Design of antireflective coatings for AZO low infrared emissivity layer

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In this letter, we investigate the structural, optical, and electrical properties of Al-doped ZnO (AZO) thin film coating prepared by direct current (DC) facing-target sputtering method at room temperature, of which the average optical transmittance is 81% between 400 and 700 nm while the sheet resistance is about 10 Ω/□. Then, based on this AZO coating, in order to enhance the transmittance, interfacial adhesion strength and weathering resistance, two kinds of antireflective coatings are designed for different application purposes. For the two kinds, the highest transmittances in the visible region (400–700 nm) can reach 86.9% and 81.8%, respectively. The design is performed using Macleod Optical Design software.

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Transparent conductive oxides (TCO) thin films have been used in many industrial applications, such as solar cell[1,2], flat panel display[3], and light emitting diode[4], due to their transparency in the visible wavelength region, large exciton binding energy and low resistivity. As a direct band gap semiconductor, ZnO has band energy of more than 3.3 eV and more than 60 meV of exciton binding energy. Nowadays, Al-doped ZnO (AZO) thin films as a promising n-type TCO are intensively researched to replace indium tin oxide (ITO) thin films, because they are non-toxic and cost-effective[5]. Superior AZO films with small resistance of about 10^{-4} Ω·cm not only have wide application prospects in flexible displays and thin film solar cells as transparent conductive electrodes[6], but also play an important role in infrared stealth field as transparent low-emissivity semiconductors. Low infrared emissivity coatings are widely used in civil and military applications such as heat loss control industry or camouflaging military equipments[7,8].

AZO thin films can be prepared by a variety of techniques such as magnetron sputtering[5,9], chemical vapor deposition, pulsed-laser deposition[10], molecular beam epitaxy, spray-pyrolysis[11], and (electro-) chemical deposition[12]. However, Sputtering method becomes the most industrially advanced deposition technique for AZO, since it leads to the best AZO films with high conductivity and transparency and also allows large area depositions. In this letter, we prepare low infrared emissivity AZO thin films with a sheet resistance about 10 Ω/□ through conventional direct current (DC) facing-target magnetron sputtering method with no attention to heat the substrate. The average optical transmittance is 81% between 400 and 700 nm. However, a single AZO film is not able to give high transmittance values and has poor weathering resistance[13]; therefore a more complex structure is necessary. Two kinds of antireflective coatings have been proven to enhance the transmittance of AZO films: in the first kind, AZO thin film is used as inner layer; in the second kind, AZO thin layer is chosen as top layer. The average transmittances in the visible region (400–700 nm) can reach 86.9% and 81.8 %, respectively.

The AZO thin films were deposited on glass substrate by the conventional DC facing-target sputtering method. Figure 1 shows a schematic of the facing target cathode. Two identical Al_2O_3 doped ZnO targets were placed in parallel at distance of 80 mm (target-to-target distance). The weight ratio of Al_2O_3 to ZnO (both 99.99% pure) was 3%. The base pressure of sputtering chamber was 6×10^{-4} Pa, sputtering pressure was in an argon atmosphere of 0.5 Pa and the sputtering power was maintained at 500 W. In addition, the substrate location was out of the high-density plasma region leading to a low-temperature deposition of AZO.

The electrical properties of AZO were examined by four-point probe system. The optical transmittance of the samples was measured by a ultraviolet-visible (UV-Vis) spectrophotometer. To investigate the crystalline structure, X-ray diffractometer with Cu Kα source measurement was also carried out. The thickness and refractive index of the thin films were measured by spectroscopic ellipsometer.

The design of optical layers includes two parts: in the first design, AZO thin film is used as inner layer and as top layer in the second design. The thickness of films is specified by optical thickness and the centre wavelength is 550 nm.

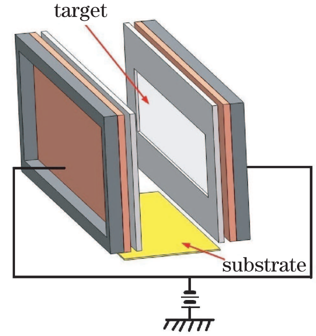

Fig. 1. Schematics of the facing target cathodes.

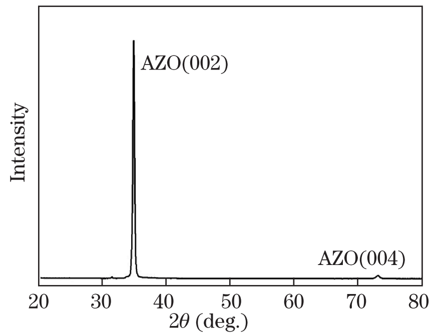

Fig. 2. XRD pattern of AZO film obtained DC face-target magnetron sputtering method.

Figure 2 shows the X-ray diffraction (XRD) pattern of the AZO thin film. The AZO film grown on the corning glass substrate exhibits two peaks located at 34.6° and 72.8°, respectively. According to the Joint Committee on Powder Diffraction Standards (JCPDS) card, the two peaks correspond to wurtzite hexagonal ZnO (0002) and (0004), respectively[14−16]. Hence the film has a single hexagonal phase with highly preferred orientation of the c-axis perpendicular to the substrate.

As we all know, the resistance and transmittance are important for transparent electrode. In this letter, the prepared AZO sample has a sheet resistance of about 10 Ω/□, which also means a low infrared emissivity[17]. Figure 3 shows the optical transmission and reflectance properties of the AZO film. Compared with the transmittance of bare glass substrate, all AZO show lower optical transmittance in the visible wavelength region of 400–700 nm. The transmittance of the AZO/glass sample varies from 72% to 86%, and the average optical transmittance is 81% between 400 and 700 nm.

