PROCESSES CONTROL FOR OXIDE LAYER DEPOSITION IN ROLL-TO-ROLL VACUUM MACHINES

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Abstract

Vacuum deposited coatings of metal oxides have wide usage in various applications. When used in roll-to-roll processing with magnetron sputter deposition, stable conditions must be maintained over a long period of time. Hysteresis with pressure variation is one cause of property/composition changes during processing. This paper shows that even by increasing the pumping speed hysteresis problems are not overcome. For stable oxide deposition by reactive deposition in the transition mode good operational control is necessary. For stable operation it is necessary to consider hysteresis associated with high and low pumping speeds. This control may be obtained with feedback from: PEM, λ-probe, mass spectrometer, plasma impedance, etc. In magnetron discharges, power may be kept constant by keeping voltage constant and controlling current by controlling the partial pressure of oxygen (by oxygen flow) in the chamber. With power held constant hysteresis associated with oxide deposition is absent. This paper shows the results of statistical analysis of stability of the reactive deposition process as a function of quality of the control system and its response time.

Introduction

Pumping speed impact on reactive magnetron sputtering has been considered in many works. There are several surveys where these issues have been discussed in detail [1, 2, 3]. Investigators pay great attention to the pumping problems, because not only coating quality and process control capabilities are contingent on the pumping speed, but this issue also determines the vacuum equipment costs, a sizable proportion of which is the pumping means pricing. Metallic or ceramic targets are used when depositing oxide and nitride coatings by magnetron sputtering. Simple technique of the metallic targets manufacture and low costs are their advantages, it is also possible to operate them at high power densities, increasing productivity. In some cases a conductive ceramic target is nonexistent in nature, e.g. for silicon oxide. The ceramic targets advantage shows itself in the deposition process stability, process control simplicity.

Achievements in the magnetron sputtering process control at a transition mode provide stable operation and high deposition rates. The process control quality or the process stability is contingent on feed-back and a control method and may assess quantitatively on the basis of statistic analysis of the process parameters.

Experiment

The experiments were carried out at a laboratory vacuum machine UV80 for coating webs up to 600 mm wide with pumping by diffusion pumps, which provided the pumping speed up to 3.4 m^3/s, the chamber volume is 3.3 m^3, ultimate vacuum is 1×10^{-5} Torr [4]. We investigated reactive sputtering of silicon and titanium. A Puls DC power supply, 10 kW, frequency 0 – 350 kHz was used during the work. Silicon was sputtered at frequency of 100 kHz, titanium was sputtered at DC and at frequency of 25 kHz.

The process control system, which had been developed relating to the machine UV80, enables conducting the process in the transition mode with feed-back of voltage, current, power, partial pressure (λ-probe Zirox XS22.3), providing collection, storage and processing information on the process flow with periodicity 1 s.

Statistical methods of the results processing may be used for assessment of the process control quality. A parameter of interest is selected from the table of the process information for the final time period (no less than 1 minute). As there are many measurements (> 30), in estimated average x_{ave} we used the confidence interval $\pm\ 2\sigma$ with accuracy α = 0.954.

It is possible to consider the value of the confidence interval relative deviation from the average (in percents)

$$\varepsilon = \pm 2\sigma / x_{ave} \quad (1)$$

as the control quality measure or the process stability measure. It is evident that the lower value ε is, the higher the process stability and the control quality are.

Program Excel was used for calculation, it enables automatic obtainment of complete information on the control quality or the process stability numerically.

Two hysteresis types and the process control

The pumping speed exercises great influence over the reactive magnetron deposition of oxide coatings. At constant sputtering power and manual control of the oxygen flow (gradual increase and subsequent decrease of the flow) hysteresis availability is characteristic for voltage, current and coating thickness. Hysteresis is determined by dynamic interrelation of the pumping speed and argon and reactive gas feeding into the vacuum chamber, it also depends on sputtered material and the chamber design peculiarities. The magnetron discharge voltage hysteresis is considered below uppermost because it does not require special equipment for the process observation on a real time basis, the discharge is not interrupted, operation in the transition mode is easily controlled and provides positive capability of reproducing, which is important for commercial equipment.

When the chamber pumping speed varies, reactive gas pumping also changes and the voltage hysteresis nature changes while sputtering Si and Ti, Fig. 1, 2.

Silicon reactive sputtering at frequency of 100 kHz provides the process stable flow, practically without arcing. At DC sputtering instability occurs in the form of arcing, especially when sputtering power is increased.

