Research of the influence of bias voltage on the structure and residual stress in Ta/W coatings applied on a copper substrate by inverted magnetron

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Abstract. This paper presents the results of studying the effect of the bias voltage on a copper tube substrate on the texture and residual stresses in 4-layer Ta/W/Ta/W coatings deposited with an inverted magnetron. It is shown that, in contrast to monolayer coatings of Ta and W, in which residual stresses exceeding 2 GPa are formed on the substrate at high voltages on the substrate, stress relaxations occur in the 4-layer coating in alternating layers differing in LTEC values and in the outer W-layer of stress practically absent.

Thin films and coatings made of tungsten are of considerable interest for use in various fields: microelectronics [1] (including spintronics [2, 3]), thermo photovoltaic converters in power engineering, high-temperature nanophotonics [4] or others. One of the possible areas of application W coatings are thermal barrier coatings for parts of future thermonuclear reactors, such as ITER [5], which will be exposed to extreme thermal loads and ion bombardment, as well as for other thermally loaded products, such as combustion chambers of rocket engines. The key problems for such coatings are thermo mechanical stability with respect to delamination, oxide formation and diffusion at high operating temperatures, and the achievement of long-term high-temperature grain stability [4].

For deposition of W, pulsed laser deposition, atomic layer deposition, and sputtering by planar DC magnetrons are commonly used. In [5, 6], we showed that for the creation of thin-walled small-sized axis symmetric shell structures from layered composites, for example, pipe products with different surface profiles, a system of sequentially located inverted magnetrons and one ordinary cylindrical magnetron used for cleaning substrates is very effective. The system allows the formation of layered composite shells by spraying various layers onto a mandrel (for example, copper), which is subsequently etched away.

It is known that the deposition of W-coatings on Cu substrates, which have a significantly higher thermal coefficient of linear expansion (TCLE), leads to huge residual stresses, reaching -5 GPa. The magnitude of these stresses, in addition to the difference in the LTEC and Young's modulus, depend on the temperature of the coating process, which is determined by the energy parameters of the deposition process, primarily the voltage on the substrate. In our works [7, 8], we investigated the effect of voltage on the substrate on the formation of texture and residual stresses in monolayer W and Ta coatings deposited on a Cu substrate by an inverted magnetron. In our works [7, 8], we studied the effect of voltage on the substrate on the formation of texture and residual stresses in monolayer W and Ta coatings deposited on a Cu substrate by an inverted magnetron. At high voltages from -100 to -300 V, high compressive residual stresses exceeding 2 GPa were found in the coatings, which led to
disruption of adhesion and destruction of coatings. In [9], when studying the formation of residual stresses in monolayer coatings of Mo and Nb, high residual compressive stresses were also found. However, in a multilayer (n = 120) Mo / Nb coating 800 μm thick, the stresses in the outer layers did not exceed 600 MPa. In this regard, in this work, we investigated the effect of the voltage on the substrate on the residual stresses, as well as on the texture and crystal lattice distortions of coatings of 4-layer Ta/W/Ta W coatings deposited using an inverted magnetron.

Spraying was carried out on a specialized installation MRM-1, presented in [6]. Argon with a purity of at least 99.9% was used as a working gas. The cathode material is W purity ≈ 99.9%, the inner diameter and length of the cathode were 37 mm and 24 mm. A copper tube M-1 with a diameter of 10 mm and a length of 20 mm was used as a substrate. Before sputtering, the tube was polished and degreased. Then the substrate was installed on the rod of vertical movement of the samples in the chamber and the chamber of the setup was evacuated to a residual pressure of 10⁻³ Pa. Before spraying, for 30 minutes, cleaning with a glow discharge plasma was carried out at an argon pressure of 5 Pa and a voltage on the substrate of 1100 V. Then, tungsten was deposited at various bias voltages on the substrate according to the modes shown in table 1. The cathode axis and periodically completely left the cathode region, alternately completely leaving its ends. The total spraying time for all samples is 8 hours. Each layer was applied for 2 hours. alternation of layers: Ta-W-Ta-W.

