

## Comparative analysis of electrophysical properties of ceramic tantalum pentoxide coatings, deposited by electron beam evaporation and magnetron sputtering methods.

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**Abstract.** Ta<sub>2</sub>O<sub>5</sub> ceramic coatings have been deposited on glass substrates by e-beam evaporation and magnetron sputtering methods. For the magnetron sputtering process Ta target was used. X-ray diffraction measurements show that these coatings are amorphous. XPS survey spectra of the ceramic Ta<sub>2</sub>O<sub>5</sub> coatings were obtained. All spectra consist of well-defined XPS lines of Ta 4f, 4d, 4p and 4s; O 1s; C 1s. Ta 4f doublets are typical for Ta<sub>2</sub>O<sub>5</sub> coatings with two main peaks. Scanning electron microscopy and atomic force microscopy images of the e-beam evaporated and magnetron sputtered Ta<sub>2</sub>O<sub>5</sub> ceramic coatings have revealed a relatively flat surface with no cracks. The dielectric properties of the tantalum pentoxide coatings have been investigated in the frequency range of 100 Hz to 1 MHz. The electrical behaviour of e-beam evaporated and magnetron sputtered Ta<sub>2</sub>O<sub>5</sub> ceramic coatings have also been compared. The deposition process conditions principally effect the structure parameters and electrical properties of Ta<sub>2</sub>O<sub>5</sub> ceramic coatings. The coatings deposited by different methods demonstrate the range of dielectric parameters due to the structural and stoichiometric composition changes

### 1. Introduction

The interest in dielectric materials and coatings applications has considerably increased in various areas of science and technology in recent years. The high dielectric parameters, band gap and low values of dielectric loss tangent are very important properties for various applications such as electronic devices, thin-film capacitors and biomedicine [1-3]. Dielectric oxides are known for their good optical properties but relatively little has been published on their electrical characteristics [4-6]. Dielectric oxide layers were proposed for double insulator thin-film electroluminescent devices [7]



and a complete EL-device has been made with Ta<sub>2</sub>O<sub>5</sub> insulators [8]. Dielectric coatings with electret properties are applied in medicine for stimulation of positive biological processes by short-range quasi-static electric field actions. Tantalum and tantalum-based compounds are promising for biomedical applications. Ta-based materials demonstrate high fracture toughness, corrosion and wear resistance, and chemical stability. The results of test on implantation of Ta in both soft and hard tissue of rats showed the good biocompatibility and osteogenesis of this metal [9]. TaC and TaN materials possess relatively high hardness due to the covalent nature of their bond [10] and demonstrate high thermal stability and superior corrosion resistance [11]. The blood compatibility of TaN films was shown to be better than that of Ta [12]. The deposition and treatment processes may change the coating's structure from amorphous one (for as-deposited films) to an orthorhombic phase, the surface topography, from smooth to nano-crystalline and improve the crystallinity of the tantalum pentoxide coatings [13]. The research of the dielectric properties of Ta<sub>2</sub>O<sub>5</sub> ceramic coatings deposited on glass substrates by means of e-beam evaporation and magnetron sputtering methods was presented.

