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NANOSIZED COLUMNAR MICROSTRUCTURE AND RELATED PROPERTIES OF AMORPHOUS Al_2O_3 THIN FILMS

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Amorphous Al_2O_3 films deposited by electron-gun (e-gun) at different vapour incidence angles with respect to the substrate normal were studied. A nanosized columnar growth morphology was revealed by electron optical imaging. Three different methods were used in order to investigate the dependence of the physical properties of Al_2O_3 films on their microstructure: a Knoop prism indentation for microhardness measurements, optical spectrophotometry for refractive index calculation and a modified pulsed transient heat flow method for evaluation of the thermal diffusivity of the films. It is shown that the microstructural inhomogeneity of the samples strongly influences the properties studied, the values of microhardness, thermal diffusivity and refractive index being decreased with the increase of vapour incidence angle.

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1. Introduction

The aluminum oxide is known as a refractory material with good chemical stability and extremly high hardness. Especially for the alumina thin films these properties are very important for their technological applications in micro- and opto-electronics, sensor techniques and catalysis. A variety of vacuum deposition methods for preparation of Al_2O_3 films are available. It is well known [1] that applying electron gun deposition good quality amorphous alumina thin films could be obtained. However, the preparation conditions determine the growth morphology and microstructure of the films thus changing their physical and chemical properties. For instance, varying only one deposition parameter – the vapour incidence angle, strongly influences the film microhardness, thermal and electrical conductivity as well as the refractive index.

It is the aim of the present work to reveal the growth morphology of e-gun deposited Al_2O_3 thin films prepared under different vapour incidence angles in order to check the relationship between the microstructure and the physical properties of the films.

2. Experimental

The experiments were carried out with 1000-2000 nm thick Al_2O_3 films vapour deposited onto pre-cleaned glass substrate at background pressure of the order of 1×10^{-3} Pa. The samples were obtained by e-beam evaporation of Al_2O_3 tablets (99.99%) at low mean deposited rate - 0.07 nm/s, in order to prevent the thermal decomposition of the target. The substrates were positioned at different vapor incidence angles $\alpha = 0^o, 40^o, 60^o, 70^o$ and 80^o between the collimated vapour beam and substrate normal. A scanning electron microscopy (SEM 515 Philips) is applied for imaging of the growth profiles of the samples. The microhardness (MH) of the alumina thin films was measured by Knoop

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indentation method at different loading providing approximately equal prism indentation size. At least 10 indentations were made for evaluation of each mean MH value.

A modified transient heat flow method was applied in order to determine the thermal diffusivity of the alumina thin films [2]. The samples investigated were sandwiched between vacuum deposited thin planar thermocouple Bi/Sb underneath and photothermal absorbing metal layer (Sb) at the top. The thickness of both, the thermocouple and the metal layer, was about 200 nm. The thermal response of the samples during its heating via Nd:YAG laser pulse (λ=1064 nm, τ=12ns) was detected by the thermocouple, its thermoelectric power being monitored by means of differential recording double oscillograph. A simple relation

$$a=d^2/(2t) \quad (1)$$

between thermal diffusivity coefficient – *a*, the film tickness *d* and the time *t* for measuring the maximal thermoelectric power was used in order to calculate the thermal diffusivity of the Al_2O_3 films.

The optical properties were studied using the sample transmissivity measured by Cary 5 UV-VIS-NIR spectrophotometer in the wavelength range 320-800 nm. The effective refractive index n was evaluated from the minimax envelope of transmission spectra beyond the absorption edge using Svanepoel's method [3] and a computer program [4].

3. Results

All Al_2O_3 samples obtained and studied in the present paper are clear and transparent having a very good adhesion to the glass substrate. In addition, the electron diffraction spectra show that the alumina films are amorphous at all vapour deposition angles.

Fig. 1 (a-d) presents scanning electron micrographs of growth profiles of about 2000 nm thick Al_2O_3 films e-beam deposited at different vapour incidence angles α. It is clearly seen that the SEM imaging reveals a typical columnar growth at normal and oblique vapour incidence. The figure demonstrates also that the columns run through the entire film volume from the substrate to the free surface of the samples. Besides, the individual columns are separated from low density material and a free intercolumnar volume could be distinguished at high α values. Also, in obliquely deposited films, a column inclination toward substrate plane is observed. It should be noted here that both the vapour incidence angle α and the column inclination β measured with respect to the substrate normal, satisfied the so called tangent rule established for many crystalline as well as for amorphous films [5]

$$tg\,\alpha = 2tg\,\beta. \quad (2)$$

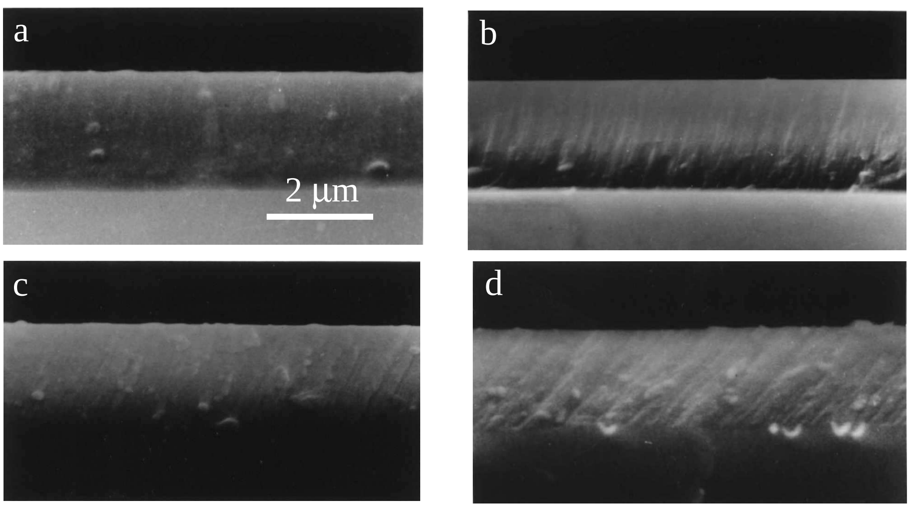

Fig.1 Scanning electron micrographs of amorphous alumina films e-gun deposited at: a) 0 °; b) 40 °; c) 60 °; d) 70 ° vapour incidence angles.

