KFKI-1981-22

F. PÁSZTI
L. POGÁNY
G. MEZEY
E. KÓTAI
A. MANUABA
L. PÓCS
J. GYULAI
T. LOHNER

BLISTERING AND EXFOLIATION INVESTIGATIONS ON GOLD BY 3.52 MeV HELIUM PARTICLES

Hungarian Academy of Sciences

CENTRAL RESEARCH INSTITUTE FOR PHYSICS

BUDAPEST

2017

KFKI-1981-22

BLISTERING AND EXFOLIATION INVESTIGATIONS ON GOLD BY 3.52 MeV HELIUM PARTICLES

F. Pászti, L. Pogány, G. Mezey, E. Kótai, A. Manuaba, L. Pócs, J. Gyulai, T. Lohner

Central Research Institute for Physics
H-1525 Budapest 114, P.O.B. 49, Hungary

Submitted to Journal of Nuclear Materials

HU ISSN 0368 5330
ISBN 963 371 799 X

ABSTRACT

The mechanism of blister formation was investigated on cold-rolled gold target by 3.52 MeV helium ion bombardment. The critical dose was found to be $6x10^{17} He^{+}/cm^{2}$ under present experimental condition.

To study the inner morphology of the blisters, they were opened mechanically. Based on these observations several new features are reported.

A speculation of high-energy blister formation is discussed, based on the fact that the diameter increases with sudden size changes. It is pointed out that in MeV energy region this formation could be exfoliation rather than blister confirming the previous investigations.

АННОТАЦИЯ

При исследовании процесса образования блистеров /вздутий/ на поверхности прокатанного золота при облучении их ионами гелия с энергией 3520 кэВ, было установлено, что доза, необходимая для блистеринга в данных экспериментальных условиях, составляет $6x10^{17}$ ион/см^{2}.

Для изучения внутренней структуры блистеров, они были вскрыты механическим путем. На основании этих исследований был написан отчет об обнаруженных новых свойствах.

На основании наших наблюдений о том, что увеличение диаметра происходит скачкообразно, нами развита модель образования блистеров при большой энергии. В соответствии с результатами предыдущих исследований, обнаружено, что эти деформации поверхностей скорее похожи на шелушение, чем на блистеринг.

KIVONAT

3.52 MeV energiáju hélium ionokkal hidegen hengerelt arany céltárgyak felületét bombázva tanulmányoztuk a hólyagosodás (blistering) folyamatát. Az adott kisérleti körülmények esetén a kritikus dózist $6x10^{17}$ He^{+}/cm^{2}-nek találtuk.

A hólyagok belső szerkezetének tanulányozása érdekében azokat mechanikai uton kinyitottuk. E vizsgálatok alapján több uj tulajdonságról számolunk be.

Azon megfigyeléseinkre alapozva, hogy az átmérő ugrásszerüen nő, megfontolásokat tettünk a nagyenergiáju hólyagok kialakulásával kapcsolatban. A korábbi kutatások eredményeivel összhangban azt tapasztaltuk, hogy a felületek észlelt deformációja inkább hámlás (exfoliation) mint hólyagosodás.

Introduction

In the future CTR machines the basic fusion process will be the $^{3}H(d,n)^{4}He$ nuclear reaction. As a consequence of this, a great number of helium particles reach the first wall with the energy maximum of 3.52 MeV [1,2]. Although the first wall erosion is subjected to extensive studies, relatively few experiments were done to investigate blistering of materials by helium ion bombardment in MeV energy region.

The conclusion of previous works [3,4] can be summarized as the following:

i/ only one huge blister was formed by helium irradiation and it covered almost the total bombarded area;

ii/ the relationship between the blister skin thickness and blister diameter based on low energy experiments seems to be not valid at high bombarding energies;

iii/ the higher the irradiation dose was used the larger blister size was found but no data are available on the dependence of the diameter on dose;

iiii/ no attempt was done to study the internal structure of the unraptured blisters neither in the MeV region nor at lower energies.

This paper tries to answer these open questions. Some examples are shown for the morphology of the interior of huge blisters.

