The target heating influence on the reactive magnetron sputtering process

A Bondarenko, A Kolomiytsev and V Shapovalov
St. Petersburg State Electrotechnical University “LETI”, 5 Prof. Popov Street, St. Petersburg 194354, Russia
E-mail: stopnastia@gmail.com

Abstract. A physicochemical model for the reactive magnetron sputtering of a “hot” target is described in this paper. The system consisting of eight algebraic equations was solved for a tantalum target sputtered in an O$_2$ environment. It was established that the hysteresis effect disappears with the increase of the ion current density.

1. Introduction
The reactive magnetron sputtering technology is one of the most commonly used methods of the deposition of thin films [1]. By changing the reactive gas flow rate and the current density on the target, it is possible to deposit both metallic and stoichiometric thin films. Using the magnetron with a “hot” target opens new possibilities for the reactive sputtering, since the temperature of the target can be led up to its melting point [2, 3].

For the effective application of the reactive sputtering technology, it is necessary to establish its general characteristics and to figure out the relations between the dependent and independent variables. This research was carried out both experimentally and by simulations. All notorious models for the reactive sputtering technology do not consider the target heating effect and are based on two basic assumptions [4–6]:

- two processes occur on the target surface: the formation of a thin layer of the metal/reactive gas compound and the sputtering of this compound by the argon ions;
- the deposition of both metal and compound particles sputtered from the target and the chemical reaction occur on the substrate and the walls of the vacuum chamber.

2. Model
The physicochemical model offered in [5, 6] describes this process in the most correct way. We are going to use this model considering the effect of the heating of the target surface.

The metallic (M) target is sputtered in an Ar + X$_2$ environment, where X$_2$ is a chemically active gas. The stationary state of the process is provided by nonisothermal conditions, since the substrate and the walls of the vacuum chamber have constant temperatures $T_s$ and $T_w$, respectfully. The target temperature $T_t$ depends on the power density at the target surface (or the ion current density $j_i$). Furthermore

- the discharge current density represents a summation

$$j = (1 + \gamma) j_i + j_f,$$

$$j_f,$$
where $\gamma$ is the ion-induced electron emission coefficient; $j_T$ is the thermal electron emission current density (Richardson’s law);

- the $\text{M}_m\text{X}_n$ compound appears on all of the surfaces as a result of a chemical reaction:

$$
\text{M} + \frac{n}{2m} \text{X}_2 \leftrightarrow \frac{1}{m} \text{M}_m\text{X}_n,
$$

where $k(T)$ is the reaction rate coefficient (Arrhenius equation). The reaction (2) happens due to the physical adsorption of the $\text{X}_2$ molecules, which is described by the Langmuir isotherm. As a result, the consumption of the $\text{X}_2$ gas occurring on the target, substrate and walls surfaces is denoted as $Q_t$, $Q_s$ and $Q_w$, respectively;

- the target surface is sputtered by the argon ions and evaporated according to the Hertz–Knudsen law, hence the metal $F_{1t}$ and the compound $F_{2t}$ flux rates can be expressed as

$$
F_{1t}(j_t) = F_{1sp}(j_t) + F_{1ev}[T_i(j_t)];
$$

$$
F_{2t}(j_t) = F_{2sp}(j_t) + F_{2ev}[T_i(j_t)].
$$

The first addends in the right part of equations (3) and (4) describe the sputtered particles, the second ones describe the evaporated particles;

- the temperature influence on such parameters of the process as the sputter yield and the ion-induced electron emission coefficient is neglected.

