Study of the bias voltage influence on the structure, texture and residual stresses in Ta coatings deposited on a copper substrate of inverted magnetron

A S Lenkovets, A A Lozovan, S Ya Betsofen, A V Bespalov, I A Grushin and N A Ivanov

MAI — Moscow Aviation Institute (National Research University), Orshanskaya 3, 109383 Moscow, Russia

E-mail: loz-plasma@yandex.ru

Abstract. The results of the study of the bias voltage effect on the structure, texture and residual stresses of Ta-coatings deposited using an inverted magnetron on a copper substrate are presented. It is shown that variations in residual stresses are non-monotonic, which is determined by the complex effect of the bias voltage on the process temperature, which affects both the formation of mechanical and thermal stresses and their partial relaxation.

1. Introduction

Ta coatings have long been used in various industries, including for the formation of protective coatings [1], the creation of a diffusion barrier between the metallization of the upper part of copper and a silicon-based substrate [2], etc. Due to the high melting point of 3292 K, it is more often deposited by sputtering.

In [3, 4], we showed that to create thin-walled small-sized axisymmetric shell structures from laminated composites, for example, tubular products with different surface profiles, systems of sequentially located inverted magnetrons are used. The system allows the formation of layered composite shells by depositions various layers onto a mandrel, which is subsequently etched. In this work, Ta was sprayed, as before, on a M1 grade copper substrate (tube with a diameter of 10 mm and a length of 20 mm).

The application of Ta coatings on Cu substrates should inevitably be accompanied by the formation of high residual stresses due to a significant difference in thermal expansion coefficient, and the sign of the stresses should be negative, since the thermal expansion coefficient of copper is higher than that of the Ta coating. In addition, an additional contribution to the stress contributes to the mechanical action on the substrate, which are similar to shot-blasting and are also accompanied by the formation of compressive stresses as the reaction of the subsurface layers to the crushing of the surface layer of the substrate by ion flow. The magnitudes of these stresses, in addition to the difference in the values of the thermal expansion coefficient and the Young modulus, depend on the temperature of the coating process, which is determined by the energy parameters of the sputtering process, primarily the magnitude of the voltage on the substrate. Therefore, in this work, we studied the effect of bias stress on residual stresses, as well as on the structure, texture, and distortion of the crystal lattice of coatings. Before depositions, the substrate was polished with sandpaper of various degrees of graininess to the
10th surface roughness class, then rubbed with alcohol and installed in the chamber on the rod of the movement mechanism. The installation was pumped out to a pressure of $2 \times 10^{-2}$ Pa.

Then, Ta was deposition at various bias voltages on the substrate. Depositions modes are shown in table 1.

### Table 1. Depositions modes Ta.

| Sample | Bias voltage $U_b$, V | Cathode current $I_c$, A | Cathode voltage $U_c$, V | Depositions time, $t_d$, hour | Working gas pressure, $P_{Ar}$, Pa | Surface temperature $T_s$, °C |
|--------|----------------------|--------------------------|--------------------------|-----------------------------|-----------------------------------|-----------------------------|
| 1      | 0                    | 1                        | 276-285                  | 3                           | 0.2                               | 420                         |
| 2      | 50                   | 1                        | 267-277                  | 3                           | 0.2                               | 430                         |
| 3      | 100                  | 1                        | 256-264                  | 3                           | 0.2                               | 445                         |
| 4      | 150                  | 1                        | 257-264                  | 3                           | 0.2                               | 502                         |
| 5      | 200                  | 1                        | 266-275                  | 3                           | 0.2                               | 520                         |
| 6      | 250                  | 1                        | 255-262                  | 3                           | 0.2                               | 532                         |
| 7      | 300                  | 1                        | 253-260                  | 3                           | 0.2                               | 560                         |

2. Materials and methods
The residual stresses were estimated using the $\sin^2 \psi$ method on a DRON-4 X-ray diffractometer in CuKα radiation using a reflection (321). Based on the determination of the line position, the lattice parameters were calculated and the dependences $a^2_\psi$ on $\sin^2 \psi$ were calculated. The residual stress was determined from the slope of the straight line ($\tan \alpha$), the least squares method from the equation:

$$\sigma_\psi = \tan \alpha \left[ \frac{E}{1+\nu} \right]_{321} / a_0$$

(1)

The value of $[E/(1+\nu)]_{321}$ was calculated from the relation:

$$(1 + \nu) / E_{321} = S_{11} - S_{12} - 3JG$$

where $J=S_{11}-S_{12}-0.5S_{44}$ - anisotropy parameter; $G = (h^2k^2+k^2l^2+h^2l^2)/(h^2+k^2+l^2)^2$ - orientation factor; single crystal elastic constants for Ta: $S_{11}=0.686$ 10$^{-2}$ GPa$^{-1}$, $S_{12}=-0.258$ 10$^{-2}$ GPa$^{-1}$, $S_{44}=1.212$ 10$^{-2}$ GPa$^{-1}$; $(1+\nu)/E_{321}=0.691$ 10$^{-2}$ GPa$^{-1}$.