Experimental

3.52 MeV $^4He^+$ particles were used from a 5 MeV Van de Graaff accelerator. As a good model material, cold-rolled gold of chemical purity was chosen as target. Gold plates were chemically cleaned in H_2SO_4 and clean alcohol and distilled water. The bombardment was done perpendicular to the surface on a spot size of 1 mm^2. The current was kept as low as 100 nA, so the estimated temperature rise was below 9^oC, that is the target was on room temperature during experiments. The bombarding dose was measured by standard secondary electron suppresion and current integration. The vacuum was kept at $5x10^{-5}$Pa. A special cold trap system minimized the hydrocarbon deposition onto surface. No color change was experienced even for the highest dose.
To determine the critical dose a binocular system with a magnification of ten was applied to in-situ observation during bombardment. This system was able to detect blisters with a minimum diameter of 50 μm as an intensively glistening spot. Anyway, the smallest observed diameter was about 90 μm in present experimental conditions. Blisters were produced with five different doses /Table 1./.

The blisters were investigated by a JEOL-JSN-35 type scanning electron microscope with a lateral resolution of 10 nm. To investigate the structure inside, the blisters were opened mechanically by a wolfram pin and a detailed study of both the bottom of the blister and the inner side of lid was done. Scanning electron micrographs were made with different magnification and tilt angles. For calibration a standard with 1.102±0.002 μm scale served.

Results

Our first observation concerned the critical dose which was found to be 6×10^{17} $^4He^+/cm^2$ with a blister diameter of about 90 μm. After prolongated irradiation, the higher the dose was applied, the larger diameter was formed suggesting, that the only limiting parameter is the area of the bombarded spot.

The shape of the blisters, just after appearance were regular like in the previous studies. At higher doses, however, serious alteration was observed. Strange, dome like structure of more than one level was exhibited. Two, three and four level blisters were found for increased helium dose. The second and higher levels were grown with smaller diameters on the top of the biggest one. Fig. 1 shows a three-level structure of sample B.

On samples C,D,E several slips were experienced along crystal planes and some pieces of the cover moved parallel to each other. One may speculate that helium gas that produced the blister could escape through these slips, so spontaneous rapture might not be expected during further bombardment /Fig. 2 taken on sample D/.

After opening the blisters mechanically, both on the bottom of blister and the inner side of lid well-separated, quasi-circular regions were observed on SEM micrographs bordered by bright zones caused by enhanced secondary electron emission of roughened surface. This can be seen on Fig. 3 taken on sample B.

The number of regions increased with helium dose. These regions were characterized by an average diameter. The error of the diameter was defined as the maximum alteration from the circular shape. The diameters of different regions for blisters are summarized in Table 1.

Table 1.

Diameters of quasi-circular regions in μm units

Sample	Dose	Regions			
	x 10^{18} ions cm^{2}	No 1	No 2	No 3	No 4
A	0.81	83±8	200±16	-	-
B	0.92	88±5	233±17	391±25	-
C	1.37	91±8	225±25	270±42	504±25
D	1.68	88±5	138±8	300±15	580±65
E	2.06	?	?	?	?

At sample E /for highest dose implantation/ the regions were smeared out. It is apparent, that the size of the same region in different blisters is approximately the same, especially for the first one. It must be emphasized that regions on the bottom of blisters and the inner side of lids are mirror image of each other and they are in one-to-one correspondence with the levels on the cover (see Fig. 3.). So one can conclude that this structure is correlated in some way with the different stages of blister evolution. Note, that the diameter of the first region is equal with that of blister observed optically at critical dose.

Applying higher magnification to investigate the border zones, SEM micrographs show crater-like structures consisting of splitted lamellas bending outward on the bottom of blister and inward on the wall of the skin (Fig. 4., Fig. 5. and Fig. 9.).

Besides, in different regions, different degree of surface roughness was observed both on the bottom of blisters and the inner side of lids. The roughest is the region No 1. and the roughness decreases going outward from the centre. For example Fig. 5. shows the border zone between regions No 1. and No 2. on the lid and Fig. 9. on the bottom of sample B, respectively.

On the bottom of blisters several dips of quasi-circular shape in the diameter range of 10-90 μm were observed. The material missing from them was found on the inner side of lid. The 0.5 μm depth of these formations are surprisingly uniform (Fig. 6.).

