Jan WALKOWICZ*, Aleksandr ZYKOV, Stanislav DUDIN**, Stanislav YAKOVIN**, Rafał BRUDNIAS***

ICP ENHANCED REACTIVE MAGNETRON SPUTTERING SYSTEM FOR SYNTHESIS OF ALUMINA COATINGS

UKŁAD REAKTYWNEGO ROZPYLANIA MAGNETRONOWEGO DO OSADZANIA POWŁOK TLENKU ALUMINIUM WSPOMAGANY WYŁADOWANIEM INDUKCYJNYM RF

Key-words:

reactive magnetron sputtering, inductively coupled plasma, alumina coatings

Słowa kluczowe:

reaktywne rozpylanie magnetronowe, plazma wyładowania indukcyjnego, powłoki tlenku aluminium

* Institute for Sustainable Technologies – National Research Institute, ul. Pułaskiego 6/10, 26-600, Radom, Poland.

** Department of Physical Technologies, Kharkiv National University, 31 Kurchatov Ave., Kharkiv – 61108, Ukraine.

Summary

The investigation results of the DC magnetron sputtering system for synthesis of high-quality aluminum oxide coatings are presented. In the system oxygen, activated by an independent Inductively-Coupled Plasma (ICP) source, was introduced into an aluminum sputtering chamber as an alternative to conventional reactive sputtering techniques employing the injection of ground state molecular oxygen O_2. Behaviour of deposition process characteristics were investigated with and without the activation of the reactive species.

INTRODUCTION

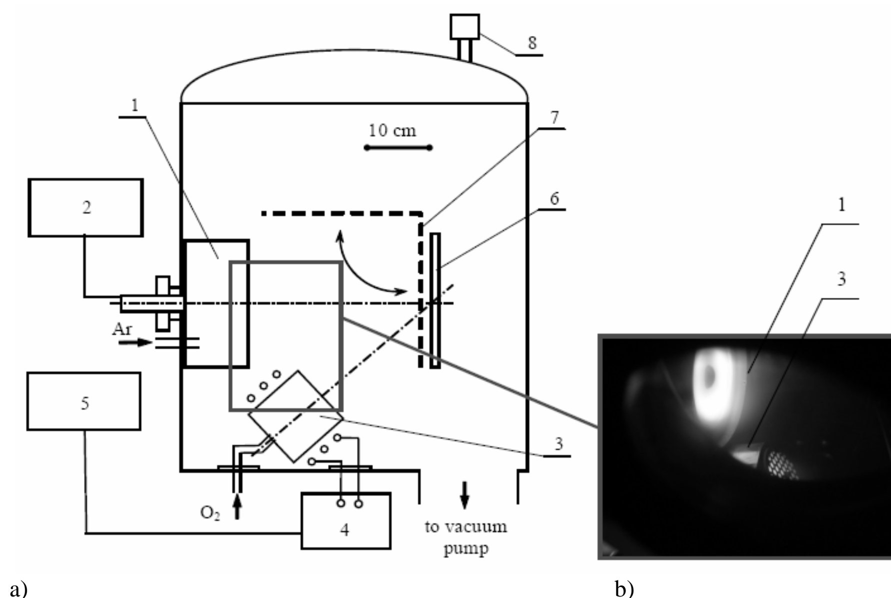
Aluminum oxide thin films are widely used in many mechanical, optical and microelectronic applications because of their excellent properties **[L. 1]**, in terms of chemical inertness, mechanical strength and hardness **[L. 2]**, transparency, high abrasive and corrosion resistance, as well as insulating and opticalb properties. However the synthesis of dielectric coatings, in particular the oxides such as TiO_2, Al_2O_3, Ta_2O_5, by DC magnetron sputtering encounters some serious difficulties for the simple reason that the films being processed do not conduct current. This leads to the target passivation, intensive arcing, and the instability of discharge (phenomenon of "disappearing anode"), that essentially constricts the "window" of technological parameters and reduces the coatings quality **[L. 4]**.