AZO thin film coatings are usually in the form of one-layer as a transparent electrode[16,18]. As the result, the transmittance in visible wavelength range is less than 85%, which may limit its application. So we should enhance the transmittance in the visible range of light spectrum and weathering resistance of AZO coatings for our thin film, which is of crucial importance for the applications. Using antireflective coatings can be a good way to enhance the transmittance of AZO coatings and choosing some hard coating as one antireflective layer in the layer stacks also may improve the weathering resistance of AZO thin film, hence extend their application purposes. With the use of Macleod Optical Design software, two kinds of antireflective coatings have been designed. The details of designed coating are shown in Table 1 and the optical transmittance of all the samples are presented in Fig. 4.

In the first kind of coatings, AZO thin film is used as inner layer and low refractive index material SiO_2 and Al_2O_3 are used as anti-reflection layers for optical optimization. Both SiO_2 and Al_2O_3 have high transmittance in the infrared wavelength region, so they will not affect the infrared emissivity of AZO thin film. The coating system can be described as follows: glass/AZO/SiO_2 and glass/Al_2O_3/AZO/SiO_2. As shown in Table 1, the average transmittance of glass/AZO/SiO_2 coating system can reach 86.8% in the visible region (400–700 nm) and the highest transmittance is 90.4%, which has a significant optical improvement compared with the single AZO thin film. For the three layer coating system, Al_2O_3 was used as a buffer layer between glass and AZO thin film, which can improve the adhesion strength between the substrate and AZO thin film. The highest and average transmittances are 90.5% and 86.9%, respectively. The SiO_2 layer used as a top layer can protect the AZO thin film from the environmental corrosion. These coatings can be used for window glass of aviation cockpit which requires low infrared emissivity and high conductivity.

The second type of coatings, AZO thin layer is chosen as top layer with three layer stacks: glass /TiO_2/AZO and glass/Al_2O_3/TiO_2/AZO. The layer of TiO_2 was used as the high refractive index material, SiO_2 as the low refractive index material and Al_2O_3 as a buffer layer. The results from Table 1 show that the average transmittance in the visible region (400–700 nm) has a value of 80.8% for the two layer system (glass/TiO_2/AZO) and 81.8% for the three layer system (glass/Al_2O_3/TiO_2/AZO). The highest transmittance of the two layers system is 88.1%, which is higher than 86% of the single AZO thin film. For the glass/Al_2O_3/TiO_2/AZO system, the lowest transmittance is 75.8%, which is higher than 72.8% obviously. They can be used as transparent conductive electrode in display devices and anti-static layer in window glass of aviation cockpit.

In conclusion, we prepare AZO thin films through DC face-target magnetron sputtering method. We demonstrate that the face-target magnetron sputtering is a promising low-temperature sputtering method. The AZO thin film grown on the glass substrate shows a sheet resistance of 10 Ω/□ and an average transparency of 81.2% between 400 and 700 nm. In order to enhance the

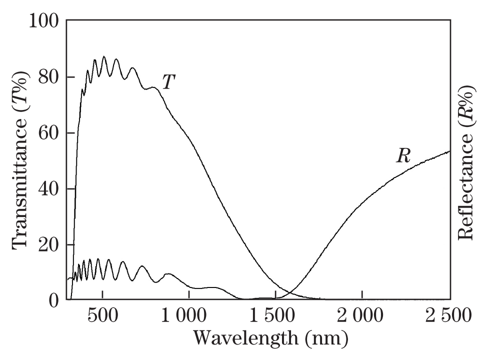

Fig. 3. Optical transmittance and reflectance of AZO films.

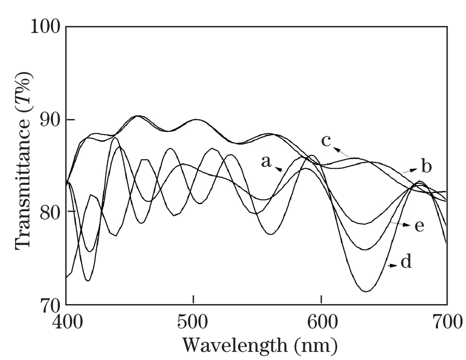

Fig. 4. Optical transmittance of the designed antireflective coatings in the visible region (400–700 nm).

Table 1. Designed Layer Stacks and Their Transmittance Results

	Layer Structure	Highest Transmittance	Lowest Transmittance	Average Transmittance
a	Glass/13.6AZO	86%	72.8%	81.2%
b	Glass/13.6AZO/0.9SiO_2	90.4%	81.2%	86.8%
c	Glass/1.86Al_2O_3/13.6AZO/ 0.89SiO_2	90.5%	82.2%	86.9%
d	Glass/1.83TiO_2/ 13.6AZO	88.1%	71.5%	80.8%
e	Glass/0.91Al_2O_3/1.87TiO_2/ 13.6AZO	87.1%	75.8%	81.8%

The thickness of films is specified by optical thickness and the central wavelength is 550 nm. The coefficient before the material is the ratio of central wavelength.

transmittance and interfacial adhesion strength and weathering resistance of AZO coatings, two kinds of antireflective coatings are designed. In the first kind of coatings, AZO thin film is used as inner layer and low refractive index material SiO_2 and Al_2O_3 are used as anti-reflection layers for optical optimization and the average transmittance reaches 86.8% and 86.9 % in the visible region (400–700 nm). The second type choosing AZO thin layer as top layer with three layer stacks, the average transmittance reaches 80.8% and 81.8 % in the visible region (400–700 nm).

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