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Antireflective bilayer coatings based on Al_2O_3 film for UV region*

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Bilayer antireflective coatings consisting of aluminium oxide Al_2O_3/MgF_2 and Al_2O_3/SiO_2 are presented in this paper. Oxide films were deposited by means of e-gun evaporation in vacuum of 5×10^{-3} Pa in the presence of oxygen, and magnesium fluoride was prepared by thermal evaporation on heated optical lenses made from quartz glass (Corning HPFS). Substrate temperature was maintained at 250 °C during the deposition. Thickness and deposition rate were controlled with a thickness measuring system Inficon XTC/2. The experimental results of the optical measurements carried out during and after the deposition process have been presented. Physical thickness measurements were made during the deposition process and resulted in 44 nm/52 nm for Al_2O_3/MgF_2 and 44 nm/50 nm for Al_2O_3 /SiO_2 system. Optimization was carried out for ultraviolet region with minimum of reflectance at 300 nm. The influence of post deposition annealing on the crystal structure was determined by X-ray measurements. In the range from ultraviolet to the beginning of visible region, the reflectance of both systems decreased and reached minimum at 290 nm. The value of reflectance at this point, for the coating Al_2O_3/MgF_2 was equal to $R_{290nm} = 0.6$ % and for Al_2O_3 /$SiO_2 R_{290nm} = 1.1$ %. Despite the difference between these values both are sufficient for applications in the UV optical systems for medicine and UV laser technology.

Keywords: *Al_2O_3/MgF_2, Al_2O_3/SiO_2; antireflective coatings; optical properties.*

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1. Introduction

Antireflective coatings (AR) are regarded as the basic thin films applied in optics. The coatings provide numerous benefits including increased transmissibility, reduced glare and surface reflection. The family of thin antireflective films is applied in several devices, like photovoltaic panels [1, 2], lenses [3], displays, cameras [4, 5], spyglasses, lasers [6] . Most of the films are designed for visible region but many devices are used in infrared or ultraviolet regions.

Vast range of oxide and fluoride materials are used to produce successfully high standard antireflective coatings [7]. The first group are materials with high refractive indices, such as Er_2O_3 [8], HfO_2 [9] and Ta_2O_5 [10]. The second group are materials with medium refractive indices, such as Al_2O_3, LaF_3, NdF_3 [11, 12]. The last group represents oxide and fluoride SiO_2 and MgF_2, with low refractive indices [12, 13]. Except optical properties, materials for antireflection coatings should have good mechanical features, such as hardness, adhesion to substrate and chemical resistance.

Quartz glass is the most common material used for producing optical parts for UV region. In case of a single lens, which is produced from this material, transmittance is equal approximately to 92.8 %. In case of a system composed from five lenses the transmittance falls to about 69 %. When optical parts are made of high index glass (e.g. n = 1.8) transmittance of the same system falls to about 41 %. This fact signifies that antireflective coating becomes obligatory to prevent any

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optical system from lowering the quality of the optical transmittance.

2. Result and discussion

The deposited antireflective coating system was composed of two layers Al_2O_3/MgF_2 and Al_2O_3/SiO_2. The optimal thickness of the films was determined by a well known Quarter Wavelength Optical Thickness (QWOT) method. The physical thickness of the QWOT film is in a direct relation to the refractive index of the material that is being used. Refractive indices reported by the producer were equal to n = 1.63 for Al_2O_3 [14], n = 1.46 for SiO_2, and n = 1.38 for MgF_2.

Description of the simulation methods for the transmittance and the reflectance spectra has been presented in [2, 15, 16]. In the case of our experiments, QWOT analysis was supported by simulation of transmittance and reflectance characteristics based on refractive indices and variation of film thicknesses for all films. The results were obtained for a constant thickness of the MgF_2 film and a dedicated variation of the Al_2O_3 film. Almost identical simulations were performed for all other materials used for Al_2O_3/SiO_2 antireflective coatings production.

The deposition methods, such as sputtering and evaporation, are often used to prepare antireflective coatings.