In this paper the results of development of the new DC magnetron sputtering system for synthesis of high-quality oxide coatings are presented. The basic idea of the system consists in separation of two main constituent processes: metal target sputtering by DC magnetron discharge in inert gas, and activation and transport of reactive gas by additional plasma source based on RF inductive discharge. The key novelty of the present system comparing to known designs is the operation pressure range $(0.7–3.5)\times10^{-3}$ mbar where motion of particles may be treated as free fall. It allows the distance magnetron – substrate holder to be increased up to 30–40 cm, which leads to significant increase in the deposition area at acceptable deposition rate.

EXPERIMENTAL

Experimental setup

Deposition processes were performed in the Balzers BA-510A vacuum system equipped with high vacuum pumping system consisting of oil diffusion pump, Roots' pomp and sliding-vane pump. A schematic layout of the magnetron and ICP source in the sputtering chamber is shown in **Fig. 1**.



a) b)

Fig. 1. Experimental setup: a) arrangement of main elements of the deposition system, b) DC magnetron and ICP source during deposition process; 1– magnetron, 2 – DC power supply, 3 – ICP source, 4 – RF matchbox, 5 – RF power supply, 6 – substrate holder, 7 – substrate shield, 8 – vacuum gauge

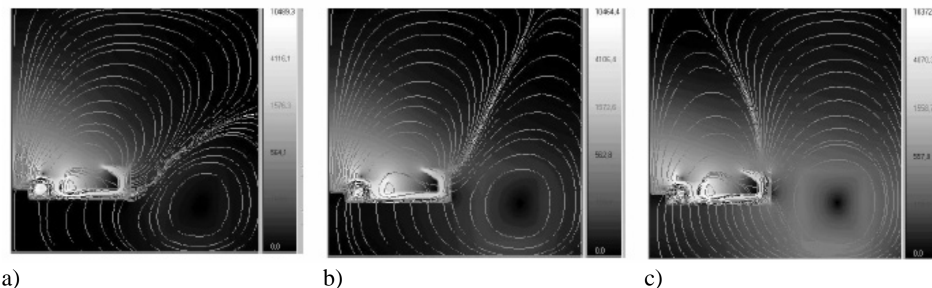
Rys. 1. Układ eksperymentalny: a) rozmieszczenie głównych elementów układu osadzania, b) magnetron DC i indukcyjne źródło plazmy ICP podczas procesu osadzania; 1 – magnetron, 2 – zasilacz DC, 3 – źródło ICP, 4 – układ dopasowania wyładowania RF, 5 – zasilacz generatora RF, 6 – uchwyt podłoży, 7 – ekran uchwytu podłoży, 8 – czujnik próżni

Power to the sputtered cathode was applied using 10 kW DC power supply (2), produced by ITeE-PIB, operated either in current or voltage regulation mode. Oxygen for the reactive deposition was delivered through the ICP source (3). Argon, used as the sputterring gas in all

deposition experiments, was fed to the chamber independently of the reactive oxygen – directly on the magnetron target. Flows for both argon and oxygen were regulated using BETA ERG mass flow controllers operated by two-channel process control unit. Pressure monitoring in the deposition chamber was accomplished using Balzers PKR-250 Pirani / Cold Cathode gauge (8). The substrates were mounted on a shielded substrate holder (6) that allowed to deposit up to 12 samples per pumping cycle.

Low pressure magnetron

The deposition of coatings at pressure range of 10^{-3} mbar allows to execute collisionless free-fall regime of deposition process, and to increase essentially the deposition rate at considerable distances from the target. In presented experiments a newly developed low-pressure magnetron was used. The magnetron construction was developed and optimized with the use of computer simulation, of resultant magnetic field configuration in magnetrons with double magnetic system, carried out in Kharkov National University **[L. 5, 6]**. Depending on relation between external magnetic field, generated by the solenoid located around the target periphery, and internal magnetic field, generated by a permanent magnet located underneath the target, it is possible to realize three types of magnetron discharge (**Fig. 2**).

