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Influence of the method of gas supply to the chamber on the processes of reactive magnetron sputtering of Ti-Al composite target

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The paper demonstrates study results of the processes of reactive magnetron sputtering of a Ti-Al composite target in an Ar/O_2 gas mixture with various methods of gas supply to the chamber. It has been established that, regardless of the method of gas supply, when changing the oxygen concentration in the Ar/O_2 gas mixture, the ratio of the Al and Ti content in the deposited $Ti_{1-x}Al_xO_y$ films changes. The metal content ratio in the deposited films varies in proportion to the change in the ratio of the control lines intensities of the optical emission of aluminum AlI (396.15 nm) and titanium TiI (395.82 nm) in plasma. This makes it possible to effectively use the method of optical emission spectroscopy to predict and control the content of metals in deposited films. Based on studies of the dielectric characteristics of deposited $Ti_{1-x}Al_xO_y$ films, it was discovered that with separate gas supply (Ar is supplied to the target region, O_2 is supplied to the substrate region), in contrast to the joint gas supply (the Ar/O_2 gas mixture is supplied to the target region), it is possible to deposit dielectric films in the transient mode at relatively high sputtering rates. In this case, the oxygen supply to the substrate region makes it possible to increase the oxygen content in the films up to 60−64%.

Keywords: preactive magnetron sputtering, composite target, thin films, titanium-aluminum oxide, elemental composition, permittivity, dielectric loss tangent.

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Introduction

One of the urgent problems of modern microelectronics is the introduction of new materials with special properties into technological processes [1–6]. The group of such materials includes complex multicomponent oxides, which, depending on the composition and structure, have unique properties. Examples are wide-gap oxide semiconductors, high-temperature superconductors, ferroelectrics, ferromagnets, solid electrolytes [3]. Currently we begin such materials use in modern integrated circuits as functional layers of memory elements, metal-oxide-semiconductor (MOS) transistors, integrated sensors and power supplies, capacitor and resistive structures, as well as diffusion-barrier and coating layers [1,4,5].

The functional properties of complex oxide films now are mainly improved due to the expansion of their spectrum and the complication of the composition [2]. However, practice shows that the properties of such materials are extremely sensitive to the elemental composition and structural arrangement. This imposes stringent requirements on the process of formation of complex oxide films. The deposition method should provide an accurate and reproducible from process to process repetition of the structure, elemental and phase composition of the applied layers. In this case, the thickness of individual layers must be maintained with an accuracy of several nanometers.

For the formation of films of complex oxides, the method of reactive magnetron sputtering of composite (mosaic) targets has great prospects [7], i.e. targets consisting of a matrix of one metal with inserts of other metals. The method makes it possible to obtain multicomponent films with an arbitrary number and content of elements using a single magnetron, provides high film deposition rates, their high chemical purity, density, and adhesion to the substrate. However, during the reactive magnetron sputtering of films of binary compounds the elemental composition of the sputtered films has a strong dependence on the partial pressure of the reactive gas, which is associated with the formation of a compound film on the target surface [8]. Therefore, most often during reactive sputtering, films are applied in the „oxide“ state of the target (high oxygen concentrations) [9,10]. In this case, the mixture of gases (Ar/O_2) is fed into the chamber or directly into the discharge zone. Operation in the oxide state allows, in some cases, to get rid of the process instability. However, in this mode it is possible to form films at low deposition rates only. During reactive sputtering of composite targets, the processes in the discharge become more complicated, and the composition of the deposited films should depend not only on the rate of sputtering of each of the target components, but also on the rate of their oxidation. This requires precise control of the sputtering process.

Improving the controllability of the reactive sputtering and the composition of deposited films during sputtering of composite targets is possible by reducing the effect of reactive gas (oxygen) on the processes of compound formation on the target surface. This can be achieved by reducing the partial pressure of the reactive gas in the sputtering zone. Such effect can be obtained using high-vacuum magnetron sputtering [11] and/or spatial separation of sputtering and film oxidation processes due to separate gas supply. With separate gas supply, the target is sputtered in inert gas (Ar), which is supplied directly to the magnetron discharge region, and the reactive gas is supplied to the substrate region, where the sputtered flow condenses, and the compound film is formed. However, there are practically no publications that would analyze the features of the formation of multicomponent thin films using various methods of supplying work gases in the chamber and their advantages.

Thus, the purpose of this paper was to study the processes of reactive magnetron sputtering of a Ti-Al of composite target in Ar/O_2 work gases and the influence of the method of gas supply into the chamber on the controllability of the sputtering process and properties of deposited titanium-aluminum oxide films.