Using the law of mass action the way it was done in [5] and taking into consideration (1)–(4) we obtained a system of eight algebraic equations for one reactive gas. These equations describe

1) the balance on the surfaces of

- the target:

$$
F_{ch}[T_i(j_s)] + F_{2i} = 0;
$$

2) the substrate and the walls:

$$
F_{ch}(T_i) + F_{2i} = 0, \quad i = s, w;
$$

3) the reactive gas flow on all of the three internal surfaces of the vacuum chamber:

$$
Q_i = \frac{n}{2} F_{ch}(T_i) A_i, \quad i = t, s, w;
$$

4) the balance of the reactive gas flows:

$$
Q_o = Q_t + Q_s + Q_w + Q_p;
$$

The following parameters were used in (5)–(9): $F_{ch}(T_i)$ is the temperature dependable flux rate of the $\text{M}_m\text{X}_n$ compound formed due to the chemical reaction (2) on the $i$-surface; $F_{1i}(j_s)$ and $F_{2i}(j_s)$ are the flux rates of the metal and the compound particles sputtered from the target and deposited on the $i$-surface; $c^0 = 2.5 \cdot 10^{25}$ Pa$^{-1}$m$^{-3}$ is a coefficient converting the flow $Q_p$ from [Pa · m$^3$/s] to [s$^{-1}$]; $S_p$ is the system pumping speed; $p$ is the oxygen partial pressure.

The system of algebraic equations (5)–(9) contains two independent variables: the reactive gas flow and the ion current density. The unknowns are the partial pressure of the reactive gas $\text{X}_2$, the gas flows $Q_t$, $Q_s$, $Q_w$ and $Q_o$ and the fraction of each surface covered with the $\text{M}_m\text{X}_n$ compound. The system has to be solved numerically. In particular, when the surface temperature is so small that $j_T \ll \gamma j_s$ (1), $F_{1ev} \ll F_{1sp}$ (3) and $F_{2ev} \ll F_{2sp}$ (4), this system of equations describes the reactive sputtering process when $T_i \neq f(j_s)$. 
3. Simulations
The system of equations (5)–(9) was numerically solved for the magnetron with a tantalum target sputtered in the Ar + O$_2$ environment. The power density dependence of the target temperature was obtained by solving a thermal task. Using this dependence and the experimental current-voltage characteristics we determined the relation connecting the target temperature and the ion current density:

$$T_i \approx 293 + 1517 \left(1 - e^{-0.00438j_+}\right).$$

Considering (10) the equations (5)–(9) were solved with the ion current density ranging from 0 to 200 A/m$^2$. Figure 1 shows the oxygen partial pressure vs. the supply of the reactive gas for different ion current densities. Two characteristic curve segments are observed:

- first – when the oxygen flow is small, the pressure is close to 0;
- second – the pressure is proportional to the gas flow.

Similar dependences were often obtained experimentally during the reactive sputtering of a “cold” target [5]. The first segment of the curve is related to the metal mode of the target, the second one stands for the oxide mode. But the main feature of the sputtering process observed during these experiments is the hysteresis effect. The simulations result reflecting this effect is shown in figure 1(a). During the sputtering of a “cold” target the increase of the ion current density does not change the nature of the sputtering process curves. Both instability points of the $p = f(Q_0)$ curve shift to higher $Q_0$ values proportionally to $j_+$. The model predicts the elimination of the hysteresis effect at a certain critical value of the ion current density (figure 1(b)).

![Figure 1](image)

**Figure 1.** The oxygen pressure vs. the oxygen flow for $j_+$ (A/m$^2$): (a) – 160; (b) – 170; (c) – 180.

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References
[1] Levitskii V S, Shapovalov V I, Komlev A E et al 2015 Techn. Phys. Lett. 41 1094–6
[2] Lapshin A E, Levitskii V S, Shapovalov V I et al 2016 Glass Phys. Chem. 42 (in print)
[3] Shapovalov V I 2015 Nanoingeneria 11 22–34
[4] Särhammar E, Nyberg T and Berg S 2015 Sur. Coat. Technol. 279 39–43
[5] Barybin A A and Shapovalov V I 2007 J. Appl. Phys. 101 054905
[6] Barybin A A, Zavyalov A V and Shapovalov V I 2012 Glass Phys. Chem. 38 396–401