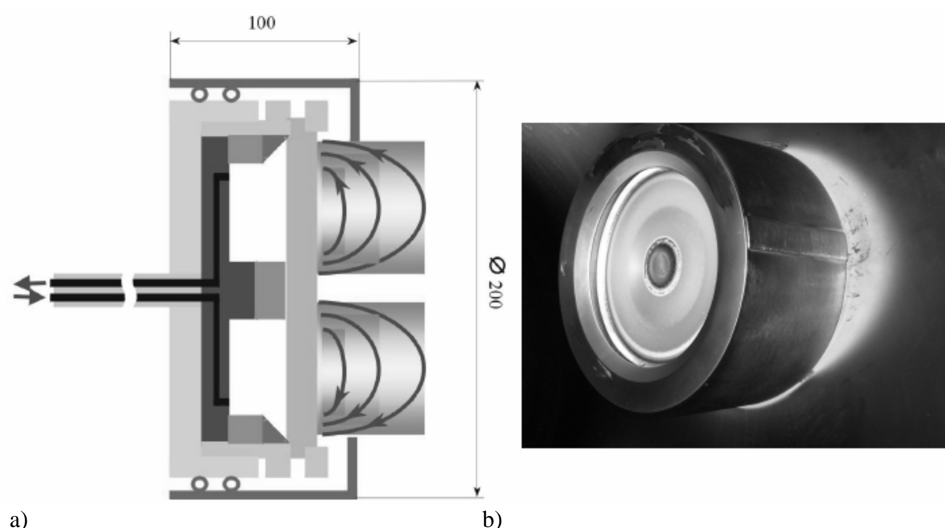


a) b) c)

Fig. 2. Computer simulation of the resultant magnetic field configuration of low pressure magnetron: a) unbalanced magnetron of the first type – weak „external" magnetic field, b) balanced magnetron – medium „external" magnetic field, c) unbalanced magnetron of the second type – strong „external" magnetic field

Rys. 2. Symulacja komputerowa konfiguracji wypadkowego pola magnetycznego w magnetronie niskociśnieniowym: magnetron niezbalansowany pierwszego rodzaju – słabe „zewnętrzne" pole magnetyczne, magnetron zbalansowany – średnie „zewnętrzne" pole magnetyczne, magnetron niezbalansowany drugiego rodzaju – silne „zewnętrzne" pole magnetyczne

The switch-off pressure for magnetron discharge depends on the ratio of magnitudes of external and internal magnetic fields. This ratio defines a configuration of the resultant magnetic field over the magnetron target or, in other words, the degree of the magnetron balance. Minimum pressure, at which the existence of magnetron discharge is still possible, is in analysed configurations lower by an order of magnitude than in the case of traditional magnetron, i.e. when the current in external coil is equal to zero. Based on the obtained results the low pressure magnetron 200 mm in diameter with permanent magnets and minimum working pressure less then 0.7×10^{-3} mbar was developed, manufactured and mounted in the Balzers BA-510A vacuum chamber (**Fig. 3**). A pure aluminium target 170 mm in diameter served as the sputtered aluminium source. Magnetic force lines configuration of the developed magnetron corresponds to the balanced type of magnetron (**Fig. 2b**).



a) b)

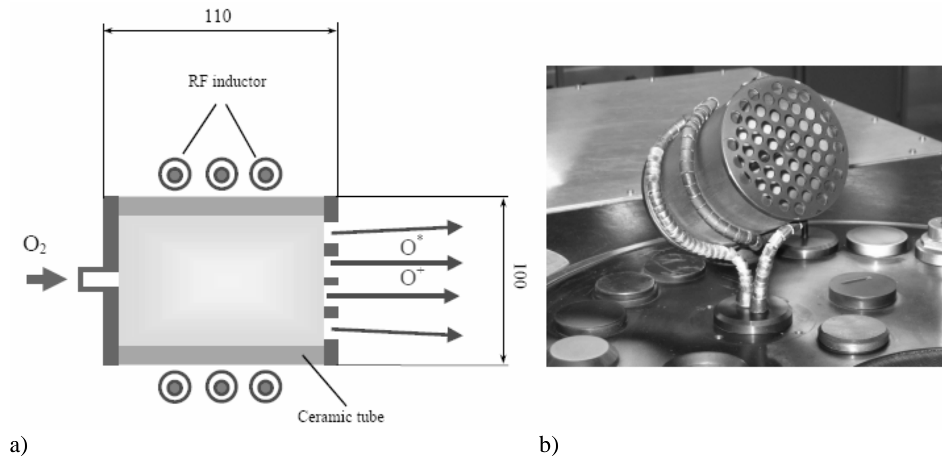
Fig. 3. Low pressure DC magnetron used for Al_2O_3 coatings deposition: a) pictorial diagram of the magnetron, b) the magnetron installed in Balzers BA-510A vacuum chamber