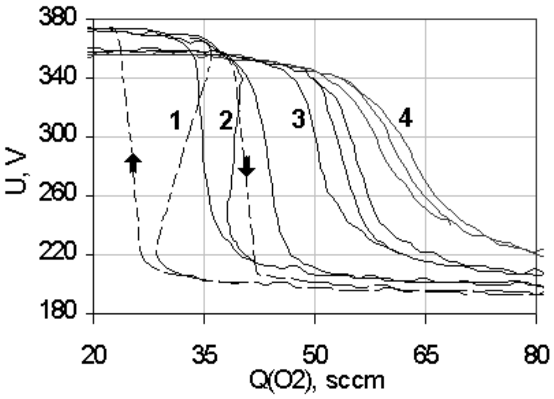

Fig. 1. Voltage hysteresis loop dependence on the pumping speed, SiO_2, Puls DC. 1 – 0.4 m^3/s, 2 – 0.8 m^3/s, 3 – 1.7 m^3/s, 4 – 3.4 m^3/s.

At the low pumping speed the voltage hysteresis loop for Si is characterized by classicality, transition from the metal mode to the oxide mode and vice versa occurs in the form of an avalanche at small changes of the oxygen flow into the chamber and oxygen flow Q_1, at which the metal mode/oxide mode transition starts, is higher than flow Q_2, at which the opposite oxide mode/metal mode transition starts (loops at pumping speeds 0.8 m^3/s and 0.4 m^3/s, the first mode). During the process control the voltage derivative by oxygen flow in the transition mode on the hysteresis loop curve is

$$\frac{\partial U}{\partial Q} > 0 \qquad (2).$$

When the pumping speed is increased, a smooth change of the discharge voltage occurs at transition from the metal mode to the oxide one and vice versa, though hysteresis remains, but the relation between Q_1 and Q_2 flows is opposite (hysteresis loops at the pumping speeds 3.4 m^3/s and 1.7 m^3/s, the second mode) and it is impossible to identify clearly the flows Q_1 and Q_2, the voltage derivative by the oxygen flow becomes negative

$$\frac{\partial U}{\partial Q} < 0 \qquad (3).$$

We are of the opinion that it is necessary to discern hysteresis in the first and second modes. It is possible to speak about classic hysteresis in the first mode at the low pumping speed, Fig. 2. In the second mode hysteresis exists, but it is essentially different from the classic one on its influence on the process stability.

In both modes the process control with feed-back is necessary for stable process flow in the transition mode. In the first mode after control switching off the process transits from the transient mode to the metal or oxide mode during several seconds, i.e. the normal process flow is interrupted. In the second mode the process is more stable and after control switching off only in course of time some drifting between minimum and maximum voltage values occurs at the set oxygen flow value, namely this drifting is controlled and eliminated by the control system. The narrower hysteresis loop is, the less voltage vary will be after control switching off.

Thus, when the pumping speed is increased, as it follows from Fig. 1 for SiO_2, the process transits from the first mode to the second one, the flow values Q_1 and Q_2 are compared and the derivative is increasing and passes through value

$$\frac{\partial U}{\partial Q} = \propto \qquad (4).$$

Close by transition from the first mode to the second one the control system operates unsteadily and cannot cope with the process stabilization. In this case the set voltage should be maintained by extremely small changes of the oxygen flow, it is fairly difficult to implement such task with standard equipment. When silicon oxide and other coatings with similar hysteresis loop are deposited, it is necessary to avoid the conditions, which are close to (4). It may be provided e.g. by changing the pumping speed or another process parameter.

Character of the discharge voltage hysteresis loop at reactive sputtering of titanium, Fig. 2, is different from the corresponding curve of silicon. This difference may be explained by different factors of ion-electron emission during titanium and silicon sputtering. When silicon is sputtered, the factor of the secondary ion-electron emission is higher at oxide sputtering in comparison with pure silicon. For titanium the relation is opposite: the factor of the secondary ion-electron emission is higher at pure titanium sputtering in comparison with its oxide.

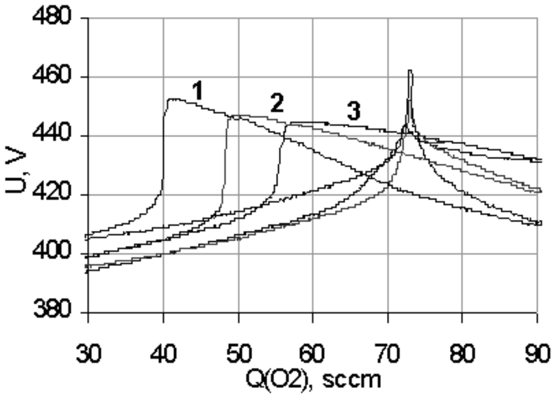

Fig. 2. Voltage hysteresis loop dependence on the pumping speed, TiO_2, Puls DC, 25 kHz. 1 – 0.8 m^3/s, 2 – 1.7 m^3/s, 3 – 3.4 m^3/s.

