THE STRUCTURE OF POWDER POLYCRYSTALLINE TUNGSTEN AFTER TREATMENT IN PLASMA OF Ar GLOW DISCHARGE

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UDC 539.L4: 621.039.616

The interaction of the plasma of Ar normal glow discharge with powder polycrystalline tungsten is investigated by the methods of scanning electron microscopy, X-ray microanalysis, and X-ray structure analysis, and its microhardness is determined. The effective influence of low-energetic Ar ions both at the material surface and on its volume is showed. The depth of changes of the structure runs into 100—120 μm. The sizes of the new inclusions at the surface run into 10 μm and in the volume down to 5 μm.

1. Introduction

Studying the interaction of charged particles with a solid body is agreed-upon a standpoint that a modification of the material occurs only at enough large energies of the particles ($\geq 10^4$ eV).

It is established, however, that the intensive modification of crystalline materials occurs at energies of the accelerated particles of less than 5 keV, i.e. under conditions of the gas glow discharge [1,2]. In particular, as shown in [2], the bombardment by low-energy ions of an inert gas can substantially change the mechanisms of the processes which take place on the surface.

The characteristics of regimes of the treatments of products are easily realized on the practice and find gradually the industrial application in the technologies aimed at changes in the structure and properties, above all in the metals and alloys. All the results obtained are related, above all, to monolithic materials, whereas powder materials were not sufficiently studied.

Thus, the present work is devoted to studying the influence of the Ar glow discharge plasma (GDP) on the structure and properties of powder polycrystalline tungsten (poly-W).

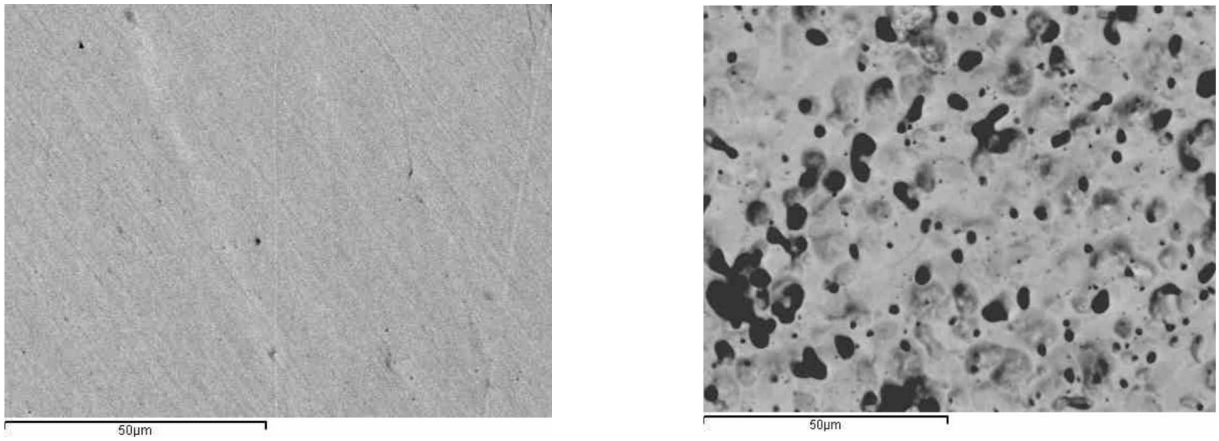
2. Experimental Technique

In our work, the irradiation is performed by the plasma of Ar normal glow discharges. The plasma voltage of a discharge was (3.0±0.5) kV, and the current density was $\approx$ 1 mA/cm^2. The mean energy of Ar ions was 300—600 eV, and the exposure dose was 4×10^{23} m^{-2}. The temperature in the working volume was controlled by a thermocouple and varied at the surface of a specimen from 400 to 450 K.

The objects of investigation were powder polycrystalline W of cubic form, which was cut from a W coarse-grained powder with a purity of 99.95% (10×10 mm in size) [4]. The working surfaces of the specimens were ground and then electro-polished to remove the disturbance layers. The quality control of the electro-polishing was carried out by the X-Ray method, and the quality of the surface was determined with assistance of an optical microscope.