Both on the skin and bottom a network of cracks was found due to the radiation hardening (Fig. 7.).

The effect of radiation hardening can also be studied on the side view micrographs of the lid of sample D after the opening (Fig. 8.). Cracks together with thin, ≈0.8 μm hardened layer with sharp border on the originally inner side and the thick, plastic external cover can be observed.

Unsuccesful attempts were done to measure the original thickness of blister skin because of its plastic elongation during opening up and blister formation. The measured values were about 4 μm and the tabulated range of $^4He^+$ in gold at the energy used for bombardment is R_p=5.5 μm with ΔR_p=0.61 μm [5].

Perhaps the most interesting observation is the appearance of secondary blisters with diameters of 0.6-3.5 μm (Fig. 9. and Fig. 10.). Their density increased with the dose. For example, inside the sample A only four small blister was found.

For the other samples, the density of secondary blisters was the highest near to the region No 1. and decreased abruptly from region to region going outward from center. On the inner side of the cover of sample E a number of small blisters were also found (see Fig. 11.).

The Fig. 12. summarizes the present observations.

Discussion

The first conclusion is that these results confirm, that the relationship between blister skin thickness and diameter experienced at lower bombarding energy [6, 7] is not valid anymore at high energies. The maximum diameter was about 0.8 mm at present experiments. This tendency was observed earlier for 2 MeV $^4He^+$ ions on Zr and SS-316 materials [3, 4].

From the experimental observations, however, one can speculate the mechanism of the high energy blister (or exfoliation) formation. Several indirect evidences support that the blister grows by sudden size changes. In other words, the diameter increase consists of quasi-equilibrium and expansion stages. Expansion takes place unhindered.

In the quasi-equilibrium stage the edge of blisters is not able to be splitted up, although it is subjected to large mechanical stresses arising from the extremly high gas pressure inside. These forces try to move the edge of the lid away from the center. Such a stress is supposed to cause the splitted lamellas at the bordering zones of different regions.

The first expansion coincides with the appearance of the blister at the critical dose and corresponds to the region No 1.

The splitting up in the expansion stage keeps going on till gas pressure decreases to the point where the quasi-equilibrium is reached.

The new expansion is initiated by helium-rich regions around the first one. If a second blister appears next to the first such a way that its diameter would overlap the first one, additional forces help to split and lift up the edge at the lamellas and the expansion goes on around the first formation till the new equilibrium stage is reached.

As a result of plastic deformations, the lid preserves the shape of the smaller blister of the earlier stage of evolution (more than one level structure). This idea seems to be confirmed by the good agreement of diameters of same regions in different blisters (Table 1.).

The different surface roughness in different regions and the presence of secondary blisters, also support these considerations, together with the assumption that during formation, the cover becomes thinner than the range of helium particles. Near to the central region most of the bombarding particles go through it with low energy and large energy spread due to energy loss and straggling inside the skin. This synergestic beam could initiate forward sputtering on the cover and a normal one on the bottom. Besides, the highest energy part could be implanted into the bottom. The remaining portion of the beam with the lowest energy stops inside the skin. These implantations should be responsible for the formation of small blisters. This is the reason why the secondary blisters appear on the bottom first.

We would like to emphasize that these latter phenomena are the consequence of such a multiple energy implantation that is similar to the radiation which would strike the surfaces of structural components of fusion reactors i.e. broad energy spectrum with the variety of the angle of incidence. In our knowledge, no experiments were performed under such a realistic conditions.

Finally, we have to point out the significance of the investigation of the inner morphology of unruptured blisters, because the increase of the bombarding dose, in our experiences, smears out fine, structural informations characterizing the early stages of blister evolution.

Conclusion

Recent SEM studies which perfomed on mechanically opened blisters gives a number of new information about the inner structure of this kind of formations. Based on these observations we suggest a new model of high energy blistering - or rather exfoliation - phenomenon. Although more experiments are necessary to clarify what is going on, we think, this work together with our speculations give some stimulus for further investigations to clarify this essential process in future CTR machines. Furthermore, at this time it is not clear yet how many of our observations could be generally accepted and what part of them is specific to the gold. This is why we suggest to repeat similar experiments on wide variety of other materials too.