1. Experiment

The study of the processes of reactive magnetron sputtering of the composite target was carried out at the FERO-001 unit for magnetron sputtering of thin films. Experimental unit scheme is presented in Fig. 1. The unit was made on the basis of the vacuum station VU-1B. The vacuum unit chamber is equipped with MARS.011-80 magnetron sputtering system with target Ø 80 mm using ion source based on EHPM.100.005 end-face Hall accelerator, and a rotatable substrate holder. The magnetron had its own gas distributor, which was integrated into the cathode assembly and injected gas directly into the magnetron discharge zone. The rotatable substrate holder was located at a distance of 120 mm from the target surface. To increase the uniformity of the thickness of the deposited films the axis of the substrate holder was shifted relative to the magnetron axis by 100 mm. The substrate rotation speed was 24.5 rpm. An additional ring gas distributor was installed along the perimeter of the substrate holder. This made it possible to supply reactive gas (O_2) directly to the substrate region.

During the experiments two methods of supplying work gases to the chamber were used: 1. Joint gas supply. The mixture of gases (Ar/O_2) was supplied to the gas distribution system of the magnetron (Joint Gas Supply or JGS);

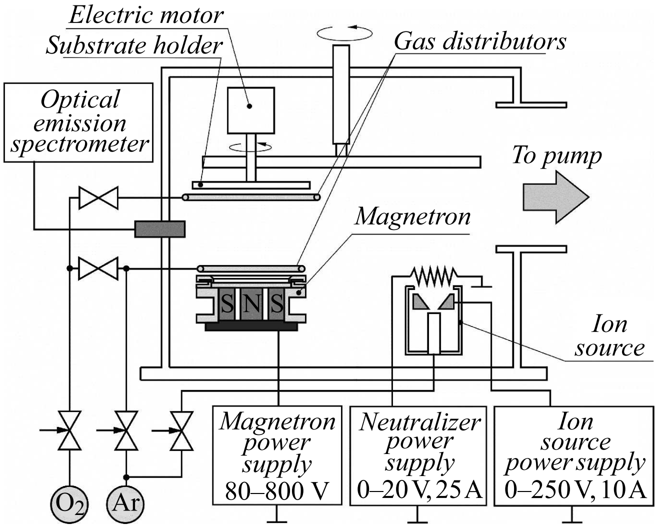

Figure 1. Scheme of experimental unit for magnetron sputtering of thin films FERO-001.

2. Separate gas supply. The inert gas (Ar) was supplied to the gas distribution system of the magnetron, and the reactive gas (O_2) was supplied to the gas distributor in the substrate region (Separate Gas Supply or SGS).

The consumption of work gases was controlled using automatic gas flow regulators RRG-1. A Ti-Al composite target Ø 80 mm and 5 mm thick was used for sputtering. The target consisted of three parts: a central Al insert 18.5 mm wide and two side Ti segments. At that, the dimensions of the Al inserts were chosen so that the Al/Ti ratio in the deposited films, calculated according to the method proposed in the article [12], was 4:6.

Titanium-aluminum oxide films were deposited on substrates of highly doped single-crystal silicon EKES 0.01 (100). During the experiments the substrates were placed on the substrate holder. The chamber of the vacuum unit was evacuated to a residual pressure of 10^{-3} Pa, and the substrates were ionically cleaned. To do this, the substrate was fed to the cleaning zone, and Ar was supplied to the ion source. The argon flow was $Q_{\mathrm{Ar}} = 30$ sccm. The cleaning time, voltage and discharge current in all experiments were constant and amounted to 6 min, 80 V, 7.0 A, respectively. Then the substrate was brought into the sputtering zone, and the films were sputtered. The sputtering of Ti-Al composite target was made by the method of pulsed reactive magnetron sputtering in Ar/O_2 gases. The oxygen content in the chamber (Γ_{O_2}) varied from 0 to 90% at a total flow rate of 60 sccm. At that pressure in the chamber was 0.08 Pa. The magnetron was powered by APEL-M-5PDC-800A-2 power supply (LLC „Applied Electronics“, Russia) with power 5.0 kW. The magnetron was powered by a unipolar pulsed current (pulse repetition rate $F = 10$ kHz, duty cycle $D = 80\%$). The magnetron discharge current in

Spectral lines used to control the plasma composition during reactive magnetron sputtering of Ti-Al targets

Line	λ, nm	Transition type
AlI	396.15	$3s^24s-3s^23p$
TiI	395.82	$3d^3(^4\mathrm{F})4p-3d^24s^2$
ArI	750.39	$3s^23p^5(^2\mathrm{P}^\circ_{1/2})4p-3s^23p^5(^2\mathrm{P}^\circ_{1/2})4s$
OI	777.15	$2s^22p^3(^4\mathrm{S}^\circ)3p-2s^22p^3(^4\mathrm{S}^\circ)3s$

all experiments was maintained constant and was equal to $I_t = 1.5\,\mathrm{A}$. In this case, the discharge voltage (U_t), depending on the oxygen concentration and the method of gas supply, varied from 300 to 520 V. The thickness of the deposited films was about 100 nm and was controlled by the sputtering time.

