the area 2 behind the tapered part of the slit, respectively (see Fig. 4c). They are also seen on the hydrogen bombarded surfaces to fluences between 5×10^{17} and 5×10^{18} particles/cm^2 where no surface structural change caused by blister formation is observed. Thus the bombarded area is easily identified by observing two circular boundaries even when no blisters appear on the surface at low fluences. Appearance of the annular region on the target may be explained as follows: Since the area 2 is exposed to the secondary electrons caused by ion bombardment, the chemical composition of the area 2 becomes different from that of the area 3 which is completely in contact with the slit. Therefore, in area 2 the number of emitted secondary electrons produced by the primary electron beam of SEM should become different from that in area 3. Two circular boundaries are seen on the helium bombarded surfaces. No blisters appear at fluences below 5.0×10^{18} particles/cm^2. A few blisters appear locally at a fluence of 1.0×10^{19} particles/cm^2. They were formed in the area exposed to the beam with maximum intensity, since the beam hit the target less uniformly. Hydrogen bombardment to a fluence of 2.5×10^{19} particles/cm^2 produces blisters (Fig. 4b). Therefore the critical fluence for blister formation is determined to be approximately 1.0×10^{19} particles/cm^2.

Figure 5 shows the SEM micrographs of surface A after helium bombardment to different fluences. General features for blister formation are very similar to the results obtained by Erents and McCracken$^{1)}$. Although blisters are not observed below 5.0×10^{17} particles/cm^2, they appear at 7.5×10^{17} particles/cm^2 (Fig. 5a). Therefore the critical fluence for blister formation lies between 5.0×10^{17} and 7.5×10^{17} particles/cm^2 for 100 keV helium which is more than one order of magnitude lower than that for 100 keV hydrogen. The numbers of blisters and of flakes of blister cover increase as the fluence increases (Fig. 5b). In Fig. 5c is shown the typical blister cover whose thickness is obtained to be 0.36 μm. At the fluence of 1.0×10^{19} particles/cm^2 many cracks are observed (Fig. 5 d, e and f). The size of an area enclosed with some cracks is distributed between 10 μm and 100 μm, which is nearly equal to the grain size of molybdenum. This has not previously been reported. It is of interest to note that blisters near



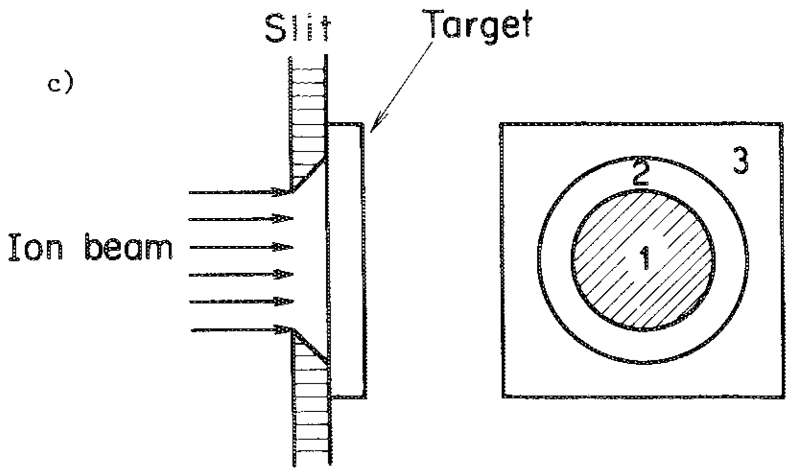


Fig. 4 a) and b) SEM micrographs of electro-polished surface after bombardment with 200 keV H_2^+ ions to a fluence of 2.5×10^{19} particles/cm^2.
c) Schematic representation of arrangement of target and slit.