References

[1] L.M. Hivley, G.H. Miley, Nucl. Fusion 17 /1977/ 1031.

[2] G.H. Miley, L.M. Hivley, J. Nucl. Mat. 76-77 /1978/ 389.

[3] D.K. Sood, M. Sundararaman, S.K. Deb, R. Krishnan and M.K. Mehta, IEEE Trans. Nucl. Sci. NS-26 /1979/ 1308.

[4] D.K. Sood, M. Sundararaman, S.K. Deb, R. Krishnan and M.K. Mehta, J. Nucl. Mat. 79 /1979/ 423.

[5] J.F. Ziegler, The stopping and ranges of ions in matter, Vol. 4, Pergamon Press, New York, 1977.

[6] M. Risch, J. Roth and B.M.V. Scherzer, Proc. Int. Symp. on Plasma Wall Interaction, Jülich, 1976 /Pergamon Press, Oxford 1977/ p. 391.

[7] S.K. Das, M. Kaminsky and G. Fenske, J. Nucl. Mat. 76-77 /1978/ 215.

Fig. 1. The three level structure on sample B before opening

Fig. 2. A slip on the cover of sample D before opening

Fig. 3. Sample B after opening mechanically. Quasi-circular regions can be seen both on the bottom of blister and the inner side of skin.

Fig. 4. Lamellas on the border zone between region No 1. and No 2. of sample D.

Fig. 5. The No 1. and No 2. region on the inner side of skin on sample B. In the different regions, different degree of surface roughness can be observed.

Fig. 6. Micrograph taken on sample E, showing missing material from the bottom of the blister and the corresponding material on the inner side of the lid.

Fig. 7a and 7b. Cracks due to the radiation hardening on the bottom of sample D. Thick paralell lines are the prints of surface scratches. Besides, border zones between regions, quasi-circular dips with the depth of 0.5 μm can also be observed.

Fig. 8. The effect of radiation hardening on blister skin. The micrograph was taken on side view of the lid of sample D. The cracks and the thin hardened layer were on the inner side.

Fig. 9. Secondary blisters on the bottom of sample B next to the border of regions.

Fig. 10. Secondary blisters inside the sample D.

Fig. 11. Secondary blisters on the inner side of skin of sample E.

Fig.12. A schematic drawing summing up the observations.

1. The edge of blister, border zone of region under formation
2. Quasi-circular dip and missing material on the skin
3. Secondary blister on the bottom of blister
4. Border zone of a region
5. Secondary blister on the skin
6. Surface scratch and its print on the bottom
7. Radiation hardening crack
8. Slip on the cover
9. Soft, thick layer of the cover
10. Thin, hardened layer of the cover
11. Inner volume of blister
12. The bottom of the blister

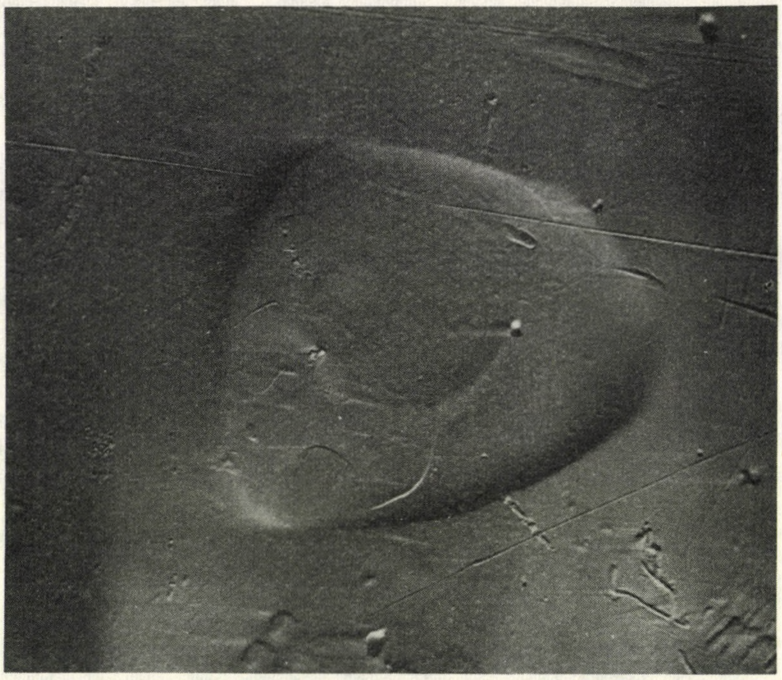

Fig. 1.