To control the process of reactive magnetron sputtering the method of optical emission spectroscopy (OES) of magnetron discharge plasma was used. The optical spectra of plasma radiation were recorded using *S150 Duo* spectrum analyzer (LLC „SOL Instruments", Republic of Belarus). This spectrometer made it possible to record spectra in the range 290–865 nm with spectral resolution of less than 0.15 nm and an absolute error on the wavelength scale of ±0.06 nm maximum. To record the emission spectra, the optical emission spectrometer was connected to the camera with a light guide, and the tip of the light guide was directed to the plasma through a viewing window. Plasma emission lines were identified using Tables of spectral lines [13].

The thickness of the deposited layers was determined using an optical interferometric profilometer POI-08. The elemental composition of the deposited films was analyzed by energy dispersive X-ray spectroscopy (EDX) using Hitachi S-4800 high-resolution field emission scanning electron microscope with Bruker Quantax 200 EDX spectrometer. The distribution of the elemental composition of the coatings through the thickness was studied by atomic emission spectroscopy (AES) in optical-glow discharge spectrometer GD Profiler 2 (HORIBA Jobin Yvon SAS).

The dielectric characteristics of deposited titanium–aluminum oxide films were measured on test MOS structures. To do this, the upper Ni electrode was deposited on the oxide film by ion-beam sputtering through a mask. The area of the capacitor plate was 0.096 mm^2. The capacitance and dielectric loss tangent of the films were measured using E7-20 immittance meter at frequencies of 1.0kHz and 1.0MHz. The permittivity values were calculated based on the thickness of the dielectric layer and the capacitance of the capacitor structure.

2. Results and discussion

The discharge characteristics of the magnetron were studied during reactive sputtering of Ti-Al composite target. Fig. 2 shows the dependences of the magnetron discharge voltage on the oxygen concentration in the Ar/O$_2$ mixture of gases for joint (JGS) and separate (SGS) supply of work gases to the chamber. The dependences were obtained in the discharge current stabilization mode $I_t = 1.5\,\mathrm{A}$. When the target was sputtered in the Ar medium, the magnetron discharge voltage U_t was 345 V. In case of joint gas supply and oxygen addition (Fig. 2, JGS curve), the discharge voltage increased and reached a maximum $U_t = 383\,\mathrm{V}$ at $\Gamma_{\mathrm{O}_2} = 10\%$. With a further increase in the oxygen concentration, the discharge voltage initially sharply decreased to 345 V at $\Gamma_{\mathrm{O}_2} = 20.8\%$, and then a slow decrease was observed to 328 V at $\Gamma_{\mathrm{O}_2} = 70.8\%$. At oxygen concentrations above 40%, the discharge voltage was practically independent of Γ_{O_2}. At Γ_{O_2} over 72% the magnetron discharge became unstable due to formation of a large number of microarcs on the target surface.

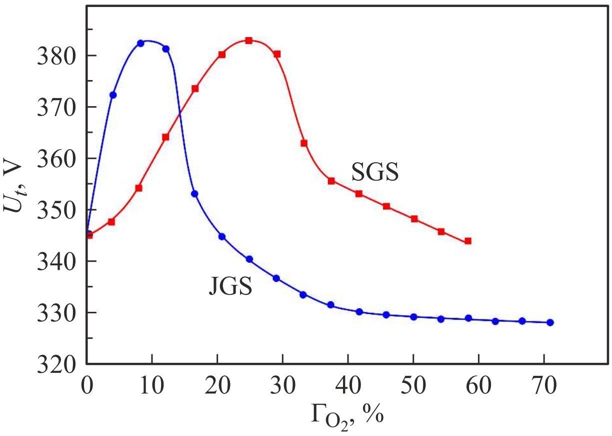

Figure 2. Magnetron discharge voltage vs. oxygen concentration in the Ar/O$_2$ mixture of gases for joint (JGS) and separate (SGS) supply of work gases to the chamber.

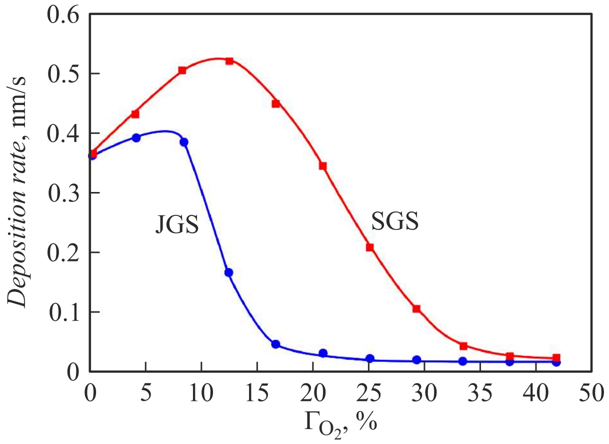

Figure 3. Deposition rate of titanium-aluminum oxide films vs. oxygen concentration in the Ar/O$_2$ mixture of gases at joint (JGS) and separate (SGS) supply of work gases to the chamber.