Rys. 3. Niskociśnieniowy magnetron DC wykorzystywany w procesach osadzania powłok Al_2O_3: a) schemat magnetronu, b) magnetron w komorze próżniowej stanowiska Balzers BA-510A

ICP source

In the presented work a simple ICP plasma source was used (**Fig. 4**). A ceramic tube 100 mm in diameter and 120 mm in length was used as RF

discharge chamber. The discharge chamber of the source was closed with a grid (with the holes diameter of 8 mm) applied for plasma contraction and creation of a pressure drop between the source chamber and the deposition chamber.



a) b)

Fig. 4. The ICP source used for Al_2O_3 coatings deposition: a) pictorial diagram of the source, b) the source installed in Balzers BA-510A vacuum chamber

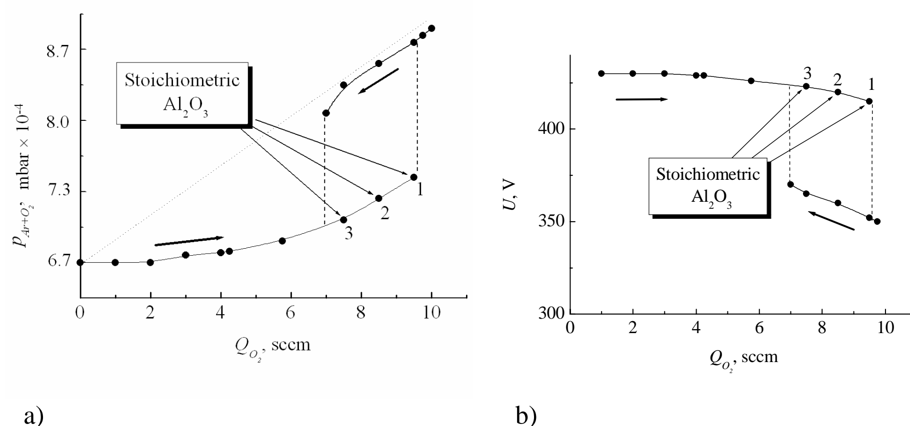
Rys. 4. Indukcyjne źródło plazmy ICP wykorzystywane w procesach osadzania powłok Al_2O_3: a) schemat źródła, b) źródło w komorze próżniowej stanowiska Balzers BA-510A

TECHNOLOGICAL BASELINE

Process parameters characteristics

The inherent feature of the reactive deposition processes executed with the use of conventional magnetron systems is the existence of discharge instabilities that are connected with the phenomenon of the target poisoning **[L. 7–10]**. The problem was also studied within presented work for the processes of Al_2O_3 coatings deposition with the use of ICP enhanced reactive magnetron sputtering system (**Fig. 5**).

Fig. 5a shows typical oxygen partial pressure hysteresis characteristic of the reactive deposition process. Presented graph clearly shows the dynamic effects that take place in a reactive sputtering process of aluminium as the oxygen partial pressure is increased. At low flows, the pumping of oxygen is aided by the reaction with aluminium on surfaces within the deposition chamber. Beyond a critical point 1 (**Fig. 5a**) where