Fig. 2.

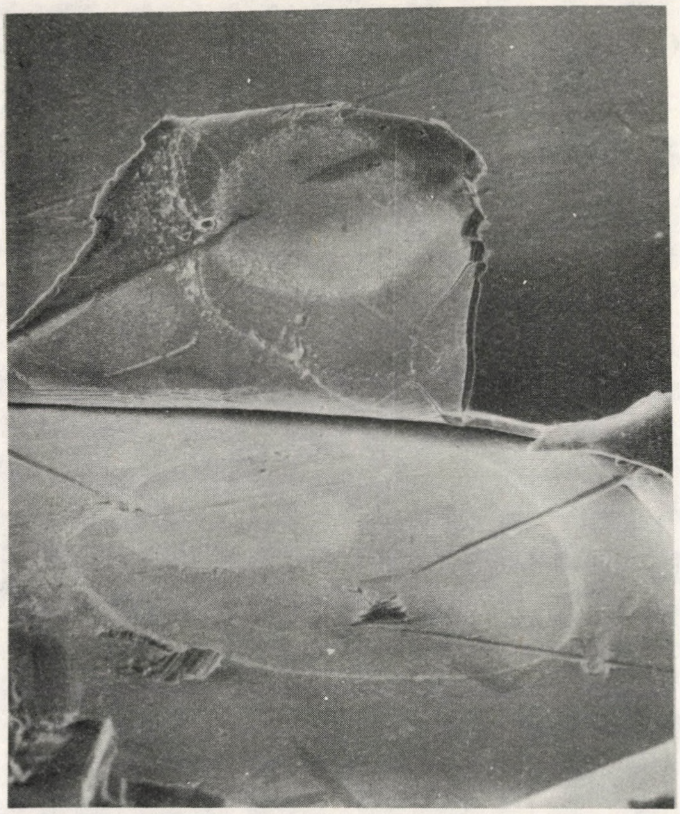

Fig. 3.

100 μm

Fig. 4.

1 μm

Fig. 5.

10 µm

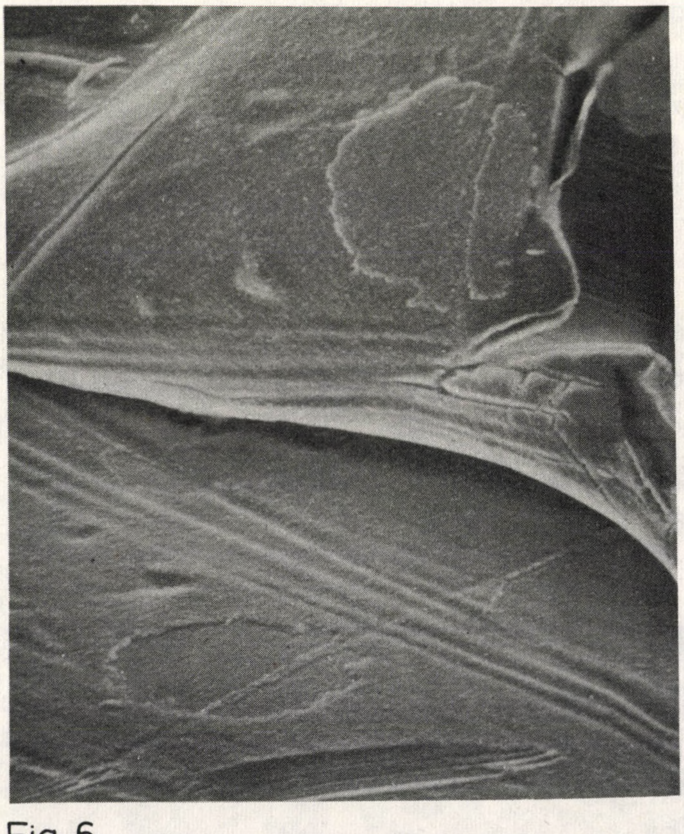

Fig. 6.