It follows from Fig. 1 and 2 that when the pumping speed is increased, the hysteresis loop width changes. Dependence of the hysteresis loop width D on the pumping speed for Si at the medium part of Fig. 1 at the level of ~ 300 V and for Ti at the level of ~ 430 V of Fig. 2 is presented in Fig. 3.

If the curve parts of the first and second modes for Si from Fig. 3 are extrapolated, it is possible to estimate the chamber pumping speed, at which transition from the first mode to the second one occurs. It is visible from the represented data that transition from the first mode to the second one occurs at the chamber pumping speed ~ 1 m^3/s. As far as the process is carried out at the constant argon flow, transition from the first mode to the second one is determined by the oxygen flow and in this case the oxygen pumping speed will be critical [1, 2].

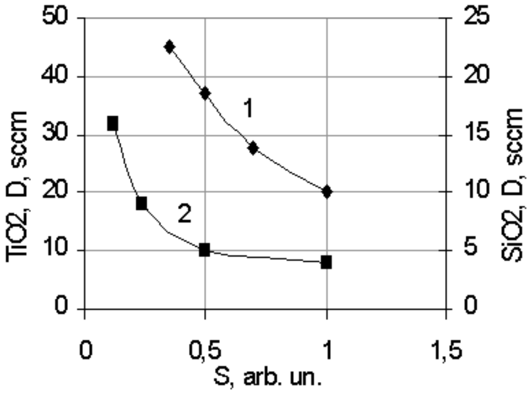

Fig. 3. Dependence of the voltage hysteresis loop width on the pumping speed. 1 – TiO_2, 2 – SiO_2.

At the chamber pumping speed S>1 m^3/s classic hysteresis will be eliminated. Virtually it is possible to eliminate only classic hysteresis by increasing the pumping speed, hysteresis will remain even at high pumping speed, Fig. 3.

As it is visible from Fig. 3, classic hysteresis for Ti also occurs at high pumping speed. In this case it is impossible to speak about critical pumping speed.

Titanium reactive sputtering was carried out at frequency of 25 kHz, during DC sputtering there were arcing instabilities. During titanium reactive sputtering the hysteresis loop form is sensitive to sputtering frequency, when sputtering frequency was increased to 50 kHz, the hysteresis loop degenerated, but discharge voltage in the metal mode differed from the oxide mode only by 10 - 20 V, and when frequency was increased to 100 kHz, the hysteresis loop orientation changed in inversed manner to have the form, which is characteristic for Si. In this case the dynamic deposition rate was low (Fig. 4).

We considered the pumping problems in terms of voltage hysteresis. Similar results were received when hysteresis of reactive gas partial pressure and other values was observed.

The mode at the high pumping speed is more stable, but the pumping speed increase requires tangible cost and complicates the design. Necessary coating properties are basic criterion, these properties should be maintained. If necessary properties are received without increasing pumping, there is no necessity to enhance pumping. Up-to-date methods of the process control provide stable operation irrespective of hysteresis.

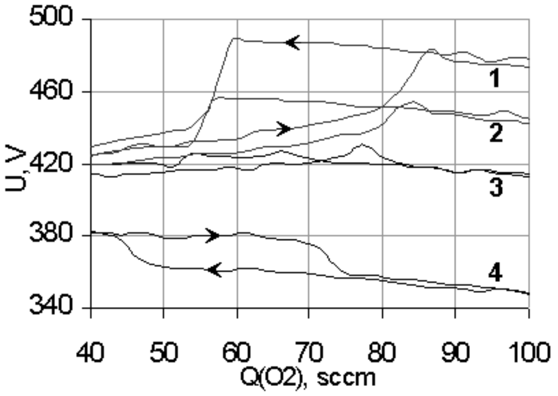

Fig. 4. Hysteresis loop alteration when frequency of the Puls DC power supply was changing. 1 – DC, 2 – 25 kHz, 3 – 50 kHz, 4 – 100 kHz.

When SiO_2 was deposited by reactive magnetron sputtering of Si(Al, 10%) target with the transient mode control, the dynamic rate of 45 nm×m/min was achieved with the single magnetron, which was powered by Puls DC power supply of linear power density of 55 W/cm. The dynamic rate of 80 nm×m/min was achieved in the dual mode with MF power supply of linear power density of 130 W/cm.