The investigation of the structure of specimens was performed with a diffractometer in Cu-radiation. The data on the distribution of the diffracted irradiation intensity were obtained by the θ-2θ scanning method with the output to a computer.

The investigations of the surface of specimens were performed by scanning electron microscope (SEM) (Cam Scan 4D) in the regime of backward scattered electrons. The chemical microanalysis of the elements was carried out with an X-ray microanalyzer with a device EDX to SEM and the software Inca-2000. Using a Polyvar Met optical microscope with a device Micro Duromat 4000E (load 20 g, time delay 20 s), the microhardness was measured.



a **b**

Fig. 1. Surface morphology of W powder *a* — before treatment; *b* — after treatment in GDP

3. Results and Discussion

The investigations performed by the SEM method have showed the presence of a radiation erosion caused by the cathode sputtering (Fig. 1). The average size of the craters and etch pits is 5—10 μm.

The X-ray investigations of the action of Ar GDP have showed the next changes of the structure of the under-surface layers of powder poly-W.

The lattice parameter determined by the allowed doublet of the 321 line (Fig. 2) with a mistake not more than 6×10^{-5} nm has increased from 0.31621 in the initial state to 0.31647 nm after the irradiation. Under the normal conditions, the solubility of the inert gases in metals doesn't exceed 10^{-18}—10^{-19} at.% [3], and the atomic radius of Ar is well beyond the atomic radius of W. Then Ar implanted from GDP creates the supersaturated solid solution of the substitution type in W or the subtraction one at high concentrations (up to $\sim 0.1\%$).

The obtained supersaturated solid solution is metastable, as atoms of the inert gas are energetically favorable to join the lattice defects in view of the complexes or submicropores. Therefore, during the irradiation, the decomposition of the solid solution and the migration of Ar to defects of the crystalline lattice (grain boundaries, dislocations, pores, etc.) followed by decreasing the material density [4]. In addition, as well known, the ion bombardment of the metals and alloys causes the generation of a large amount of radiation defects in the neighborhood of surface, by creating the favorable condition for the display of the effects of radiation-stimulated diffusion. It was also noted that the

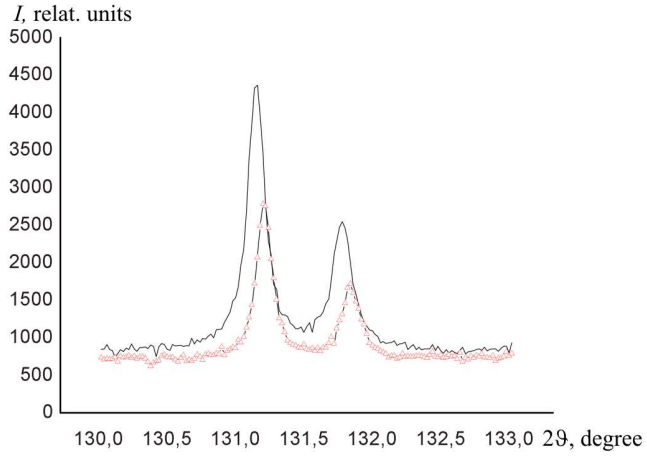


Fig. 2. Diffraction lines (321) of the initial poly-W and plasma-treated one: △ — initial state; ○ — irradiated in GDP

intensity of the radiation-stimulated diffusion of the own and impurity defects (of the Ar ions implanted from GDP and radiation vacancies) in the poly-W depends strongly on the parameters of a treatment (particle flux density, energy and state of a solid body) [5]. The appearance of the elevated concentration of defects (point-like and linear ones) takes place as a supplementary contribution to the common diffusion coefficient.