The coatings deposition processes were performed in a vacuum evaporation equipment (NA501A type) at a base pressure below 5×10^{-3} Pa in a water cooled vacuum bell jar. Quartz glass substrates (Corning HPFS) with a refractive index n = 1.46 were coated with bilayer filters. Conditions during the experiment were as follows: substrate temperature T = 250 °C, pressure $p = 5 \times 10^{-3}$ Pa. Thin films of Al_2O_3 and SiO_2 were deposited from an e-gun evaporator. Additionally, during the deposition of Al_2O_3, O_2 was dosed. MgF_2 was deposited from a tantalum boat by thermal evaporation method. The multicrucible, linear e-gun from Varian-H and the boat type tantalum evaporator are shown in Fig. 1. The film thickness and the deposition rate measured with a

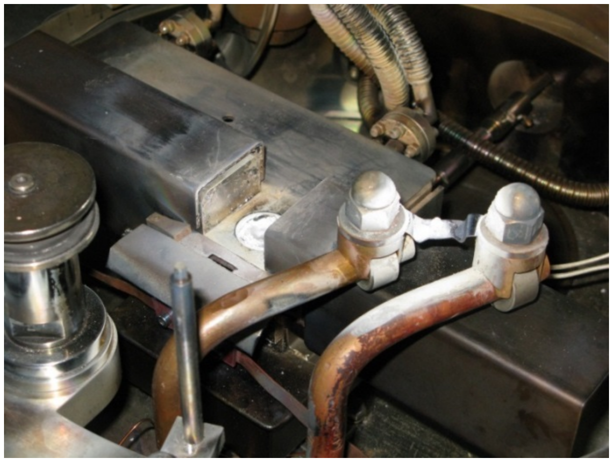

Fig. 1. The boat type evaporator and multicrucible linear e-gun from Varian.

thickness controller type Inficon XTC/2. The final thicknesses were equal to 44 nm, 50 nm and 52 nm for Al_2O_3, SiO_2, MgF_2 and evaporation rates for these materials were equal to 0.6 nm/s, 0.6 nm/s, and 0.8 nm/s, respectively. Film thicknesses, densities and crystalline structure were evaluated from X-ray reflection (XRR) and grazing incidence X-ray diffraction (GI-XRD) patterns were measured with a PANalytical X'pert Pro MPD X-ray diffractometer.

Samples were annealed after deposition in air at 600 °C. The Al_2O_3 layer after annealing still remained amorphous as shown in Fig. 2 but the MgF_2 top layer crystallized already at this temperature [17, 18]. Fig. 3 shows the X-ray reflectometry patterns for the studied bilayers. The X-ray reflectivity curves are offset for clarity. In all cases pronounced Kiessig fringes arising from the finite thickness of the films were observed, indicating well-defined interfaces.

The experimental reflectivity curves were fitted using the Parratt recursion and the least squares method with segmented fitting algorithm. The initial model of the sample was created in accordance with the nominal composition and thickness of the deposited layers. The fitted parameters were the substrate and layers roughness, the density and thickness of subsequent Al_2O_3 and top layers. During the fitting procedure the roughness

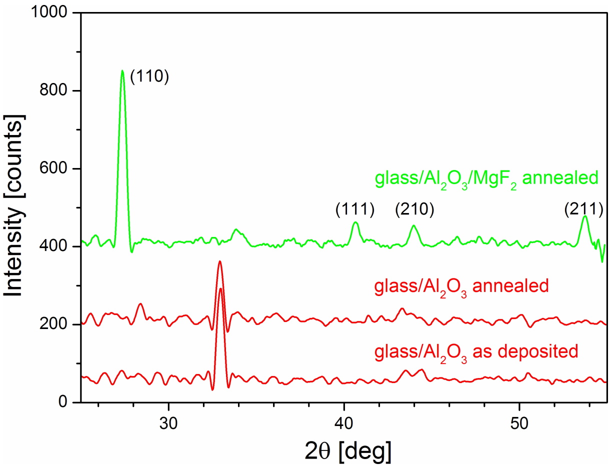

Fig. 2. X-ray diffraction spectrum of Al_2O_3/MgF_2 (thickness of 44 nm/52 nm) and Al_2O_3/SiO_2 (thickness of 46 nm/51 nm) as-deposited and after 1 h annealing at 600 °C in air.