During separate gas supply (Fig. 2, SGS curve), the shape of the $U_t(\Gamma_{\mathrm{O}_2})$ curve did not change, but the extrema shifted to higher oxygen concentrations in the chamber. The

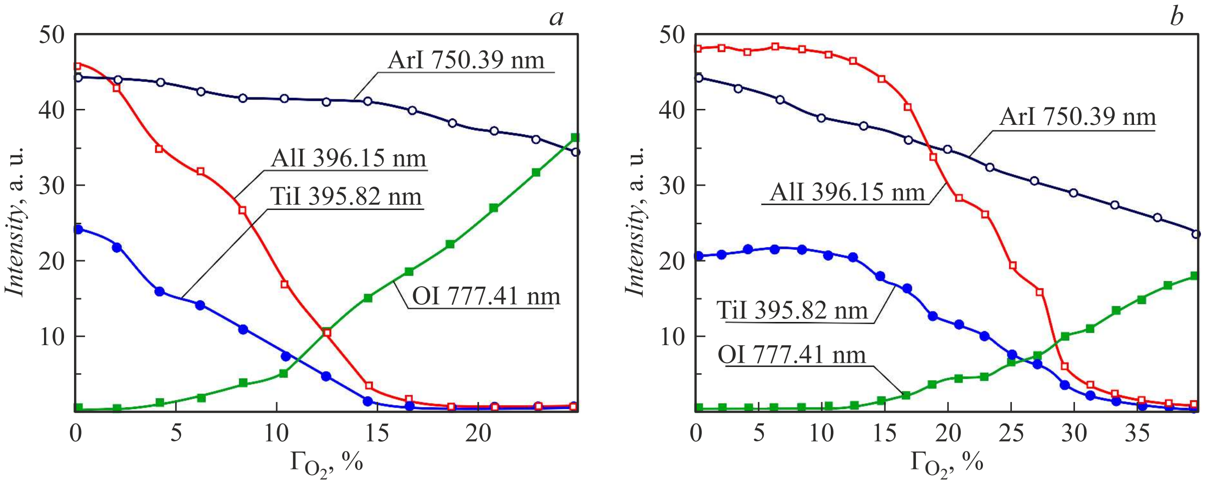

Figure 4. Dependences of the control lines intensity of TiI, AlI, ArI and OI of plasma emission on the oxygen concentration in the Ar/O_2 mixture of gases during joint (*a*) and separate (*b*) gas supply.

maximum voltage was observed at oxygen concentrations of 25%, and the transition to the region with a slow voltage drop was at Γ_{O_2} over 37.5%. The magnetron discharge stably existed at oxygen concentrations up to 59%.

The film deposition rate also depended on the gas supply method. When the Ti-Al target was sputtered in the Ar medium, the deposition rate was 0.37 nm/s. During joint gas supply and addition of oxygen (Fig. 3, curve JGS), the deposition rate initially increased to 0.4 nm/s at $\Gamma_{O_2} = 7\%$ and then sharply decreased to 0.03 nm/s at $\Gamma_{O_2} = 16.7\%$. With a further increase in Γ_{O_2}, the deposition rate was almost independent of the oxygen concentration. During separate gas supply (Fig. 3, curve SGS) and increase in the oxygen flow, the deposition rate increased to 0.52 nm/s, and the maximum deposition rate was observed at $\Gamma_{O_2} = 12\%$. The transition to plateau of the curve with low deposition rate occurred at oxygen concentrations above 40%.

OES studies of the emission spectra of plasma during sputtering of Ti-Al target in Ar medium showed that the spectrum contained lines of excited argon atoms ArI, which were in the wavelength range 690−1050 nm, of excited titanium atoms TiI — in the wavelength range of 390−530 nm, of titanium ions TiII — in the wavelength range of 320−380 nm and a double line of excited aluminum atoms AlI — at wavelengths of 394.40 and 396.15 nm. When oxygen is added and the target is sputtered in Ar/O_2 medium of work gases, triple lines of excited oxygen atoms OI at wavelengths of 777.1−777.6 nm and 844.63−844.64 nm were identified in the spectrum. When analyzing the sputtering processes of the Ti-Al target for the sputtered metals and work gases, the most intense lines of aluminum AlI (396.15 nm), titanium TiI (395.82 nm), argon ArI (750.39 nm) and oxygen OI (777.15 nm) (see table) were selected as control lines.

Studies of the dependences of the intensity of the control lines of plasma emission on the oxygen concentration in the chamber showed that, regardless of the method of gas supply, with increase in Γ_{O_2} the lines intensity of titanium I_{Ti}, aluminum I_{Al}, argon I_{Ar} decreased, and the line intensity of the oxygen I_{O} increased (Fig. 4, *a*). During joint gas supply the decrease in the line intensity of the titanium and aluminum was observed even with a small addition of oxygen, and their intensity decreased almost linearly to $\Gamma_{O_2} = 16.7\%$. At high oxygen concentrations the Ti and Al lines had low intensity, which did not depend on the oxygen concentration, and at Γ_{O_2} over 30% the lines could not be identified at the noise level. Since argon does not participate in chemical reactions, the intensity of the I_{Ar} line varied in proportion to the Ar concentration in the chamber. Two regions could be distinguished in the dependence of the oxygen line intensity on Γ_{O_2}. In the first region from 0 to 10% the intensity of the OI line slowly increased, which is due to the high rate of oxygen absorption by the target surface and the growing film. In the second region (over 10% of O_2), the line intensity increased almost in proportion to Γ_{O_2}.

