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POST-IRRADIATION HELIUM BLISTERING IN COPPER, AUSTENITIC STEELS AND ALLOYS OVER THE TEMPERATURE INTERVAL 0.65-0.95T_{melt}

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The specimens of copper, austenitic steel 08Cr16Ni15Mo3Nb and nickel-rich 03Cr20Ni45 alloys were irradiated by α-particles (50 MeV, 0.3...3.6 · $10^{21}m^{-2}$) through the special filter-mask and subjected to following thermal treatment at temperatures up to 0.95 · T_{melt} for 1...100 h . The helium behavior in the materials was investigated by the methods of scanning electron microscopy and profilimetry of a surface. It was shown that the development of helium bubbles in a closed bulk of the specimens during high-temperature annealing leads to appearing steps on irradiated surfaces followed by flacks and blisters. Migration of helium bubbles in the field of stress gradients causes the nucleation and growth of the long-distance gas-filled cracks. Accumulation of helium bubbles in the cracks leads to lifting of surface layers and formation of a dome-shaped step on the irradiated surface, i.e. the bulk blistering occurs.

INTRODUCTION

Copper (99.99 wt.%), the 08Cr16Ni15Mo3Nb stainless steel and the Cr20Ni45 nickel enriched alloys were taken as objects for helium behavior investigation. Specimens with thickness of 1 mm were irradiated by α-particles at 40 and 50 MeV in the isochronal cyclotron of the Institute of Nuclear Physics NNC RK to fluences of 0.3x10^{21} ...3.6x10^{21} m^{-2} through a special filter-mask with diameters of circular holes 0.3...3.0mm. The irradiation temperature didn't exceed 375 K. The scheme of irradiation and the following study is presented in fig.1. Prior to irradiation, the specimens were subjected to the appropriate thermal treatment. Copper was annealed in vacuum at 1325 K for 3 h, in this case the mean size of grains exceeded 0.5mm; stainless steels and alloys were subjected to austenization at 1425 K for 2 h, the mean size of their grains was about 70μm. After performance of the successive 50K-isochronal and isothermal annealings at temperatures 675...1325 K surfaces of irradiated specimens were studied with the profilometer.

Thereafter the specimens were cut up normally to a surface, planished, polished and subjected to electrolysis and chemical etching treatment to reveal sections of helium cavities with the aid of the scanning microscope REM-200.

RESULTS AND DISCUSSION

Profilometry investigations of copper specimens with the mean concentration of 0.02...0.14 at.% in a straggling zone showed that after annealing at temperatures higher then 0.8· T_{melt} the step with a height up to several tens of microns appeared on the irradiated surface. In so doing the shape and size of the step copied the shape and size of the mask holes. Special metallography studies allowed revealing the primary role in a step formation of a swelled layer containing helium. This layer is located in a straggling zone at the depth of 0.36mm. The obtained results showed clearly that a step formation occurred as a result of displacement of interlayer, between the doped layer in a straggling zone and the irradiated surface, from the other undeformed environment. This gives the possibility to conclude that the step on a surface being nearest to the doped layer forms as a result of shear deformation of this interlayer along the cylindrical surface separating it from the undeformed area. This takes place in the process of stress relaxation in the doped layer during post-irradiation annealing. The temperature T of the beginning of step formation and the following step behavior versus both time and temperature of post-irradiation annealing turned out to depend on the concentration of implanted helium M_{He} (T diminished with M_{He} increasing) and not to depend on a size (0.3≤ d ≤ 3.0 mm) and a depth of the doped layer location (50 ≤ Z ≤ 350 mm). Here d is a diameter of a hole in the filter-mask; Z is a depth of the layer occurrence from the studied surface. To illustrate the above mentioned we represent a curve of evolution of the step height versus the temperature of successive isochronal annealings for copper specimens with helium content M_{He} ≈0.07at.% (fig.2). Here the depth of occurrence of the doped layer is varied from 50 μm to 350 μm.