In this case, the influence of GDP in the medium of the residual atmospheric gases initiates a minor increase of the density of dislocation loops which is approximately by an order of magnitude smaller than the screw dislocation density, and the main changes come about into under-surface layers. It is considered that the reasons for the generation of screw dislocations

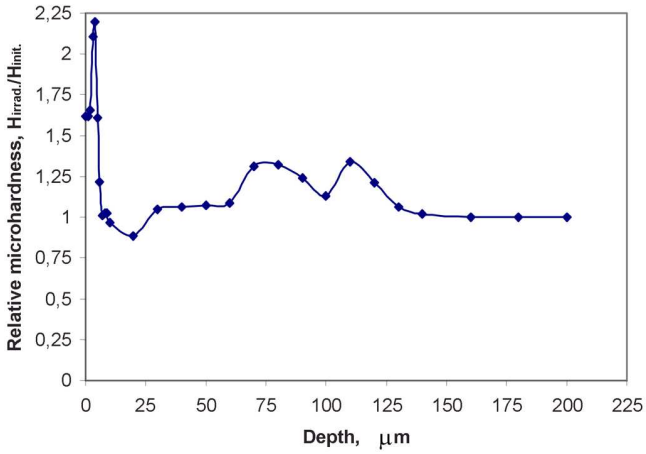


Fig. 3. Change of the relative microhardness (H_{irrad}/H_{init}) of poly-W over depth after the treatment in GDP

and structural changes are the action of shear stresses (as a result of the considerable supersaturation of the lattice metal in the neighborhood of the surface by atoms of the introduced gas), an increase in micro-stresses, and the radiation-accelerated diffusion of defects [6].

In addition to the formation of the supersaturated solid solution of Ar in W, the dislocation-type defects (edge dislocations) are created in under-surface layers at depths of at most 4 μm. The layerwise analysis of the irradiated surface (by a step of 1 μm) showed the sharp decrease of the density of chaotically distributed dislocations to the initial level. Consequently, the so-called "long-range effect" is manifested. It concerns with the formation of a developed dislocation structure into under-surface layers of the irradiated material, which must unconditionally be accompanied by its substantial strengthening.

It follows from the data shown in Fig. 3 on microhardness that the irradiation of powder poly-W by low-energetic ions of Ar induces a change of the microhardness over the depth of a specimen: the microhardness is substantially increased at the depths less than 3—5 μm. The maximal increase of the microhardness is achieved at a depth of 4 μm, and then it falls off smoothly. At a depth of 8—10 μm, the microhardness goes to a value that corresponds to the level of initial powder poly-W. The present result indicates the considerable influence of the treatment (by GDP) on the strength properties of under-surface layers of the metals. The change of the microhardness correlates with the data on a change of the dislocation density, which confirms the conclusion about the strengthening mechanism under the irradiation of metals by GDP.

It is revealed that the influence of GDP is spread to a depth of 100—120 μm (Figs. 3 and 4), i.e. not only into under-surface layer, but in the material volume as well.

It follows from the analysis of experimental data that the role of Ar implanted as defects in comparison with the induced dislocation structure, which appears under the irradiation of powder poly-W, is not substantial in the neighborhood of the surface. Beginning with a depth of 20 μm (see Fig. 3), the Ar atoms advantageously coagulate at defects of the crystalline lattice. Therefore, it can be supposed that, after the beginning of the irradiation, the gas-vacancy clusters are formed at these depths, i.e. the nucleation of microbubbles. Their presence in combination with the ongoing Ar saturation of under-surface layers of the material and the radiatively stimulated diffusion process of different defects (initial and radiation-induced ones and impurities) will lead to the formation of gas bubbles (gas-filled pores) in the volume of grains and on their boundaries. The subsequent development of the gaseous porosity will be supplied with the decomposition of the supersaturated solid solution of Ar. This leads to (as a result of hydrostatic weighting) a decrease in the W density from 17.29 to 16.83 g/cm^3. That is, the specific volume is increased by the gas swelling. Such relative small decrease of the density for the metallic powder polycrystals irradiated by the Ar plasma is linked with the effective flow of radiation-induced defects to pores of the irradiated W which were formed after the hot pressing of a powder granule.

Thus, the interaction of the Ar ions of GDP with powder poly-W doesn't stay localized in the thin under-surface layer ($\sim$10 Å) where the ions are braken (as shown in [7]), but it is spread at considerable distances from the irradiated surface. This leads to the "long-range effect" [6]. As the dose achieved 4×10^{23} m^{-2} (during three hours of irradiation), the Ar atoms diffuse to depths over 100 μm (Fig. 4,*b*). It is obvious that, in this case, the radiation-stimulated diffusion with the effective coefficient $\sim10^{-9}$cm^2/s ($x \sim \sqrt{Dt}$) takes place, which also leads to the increase of the penetration depth due to the material porosity.