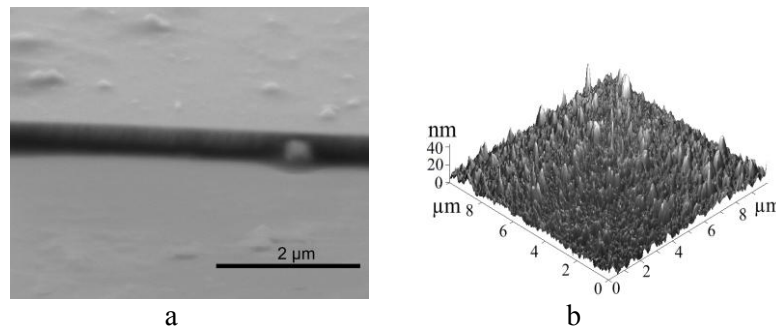
## 2. Materials and methods

The study of Ta<sub>2</sub>O<sub>5</sub> ceramic coatings structure and properties which effect on electrophysical parameters was performed. The substrates were glass samples. The main parameters of the process were described in our previous study [14]. The evaporation process was carried out at initial vacuum of  $9.3 \times 10^{-4}$  Pa, operational vacuum of  $4 \times 10^{-3}$  Pa, anode current of 50 mA and calculated evaporation power of 350 W. The deposition rate under these conditions was 30 nm/min. For magnetron sputtering process Ta target was used. The substrates were displaced far from the magnetron axes to avoid bombardment with ions accelerated by the static electric field. The operating gas was chosen between O<sub>2</sub> (ensuring the highest possible degree of oxidation, but having the lowest deposition rate), Ar (ensuring the highest deposition rate, but the oxidation is only via residual gas of the vacuum system – high vacuum – not UHV) and mixture of 50% O<sub>2</sub> and 50% Ar – the intermediate case. Typical operating pressure was about 4 Pa and operating power was 80 W. The bias at such conditions was about 300 V. The e-beam evaporated (EB) and magnetron sputtered (MS) Ta<sub>2</sub>O<sub>5</sub> ceramic coatings structure and composition were investigated by means of photoelectron spectroscopy XPS and X-ray diffraction XRD methods. X-ray diffraction profiles of Ta<sub>2</sub>O<sub>5</sub> were observed by means of diffraction device “DRON-3” with filtered Cu-K $\alpha$  radiation. X-ray photoelectron measurements have been carried out on the ESCALAB MkII (VG Scientific) electron spectrometer at a base pressure in the analysis chamber of  $5 \times 10^{-8}$  Pa using anode AlK $\alpha$  X-ray source with excitation energies of 1486.6 eV (160 W). The spectra are recorded at the total instrumental resolution (as it was measured with the FWHM of Ag3d<sub>5/2</sub> photoelectron line) of 1.18 eV for AlK $\alpha$  excitation source. The energy scale has been calibrated by normalizing the C1s line of adsorbed adventitious hydrocarbons to 285.0 eV. The processing of the measured spectra includes a subtraction of X-ray satellites and Shirley-type background. The peak positions and areas are evaluated by a symmetrical Gaussian-Lorentzian curve fitting. The relative concentrations of the different chemical species are determined based on normalization of the peak areas to their photo ionization cross-sections, calculated by Scofield [15].

The surface topography and roughness parameters of the tantalum pentoxide ceramic coatings were evaluated by scanning electron microscopy (SEM) and atomic force microscopy (AFM) methods. The thickness parameters were obtained from the SEM cross-section measurements. The analysis of capacity, dielectric constant, parameters of dielectric loss tangent depending on the frequency of electromagnetic field for oxide ceramic coatings deposited in the different sputtering regimes was made by LCR-meter BR-2876. The research of near-surface electric field distributions and surface potentials were made. The correlation of surface characteristics and field distributions was analyzed.

## 3. Results and discussion

The surface properties of e-beam evaporated and magnetron sputtered Ta<sub>2</sub>O<sub>5</sub> coatings were investigated. The SEM surface images show no cracks on the surface of the tantalum pentoxide



**Figure 1.** Surface topography of the e-beam evaporated  $\text{Ta}_2\text{O}_5$  coatings by SEM (a) and AFM (b).

ceramic coatings in a good agreement with Atomic Force Microscopy (AFM) measurements, which show relatively flat surfaces (figure 1).

The structure of  $\text{Ta}_2\text{O}_5$  coatings was investigated by means of the XRD method. X-ray diffraction profiles of as-deposited  $\text{Ta}_2\text{O}_5$  coatings demonstrated an amorphous nature for both e-beam evaporated and magnetron sputtered  $\text{Ta}_2\text{O}_5$  coatings, no peaks were observed. XPS survey spectra of the e-beam evaporated and magnetron sputtered  $\text{Ta}_2\text{O}_5$  films were obtained. All spectra consist of well-defined XPS lines of Ta 4f, 4d, 4p and 4s; O 1s; C 1s. All binding energies of the high-resolution spectra were calibrated with the C 1s binding energy of 285.0 eV. Ta 4f doublets are typical for  $\text{Ta}_2\text{O}_5$  coatings and have two peaks: Ta 4f<sub>7/2</sub> at ~ 26.2 for e-beam evaporated and 26.5 eV for magnetron sputtered layers and Ta 4f<sub>5/2</sub> whose binding energy higher by 1.9 eV. The Ta 4f lines of the films deposited agree well with the Ta 4f doublet representative of the Ta-O bond in  $\text{Ta}_2\text{O}_5$ . The Ta/O ratio estimated from the spectra was in the range 0.41 for magnetron sputtered and 0.37 for e-beam evaporated  $\text{Ta}_2\text{O}_5$  coatings (table 1). The results demonstrate that magnetron-sputtering process improves the surface atomic concentration and the stoichiometry of the  $\text{Ta}_2\text{O}_5$  films (Figure 2). The O1s spectra further support this assumption. The O 1s peaks of the deposited layers are centered at binding energies of 530.8 eV for e-beam evaporated and 530.6 eV for magnetron sputtered coatings.