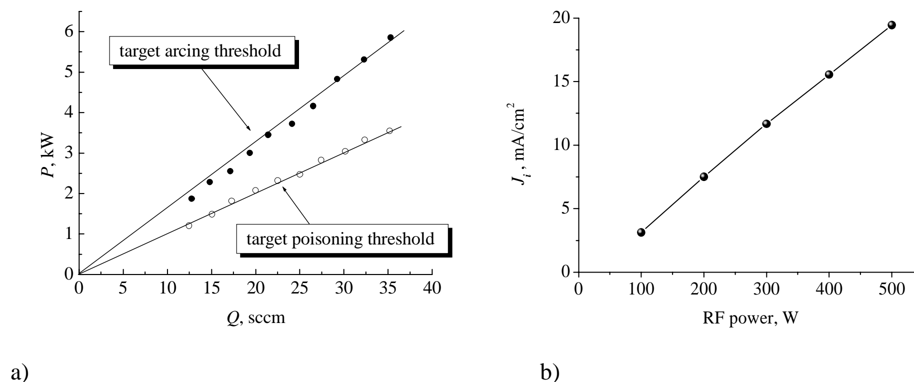
a) b)

Fig. 5. Hysteresis of the process parameters with changing flow rate of the reactive gas: a) oxygen partial pressure hysteresis, b) cathode voltage hysteresis

Rys. 5. Histereza parametrów procesu przy zmiennej szybkości przepływu gazu reaktywnego: a) histereza ciśnienia cząstkowego tlenu, b) histereza napięcia na katodzie magnetronu

the stoichiometric compound is produced, the gettering process within the reaction chamber is saturated and the pumping capacity is overwhelmed leading to a rapid increase in oxygen partial pressure. In the oxygen saturation region of the curve, all surfaces in the reaction chamber (including the magnetron target) are covered with an insulating layer of alumina. Most noteworthy is the state of the target in this region as it is said to be "poisoned" or "passivated" due to the presence of an oxide layer on the surface. When passivation occurs, deposition rate drops dramatically, arcing intensity increases, and severe instabilities start to occur. This is explained by the increase in secondary electron emission from the oxide surface illustrated in the complementary target voltage hysteresis curve shown in **Fig. 5b**. Here, the target voltage is monitored at fixed current to the cathode of 2.5 A. As the oxygen flow is increased and chamber surfaces become oxidized, the transition of the target from the metallic mode (450 V) to the passivated mode (340 V) occurs very rapidly. The dependence of minimum DC power of magnetron operation in metallic mode was measured versus oxygen flow rate (**Fig. 6a**). Apparently from the figure it is practically linear with the factor of about 10–12 sccm/kW. In **Fig. 6b** the dependence of ion current density J_i at

the output of the ICP source on RF power is presented. Preliminary measurements showed, that at pressure less than 3.5×10^{-3} mbar ion and active particle flows from the source have divergent profile with a cosine-like law. This fact was taken into consideration in development of the experimental setup configuration.



a) b)

Fig. 6. Relation between DC and RF discharges power and oxygen flow characteristics: a) dependence of minimum magnetron DC power P for metallic mode operation on oxygen flow rate Q, b) dependence of ion current density J_i at the output of the ICP source on RF power

Rys. 6. Relacje pomiędzy mocą wyładowań DC i RF a charakterystykami przepływu tlenu: a) zależność minimalnej mocy P magnetronu DC w modzie metalicznym od szybkości przepływu tlenu Q, b) zależność prądu jonowego na wylocie źródła ICP od mocy wyładowania RF

Deposition of alumina coatings

In the developed method, to achieve stable conditions for aluminium oxide deposition and to provide control margin of the reactive deposition process, the effective oxygen partial pressure at the substrate was modified using two techniques simultaneously. The first one is delivery of the reactive gas directly to the substrate surface. This technique protects the magnetron target surface from poisoning by maintaining lower oxygen partial pressure in its vicinity. The second technique of sputtering conditions modification, which is of primary interest for the developed method, consists in preactivation of the reactive gas using the independent RF ICP source. This idea was successfully verified in the carried out experiments. In **Fig. 5** the limit of stoichiometric film deposition is marked by 2 and 3 and switching-on the ICP source (500–700 W) did not

change the hysteresis dependencies. Thus the principal problem with deposition of stoichiometric Al_2O_3 films when using the directed source of activated oxygen is the relative location of the magnetron, the ICP source and the substrate, which determine 3-dimensional Al_2O_3 stoichiometry region in the chamber space. In **Fig. 7a** calculated 2D stoichiometry diagram of the films synthesized in the developed system is shown in the plane passing trough the axes of the magnetron and ICP source.

