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Features of copper coatings growth at high-rate deposition using magnetron sputtering systems with a liquid metal target

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ABSTRACT

The article focuses on the study of growth conditions for metal coatings during the operation of magnetron sputtering systems with a liquid-phase target. It also discusses the trends of coating properties formation (in terms of copper example) depending on the growth conditions. The data of the experiments and calculations confirm that the appearance of intensive evaporation on the target surface allows increasing more than 10 times the deposition rate and deposited particles flux density in comparison to conventional sputtering with a cooled solid target at the same magnetron power.

In the case of the magnetron sputtering system with a liquid-phase target, energy fluxes and particles ones towards the substrate during the coating growth and the substrate heating rate are much greater than at conventional magnetron deposition with a solid target.

The combination of calculations and experiments have made it possible to reveal the structure of energy and particles fluxes towards the substrate during the operation of the magnetron sputtering system with a liquid-phase target. The contribution of these fluxes formed by different mechanisms in a wide range of magnetron power has also been found out. The analysis of structural and mechanical properties of copper films being deposited at different energy and particles fluxes ratio towards the substrate has been carried out. It has been found that at intensive evaporation the coating surface is smooth and uniform, and the size of grains decreases. The mechanical properties of coatings (adhesion and microhardness) have got higher values compared to the cases related to only sputtering.

1. Introduction

Magnetron sputtering is a commonly used technique for deposition of modifying coatings on the surface of workpieces and materials. However, despite the development level of this technology, it has a number of disadvantages such as a low coating deposition rate and low deposited particles flux density [1–3]. Regardless the power supply, the deposition rate of metal coatings is usually not more than 10 nm/s [2,4,5]. It happens due to the fact that the main mechanism of atoms emission from the target surface is the sputtering with the ions of working gas. The sputtering rate is proportional to the ion current power density applied to the target from the magnetron plasma region. Due to the fact that there are technical limitations on the magnetron power supply, the sputtering rate cannot be increased significantly.

The investigations of various research teams show that the evaporation initialization in addition to sputtering on the target surface of magnetron sputtering systems (MSS) can significantly increase the coatings deposition rate [6–11]. The evaporation rate increases almost exponentially with the rise in target surface temperature, which has a non-linear dependence on ion current power density of the magnetron discharge [12]. Moreover, the increase in the atoms emission rate from the target surface leads to the instantaneous growth of deposited particles flux density.

A substantial increase in the temperature of a target can be achieved by its thermal isolation using special inserts between the target assembly and the magnetron body with a cooled magnet system (Fig.1). As a result, the energy which comes on the target from plasma will not flow away due to heat conductivity, and the target substance can melt. In this case the crucible made from higher-melting-point material (e.g. Mo, Graphite) should be used to preserve the target shape. The coatings deposition rate can be increased by several tens of times at the same power density in comparison to sputtering cooled solid targets [9,12].

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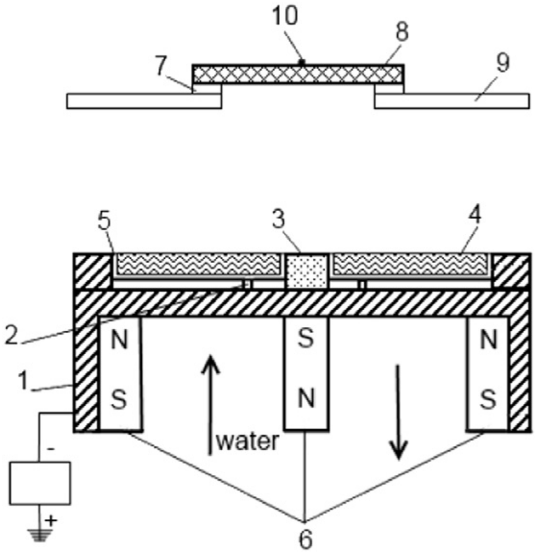

Fig. 1. Layout of the cathode assembly and substrate: 1 – magnetron body, 2 – ceramic insert, 3 – magnetic core, 4 – target, 5 – crucible, 6 – magnets cooled by water, 7 – ceramic insulator, 8 – substrate, 9 – substrate holder, 10 – the place of thermocouple mounting.

There is a demand for magnetron technologies of high performance such as deposition of protective, conductive and decorative layers on the surface of materials and workpieces. Therefore, it is necessary to make coating properties meet the operating requirements in addition to high performance.

It should be noted that in this case we deal with totally different coatings growth conditions compared to the conventional magnetron sputtering of cooled solid targets because the total energy and particles fluxes towards the substrate are much higher. That is why the structural and functional properties of coatings when using magnetrons with liquid phase targets can also be different.

