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Preparation and Characterization of Low- emissivity AlN/Ag/AlN Films by Magnetron Co-Sputtering Method

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Abstract

AlN and Ag thin films were deposited independently on the Si(100) wafers and glass slides by sputtering technique at various times to find the optimum deposition times for coating AlN and Ag films layers. The results obtained from glazing-incident X-ray diffraction (GIXRD), field-emission scanning electron microscopy (FE-SEM), and transmittance measurements showed that the optimum deposition time for coating AlN and Ag film layers were 40 min and 15 s corresponding to the film thickness of 46.7 and 19.6 nm, respectively. The optimum deposition times were used for coating AlN and Ag films in the multilayer AlN/Ag/AlN film stack. Then, the multilayer AlN/Ag/AlN film stack was deposited on the glass slide for transmittance measurement and a test glass plate with a size of 10 cm x 10 cm for infrared protection testing. The average solar transmittances in the visible range (λ = 380-780 nm) and in the near infrared range (λ = 780-2,000 nm) were found to be 48.05 and 15.17%, respectively which are comparable with those of a commercial glass.

Keywords: *AlN/Ag/AlN thin film; low-emissivity glass; magnetron co-sputtering; multilayer films; infrared protection; solar transmittance*

1. Introduction

Nowadays, tall buildings in all the big cities around the world have been constructed with walls made of glass. The glass wall without thin film coating generally transmits the solar radiation (λ 400-2500 nm) of about 80% into the building, about 4% is reflected and the rest is absorbed. The absorbed solar radiation, then will be emitted in the infrared range and cause the heat to transfer into the building. In recent years, there has been extensive research into coating glass buildings with low-emissivity materials to reduce the transmission of infrared radiation.

Low emissivity coating has high transmittance in the visible spectrum region and high reflectance in the near infrared region (Tsai et al., 2019; Sukkasem et al, 2021). Generally, low emissivity film composes of dielectric/metal/dielectric stack layers (Zhang et al., 2020). Highly conductive metals like Ag, Au, Cu and Al are used for metal layer (Zhang et al., 2020; Jang et al., 2020). Among those metals, Ag is the most popular metal employed in the stack layers. This is due to its high IR reflectance and without significant absorption of visible light (Tsai et al., 2019). For dielectric layer, the transparent oxide and sulfide films of various dielectric materials including ITO, TiO_2, SnO_2, ZnO and ZnS have been used as protective layer in dielectric-metal-dielectric multilayer structure **(**Chiba & Kaminishi, 2008). The low-emissivity coatings of dielectric-metal-dielectric multilayer structure have been widely investigated such as ITO/Ag/ITO, TiO_2/Ag/TiO_2, SnO_2/Ag/SnO_2, ZnO/Ag /ZnO and ZnS/Ag/ZnS **(**Chiba & Kaminishi, 2008; Dhar & Alford, 2013; Mungchamnankit et al., 2014; Kim et al., 2016). However, the Ag atoms can react with the oxygen of dielectric layer at high temperature and lead to poor- low-emissivity, corrosion resistance, chemical durability and thermal stability (Chokboribal et al., 2021; Kulczyk-Malecka et al., 2014; Loka, Yu & Lee, 2014).

One way to solve this problem is the coating of metal nitrides –Ag –metal nitride structure. In the past decade, TiN, TaN and TiAlN have been studied on low-emissivity coatings of metal nitride-Ag-metal nitride structure (Loka, Yu & Lee, 2014; Akepati et al., 2013; Huang et al., 2011; Huang et al., 2014)

Aluminum nitride (AlN) is a dielectric material that has attracted much interest due to its properties such as good thermal and chemical stability, large electrical resistivity, wide bandgap, and high refractive index (Zhu & Yang, 2022; Manova et al., 1998; Kumar et al., 1997; Cheng et al., 2003). Therefore, in recent years, AlN was used as a dielectric in dielectric/Ag/dielectric multilayer structure. So far, few studies on AlN/Ag/AlN film and AlN-Ag based film have been reported (Zhu & Yang, 2022; Ferrara et al., 2016; Addonizio et al., 2021).