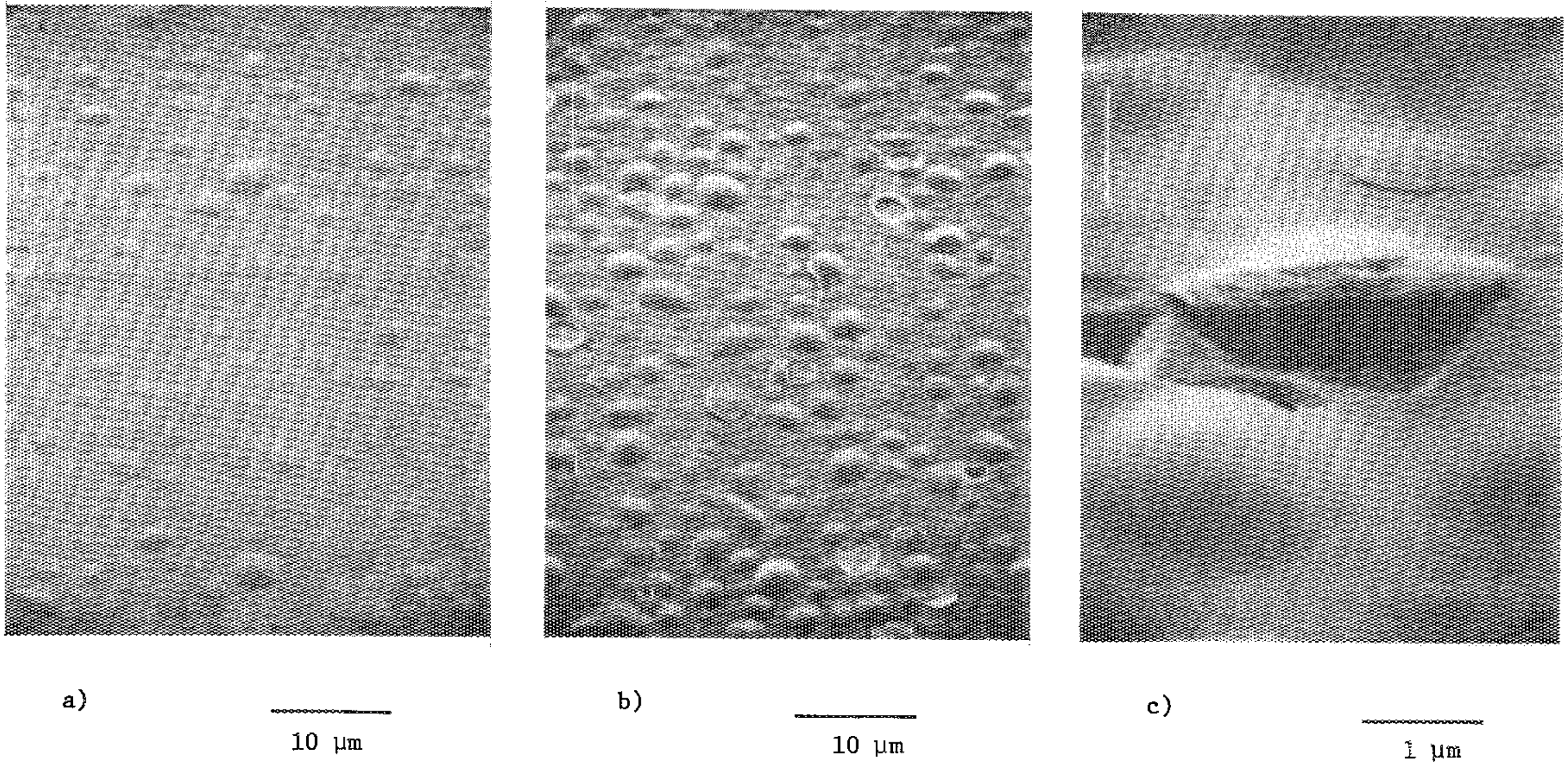


Fig. 5 SEM micrographs of electropolished surfaces after bombardment with 100 keV helium ions to fluences of 7.5×10^{17} (a), 2.5×10^{18} (b and c) and 1.0×10^{19} (d, e and f) particles/cm^2.