Unlike joint gas supply during separate gas supply (Fig. 4, *b*) and low oxygen concentrations (up to 12% O_2), the intensity of the OI line was close to zero, and the intensity of the TiI and AlI lines remained practically unchanged, which is associated with low oxygen concentration in the discharge zone and the absence of target oxidation processes. At high oxygen concentrations the intensity of I_{Ti} and I_{Al} Ti lines decreased almost linearly lines, and at Γ_{O_2} over 40% the lines had low intensity, which did not depend on the oxygen concentration.

AES studies of the elemental composition of deposited films $Ti_{1-x}Al_xO_y$ showed that the depth distribution of elements in all samples was uniform (Fig. 5). The increased oxygen content was observed in surface layers with thickness of 5−10 nm only, which is associated with the oxygen adsorption from the atmosphere and the film oxidation. The comparison of the analysis results of the elemental composition of the films by AES and EDX

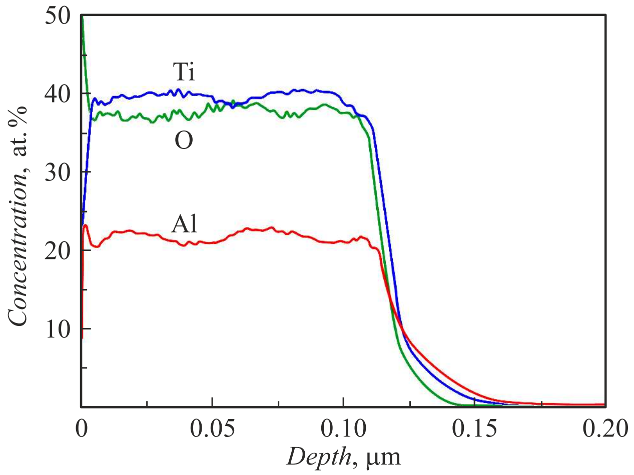

Figure 5. Depth distribution of the elemental composition of the titanium-aluminum oxide film (joint gas supply, $\Gamma_{O_2} = 10.4\%$).

methods showed that both methods had the same results for the relative content of metals in the deposited film. However, the EDX method gave higher oxygen content in the film, which, apparently is due to the fact that the EDX method determines the total integral concentration of elements, and the surface layers of the films had increased oxygen concentration.

Using the results of AES-analysis, dependences of the atomic content of elements in $Ti_{1-x}Al_xO_y$ films on the oxygen concentration in Ar/O_2 gas mixtures (Fig. 6) were plotted. Both during joint and separate gas supply the sharp increase in the oxygen content in the films (C_O) was observed at oxygen concentration in the Ar/O_2 mixture of gases up to 7%. At the same time, C_O reached 40%. With joint gas supply and further increase in Γ_{O_2} the oxygen content in the films increased to 48% and then almost did not depend on the oxygen concentration in the Ar/O_2 mixture of gases. During separate gas supply the oxygen concentration in the films continued increasing and reached 64% at $\Gamma_{O_2} = 41.7\%$. Thus, we can conclude that the oxygen supply to the film growth region during separate gas supply contributes to increase in the oxygen content in the films.

When applying films of complex oxides, the concentration of metals in the film is of particular importance. Fig. 7 shows ratio of the atomic content of metals C_{Al}/C_{Ti} in $Ti_{1-x}Al_xO\mathit{films}_y$ vs. oxygen concentration in the Ar/O_2 gas mixture. As can be seen from the Figure, in both cases with change in the oxygen concentration in the Ar/O_2 mixture of gases the ratio of the metal content in the film changed; at that regardless of the method of gas supply in curve minimum content of aluminium was observed which during joint and separate gas supply was $\Gamma_{O_2} = 12.5$ and 30%, respectively. At the same time, the ratio C_{Al}/C_{Ti} decreased from 0.52 to 0.36.