In order to compare different magnetron systems (magnetron in a set with a power supply) it is convenient to use value

$$e = DDR/w \quad (5),$$

which may be called the magnetron system coefficient of performance.

Here:

DDR – dynamic deposition rate by magnetron sputtering, nm×m/min,

w – linear power density (of single or dual magnetron), W/cm.

Then the single magnetron with the Puls DC power supply has the silicon reactive sputtering coefficient of performance $e = 0.82$, while that of the dual magnetron with the MF power supply is $e = 0.62$.

Linear density of sputtering power is limited due to the silicon target breakup at high power. Cylindrical targets have more favorable cooling conditions in comparison with planar targets, and in this case it is possible to increase linear power density.

When TiO_2 was deposited by planar magnetron reactive sputtering of the metallic target (single magnetron) with the process control in the transient mode, the dynamic rate of 30 nm×m/min was achieved at linear power density of 116 W/cm at frequency of 25 kHz with the Puls DC power supply, $e = 0.26$. There is the sputtering power density reserve for the metallic target, it is possible to increase power density 2 times. The dynamic rate of 13 nm×m/min was achieved by sputtering the ceramic target TiO_x at power density of 57 W/cm, $e = 0.23$. There is no power reserve, because the ceramic target starts breaking, when power is increased. When the ceramic targets TiO_x were sputtered in the dual mode, the dynamic rate of 30 nm×m/min was achieved at linear power density of 170 W/cm, $e = 0.18$. Power density of the cylindrical targets is somewhat higher due to the better cooling condition.

Control system operation speed

Analysis of the process control quality of silicon reactive sputtering in the transition mode in three control systems has been implemented during the work. Sputtering power was stabilized by the power supply in the systems of control by impedance and by oxygen partial pressure. In the control mode according to the system of work [4] the power supply stabilized sputtering voltage, what allowed eliminating hysteresis. Sputtering was implemented with the Puls DC power supply at 4 kW power at two values of the pumping speed 0.4 m^3/s and 3.4 m^3/s.

The control system setting provided high accuracy of maintaining the process parameters in the transition mode. Control quality or the process stability is not worse than ± 0.5 % at the pumping speed of 0.4 m^3/s and not worse than ± 0.1 % at the pumping speed of 3.4 m^3/s, what provides capability of reproduction and stabilization of the coating properties during the process. As it is obvious, accuracy of maintaining is higher, when the pumping speed is increased and when we are getting away from classic hysteresis.

Assessment of the control system rate of response is of interest. After the process control switching off we estimated time Δt of moving the value under consideration outside the range of the value confidence interval, the results are given in Table 1. Assessment accuracy is ± 1 s, as registration of the measurement results is made at the 1 s interval. Time of maintaining the controlled value magnitude after control switching off is no less than 3 s for impedance and partial pressure systems. It means that the control system has no less than 3 s for receipt of the signal from the sensor, processing the signal, transfer of the command to the operating mechanism and correction input.

Table 1.

Control mode		$S = 0.4$ m^3/s	$S = 3.4$ m^3/s
Impedance	Δt_U, s	10	3
	Δt_λ, s	7	4
Partial pressure	Δt_U, s	5	>10
	Δt_λ, s	5	>10
[4]	Δt_U, s	>60	>60
	Δt_λ, s	>60	>60

Δt_U – duration of maintaining the set discharge voltage after control switching off;

$\Delta t\lambda$ – duration of maintaining the set value of λ-probe after control switching off.

Apparently, the received values are determined by the machine design peculiarities and they will be different for other machines, but unlikely that they will be less than 1 s.

These results contradict with conclusions of other researchers, who claim that the control system operation speed should be much higher and it is necessary to use a high-speed piezoelectric valve in order to control the process [5, 6]. Our results show that the standard gas flow control valve of 1179 type of MKS Company may be used in the control system, it has time of the set flow assignment < 0.8 s (within 2 % of the set value).

The control system, which provides the process operation without hysteresis [4], compares favorably with those of impedance and partial pressure, after the control system switching off the process remains stable for an appreciable length of time (dozens of minutes) due to hysteresis absence.

Conclusions

For practical purposes it is important to discern two types of hysteresis depending on the pumping speed, what allows using less expensive pumping means without impairing coating properties and quality.

The coefficient of performance of the singular magnetrons and Puls DC power supplies systems is higher than those of the dual magnetrons and MF power supplies.

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