In this work, AlN/Ag/AlN thin films were prepared on Si wafers and glass slides by magnetron co-sputtering. Before fabrication of AlN/Ag/AlN film stack, the AlN and Ag films were deposited independently on Si and glass substrates at various deposition times to find the optimum AlN and Ag film layers in the AlN/Ag/AlN film stack.

2. Objectives

The objective of this research is to find the optimum AlN and Ag film layers in the AlN/Ag/AlN film stack. This research also aims to assess the solar transmittance of the AlN/Ag/AlN film stack deposited using a home-built DC magnetron co-sputtering, and to compare it with that of commercial glass.

3. Materials and methods

Multilayer AlN/Ag/AlN films were deposited on 10 cm x 10 cm Si(100) wafers and microscope glass slides using a home-built closed field unbalanced DC magnetron co-sputtering system. The schematic diagram of the magnetron co-sputtering system has been previously described elsewhere (Chantharangsi et al., 2015).

The Al (99.9995%) and Ag (99.99%) targets, each with a diameter of 3 inch were used as sputtering sources. The substrates were ultrasonically cleaned in acetone and methanol for 10 min and dried with nitrogen before installation in the vacuum chamber at a distance of 13 cm from the targets.

The vacuum chamber was evacuated to a base pressure of about 5×10^{-5} mbar using a pumping system consisting of a rotary pump and a diffusion pump. Prior to film deposition, the targets were sputter-cleaned in Ar plasma to remove contaminants on the target surfaces for 10 min. During the sputter-cleaning, an Ar flow rate was set at 4.5 sccm, while electrical power on the targets was kept at 0.2 A and ~300 V. After the sputter-cleaning of the targets, two experiments were carried out independently as follows:

(1) *Deposition of AlN film layer*

To find the optimum thickness of AlN film in the AlN/Ag/AlN stack, the AlN films were deposited on Si wafers and glass slides at different deposition times of 20, 30 and 40 min while the sputtering current of Al target, Ar flow rate and N_2 flow rate were fixed at 700 mA, 4.5 sccm and 0.5 sccm, respectively.

(2) *Deposition of Ag film layer*

To find the optimum thickness of Ag film in the AlN/Ag/AlN stack, the Ag films were deposited on Si wafers and glass slides at different deposition times of 5, 10 and 15 s while the sputtering current of Ag target and Ar flow rate were fixed at 200 mA, and 4.5 sccm, respectively.

After AlN and Ag films were prepared independently under various conditions, the crystalline structure of AlN film deposited on Si substrates was characterized by glazing-incident X-ray diffraction (Bruker, D8 Discover). The GIXRD patterns were recorded at a grazing incidence angle of 2° with a 2θ range from 20° to 80° and a scanning speed of 2°/min. The cross-sectional morphology of AlN and Ag films were studied by field emission scanning electron microscopy (FE-SEM : Jeol, JSM 660 LV). The transmittance of AlN and Ag films deposited on glass substrates was measured in the wavelength range of 300-2,000 nm using a spectrophotometer (Shimadzu, UV-VIS 3600).

(3) *Deposition of AlN/Ag/AlN films stack*

After AlN and Ag films were deposited at different times and characterized, the optimum deposition times for coatings AlN and Ag were obtained. Then, the multilayer film of AlN/Ag/AlN stack was deposited on a glass slide (for transmittance measurement) and a glass plate with a size of 10 cm x 10 cm x 5 mm (for infrared protection testing).

4. Results and Discussion

4.1 AlN films

The crystalline structure of AlN film was investigated by GIXRD. Figure 1 shows the GIXRD patterns of ALN films deposited on Si substrates with various times of 20, 30 and 40 min.