20 µm

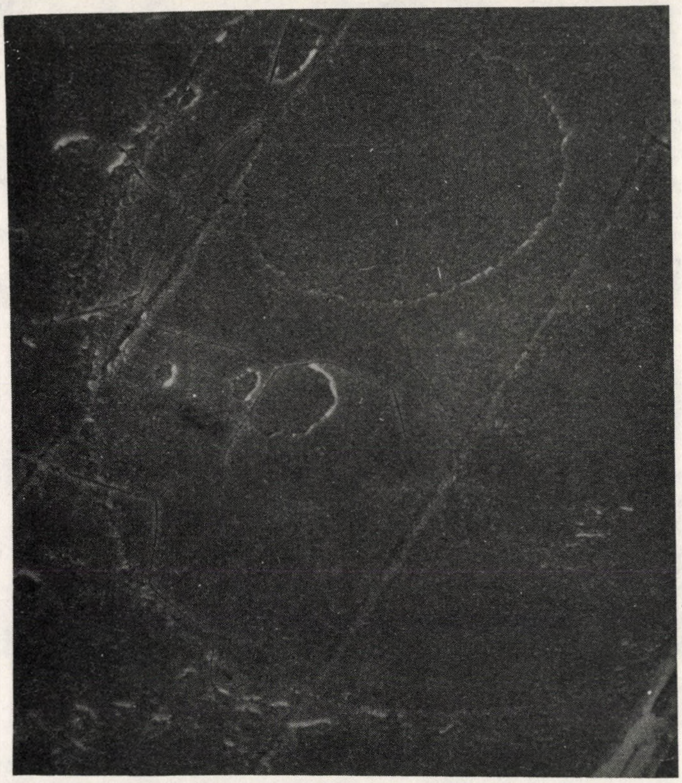

Fig. 7/a.

30 μm

Fig. 7/b.

10 μm

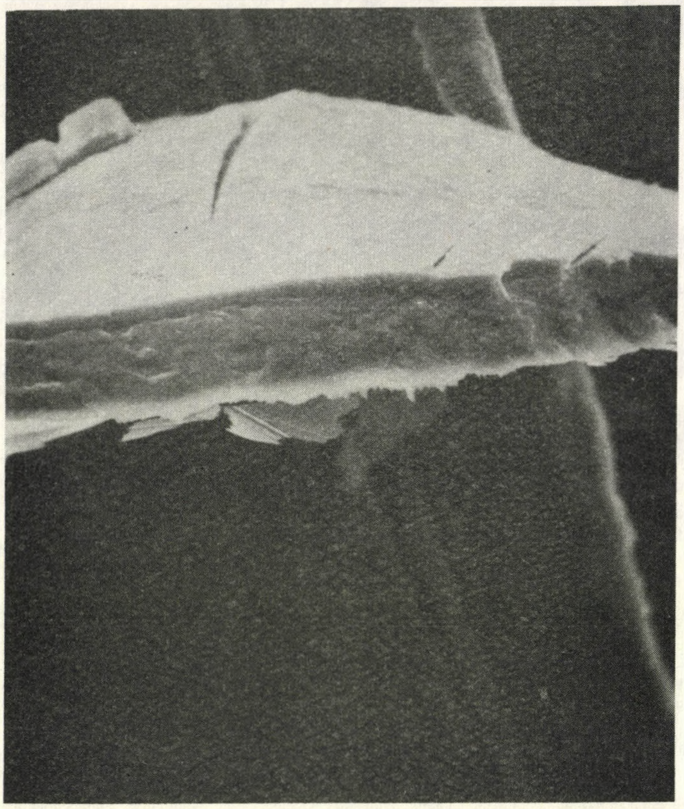

Fig. 8.

10 µm

Fig. 9.

10 µm

Fig. 10.