It is known that the substrate heating influences the following processes such as adsorption, desorption, mobility of adatoms, chemical reactions etc., which further define the coating properties. The models of coatings structure formation at sputtering cooled solid targets have already been developed [13,14]. These models use normalized temperature T_n as one of the important criterion ($T_n = T_{sub}/T_{melt}$, where T_{sub} is substrate temperature, T_{melt} is melting temperature of the target material). The impact of the ion bombardment on the coatings structure has led to the introduction of another criterion – the amount of energy that comes to the substrate fallen within one deposited atom (E_a) [15–17].This criterion is used for an analysis of coatings structure and properties obtained by sputtering cooled solid targets. However, it is not clear whether this criterion is suitable in our case.

There are a lot of works devoted to revealing dependencies between the deposition parameters and coatings properties formed by different types of MSS with solid targets, such as [18–21]. However, there is no any research that focuses on connecting deposition parameters with properties of coatings obtained by MSS with liquid phase targets.

To predict what properties the coatings might have depending on the operating parameters of MSS with liquid phase targets and to be aware of the methods of a parametric control, it is necessary to reveal the relation between the characteristics of energy and particles fluxes coming to the substrate and the peculiarity of the growth structure and functional properties of formed metal coating. This issue has not found all-around consideration in the scientific community therefore this article is to present the results of the research related to this area. Making calculations of energy and particles fluxes on the substrate, running experiments related to coatings deposition, and doing subsequent analysis of coatings properties, it has been attempted to reveal the trends in properties change depending on the operating parameters of MSS with liquid metal targets.

2. Calculation method of energy and particles fluxes

Since the energy and particles fluxes are difficult to be measured in experiments, therefore, their density has been calculated depending on the power of MSS. The atoms flux from the highly heated target surface consists of two independent components: sputtered and evaporated particles. Total rate of surface erosion can be represented as:

$$V = V_{sput} + V_{ev}, \tag{1}$$

where V_{sput} and V_{ev} are the rates of surface erosion due to sputtering and evaporation.

The models of thermal and erosion processes based on solution of heat conductivity equation at considering heat consumption for phase transitions and sputtering have been used to calculate the target temperature and evaporation rate; the boundary conditions are determined by the energy balance in the “target in the crucible” system under the influence of the ion current from magnetron plasma. The models are described in [10,22].

The sputtering rate is calculated according to the Sigmund formula for the primary knock-out mode [23]. The growth rate of the deposited coating V_{dep} in any element on the substrate with coordinates (X, Y) is influenced by the contribution of sputtered and evaporated particles and calculated by the Lambert-Knudsen law [24]:

$$V_{dep}(X,Y) = F_{dep}/n_0 = \left(F_{dep,sput} + F_{dep,ev}\right)/n_0 = \frac{L^2}{\pi}\iint_{S_{targ}} \frac{\left(V_{sput}\left(x_{targ}, y_{targ}\right) + V_{ev}\left(x_{targ}, y_{targ}\right)\right) dx_{targ} dy_{targ}}{\left(L^2 + \left(X - x_{targ}\right)^2 + \left(Y - y_{targ}\right)^2\right)^2} \tag{2}$$

Here F_{dep} is a total deposited particles flux density; $F_{dep,sput}$ and $F_{dep,ev}$ are the deposited flux density of sputtered and evaporated atoms respectively; n_0 is nuclear density of the coating, which is assumed to be equal to the target nuclear density; L is a distance between target and substrate placed in parallel; S_{targ} is target surface area. The rates of sputtering (V_{sput}) and evaporation (V_{ev}) on the element of the target surface with coordinates (x_{targ}, y_{targ}) should be expressed in m/c.

The application of this method for calculating the deposition rate is appropriate in our case since the working pressure does not exceed 0.2 Pa, and the mean free path of emitted atoms is commensurate with the distance between the target and substrate.

The energy flux density on the substrate Q_{total} is equal to the following:

$$Q_{total} = Q_{rad} + Q_{cond,sput} + Q_{cond,ev} + Q_{kin,sput} + Q_{kin,ev}, \tag{3}$$

where Q_{rad} is the flux density of heat radiation from the target; $Q_{cond,sput}$ and $Q_{cond,ev}$ are the energy fluxes densities due to condensation of sputtered and evaporated atoms; $Q_{kin,sput}$ and $Q_{kin,ev}$ are the fluxes densities due to kinetic energy of sputtered and evaporated atoms being deposited on the substrate.

The heat radiation flux density per an element of substrate surface with coordinates (X, Y) is calculated on the basis of the laws of Lambert and Stephen-Boltzmann for a gray body, taking into account the uneven distribution of the temperature on the target surface:

$$Q_{rad}(X,Y) = \frac{L^2}{\pi}\int_{S_{targ}} \frac{\varepsilon_p \sigma_{SB}\left(T_{targ}^4\left(x_{targ}, y_{targ}\right) - T_{sub}^4\right) dx_{targ} dy_{targ}}{\left(L^2 + \left(X - x_{targ}\right)^2 + \left(Y - y_{targ}\right)^2\right)^2}. \tag{4}$$

Here ε_p is specific emissivity ($\varepsilon_p = 1/(1/\varepsilon_{targ} + 1/\varepsilon_{sub} - 1)$; ε_{targ} and ε_{sub} are emissivity of the target and substrate surface respectively; σ_{SB}