Fig. 2 shows the film microhardness change when α increases. At normal vapour incidence the samples are characterized by a maximal microhardness, which gradually decreases when α increases and at α=80˚ the MH value is three times lower than the measured for the samples deposited at normal incidence.

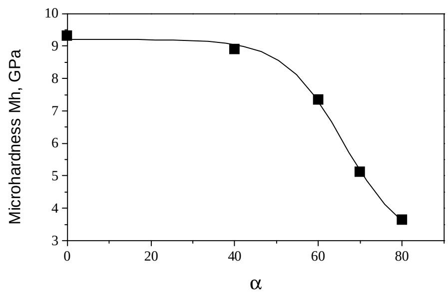

Fig. 2. Microhardness (MH) of 1300 nm thick Al_2O_3 films as dependent on the vapour incidence angle α.

It was expected that the other physical properties of the investigated alumina films should be also influenced by the revealed structural inhomogeneity. For that reason, the thermal transport in the Al_2O_3 thin films was measured for samples deposited at two vapour incidence angles – 40 ^{o} and 70 ^{o}. Following the method mentioned in experimental part it was found that the sample with higher column inclination has about two times lower thermal diffusivity than that obtained for the sample deposited at 40^o incidence angle.

Fig. 3 demonstrates the dependence of the obtained effective refractive index of the alumina thin films versus vapour incidence angle. It shows that the optical properties are also strongly influenced by the structural inhomogeneity. The n values calculated for λ= 633 nm are in the range, which is in a good agreement with the literature data n=1.51 – 1.66 for vacuum deposited alumina films [6]. As the other measured physical properties The effective refractive index shows analogous to the other measured physical properties a trend to decrease when α increases, although the n value for α=40^o is a little bit higher than the evaluated one at 0˚. A similar deviation at the same vapour incidence angle was earlier observed for the refractive index of amorphous GeS_2 films [7].

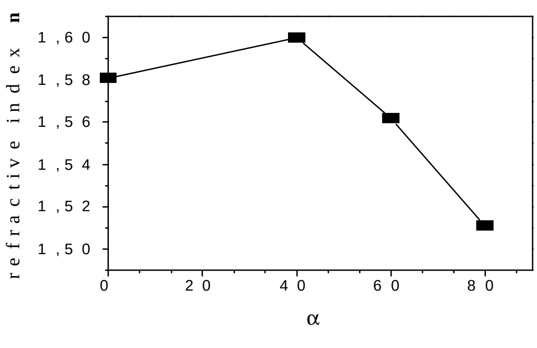

Fig. 3. Effective refractive index n of 1300 nm thick Al_2O_3 films as dependent on the vapour incidence angle α.

4. Discusion

The experimental results obtained clearly demonstrate that similarly to the vacuum deposited amorphous As_2S_3 [8], Sb_2Se_3 [9], GeS_2 [7] and ZrO_2 [10] the amorphous alumina thin films also grow in columnar manner at least under the experimental condition used. This means that Al_2O_3 thin films are structurally inhomogeneous, the individual columns being separated each other from a low density material and/or free volume. This inhomogeneity is more pronounced at higher vapour incidence angles where the growth morphology is characterized by columns inclined toward the substrate surface.

On the other hand the data obtained for microhardness, thermal conductivity as well as for the relative refractive index are sensitive function of the vapour incidence angle and therefore of film inhomogeneity. The column inclination and the film free volume are greater at large vapour incidence angle, thus leading to a decrease of microhardness, thermal conductivity and the refractive index. Therefore a correlation between these physical properties of e-gun deposited alumina thin films and column inclination is revealed.

Concerning the n data some details should be discussed. As mentioned above, the film microhardness value at α=40˚ is a little bit higher than the measured one at normal vapour incidence. As in the case of amorphous GeS_2 thin films, we have no explanation of this deviation from the tendency of n to decrease when α increases.

Further, as seen on Fig. 3 there is no n value for α=70˚. It should be stressed here, that the minimax computing program requires that the refractive index of the film to be higher than that of the glass substrate. Our experiments show that there is not interference extremes in the transmission spectra of the alumina samples deposited just at 70^o. This means that the refractive index of both, the alumina film and the glass substrate are approximately equal. Nevertheless, at higher vapour incidence angle, $\alpha=80^o$, interference extremes are again observable and the data calculated for these samples show the general trend of the refractive index change when α increases.

5. Conclusions

The present study shows that similarly to many different thin film materials the amorphous alumina thin films deposited by e-gun at different vapour incidence angles have a columnar microstructure. It was demonstrated that the structural inhomogeneity of the films strongly influences their mechanical, thermal and optical properties. The experimental data show a substantial decrease of the microhardness, the thermal diffusivity as well as the effective refractive index of the samples with increasing of the column inclination. Therefore, the method of obliquely deposition could be applied for modification of the basical physical properties of the amorphous Al_2O_3 thin films.

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