Fig.2 shows a monotonous increasing of a step height h with annealing temperature rising. The change of h depending on time of thermal annealing t in the temperature interval of 1075...1275 K was found to follows to the relation

$$h \sim t^{1/2} . \quad (1)$$

The height change depending on the initial concentration of implanted helium M_{He} for wide intervals of both temperatures and times of annealing can be described as

$$h \sim (M_{He})^{0,9} . \quad (2)$$

Definition of mean values of the concentration ρ, size D and summary volume of bubbles V over the swelling layer showed that V increased monotonously with both temperature and annealing time increasing. The volume increasing was found to be a result of increasing of mean sizes of bubbles coincidentally with decreasing of their concentration. The studies showed that changes of ρ and D with time during isochronal annealing at temperatures of 1075...1175K could be described by the following relations

$$\boldsymbol{D \sim t^{1/3}}, \qquad (3)$$

$$\boldsymbol{\rho \sim t^{-1/2}}. \qquad (4)$$

The results of study of bubble growth kinetics in copper at various concentration of implanted helium showed that dependencies of mean size and concentration of bubbles satisfied the following relations

$$\boldsymbol{D \sim (M_{He})^{0,25}}, \qquad (5)$$

$$\boldsymbol{\rho \sim (M_{He})^{0,11}}. \qquad (6)$$

Analysis of these data allows concluding about the main role of the bubble size increasing for a process of swelling. Here and thereafter the word “swelling” means the helium swelling. It should be specially noted that the summary volume of bubbles in the doped layer turned out to be equal to the volume of a step forming at a specimen surface within the 15% of experimental errors [1]. This correspondence is valid in a wide interval of temperatures and annealing times in case of the progressive change of bubbles content in the layer. Except this, it was found that the relation of the step height h to the maximal value of swelling V_m in the layer was about 0.11 μm/%.

At isothermal annealings of copper specimens for a long time at temperatures of 1175K we found another unknown phenomenon as helium bubbles accumulation in the center of a straggling zone with formation of macroscopic cracks. It turned out that the more intensive is the process of such accumulation, the higher is the temperature of annealing and the smaller is the concentration of implanted helium in the layer. Accumulation of almost all implanted helium in macrocracks results in the substantial modification of the step profile on a surface of copper specimens, i.e. the transformation from a plate profile to a dome-shaped one. It was shown that in copper single-crystals not having places, which could relieve running of the local relaxation of stresses in doped layers, such as grain boundaries, the process of bubble accumulation proceeded at higher rate. The analysis of the data obtained was made on the base of the commonly adopted criteria giving the mechanisms controlling the process of growth and migration of bubbles.

It shows that the most probable cause for bubble accumulation in the center of a doped layer is bubble migration in the field of gradients of stresses appearing in an inhomogeneous swelling layer.

The analysis of the dependencies of mean sizes and concentrations of bubbles on both temperature and annealing time and also on the concentration of implanted helium permits to conclude that Ostvald’s mechanism of resolution doesn’t govern the process of bubbles growth.

Consequently, the observed increasing of the summary volume of bubbles in copper during post-irradiation annealings is mainly caused by coalescence of bubbles during their migration along the gradients of stresses from a zone of maximal stresses to the center of a straggling zone.

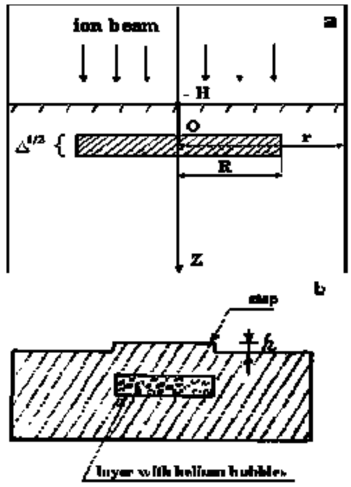


Fig.1. Appearance of a step on surfaces of specimens: a – after irradiation by α-particles, b – after irradiation and succeeding annealing

The calculations of the fields of elastic stresses were performed according to the theory of thermal elasticity. These demonstrate that the elastic stress fields appear in a swelling layer during helium implantation and following nucleation and coarsening of helium bubbles in a straggling zone as a result of following high-temperature annealings (fig.4). Here we proceeded from the assumption that stresses in a swelling layer were proportional to a local value of the relative volume of bubbles and considered such factors as the final size of a disk-shaped swelling layer and the normal distribution of bubbles over the layer section [2].