Inverse pole figures (IPF) were obtained by taking radiographs in the angular range $2\theta=35-125^0$. The pole density of 6 independent reflections hkl on the stereographic triangle: 001, 011, 013, 111, 112, 123, (Fig. 1a) was determined from the relation:

$$P_{hkl} = n \left( \frac{I_{hkl}^{\text{res}}}{I_{hkl}^{\text{null}}} \right) \sum_{i=1}^{6} \left( \frac{I_{hkl}^{\text{res}}}{I_{hkl}^{\text{null}}} \right)$$

where $I_{hkl}^{\text{res}}, I_{hkl}^{\text{null}}$ is the integral intensity of the hkl reflections for the textured and textureless (reference) sample, respectively; n is the number of independent hkl reflections ($n = 6$).

3. Experimental results and discussion
Figure 1 shows the IPF for Ta-coatings deposited on a copper substrate at bias voltages on a copper substrate from 0 to -300 V. In the absence of voltage on the substrate, the coating texture is characterized by a preferential arrangement of (321) and (110) planes parallel to the substrate. With an increase in voltage to -50 and -100 V, the texture changes dramatically and is characterized by (112) and (111) orientations, and at higher values of the bias voltage, orientation (111) dominates.

It can be noted that with increasing voltage on the substrate, the texture components are enhanced, which in the case of Ta are characterized by maximum values of the Young's modulus ($E_{111} = 217$ GPa, $E_{112} = E_{321} = 193$ GPa, but $E_{001} = 146$ GPa and $E_{310} = 160$ GPa).
Figure 1. The standard stereographic triangle of a cubic lattice (a) and IPF for Ta coatings on a copper substrate, applied at different values of the voltage on the substrate: \( U_b = 0 \) V (b); \( U_b = -50 \) V (c); \( U_b = -100 \) V (d); \( U_b = -150 \) V (e); \( U_b = -200 \) V (f); \( U_b = -250 \) V (g); \( U_b = -300 \) V (h).

In figure 2, these results are summarized as the dependence of the pole density (111) on the voltage on the substrate.

Figure 2. The dependence of the pole density for the orientation (111) (\( P_{(111)} \)) on the voltage on the substrate.
Figure 3 shows the dependences on the voltage of the half-width of the diffraction line (321), which characterize the distortion of the crystal lattice of the coatings. It can be seen that with increasing voltage on the substrate above -150 V, the of the diffraction lines width increases and, accordingly, the distortion value of the crystal lattice of the coatings increases also.

Regularities in the formation of residual stresses are also consistent with these results (figure 4). Variations of residual stresses are non-monotonic. High residual stresses (> 1000 MPa) are characteristic of small (0, -50V) values of the bias voltage. With average values of the bias voltage (-100, -150V) the residual stresses are minimal (~ 400 MPa), and at higher voltages, the residual stresses increase and exceed -1500 MPa with the bias voltage -300V (figure 4).

Figure 3. Dependence of half widths of diffraction line (321) on the voltage on the substrate.

Figure 4. The dependence of the residual stress from the voltage on the substrate.

It is likely that variations in the magnitude of residual stresses are due to multidirectional processes of their formation under the influence of processes of a mechanical and thermal nature. An increase in the process temperature with an increase in the bias voltage leads, on the one hand, to an increase in thermal stresses, and on the other hand, to their relaxation. On the one hand, these processes complicate the understanding of their laws, and on the other hand, if they are correctly interpreted, they make it possible to control the structure and stress state of the coatings.
References

[1] Maeng S, Axe L, Tyson T A and Cote P 2006 Surf. Coat. Technol. 200 5767–77
[2] Lintanf-Salaün A, Mantoux A, Djurado E and Blanquet E 2010 Microelectronic Engineering 87 373–8
[3] Lozovan A A, Lenkovets A S, Ivanov N A, Alexandrova S S and Kubatina E P 2018 Journal of Physics: Conf. Series 1121 012020
[4] Lozovan A A, Betsofen S Ya, Lenkovets A S, Grushin I A, Labutin A A and Pavlov Yu S 2018 Journal of Physics: Conf. Series 1121 012019