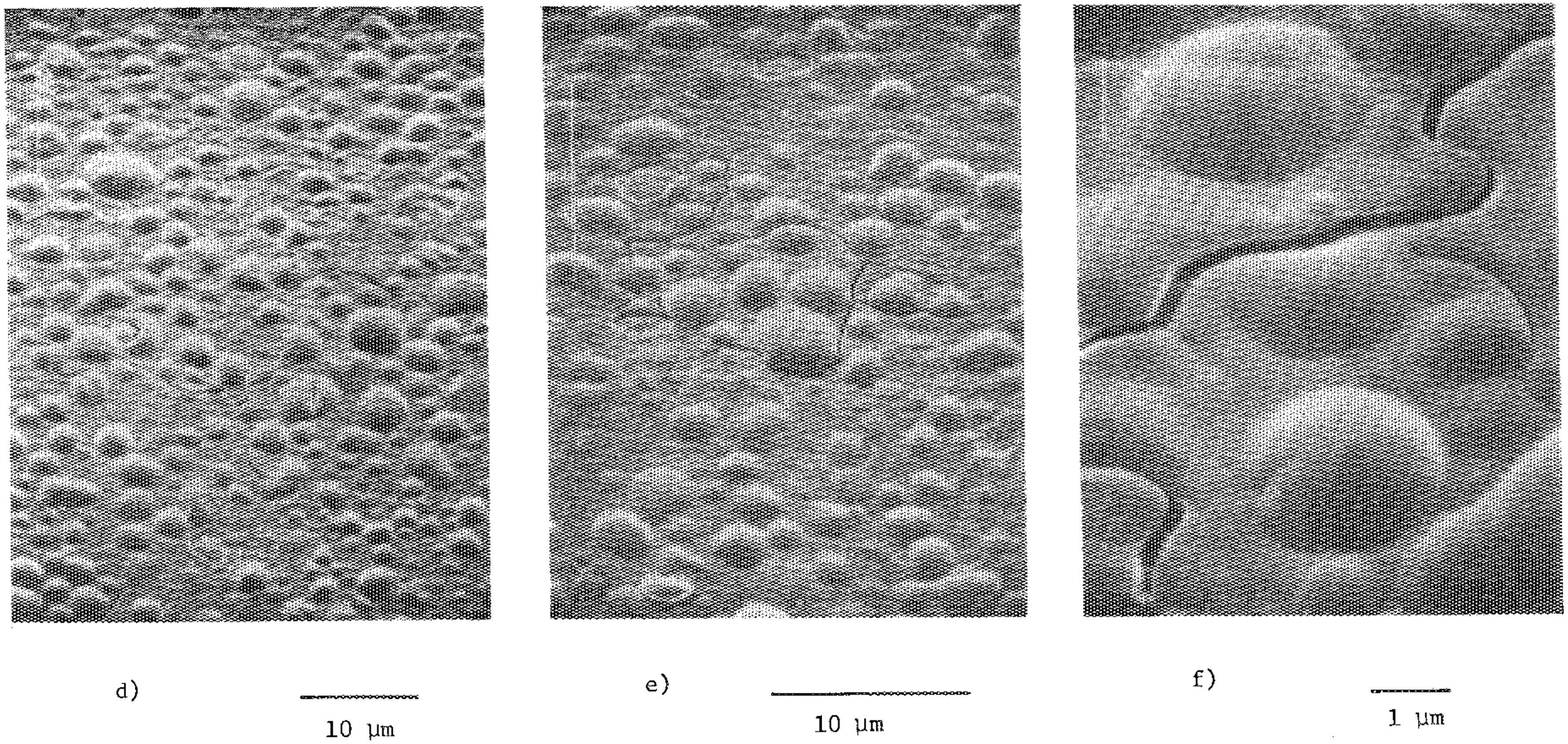


Fig. 5 (continued)

cracks do not flake. This result may be explained by considering a model that the accumulated helium gas can diffuse out through cracks.