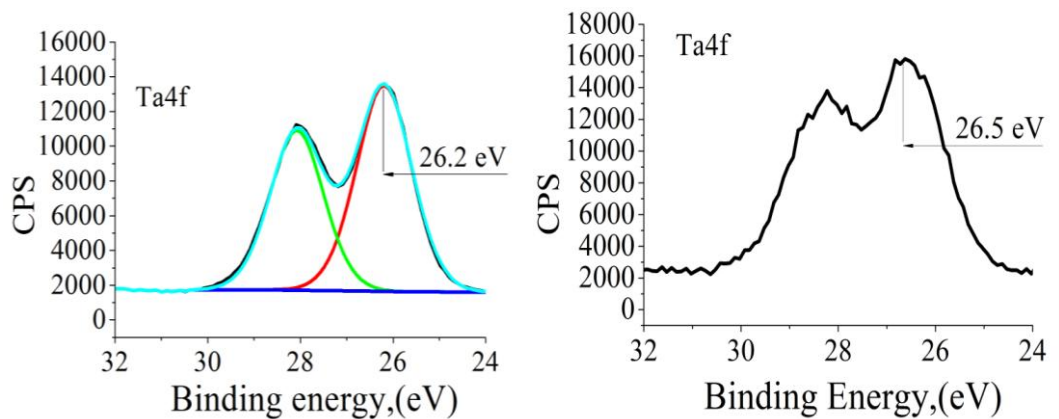
**Table 1.** Surface atomic concentrations of  $\text{Ta}_2\text{O}_5$  coatings (at.%).

Type of coating	XPS spectra lines		
	O1s	Ta4f	Ta4f/ Osl
$\text{Ta}_2\text{O}_5$ MS	7.6	14.2	0.41
$\text{Ta}_2\text{O}_5$ EB	7.4	14.1	0.37

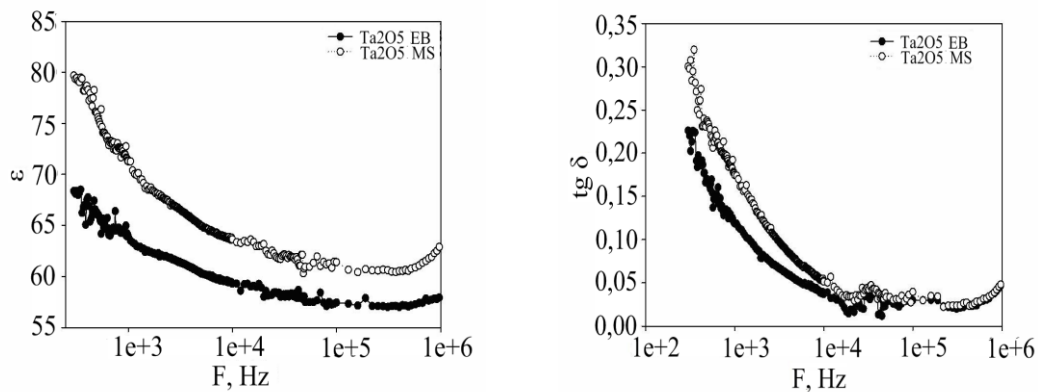
The dielectric properties of tantalum pentoxide coatings were characterized by LCR-meter BR-2876 controlled by computer. Frequencies were chosen in the range from 100 Hz to 1 MHz with an oscillation voltage of 1V. The analysis of capacity, dielectric constant  $\epsilon$  and dielectric loss  $\text{tg}\delta$  measurements as a function of the frequency for e-beam evaporated and magnetron sputtered  $\text{Ta}_2\text{O}_5$  ceramic coatings was made (figure 3).

Figure 3 shows the dielectric constant  $\epsilon$  and the dielectric loss  $\text{tg}\delta$  of e-beam evaporated in comparison with magnetron sputtered  $\text{Ta}_2\text{O}_5$  ceramic coatings. The dielectric constant is calculated on the basis of the formula for a parallel plate capacitor:  $\epsilon = dC/\epsilon_0 S$ , where  $C/S$  is the capacitance per unit area and  $d$  is the thickness of the layer.

The analysis of dielectric constant  $\epsilon$  and dielectric loss  $\text{tg}\delta$  measurements depending on the frequency for e-beam evaporated and magnetron sputtered  $\text{Ta}_2\text{O}_5$  ceramic coatings shows dielectric constant, which are steadily decrease at higher frequencies and losses, which are increasing at lower



**Figure 2.** High-resolution XPS spectra of Ta 4f and composition of the e-beam evaporated (a) and magnetron sputtered (b) Ta<sub>2</sub>O<sub>5</sub> coatings.