**Table 1. Spray mode No.1 (bias voltage 0 V).**

| Layer | U_m3, V  | I_m3, A | P_Ar, Pa | T, °C |
|-------|----------|--------|----------|-------|
| Ta    | 280-285  | 1      | 0,2      | 420   |
| W     | 290-305  | 1      | 0,2      | 430   |
| Ta    | 275-285  | 1      | 0,2      | 415   |
| W     | 290-305  | 1      | 0,2      | 430   |

**Table 2. Spraying mode No.2-4 (bias voltage -50, -100, -150, -200V).**

| Layer | U_m3, V  | I_m3, A | -Us, V  | Is, A | P_Ar, Pa | T, °C |
|-------|----------|--------|---------|-------|----------|-------|
| Ta    | 270-280  | 1      | 50 (100, 150,200) | 0,14-0,05 | 0,2      | 430   |
| W     | 290-300  | 1      | 50 (100, 150,200) | 0,14-0,05 | 0,2      | 440   |
| Ta    | 270-285  | 1      | 50 (100, 150,200) | 0,14-0,05 | 0,2      | 430   |
| W     | 290-305  | 1      | 50 (100, 150,200) | 0,14-0,05 | 0,2      | 440   |

Inverse pole figures (IPFs) were obtained by taking X-ray diffraction patterns in the angular range 2θ = 30–140°. The pole density of 6 independent reflections hkl on the stereographic triangle: 001, 011, 013, 111, 112, 123, (figure 1a) was determined from the ratio:

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P_{hkl} = \frac{n \left( \frac{I_{hkl}^{\text{ek}}}{I_{hkl}^{\text{et}}} \right)_i}{\sum_{i=1}^{n} \left( \frac{I_{hkl}^{\text{ek}}}{I_{hkl}^{\text{et}}} \right)_i}
\]

where, \( I_{hkl}^{\text{ek}} \) and \( I_{hkl}^{\text{et}} \) are the integral intensities of the hkl reflections for the textured and textureless (reference) sample, respectively; n is the number of independent hkl reflections (n = 6).

Figure 1 shows the IPF for W coatings deposited on a copper substrate at bias voltages on the substrate from 0 to -200 V. It can be seen that with an increase in the substrate voltage, changes in the coating texture occur, which are of a non-monotonic nature.
Figure 1. IPF for the outer W layer of 4-layers coatings deposited at different voltage values on the substrate: (a) a standard stereographic triangle for a cubic lattice; (b) Us = 0 V; (c) Us = -50 V; (d) Us = -150 V; (e) Us = -200 V.

In figure 2, these results are summarized in the form of the dependence of the pole density (111) on the voltage on the substrate. Figure 3 shows the voltage dependences of the half-widths of the diffraction lines (222) and (321). It can be seen that with an increase in the voltage on the substrate, the width of the diffraction lines increases, which indicates an increase in the distortion of the crystal lattice and a refinement of the subgrain structure. Figure 4a shows the values of the lattice parameters of the coatings calculated from different interplanar distances (hkl). The presented results indicate the absence of noticeable residual stresses in the coatings, which distinguishes them from monolayer coatings, in which stresses exceeding 2 GPa were found.
Figure 2. Pole density for orientation (111), (123), and (112) versus substrate voltage.

Figure 3. Dependence of the half-widths of the diffraction lines (321) and (222) on the voltage on the substrate.

It is likely that stress relaxation occurred in the 4-layer coating due to mutual compensation of thermal stresses in multilayer coatings, when the compressive stresses arising when a Ta layer is deposited on a W sublayer with a lower LTEC are balanced by tensile stresses when the next W layer is deposited on a Ta sublayer with higher LTEC, figure 4b.

Figure 4. Coating lattice parameters calculated for different interplanar distances (hkl) (a) and scheme of formation of residual stresses in a 4-layer Ta/W/Ta/W coating.

A similar situation was observed in a 120-layer Nb/Mo coating with a thickness of 800 µm [9], in which the compressive stresses were <600 MPa, while in monolayer 10 µm coatings made of the same metals the values of the compressive stresses on the copper substrate exceeded 2000 MPa. The increased lattice periods (figure 4a) of the coatings with respect to pure W (a = 0.317 nm) are possibly due to the dissolution of Ta atoms with a higher period in the W lattice (a = 0.33 nm). The lattice period of 0.318 nm corresponds to the dissolution of 5-7 at.% Ta.

1. With an increase in the voltage on the substrate, the width of the diffraction lines increases, which indicates an increase in the distortion of the crystal lattice and the refinement of the subgrain structure.

2. With an increase in the voltage on the substrate, changes in the texture of the coatings occur, which are of a non-monotonic character.
3. No noticeable residual stresses were found in 4-layer Ta/W/Ta/W coatings, which distinguishes them from monolayer coatings and is due to mutual compensation of thermal stresses in successive Ta and W layers with different LTEC values (6.3 and 4.5 $10^{-6}$ deg$^{-1}$ respectively).

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