Helium bombardment was utilized in order to examine the roughness effects on blister formation, since it leads to the formation of blisters on the surface at lower fluences than hydrogen bombardment. Typical experimental results of roughness effects on blister formation are summarized in Figs. 6 and 7. Figure 6 shows the SEM micrographs of the surfaces with different roughnesses after 100 keV helium bombardment to a fluence of 1.0 x 10^{19} particles/cm^2. Micrographs of surface A show numerous blisters, some of which flake and crack (see Fig. 6a and also Fig. 5). On the other hand, for rough surfaces abraded with emery paper blister formation is crucially reduced or suppressed. For surface B formation of large blisters is extremely suppressed, although blisters are formed (Fig. 6 b and c). Because formation of small blisters is not prevented, the surface topography changes. For surfaces C and D the blister formation is completely suppressed. As shown in Fig. 6d, the surface topography does not change when we prepare the target with sufficiently roughened surface. However, blisters appear on insufficiently roughened surface and surface topography eventually changes (Fig. 6 e and f). In Fig. 7 are shown the histograms of the size distributions of blisters formed by 100 keV helium ion bombardment of the target with surfaces A and B to a fluence of 1 x 10^{18} particles/cm^2. It can clearly be seen that the most probable blister diameter shifts to a smaller value, that is, from 2 μm in electropolished surface to 1 μm in roughened one abraded with emery paper of #1200. Total blister number counted in the area of 2.6 x 10^{-8} cm^2 is also reduced from 177 in electropolished surface to 133 in roughened one. Various values characteristic of blistering caused by helium and hydrogen bombardment are obtained in the present experiment and listed in table 2.

4. DISCUSSION

The experimental results have clearly shown that suitable surface topographies bring a noticeable effect on the reduction or suppression of blister formation. A schematic showing the effect of a surface topography on blister formation is in Fig. 8. It represents successive stages for blister formation both on a smooth surface and on a rough one consisting of many grooves. Here, we take account of two parameters; the depth of grooves,