It should be noted that, under the irradiation of polycrystals into a glow discharge, the inert gas diffuses not only on boundaries, but also into the volume of grains. The gas storage is predominantly occurs on grain boundaries and subgrains, which is caused by a great thickness of the defect layer in monocrystals. Moreover,

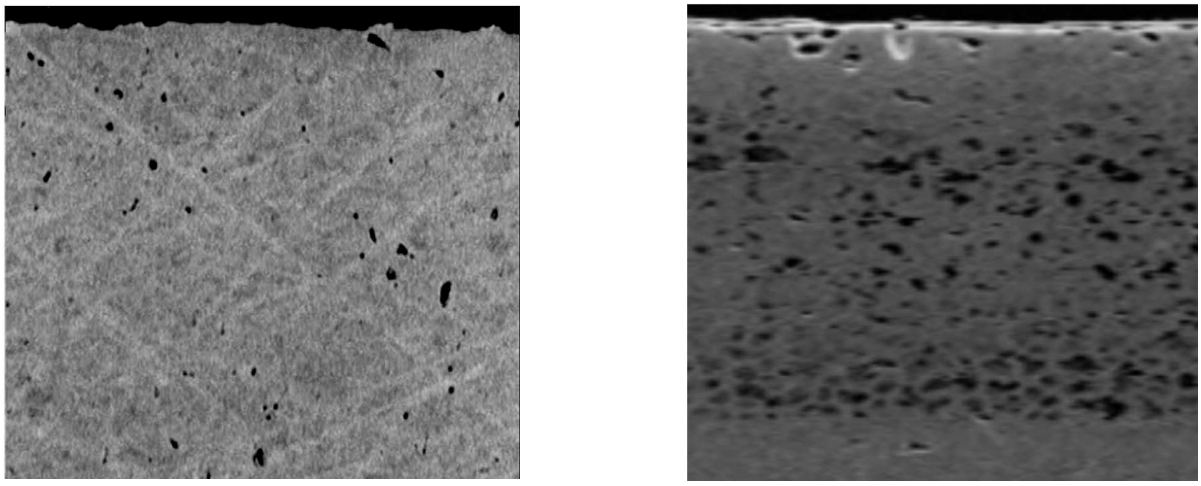


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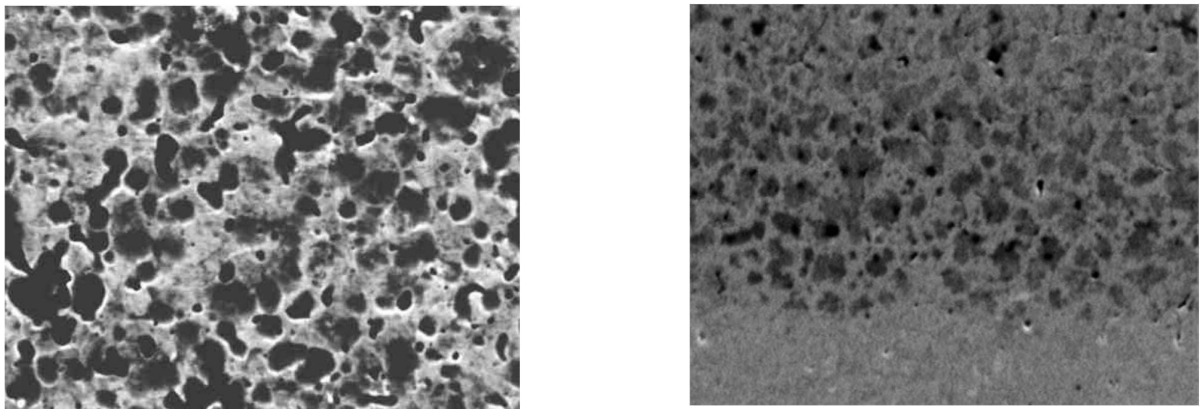
Fig. 4. Cross section of W powder *a* — before treatment; *b* — after treatment in GDP (the scale is the same, as in Fig. 1)



a **b**

Fig. 5. Structure of W after GDP: *a* — on surface; *b* — in volume (the scale is the same, as in Fig. 1)

the more the boundaries, the less deeply the defects "penetrate".

