Modelling of hot target reactive sputtering

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Abstract. The main foundations of the physicochemical models for reactive magnetron sputtering of a hot metal target in a nitrogen environment are described. The system of equations describing the model is solved for the titanium sputtering in a nitrogen environment. It is established that the width of the hysteresis loop decreases down to zero with an increase of the discharge current density.

1. Introduction
A detailed study of the cold target reactive sputtering has allowed to develop physical models of different complexity. The private physical models were proposed in the last third of the 20th century [1, 2]. In addition, a large number of studies are devoted to the development of a general model, which presents the compound formation on all surfaces of a vacuum chamber as chemisorption [3–6]. A non-isothermal physicochemical model describes the process of the cold target reactive sputtering in a more consistent way [7, 8].

2. Physicochemical model
A "hot" target means that the target is heated to a temperature at which the evaporated material flow cannot be neglected against the sputtered flow. The elements of the system are located in the Ar + N₂ mixture. Our key assumptions are described below.

In the hot target sputtering, the physicochemical processes taking place on the target surface and in the gas environment change. To describe the problem correctly, it is necessary to take into account the impact of the target temperature on these processes [9, 10].

1. The temperature of the target depends on the \( j_i \) ion current density (or on the power density radiated on the target): \( T_t = f(j_i) \).

2. The plasma-chemical reaction for the \( M_mN_n \) nitride formation on the excited target surface and surface chemical reactions on the substrate and the wall are given by an equation

\[
M + \frac{n}{2m}N_2 \leftrightarrow \frac{1}{m}M_mN_n. \tag{1}
\]

The reaction (1) is fed by the molecular flow of N₂ from the gas phase, which is described by the Langmuir isotherm. As a result, the gas consumption occurs on each surface having area \( A_i \) (figure 1); the consumption is given as a flow \( Q_i, (i = t, s, w) \).

3. The surface of the target is sputtered by argon ions and evaporated in accordance with the Hertz-Knudsen law, thus the densities of \( J_M \) metal flow and \( J_N \) compound from the target surface are expressed as follows (figure 1(a))
The first terms on the right of the equations (2) describe sputtered flows, the second ones – evaporated flows.

4. The film on the substrate and the wall is formed by the reaction (1) and material flow from the target (figure 1(b)). Its chemical composition depends on the mode of operation of the target.

5. The discharge current density is

\[ j = (1 + \gamma) j_i + j_e, \]

where \( \gamma \) – the coefficient of ion-electron emission; \( j_e \) – the current density of thermionic emission (the Richardson-Deshman law):

\[ j_e(T_e) = A T_e^2 \exp \left( -\frac{\varphi_t}{k T_e} \right), \]

where \( A \approx 120 \text{ A-cm}^{-2}\text{K}^{-2}; \varphi_t \) – electronic work function for the target material; \( k \) – Boltzmann’s constant.

6. The \( j \) discharge current density and the \( Q_0 \) volume flow rate of nitrogen are independent variables. The main dependent variable of the process is the partial pressure of nitrogen.

The analytical description of the physicochemical model contains eight algebraic equations [7, 10]:

• equations for the stationary state of the surfaces of a target, a substrate and a wall;
• equations describing nitrogen consumption by the surfaces of a target, a substrate and a wall;
• equation of high-vacuum pumping system;
• gas balance equation.

3. Simulations

The proposed model was used to study the process of the hot titanium target sputtering in the Ar + N\(_2\) environment. The dependence of the target temperature on the power density was obtained by numerical solution of the thermal problem. Using this dependence and the experimental current–voltage characteristics of the magnetron, the relation connecting the target temperature with the ion current density was determined:

\[ T_t \approx 1804 - 1464e^{-0.00446 j_e}. \]

The results of solving the system of equations are showed in figure 2. The dependence of the nitrogen partial pressure on its flow rate with the change of the current density is calculated in the range of 0 to 500 A/m\(^2\). The study of the model showed that there is a critical temperature (power density). The process of sputtering loses its hysteresis under this temperature. The region with a negative derivative disappears, but two typical regions are kept (figure 2(b), (c)) at this and higher temperatures depending on \( p = f(Q_0) \):

![Figure 1](image)

**Figure 1.** The processes on the target (a), the substrate (i = s) and the wall (i = w) (b).
• the first one corresponds to the work of the magnetron under the “metal” operating mode of the target. The pressure is close to zero on it at a low nitrogen flow;
• the second one reflects the “nitride” mode. In this mode, the pressure is proportional to the flow rate of nitrogen.

![Figure 2](image-url) Dependence of the partial nitrogen pressure on the flow for a hot target at the discharge current density (A/m²): (a) – 270 (1365); (b) – 280 (1384); (c) – 290 (1402). Corresponding target temperature in K calculated using the formula (5) is given in parentheses.

![Figure 3](image-url) Current density dependences of the nitrogen input flow values when operating modes of the cold (1) and hot (2) targets change: $Q_1$ – a metal-to-oxide mode transition; $Q_2$ – an oxide-to-metal mode transition.

It is known that the hysteresis effect remains at any current density [7] during reactive sputtering of a cold target. This parameter affects the position of the points of the target transition from one mode to another and the width of the $\Delta Q_0$ hysteresis loop. Dependences of the $Q_0$ nitrogen flow rate, when there are changes of operating mode of cold and hot target, on the current density are shown on the figure 3. As it can be seen from figure 3, these values for the cold target are proportional to the current density. And they are highly nonlinear for a hot target.

Figure 4(a) reflects the dependences $\Delta Q_0 = f(j)$. They are shown in figure 4(a) by the round markers for a cold target. The surface temperature of the target is a constant parameter in this problem. The surface temperature significantly affects the value of $\Delta Q_0$. The function $\Delta Q_0 = f(j)$ differs for a hot target as it can be seen from figure 4 (triangular markers). A smooth increase of $\Delta Q_0$ ends with an extremum and a further decrease down to zero. The critical value of the current density when the width of the hysteresis area draws in zero is equal to 458 A/m².

Further calculations shown in figure 4 allowed finding that evaporation is the basic physical mechanism initiating the loss of the hysteresis by the reactive sputtering process. Thermionic emission has a little effect on the process.

4. Conclusions
The physicochemical model of the hot target reactive magnetron sputtering makes it possible to analyse the process. For the titanium target sputtered in $Ar + N_2$ environment, two types of processes depending on the current density are possible: with and without the hysteresis. The same result was obtained for the tantalum target sputtered in $Ar + O_2$ environment [10].
Figure 4. Dependence of the hysteresis width on the current density: (a) – for hot and cold targets; (b) – Hot targ – full model, Hot targ-evap – neglecting thermionic emission, Hot targ-emiss – neglecting evaporation.

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