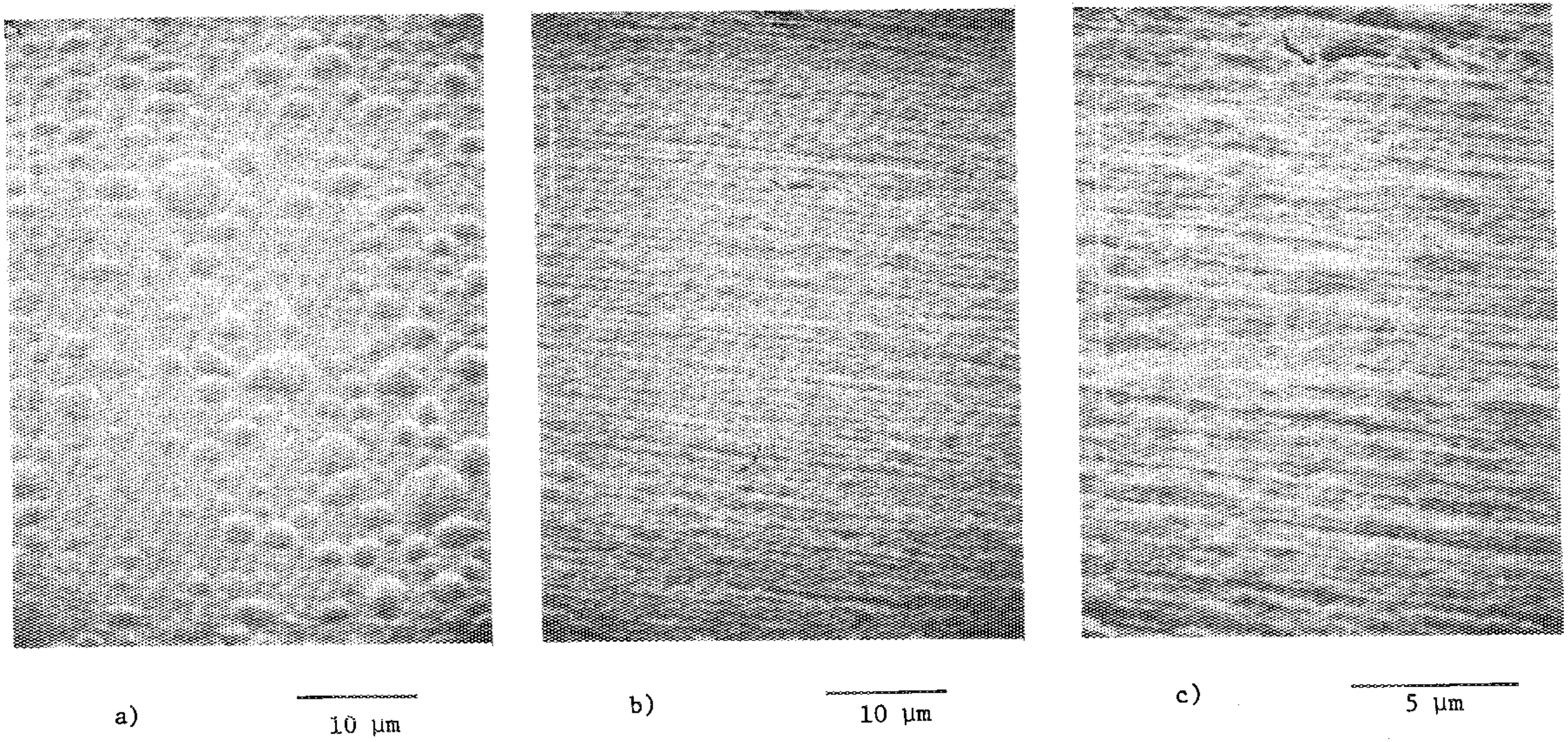