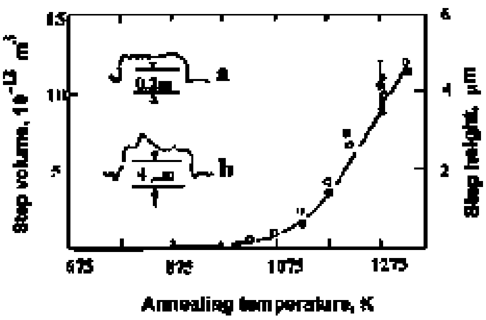


Fig. 2 Change of step height (o) and summary volume of bubbles (•) in copper specimens after successive isochronal annealings. a,b - fragments of profile diagrams of a surface after annealing at 975 K and 1325K

As can be seen from the figure, the inho-mogeneous distribution of stresses occurs over the section of the layer along the direction of α-particles, bombarded the specimen (axis OZ), and nearby the cylindrical peripheral zone of a disk-shaped layer. Under such stress gradients acting the migration and coalescence of helium bubbles in copper proceed toward the maximal stresses at temperature of 1175 K and higher, that promotes forming of long-distance cracks and blisters on the irradiated surface (fig.4). Helium bubbles accumulation in the zone of maximal stresses in the swelling layer leads to formation of micro- and macro- (long-distance) cracks in metals and alloys. With increasing of temperature and annealing time, migrating bubbles combine with a crack, that is followed by increasing of internal pressure within the crack's cavity and in some cases leads to the formation of a lentil-shaped crack and a dome on the irradiated surface, i.e. to a blister. It means that swelling of a surface can proceed under the gas pressure acting in crack cavities.

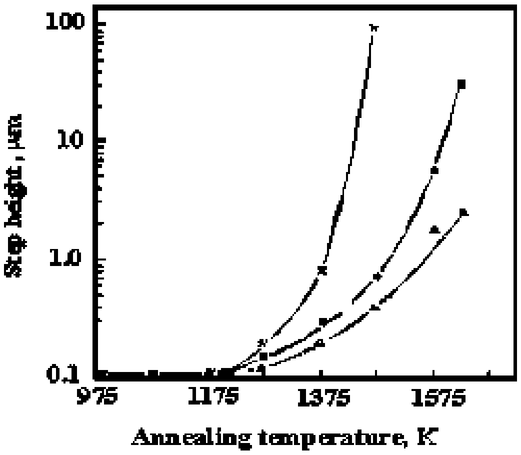


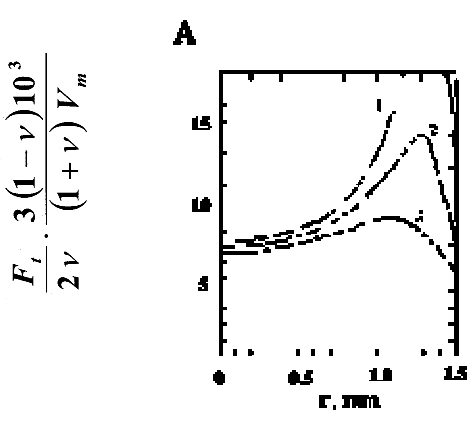
Fig. 3 Helium blistering in steel and alloys. A- Change of a step height versus the temperature of isochronal annealing in the specimens of the 08Cr16Ni15Mo3Nb austenitic steel (x), 03Cr20Ni45Mo4NbBZr (•) and 03Cr20Ni45Mo4NbY (▲) nickel alloys. B- SEM-micrographs of the structure of a zone of helium occurrence after annealing for 1h at 1475K; a refers to 08Cr16Ni15Mo3Nb, b refers to 03Cr20Ni45Mo4NbBZr, c refers to 03Cr20Ni45Mo4NbY

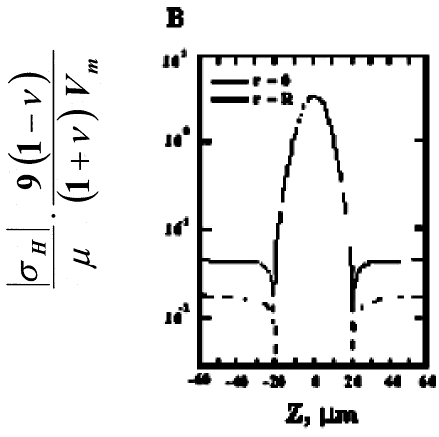
Simultaneously with copper specimens we fulfilled the comparative study of swelling in the 08Cr16Ni15Mo3Nb stainless steel; 3Cr20Ni45Mo4NbY, 03Cr20Ni45Mo4-NbBZr nickel alloys applied or recommended for application in nuclear reactors.