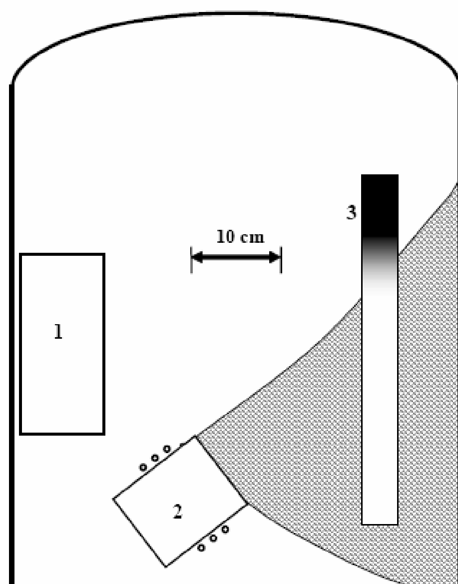


Fig. 7. 2D stoichiometry diagram of the films synthesized using oxygen activation by ICP source: 1 – magnetron, 2 – ICP source, 3 – substrate holder with film transparency distribution shown (stoichiometry region is shaded)

Rys. 7. 2D diagram rozkładu stechiometrii powłok osadzanych z wykorzystaniem aktywacji tlenu w źródle ICP: 1 – magnetron, 2 – źródło ICP, 3 – uchwyt podłoży z zaznaczonym obszarem osadzania powłok transparentnych (zacieniowano obszar stechiometrii powłok)

Deposition processes were performed after pumping the chamber to a base pressure of 10^{-5} mbar or less, and cleaning the substrates during 5–15 min with. Ar activated in ICP discharge at the pressure of $(1–3)\times10^{-3}$ mbar and RF power of 500 W. No bias was applied to the substrates during cleaning and the distance from ICP source to the substrate holder was

approximately 20 cm. The magnetron target to substrate distance was approximately 30 cm for all experiments. Because at low pressure, i.e. less than 3.5×10^{-3} mbar, ion and active particle flows from the source have divergent profile with a cosine-like law, the ICP source was located 10 cm closer to the substrate holder, than the magnetron (see **Fig. 1** and **7**). The parameters of carried out deposition processes are listed in **Table 1**, whereas basic characteristics of the deposited coatings and oblique metallographic sections of the coatings made during calotest thickness measurements are shown in **Table 2** and in the **Fig. 8** respectively.

Table 1. Parameters of Al_2O_3 coatings deposition
Tabela 1. Parametry osadzania powłok Al_2O_3

Sample No	Ar pressure [$\times10^{-3}$ mbar]	O_2 flow rate [sccm]	RF power [W]	Magnetron voltage [V]	Magnetron current [A]	Substrate bias [V]	Process duration [min]
1	3.12	28	500	700	6.5	0	45
2	2.85	26.7	500	775	6	0	45
3	2.84	23.5	500	780	6	-1000	45
4	2.84	23.5	500	780	6	-1000	45
5	2.8	23.5	500	780	6	-500	45
6	2.8	23.5	500	780	6	-500	45

Table 2. Properties of Al_2O_3 coatings
Tabela 2. Właściwości powłok Al_2O_3

Sample No	Underlayer	Thickness [μm]	Roughness Ra/Rz [μm]	Hardness H [GPa]	Young Modulus E [GPa]
1	Al	4.3	0.017/0.199	7.8	197
2	TiN	3.5	0.120/1.477	8.1	209
3	–	2.9	0.037/0.240	7.2	184
4	TiN	2.8	0.130/1.403	8.6	216
5	Al	2.4	0.013/0.143	7.3	180
6	TiN/Al	2.5	0.153/1.653	8.6	213