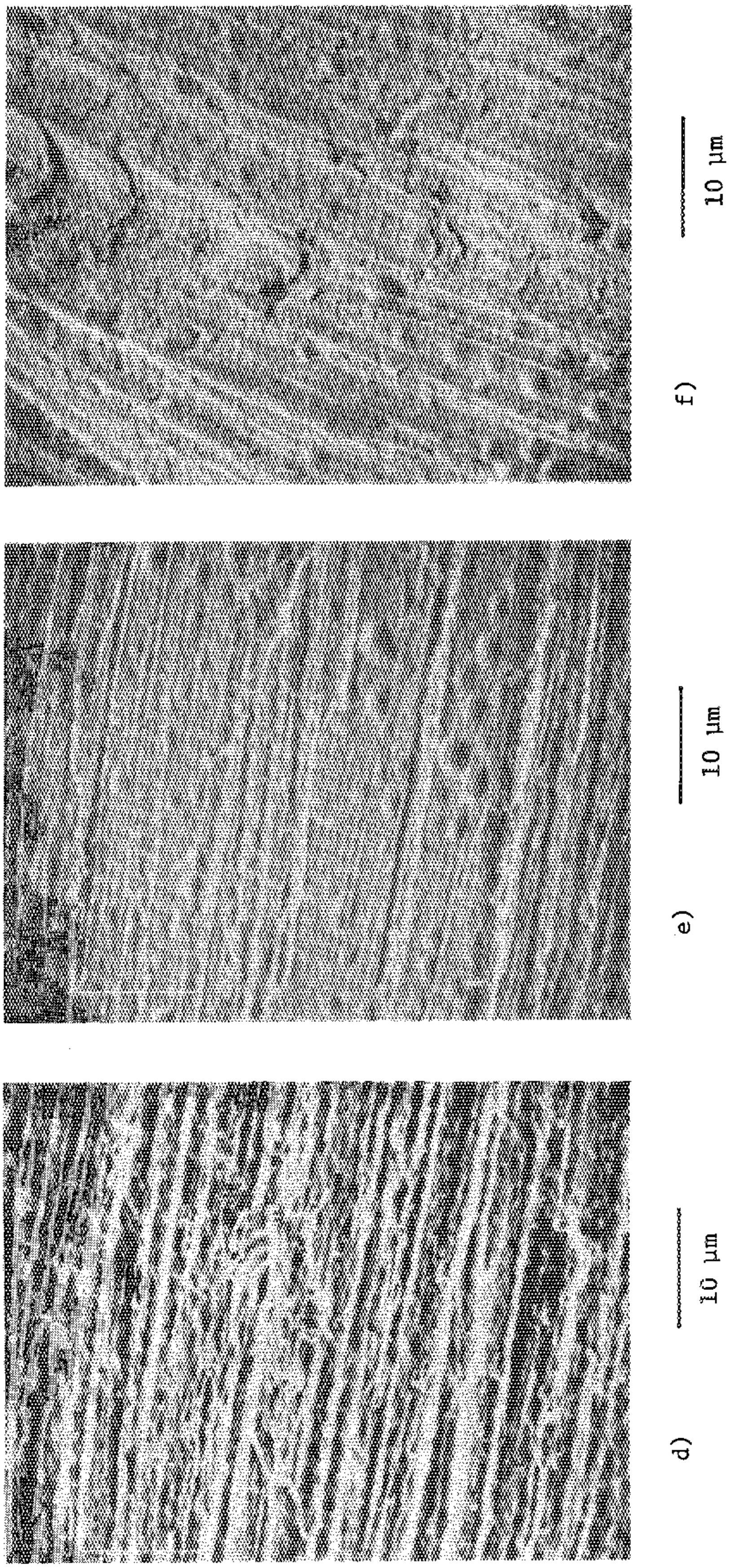