This decrease in the aluminum content is associated with the formation of oxide films on the surface of the Ti and Al parts of the target and the difference in the oxidation rates of these metals. The oxidizing activity of metals can be estimated from the Gibbs free energy of oxide formation ΔG^0. The more negative the value of ΔG^0 is, the more favorable the course of such a reaction is, and oxide formation will be observed at lower partial pressure of the reactive gas [14]. Thus, the value of the Gibbs free energy of formation of aluminum oxides Al_2O_3 and titanium TiO_2 is respectively -1582 kJ/mol and -943 kJ/mol [15]. Therefore, the course of the reaction of aluminum oxide formation is more favorable compared to titanium oxide, and with increase in the partial pressure of oxygen in the chamber, Al is first oxidized, and oxide film is formed on the aluminum part of the target. In this case, the aluminum sputtering rate decreases, and a lack of aluminum is observed in the films. With a further increase in Γ_{O_2}, oxidized aluminum practically does not absorb oxygen, and intensive oxidation of the Ti part of the target begins. At high oxygen concentrations, the change in ratio C_{Al}/C_{Ti} is determined by the sputtering coefficients of titanium and aluminum oxides. This assumption was confirmed by the results of the OES. Fig. 7 (curves calc) shows the calculated ratio C_{Al}/C_{Ti} obtained from the analysis of the intensities of the control lines of aluminum and titanium in plasma. The ratio of the metal content in the film was calculated by the method proposed in the paper [16], where it was shown that the ratio of the atomic content of aluminum and titanium in the film depends linearly on the ratio of the intensities of the control lines of aluminum and titanium in the plasma, and the dependence is described by linear function

$$\frac{C_{Al}}{C_{Ti}} = 0.221\,\frac{I_{Al}}{I_{Ti}}.$$

As can be seen from Fig. 7, the ratio of the metal content in the films changed in proportion to the change in the ratio of the aluminum and titanium line intensities I_{Al}/I_{Ti} in the plasma. This indicates that the metal content in the deposited film during reactive sputtering of the composite target is decisively affected by the processes of oxidation and sputtering of the target.

The influence of the gas supply method on the dielectric characteristics of deposited $Ti_{1-x}Al_xO_y$ films was also studied. Figs. 8 and 9 show permittivity (a) and the dielectric loss tangent (b) of titanium-aluminum oxide films versus oxygen concentration in chamber. Both during separate and joint gas supply, the formation of dielectric films occurred at a certain oxygen concentration (at Γ_{O_2} over 12.5% during joint and 25% during separate gas supply). However, such films were characterized by high dielectric losses at low frequencies (1 kHz). With a further increase in Γ_{O_2}, decrease of ε and $\mathrm{tg}\,\varphi$ occurred both at high and low frequencies, and the films deposited during separate gas supply had higher values of permittivity $\varepsilon = 18-25.2$ at frequency of 1 MHz and lower values of dielectric losses $\mathrm{tg}\,\varphi = 0.011-0.02$ at frequency of 1 kHz compared to films deposited during joint gas supply. The increase in the permittivity and the decrease in dielectric losses during

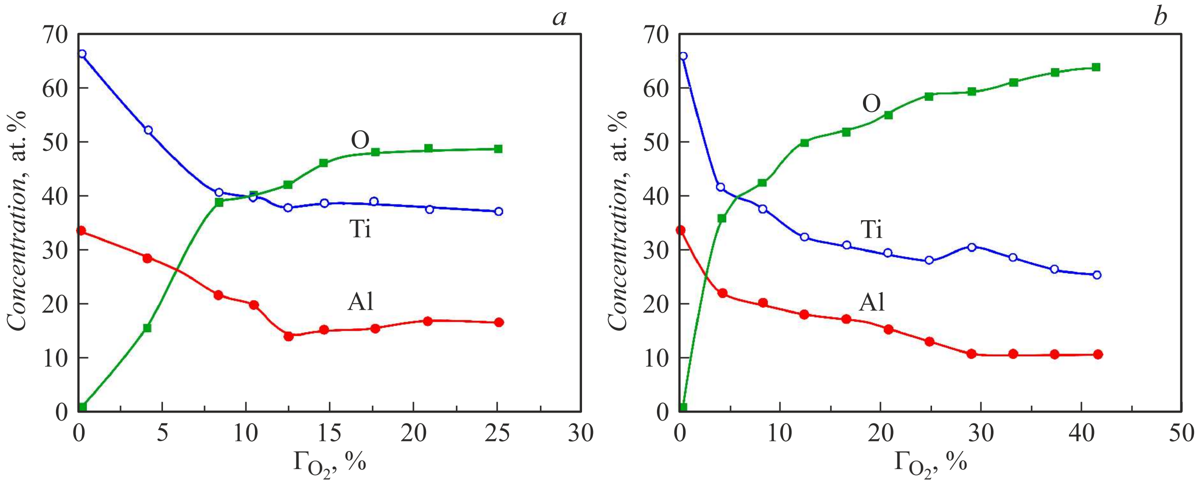

Figure 6. Atomic content of elements in deposited $Ti_{1-x}Al_xO_y$ films vs. oxygen concentration in Ar/O_2 gas mixture during joint (a) and separate (b) gas supply.