The JCPS no. 79-2497 of AlN bulk with hexagonal wurtzite structure was used as reference. The crystallites are highly oriented with their c-axis perpendicular to the substrate surface. At a deposition time of 20 min, only the (100) peak was observed. When the deposition time was increased from 20 to 40 min, the intensity of (100) peak increased which means the increasing crystallinity of the film. Furthermore, the (110) peak started to appear from the deposition time of 30 min. From GIXRD results, it can be concluded that the deposition time for coating AlN layer 40 min is better than those of films coating with deposition times of 20 and 30 min.

The transparency of AlN films were finally investigated by measuring transmittance spectra. Figure 2 shows the transmittance spectra of AlN films deposited on the glass substrates at various deposition times of 20, 30 and 40 min in the wavelength range from 300-2,000 nm. It is seen that all AlN films give rather high transmittance both in the visible range (380-780 nm) and near infrared range (NIR, 781-2,000 nm). However, the lowest transmittance (i.e., highest reflectance) in the NIR range was obtained from AlN film deposited for 40 min. Therefore, the optimum deposition time obtained from transmittance spectra was 40 min which agree with GIXRD results. Hence, it will be further used for coating AlN film in the AlN/Ag/AlN stack.

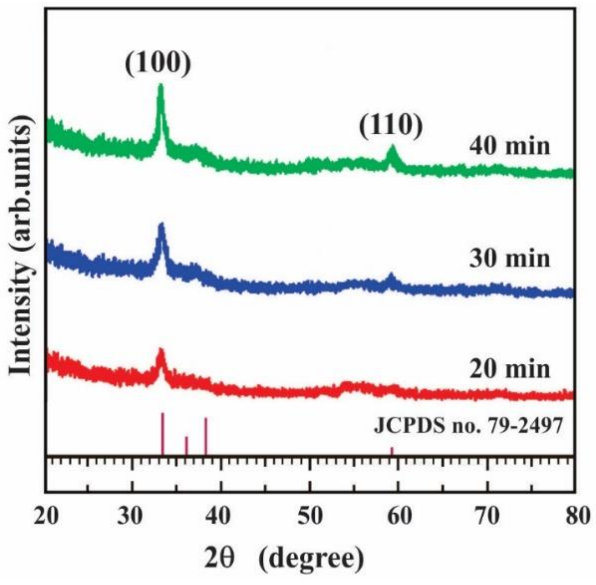

Figure 1 GIXRD patterns of AlN films deposited on Si (100) substrates with various times of 20, 30 and 40 min.

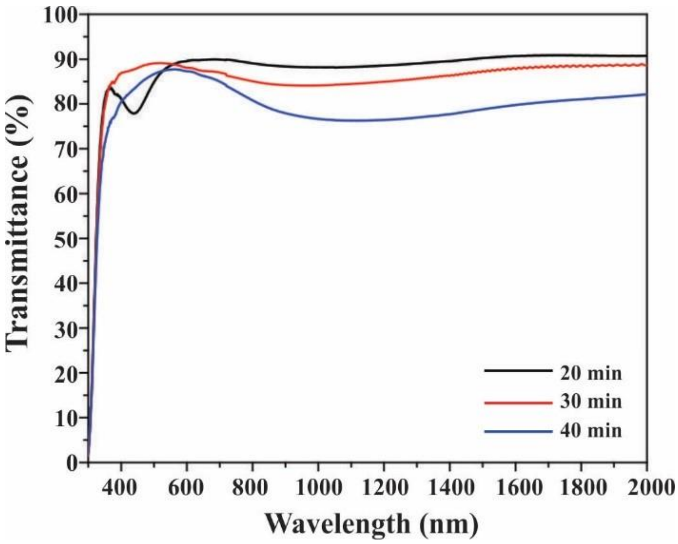

Figure 2 Transmittance spectra of AlN films deposited at various times of 20, 30 and 40 min.