Fig. 6 Typical SEM micrographs of electropolished surface (a) and rough surfaces abraded with emery paper of #1200 (b and c), #400 (d and e) and #100 (f) after bombardment with 100 keV helium ions to a fluence of 1.0×10^{19} particles/cm^2.



d)

e)

f)

Fig. 6 (continued)

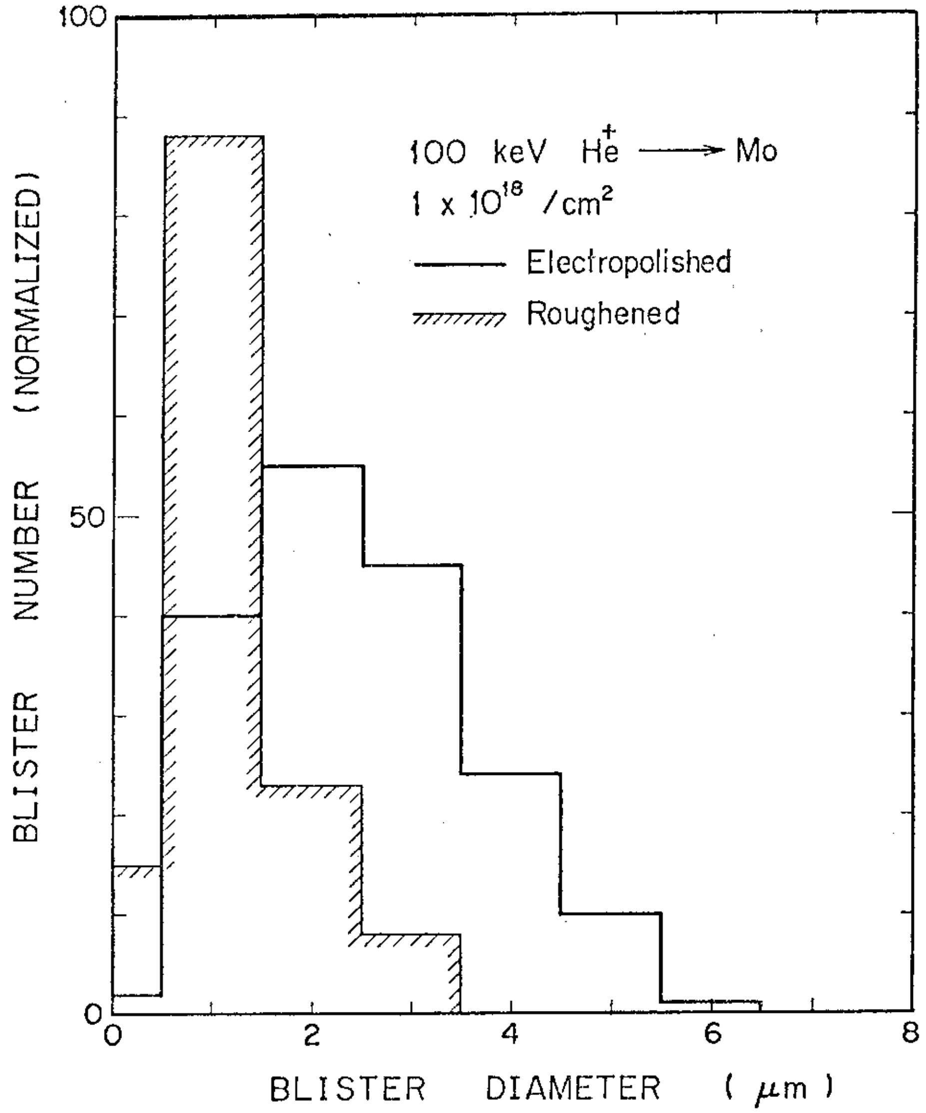


Fig. 7 Size distributions of blisters formed by 100 keV helium ion bombardment of molybdenum with electropolished surface (━) and rough surface abraded with emery paper of #1200 (▨).

Table 1 Typical topographic values obtained from SEM micrographs of three grades of roughened surfaces. Here three surfaces roughened with emery paper of #1200, #400 and #100 are named surfaces B, C and D, respectively.

	Surface B	Surface C	Surface D
distance between the two neighbouring grooves (μm)	< 3	< 3	< 3
depth of grooves (μm)	0.1 - 1	1 - 4	5 - 15

Table 2 Typical experimental values characteristic of blister formation in polycrystalline molybdenum.

	100 keV helium	200 keV H_2^+
critical fluence (particles/cm^2)	7.5×10^{17}	1.0×10^{19}
blister diameter (μm)	2.5 ± 0.2	4.0 ± 0.2
thickness of blister cover (μm)	0.36 ± 0.02	—
projected range (μm)	0.3524*	0.5170*

* calculated values (K. Sone and K. Shiraishi; JAERI-M 6094 (1975))