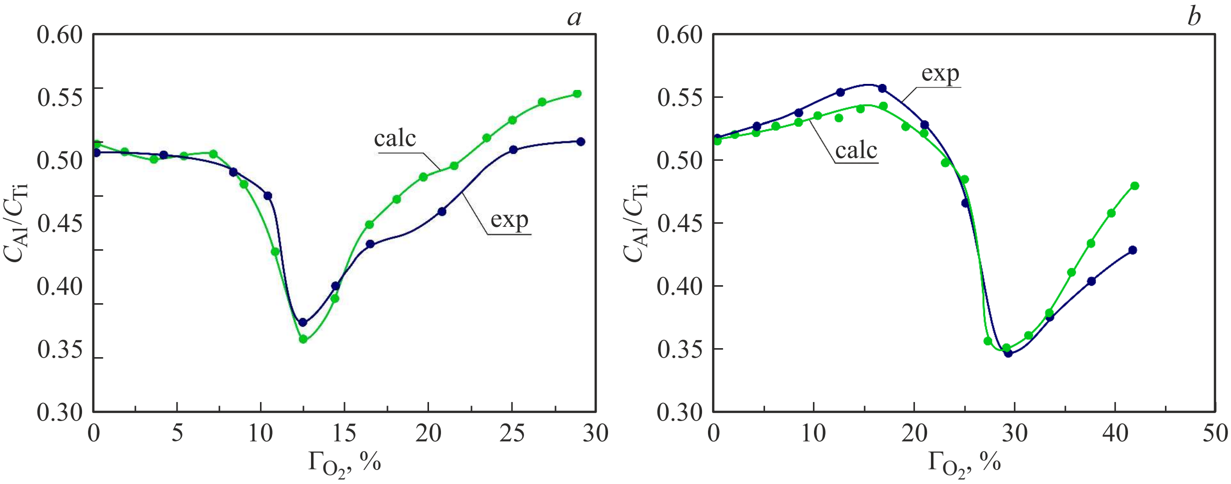

Figure 7. Experimental (exp) and calculated (calc) ratio of atomic content of metals in the deposited films vs. oxygen concentration in the chamber during joint (a) and separate (b) work gases supply into the chamber.

separate gas supply, apparently, are associated with a lower aluminum concentration in the films and a more complete oxidation of titanium and aluminum (Fig. 6, b and 7, b). Note that the dependence $\varepsilon(\Gamma_{O_2})$ with separate gas supply at frequency of 1 MHz exhibited a maximum permittivity $\varepsilon = 25$ at $\Gamma_{O_2} = 29\%$, which coincided with the minimum aluminum concentration.

The analysis of the obtained results allows us to conclude that during reactive magnetron sputtering, the content of elements in the deposited films (both metals and oxygen) depends on the oxygen concentration in the chamber and the gas supply method. As Γ_{O_2} changes, the ratio of aluminum and titanium in the films changes from 0.36 to 0.55. Assuming that $Ti_{1-x}Al_xO_y$ films are a mixture of saturated oxides TiO_2 and Al_2O_3, then the oxygen content in the films at this ratio of metals should vary from 64.57 to 65.16 at.%. However, during joint gas supply the maximum oxygen content in the films, even at high oxygen concentrations in the Ar/O_2 gas mixture, was less than 50 at.%. During separate gas supply the oxygen content in the films reached 60–64 at.%, i.e. films of almost saturated oxides were formed.

The formation of $Ti_{1-x}Al_xO_y$ dielectric films with low dielectric losses was observed at Γ_{O_2} over 16% during joint gas supply and 27% during separate gas supply. These oxygen concentrations correspond to the initial region of a sharp drop in the discharge voltage. However, during joint gas supply and Γ_{O_2} over 16%, a reactive mode of the sputtering is observed, which is characterized by low film deposition rate (0.02–0.04 nm/s) due to the target surface oxidation. During separate gas supply the deposition of dielectric films with low losses is possible in the transient mode of the process, when the target is partially covered with oxide, and the deposition rates are of about