In the case of a powder material (porosity 5÷15% [8]), the migration of Ar ions occurs mainly by the grain boundaries. The effective aggregation of Ar and impurities in the pores of a powder material leads to the appearance of a developed structure both at the material surface and in its volume (Fig. 5). It is proposed that, after a treatment in GDP, the structure presents the aggregation of defects in view of the gas-filled pores containing oxygen and argon predominantly. The sizes of these defects at the sample surface of W run into 10 μm (Fig. 5,*a*) and in the volume down to 5 μm (Fig. 5,*b*).

The Ar distribution in the neighborhood of the surface and in the volume of W after the treatment in GDP is characterized by its aggregation in the defect areas created at the expense of both the appearance of new impurities and the diffusion redistribution of intrinsic ones. The supersaturation of powder poly-W by Ar ions leads to the formation of gas bubbles in volume, which causes the additional radiation-induced erosion at the surface (Fig. 5). Such gas-filled pores become guides for the effective flows of oxygen and, perhaps, of another impurities in the material due to the radiation-stimulated diffusion.

4. Conclusions

The influence of the Ar GDP causes a substantial radiation erosion of the surface.

As a result of the irradiation of powder poly-W by the low-energetic plasma of Ar glow discharges, the next changes of the structure of under-surface layers occur: the formation of a metastable solid solution of the inert gas; an increase of the defect density of the dislocation type (at a depth less than 4 μm); an increase of the microhardness at a distance less than 8 μm from the surface with a sharp maximum at a depth of 3—4 μm.

The interaction of the plasma of an Ar normal glow discharge with powder poly-W is spread to a depth of 100—120 μm (from the treated surface).

The sizes of the new inclusions run at 10 μm at the sample surface of W and down to 5 μm in the volume.

The structural changes in powder poly-W after the treatment in GDP present the aggregation of defects in view of the gas-filled pores containing oxygen and argon predominantly.

1. *Gurevich M.E., Juravlev A.F., Krushinskaya L.A., Larikov L.N.* // Metallofiz. — 1990. — **12**, N6. — P. 47—50.
2. *Nishijima Dai, Ye M.Y., Ohno N., Takamura S.*// J. Nucl. Mater. — 2004. — P.329—333; Pt. 2. — P.1029—1033.
3. *Komarov F.F.* Ion Implantation in Metals. — Moscow: Metallurgiya, 1990 (in Russian).
4. *Larikov L.N., Gurevich M.E., Tishkevich V.M. et al.* // Ukr. J. Phys. — 1979. — **24**, N5. — P. 658—661.
5. *Gerzriken D.S., Mazanko V.F., Tishkevich V.M.* // Metallofiz. Noveish. Tekhn. — 2003. — **25**, N5. — P. 621—633.
6. *Sharkeev U.P., Didenko A.N., Kozlov E.V.* // Izv. Vuz. Fiz. — 1994. — **37**, N5. — P. 92—108.
7. *Larikov L.N., Krasilnikov V.S.* // Preprint IMF79.9, Kyiv — 1979. — P. 13.
8. *Structure* and Strength of Powder Materials /Ed. by S.A. Firstov — Kyiv: Naukova Dumka, 1993 (in Russian).

Received 24.05.06

СТРУКТУРА ПОРОШКОВОГО ПОЛІКРИСТАЛІЧНОГО ВОЛЬФРАМУ ПІСЛЯ ОБРОБКИ В ПЛАЗМІ ТЛІЮЧОГО РОЗРЯДУ АРГОНУ

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Р е з ю м е

Методами растрової електронної мікроскопії, рентгенівського мікроаналізу, рентгеноструктурного аналізу та мікродюрометрії досліджено взаємодію плазми нормального тліючого розряду аргону з полікристалічним порошковим вольфрамом. Показана ефективна дія низькоенергетичних іонів аргону як на поверхню матеріалу, так і на його об'єм. Глибина структурних змін досягає 100—120 мкм. Розміри нових включень на поверхні досягають 10 мкм, а в об'ємі — до 5 мкм.