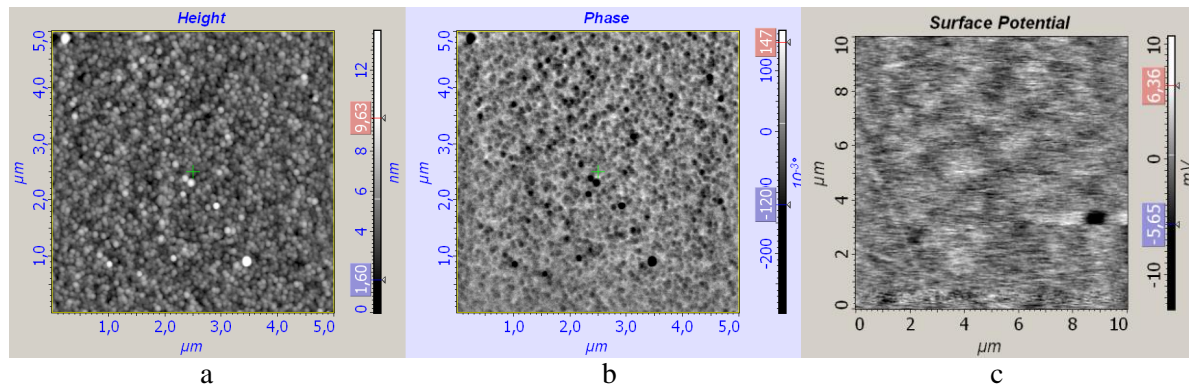


**Figure 3.** Dielectric constant  $\epsilon$  (a) and dielectric loss  $\text{tg}\delta$  (b) measurements as a function of the frequency for e-beam evaporated and magnetron sputtered Ta<sub>2</sub>O<sub>5</sub> ceramic coatings

processes. The coatings demonstrate high dielectric parameters: the dielectric loss  $\text{tg}\delta < 1$  for all Ta<sub>2</sub>O<sub>5</sub> ceramic coatings. The difference in apparent dielectric constant can be explained by the different structure and composition parameters of tantalum pentoxide ceramic coatings deposited by e-beam evaporation and magnetron sputtering methods. A dielectric constant obtained for e-beam evaporated layers are substantially lower than for magnetron sputtered, due to the degradation of the structural properties and the impurities penetration during evaporation process. Electrical properties are structurally sensitive and structural variation leads to significant changes of electrophysical characteristics [13, 16, 17].

The surface was observed by scanning microprobe method, phase contrast of coating's surface scan was analyzed and surface charge distribution by Kelvin probe method was evaluated. The method was used for analysis of electric field and charge distribution on the surface of tantalum pentoxide coatings deposited by different processes. The method realized by two stages scanning of surface relief and probe measurements at resonance frequency with the constant bias between probe and surface 2V. The images were formed as a result of probe oscillation phase changing (figure 4).

The results show that surface relief has no significant differences in the case of e-beam evaporated and magnetron sputtered Ta<sub>2</sub>O<sub>5</sub> ceramic coatings. The surface potential was in the range 3 – 5 mV. The process of deposition changes the electric state of surfaces in the case of both e-beam evaporated and magnetron sputtered coatings. The effect of constant electric fields on the cell's behaviour was previously reported [18]. Osteoblast and osteoclast cells migrate in opposite directions



**Figure 4.** Surface scans of surface relief (a), phase contrast (b) and surface potential (c) of e-beam evaporated Ta<sub>2</sub>O<sub>5</sub> ceramic coatings.

by action of a constant electric field. The variation of the electric field values allows to control the cell response in vitro tests [19]. The changes in the cell membranes potential took place at the potential values about 10 mV. Therefore, the surface potential of Ta<sub>2</sub>O<sub>5</sub> ceramic coatings deposited by both e-beam evaporation and magnetron sputtering methods can effect on further cell/material response on cellular level.

#### 4. Conclusion

The tantalum pentoxide ceramic coatings have been deposited by e-beam evaporation and magnetron sputtering process in the study. The coatings were transparent and amorphous and the surface shown no cracks. The Ta 4f lines of the deposited coatings agree well with the Ta 4f doublet representative of the Ta-O bond in Ta<sub>2</sub>O<sub>5</sub>. The Ta/O ratio estimated from the spectra was in the range 0.41 for magnetron sputtered and 0.37 for e-beam evaporated Ta<sub>2</sub>O<sub>5</sub> coatings. The results demonstrate that magnetron sputtering process improves the surface atomic concentration and the stoichiometry of the Ta<sub>2</sub>O<sub>5</sub> layers. The coatings demonstrate high dielectric parameters: the dielectric loss  $\tan \delta < 1$  for all Ta<sub>2</sub>O<sub>5</sub> ceramic coatings. Electrically the e-beam evaporated tantalum pentoxide coatings have a smaller value of dielectric constants compared with magnetron sputtered. This is probably due to the more perfect structure and composition of the magnetron sputtered coatings. The results show that the electro physical properties are strongly influenced by the coating's deposition parameters. The deposition process conditions principally effect on the structure properties and electrical behavior of the e-beam evaporated and magnetron sputtered Ta<sub>2</sub>O<sub>5</sub> coatings. The coatings deposited by different methods demonstrate the range of dielectric parameters due to the structural and stoichiometric composition changes.

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