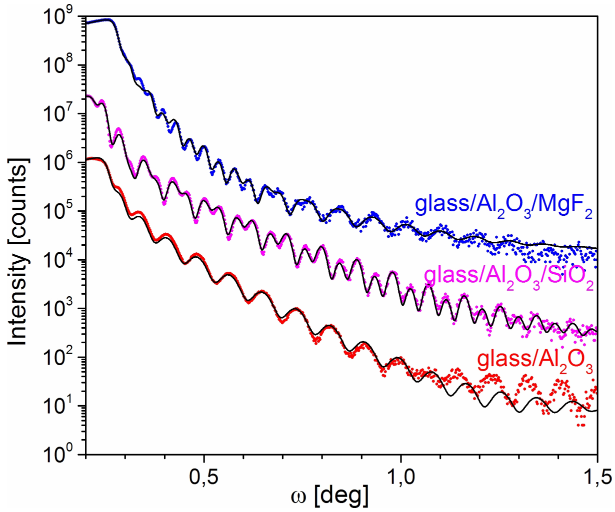

Fig. 3. X-ray reflectometry spectra together with theoretical fits for Al_2O_3/MgF_2 (thickness of 44 nm/52 nm) Al_2O_3/SiO_2 and Al_2O_3 films deposited on Si substrate after 1 h annealing at 600°C in air.

and thickness of the layers were fitted independently. The obtained results of fitting confirmed the nominal thickness of the Al_2O_3 and the top layers; the roughness of the films was approximately 1 nm. However, the position of critical angle indicates the smaller density of Al_2O_3 films (2.9 g/cm^3) than typically reported for an amorphous Al_2O_3 (3.5 to 3.8 g/cm^3) [17–19]. This effect is often observed for Al_2O_3 films deposited at low temperatures and can be explained by the fact that films deposited onto unheated substrates absorb water which stays bound in the film despite of post-deposition annealing [18, 20–22].

Optical transmittance and reflectance spectra were measured by means of spectrophotometer Shimadzu UV1601 in the range of 200 nm to 400 nm and are presented in Fig. 4 and 5. The curves of the reflectance have "V" shapes, typical of bilayer antireflective coatings. The Al_2O_3/MgF_2 system reduces reflection in a range from about 230 nm up to beginning of VIS region, with minimum of the reflectance R_{290nm} = 0.6 % at 290 nm.

The average reflectance for this area is equal to 1.7 %. It means that the antireflection coating reduces reflectance more than 50 % compared to the bare quartz glass. The second system has a little bit worse optical parameters. The Al_2O_3/SiO_2 reduces

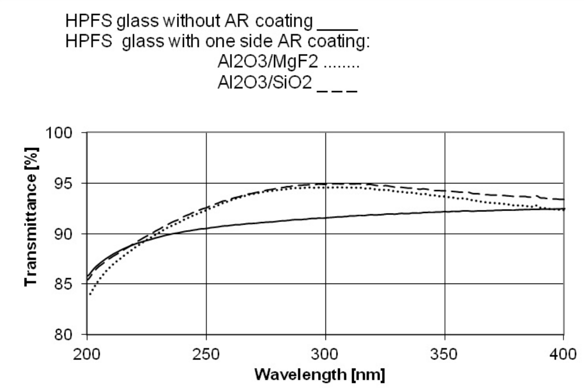

Fig. 4. Transmittance for a bare substrate HPFS glass (lower curve), HPFS glass with antireflective Al_2O_3/SiO_2 coating (upper curve) and HPFS glass with antireflective Al_2O_3/MgF_2 (dotted curve) coating optimized for UV region.

reflectance from 235 nm up to 375 nm with minimum of reflectance R_{290nm} = 1.1 % at the same wavelength.