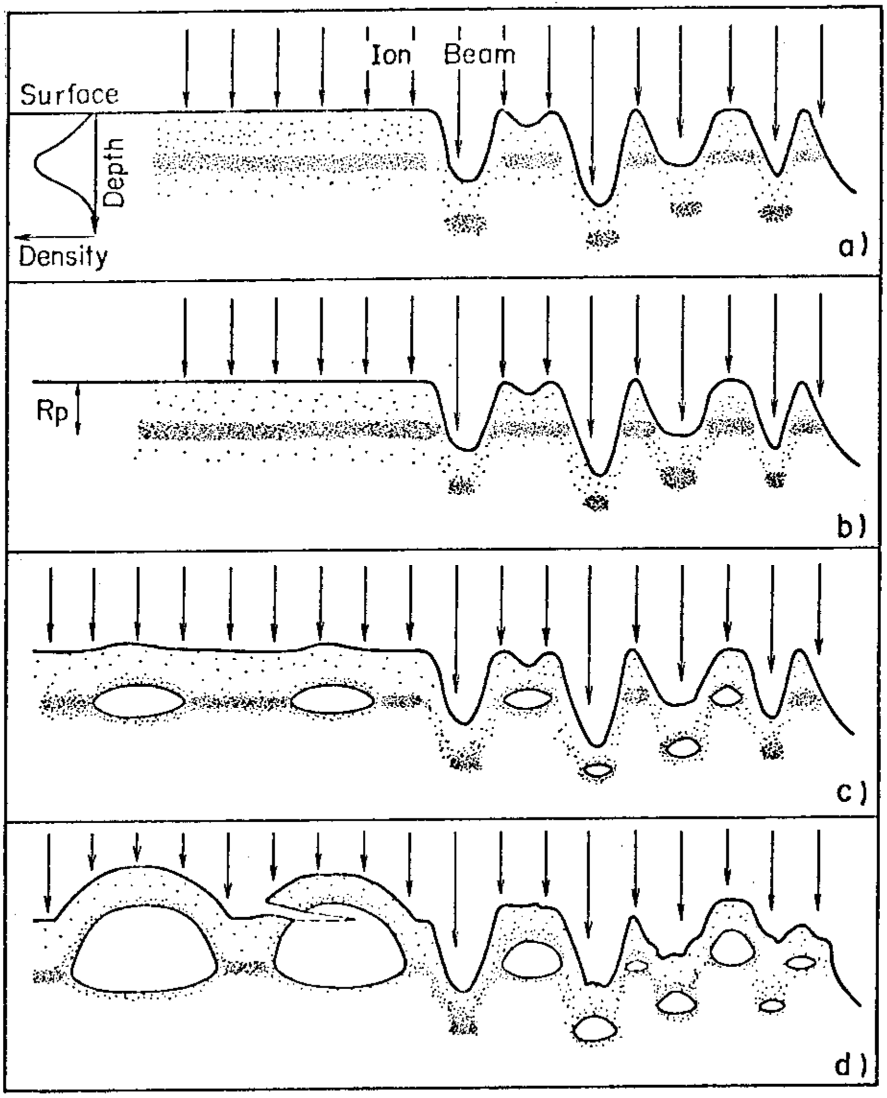


Fig. 8 Schematic representation of blister formation both in smooth and rough surfaces.

d, and the distance between the two neighbouring grooves, λ, to characterize the rough surface (see table 1). When energetic helium ions bombard the target, they are trapped in it. Near the depth corresponding to the projected range where damage is intense and helium concentration is sufficiently high, helium gas-filled-bubbles are formed if the solubility of helium in the target is very low. When target has a smooth surface the bubbles can grow to form blisters and eventually to deform the surface layer. Let us consider a rough surface consisting of many grooves whose depth d and distance λ mentioned above are larger than the thickness of the blister cover and smaller than the blister diameter, respectively. The region of bubble coalescence is divided into many parts and the growth of large gas bubbles is inhibited, which result in reducing or suppressing the blister formation. From the specific values of blisters produced in molybdenum bombarded with 100 keV helium ions, the values of the depth d and the distance λ can be determined to be larger than the thickness of the blister cover of 0.36 μm and smaller than the blister diameter of 2.5 μm, respectively (see table 2). It should be noted that the topographic values of rough surfaces used in this experiment involve the values in the above condition. Both the thickness of the blister cover and the blister diameter (D) are correlated to the projected range R_p. The former is nearly equal to Rp13), whereas the latter is related by $D/R_p \simeq 8$1). These relations are confirmed by the present experiment (see table 2). Therefore, suitable values of both the depth d and the distance λ for blister suppression can be determined from the incident energy of the projectiles. In order to suppress the blister formation caused by 100 keV hydrogen bombardment the depth d and the distance λ should be larger than 0.52 μm and smaller than 4.0 μm, respectively.

In future fusion reactors the incident particles from plasma to the wall will be of wide distributions both in energy and in angle of incidence. At rather low energies below a few 10 keV sputtering is a dominant process for surface erosion and eventually causes a change in surface topography. Therefore, further studies of the roughness effects on surface erosion should be required by the use of ion beams with polyenergetic and/or mixed ion species consisting of helium and hydrogen ions.

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