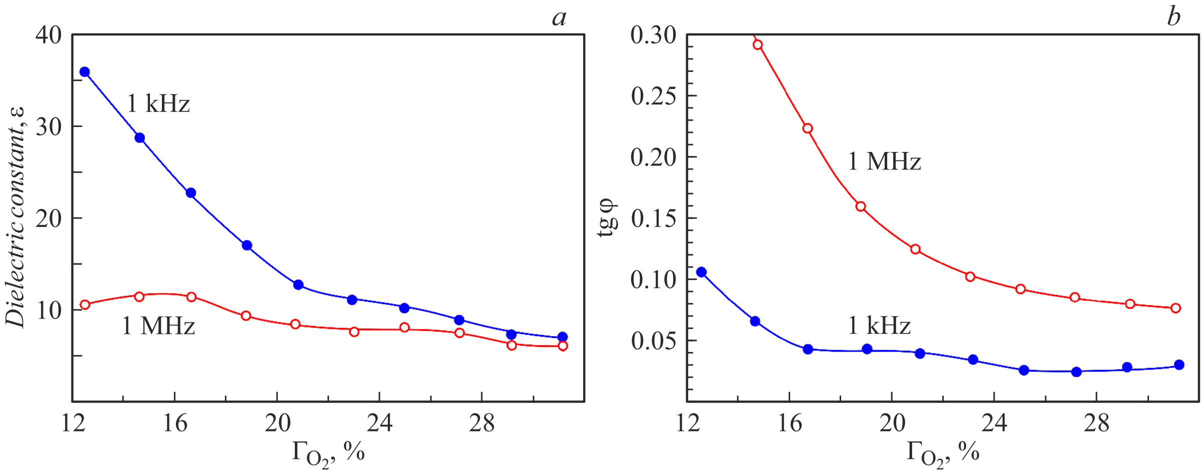

Figure 8. Permittivity (*a*) and the dielectric loss tangent (*b*) of titanium-aluminum oxide films vs. oxygen concentration in Ar/O_2 mixtures of gases during joint gas supply.

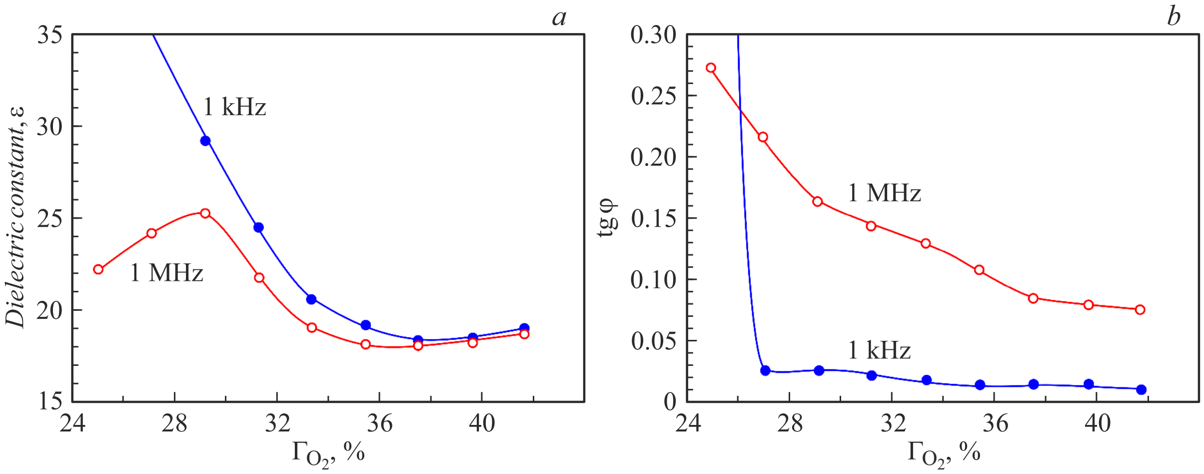

Figure 9. Permittivity (*a*) and the dielectric loss tangent (*b*) of titanium-aluminum oxide films vs. oxygen concentration in Ar/O_2 mixtures of gases during separate gas supply.

0.05−0.15 nm/s. In this case, due to increase in the oxygen concentration in the region of the substrate the films with higher oxygen content are formed. The change in the content of metals in the deposited films with change in the oxygen concentration in the chamber complicates the process of depositing films of complex oxides by the method of reactive sputtering of composite targets. However, the use of OES method to control the process makes it possible to predict the composition of deposited films with sufficient accuracy.

Conclusion

As a result of studies of the magnetron sputtering processes of Ti-Al-target in Ar/O_2 gas mixtures and the characteristics of deposited $Ti_{1-x}Al_xO_y$ films it was found that during reactive sputtering of the composite target, the content of elements in the deposited films (both metals and oxygen) depends on the oxygen concentration in the chamber and the gas supply method. The change in the metal content in the film is due to the difference in the oxidation rates of the target metals and the formation of oxide film on the surface of each of the target parts. The ratio of the metal content in the deposited films changes in proportion to the change in the ratio of the aluminum and titanium line intensities $I_{\mathrm{Al}}/I_{\mathrm{Ti}}$ in the plasma. This makes it possible to effectively use the OES method to predict and to control the content of metals in deposited films.

The use of separate gas supply makes it possible to deposit $Ti_{1-x}Al_xO_y$ films in the transient sputtering mode at relatively high sputtering rates and contributes to increase in the oxygen content in the films up to 60−64 at.% due to decrease in the partial pressure O_2 in the region of the magnetron discharge and its increase in the region of film growth. During separate gas supply and oxygen concentration in the Ar/O_2 gas mixture over 27%, titanium-aluminum oxide films with permittivity $\varepsilon = 18-25.2$ at frequency of 1 MHz and dielectric loss tangent $\mathrm{tg}\,\varphi = 0.011-0.02$ at frequency of 1 kHz were obtained. These oxygen concentrations correspond to the initial region of a sharp drop in the discharge voltage.

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Conflict of interest

The authors declare that they have no conflict of interest.

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