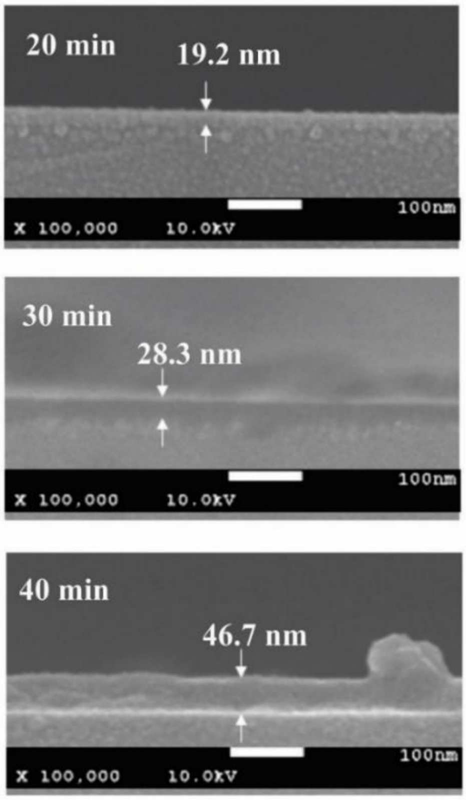

Figure 3 FE-SEM images of cross-sectional morphology of AlN films.

To investigate the optimum thickness of AlN layer, the cross-sectional morphology was carried out. Figure 3 shows the cross-sectional FE-SEM images of AlN films deposited on Si substrates with various times of 20, 30 and 40 min. The thickness of the films was found to be 19.2, 28.3 and 46.7 nm for the deposition times of 20, 30 and 40 min, respectively. Therefore, the optimum thickness corresponding to the optimum deposition time of 40 min is 46.7 nm.

4.2 Ag films

The transparency of Ag films was investigated by measuring transmittance spectra. Figure 4 shows the transmittance spectra of Ag films deposited on the glass substrates at various deposition times of 5, 10 and 15 s in the wavelength range from 300-2,000 nm. It is seen that Ag films give a medium high transmittance in the visible range (380-780 nm) and lowest transmittance in the NIR range for the Ag film deposited for 15 s. Therefore, the deposition time of 15 s will be further used for coating Ag film in the AlN/Ag/AlN stack.

The optimum thickness of Ag layer was also investigated by FE-SEM. Figure 5 shows the cross-sectional FE-SEM images of Ag films deposited on Si substrates with various times of 5, 10 and 15 s. The thickness of the Ag films was found to be 11.0, 15.7 and 19.6 nm for the deposition times of 5, 10 and 15 s, respectively. Hence, the optimum thickness of Ag layer corresponding to the optimum deposition time of 15 s is 19.6 nm.

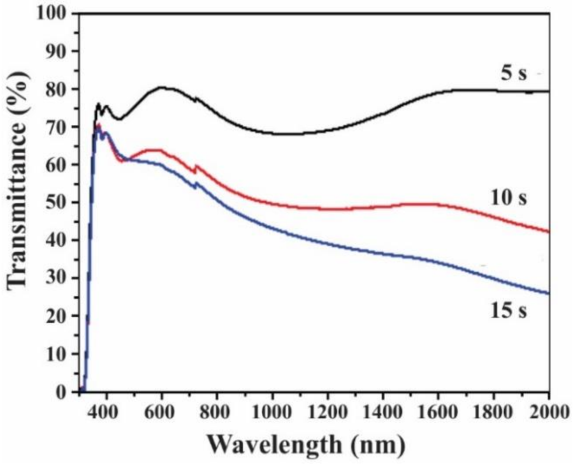

Figure 4 Transmittance spectra of Ag films deposited at 5, 10 and 15 s.

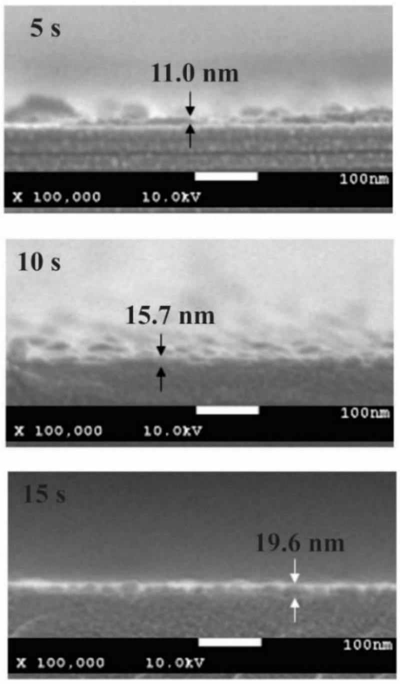

Figure 5 FE-SEM images of cross-sectional morphology of Ag films.