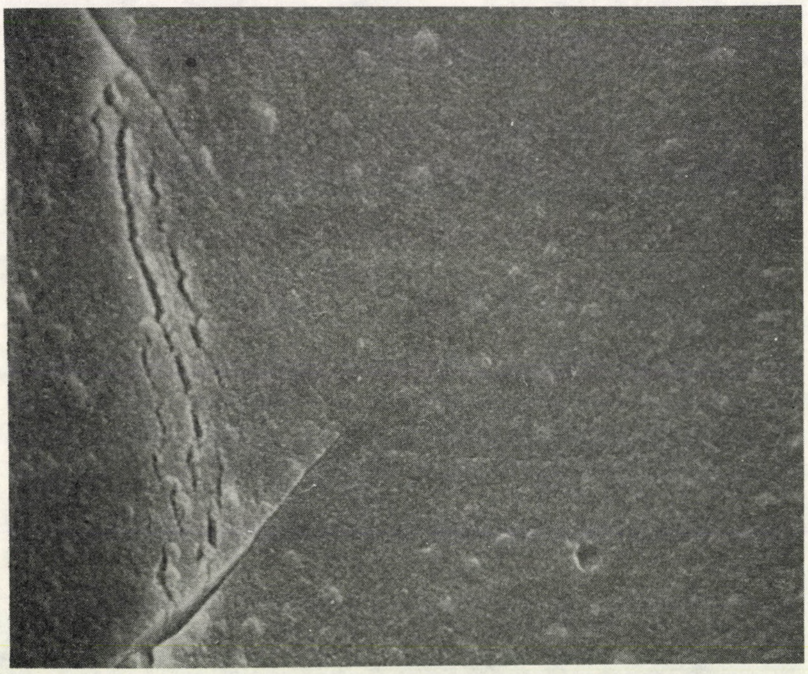

Fig.11.

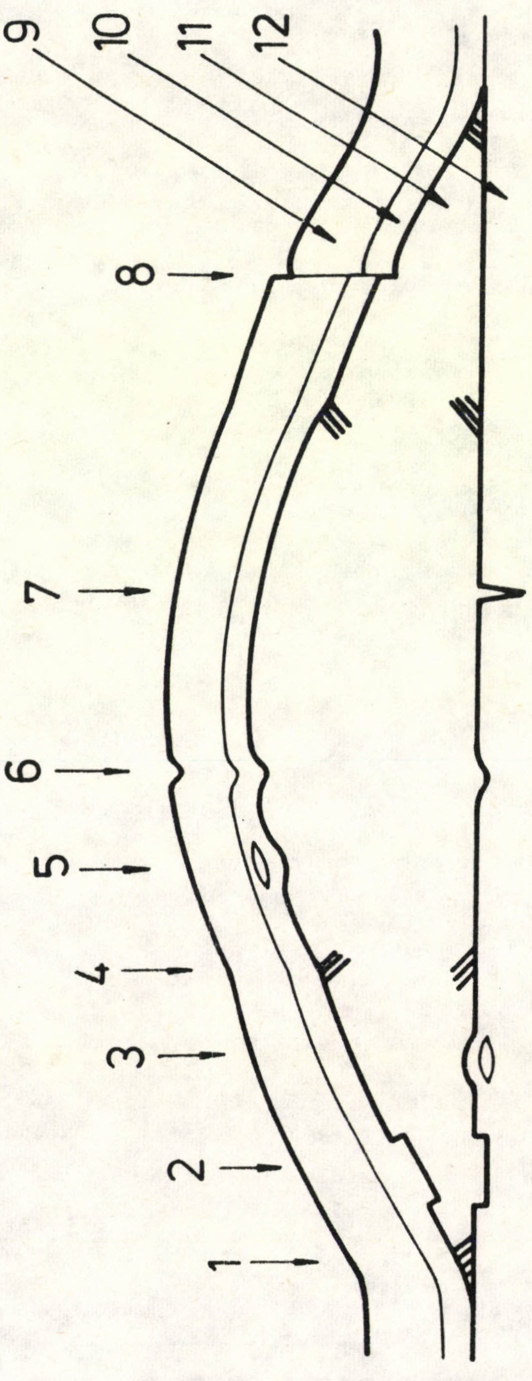

Fig. 12.

Kiadja a Központi Fizikai Kutató Intézet
Felelős kiadó: Szegő Károly
Szakmai lektor: Bakos József
Nyelvi lektor: Pócs Lajos
Példányszám: 305 Törzsszám: 81-223
Készült a KFKI sokszorosító üzemében
Felelős vezető: Nagy Károly
Budapest, 1981. április hó