The average reflectance for this area is equal to 2.1 %. The other possibility for changing the optical characteristics is the deposition at different temperatures. In our case they were 100 °C, 200 °C and 300 °C. The differences in transmittance spectra for

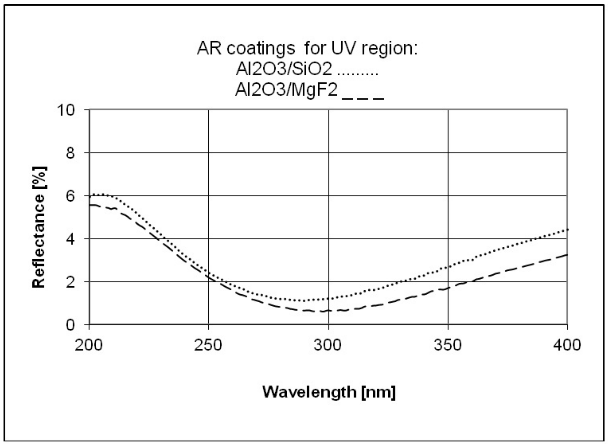

Fig. 5. Reflectance for Al_2O_3/MgF_2 and Al_2O_3/SiO_2 coatings optimized for UV region. Reflectance of the HPFS glass with antireflective Al_2O_3/SiO_2 coating (upper curve) and HPFS glass with antireflective Al_2O_3/MgF_2 coating (lower curve) optimized for UV region.

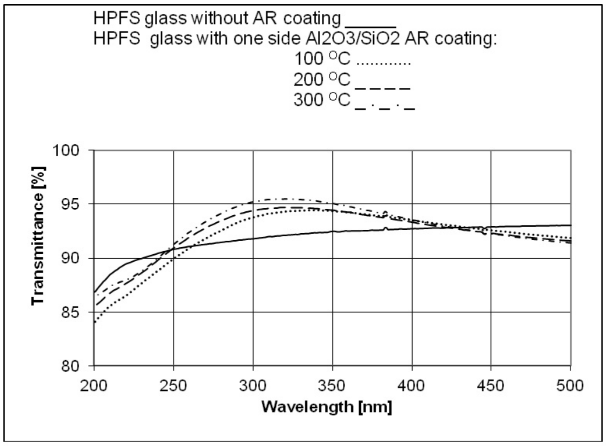

Fig. 6. Transmittance for a bare substrate HPFS glass (lower curve) and HPFS glass with antireflective Al_2O_3/SiO_2 coating deposited at different temperatures: 100 °C, 200 °C and 300 °C.

Al_2O_3/SiO_2 coating are presented in Fig. 6. One can see that the best optical properties, it means the biggest transmittance, we have obtained for the system deposited at 300 °C.

3. Conclusions

Two-layer antireflective coating systems were deposited by means of e-gun and thermal evaporation methods on quartz lenses with a diameter of 22.3 mm. Final thicknesses of the films were 44 nm/52 nm and 44 nm/50 nm for the Al_2O_3/MgF_2 and Al_2O_3/SiO_2, respectively.

Deposition rates were 0.6 nm/s and 0.8 nm/s for Al_2O_3/MgF_2 and 0.6 nm/s and 0.6 nm/s for Al_2O_3/SiO_2, respectively. Transmittance and reflectance spectra were measured. The reflectivity coefficient was equal to R_{290nm} = 0.6 % for Al_2O_3/MgF_2 and R_{290nm} = 1.1 % for Al_2O_3/SiO_2 and was much lower than for the bare quartz glass R_{290nm} = 3.6 %. In this case, transmittance of UV light for double side AR coated optical parts, made of quartz glass, at 290 nm, was near 99 % and 98 %, respectively. These values are sufficient for application in medical multilenses equipment working in UV wavelength and for UV laser technology [13, 23]. Especially optical parts dedicated for UV lasers need high quality antireflective coatings designed for only one wavelength. It seems that these thin film systems could have a large practical significance because of the good optical properties, relatively low cost and simple production technology.

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