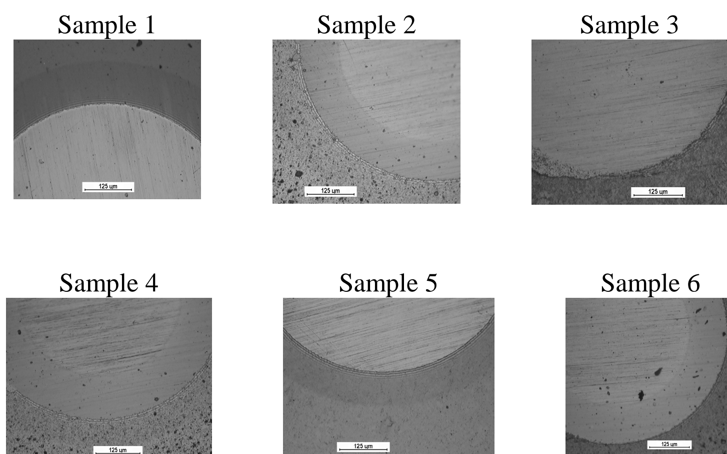


Fig. 8. Cross sections of deposited coatings made during thickness measurement
Rys. 8. Przekroje osadzonych powłok otrzymane w procesie pomiaru grubości

CONCLUSIONS

To produce stoichiometric aluminum oxide films and reliably avoid target poisoning in DC magnetron reactive sputtering processes one of two approaches must be undertaken:

1) to employ sophisticated feedback and control loops in the gas delivery system to maintain the oxygen partial pressure at a precisely specified level, or
2) to use directed source of activated oxygen so that the oxygen partial pressure at the magnetron target is measurably lower than that required to cause poisoning, while the partial pressure and reactivity at the substrate is adequate to produce the films of the desired properties.

The technique, presented in the paper, was shown to be effective in producing high quality aluminum oxide films in a DC reactive sputtering environment without the target poisoning effect. Using this technique, films deposited within the flat section of the target voltage hysteresis curve displayed stable stoichiometry over wide deposition zone with the diameter of about 30 cm. The application of developed deposition system has allowed to expand the ranges of stable deposition parameters: working pressure – $(0.3–3.5)\times10^{-3}$ mbar, magnetron discharge power 1–8 kW, power of ICP source – up to 1 kW, Al_2O_3 coatings deposition rate – up to 8 μm/hour, and to improve essentially the coatings quality.

ACKNOWLEDGEMENTS

The presented work was carried out within the program COST 532 grant M12 supported by the Ministry of Science and Higher Education of Poland.

REFERENCES

1. Dorre E., Hubner H. (Eds.): Alumina, Processing Properties and Applications, Springer, Berlin 1984.
2. Choli T.C., T Nieh.G., McAdams S.D.. Pharr G.M.: Scripta Metal. Mater. 25 (1991) 2203.
3. Ediou S.M., Smajkiewicz A., Al-Jumaily G.A.: Appl. Optics 32 (28) (1993) 5601.
4. Deshpandey C., Bunshah R.F.: Thin Solid Films. 163 (1988) 131.
5. M.Yu, I. Denysenko, S. Dudin, A. Zykov, N. Azarenkov, Phys. Plasmas 9 (2002) 11.
6. Zykov A.V., Dudin S.V., Yakovin S.D., Walkowicz J.: 10th Intl. Conf. on Plasma Physics and Controlled Fusion, Alushta, Ukraine, Book of Abstracts, p. 170, 2004.
7. Safi I.: Surface and Coatings Technology 135 (2000) 48].
8. Schütze A., Quito D.T.: Surface and Coatings Technology 162 (2003) 174.
9. Felmetsger V., Laptev P.: Vacuum 74 (2004) 403.
10. Sproul W.D., Christie D.J., Carter D.C.: Thin Solid Films 491 (2005) 1.

Recenzent:
Tadeusz BURAKOWSKI

Streszczenie

W artykule przedstawione są wyniki badań stałoprądowego magnetronowego układu rozpylającego do osadzania powłok tlenku aluminium. W prezentowanym układzie, w odróżnieniu od standardowych magnetronowych układów osadzania wykorzystujących tlen cząsteczkowy w podstawowym stanie energetycznym, do komory procesowej dostarczany był tlen wzbudzany do wyższych poziomów energetycznych w niezależnym źródle z indukcyjnym wyładowaniem RF. Zbadane zostały zmiany charakterystyk procesu osadzania, ze wzbudzaniem i bez wzbudzania składników reaktywnych.