Figure 3 represents the results of measurements of a step formed on irradiated surfaces of the specimens of the 08Cr16Ni15Mo3Nb austenitic steel and 03Cr20Ni45 nickel enriched alloys after ion implantation with fluences of $1x10^{21}m^{-2}$ and annealing for 1h in the temperature interval of 975...1625 K. It is necessary to note that the step formation begins at 1375 K for all the specimens. With annealing temperature increasing the character of subsequent behavior of the step is different for every metallic material. In temperature interval of 1175...1375 K the step behavior in the 08Cr16Ni15Mo3Nb steel and the 03Cr20Ni45Mo4NbY alloy changes abruptly, i.e. not only catastrophic increasing of growth rate proceeds, but also the transformation of the step profile from a plate to a dome-shaped one.The maximal height of a dome in the 08Cr16Ni15Mo3Nb steel reaches 0.4mm at 1475 K, that actually testifies to the bulk blistering.

The cracks appearing on a surface of a dome gives a path of helium gas emission, so the growth of the dome height stops with following increasing of temperature and annealing time.

SEM-studies of cross-sections of specimens after an-nealing at 1475 K showed in the 08Cr16Ni15Mo3Nb steel a lentil-shaped cavity forming, but in the 03Cr20Ni45Mo4NbY nickel alloy only the developed swelling.





Helium blistering in copper at 0.9 $T_{melting}$

Bubble migration in copper at 1175 K

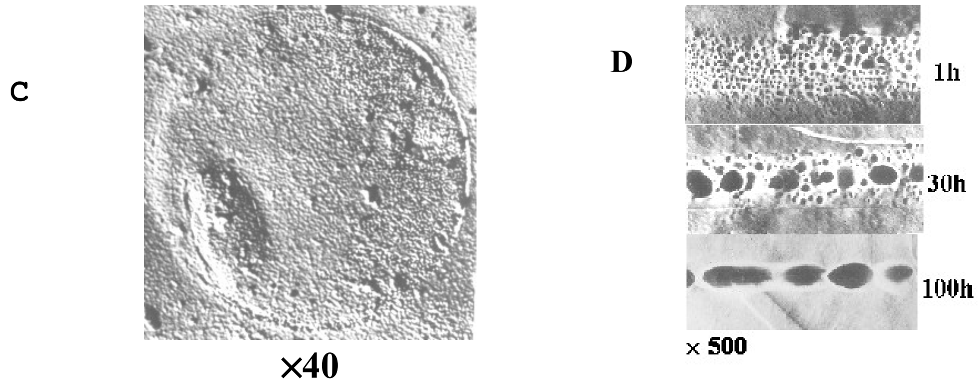


Fig.4 Results of calculation of elastic stresses and experimental data on blistering and bubbles migration. A- Dependence of Treska's function f_m *on a radial distance for various depths of helium occurrence in a swelling zone (1 – 0.005mm, 2 – 0.20mm, 3 – 0.35mm). B- Dependence of the absolute value of hydrostatic stress* σ_H *on the depth* Z *for two values of radial distance: at* $|Z| < 20\ \mu m$, $\sigma_H < 0$ *and* $|Z| > 20\ \mu m$, $\sigma_H > 0$. *C- SEM-micrographs of a surface of profilometry procedure of copper specimens subjected to isothermal annealing at 1175 K for 30 h. D- SEM-micrographs of the structure of helium doped layers in copper specimens subjected to isothermal annealing at 1175 K for 1h, 30h and 100h*

CONCLUSIONS

The results of study of helium behavior in the metallic materials after α-particle irradiation and succeeding thermal annealing provide the following conclusions.

It was found that in a straggling zone of the specimens irradiated by α-particles with initial energy of 50 MeV the strong stresses appeared during the process of helium bubble development. The relaxation of these stresses at high temperature annealing, when helium bubbles appear and grow, leads to the formation of a step on the irradiated surface.

There is a one-valued correlation between the step volume and the summary volume of helium bubbles in a swelling layer. The relation was established between the step height and the maximal value of swelling in the layer of helium occurrence.

Under the action of stress gradients helium bubbles migrate towards the maximal stresses, leading at high temperatures to the formation of long-distance cracks filled in with inert gas.

Under the conditions embarrassing the gas emission from a crack, gas forces out an interlayer of the material with thickness up to 0.35mm and forms a dome-shaped step. This testifies to the bulk blistering occurring in temperature interval of 0.8...0.95 T_{melt} .

The study showed the 03Cr20Ni45Mo4NbY alloy to be the most resistant to helium swelling and blistering among all the tested materials.

LITERATURE

V.Ph. Reutov, K.G. Farkhutdinov, H.G.Kadyrov// *Atomnaya Energia*. 1983, v.55, p. 170.