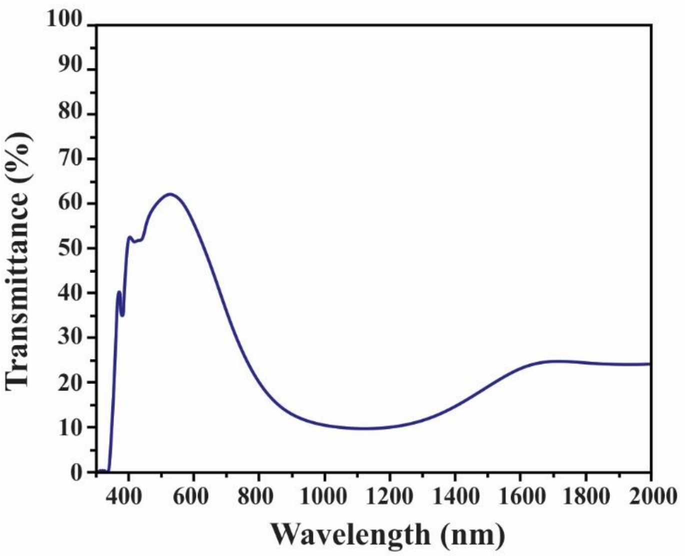

Figure 6 Transmittance spectra and of AlN/Ag/AlN film deposited with deposition times of 40 min for AlN layer and 15 s for Ag layer.

4.3 AlN/Ag/AlN films stack

After the optimum deposition times for coating AlN and Ag layers were obtained, that is 40 min and 15 s for AlN and Ag layers, respectively. Then, the multilayer AlN/Ag/AlN film stack was deposited on the glass slide for transmittance measurement and a test glass plate with a size of 10 x 10 cm^2 for infrared protection testing. The coating conditions for AlN and Ag layers in the AlN/Ag/AlN stack are the same as those of individual AlN and Ag layers.

Figure 6 shows the transmittance spectra of AlN/Ag/AlN film stack deposited on the glass substrates with deposition times of 40 min for AlN layer and 15 s for Ag layer in the wavelength range from 300-2,000 nm. The transmittance spectra were used to calculate transmittance weighed over the solar spectrum using Equation (1)

$$T(\%) = \frac{\int_{\lambda_1}^{\lambda_2} T(\lambda)\, E(\lambda)\, d\lambda}{\int_{\lambda_1}^{\lambda_2} E(\lambda)\, d\lambda} \quad (1)$$

where T(λ) is the transmittance value at the wavelength λ (obtained from transmittance spectra (Figure 6)), E(λ) is the solar spectral irradiance at wavelength λ obtained from ASTM standard number G173-03(2020) (ASTM, 2020). For visible range, λ_1 =380 nm and λ_2 = 780 nm and for NIR, λ_1 = 780 nm and λ_2 = 2,000 nm.

From Equation (1), the average solar transmittances in the visible range (λ = 380-780 nm) and in the near infrared range (λ = 780-2,000 nm) could be determined and were found to be 48.05 and 15.17 %, respectively.

4.4 Infrared protction testing of AlN/Ag/AlN film deposited on the test glass plate

The glass plate of dimension 10 cm x 10 cm x 5 mm deposited by AlN/Ag/AlN film stack at the same time of AlN/Ag/AlN deposition on the glass slide was used for infrared protection testing. For testing, a wood box with a dimension of 24 x 24 x 24 cm^3 and an open window of size 10 x 10 cm^2 was constructed as shown in Figure 7. The inner walls of the box were insulated by the ceramic fiber with a thickness of 1″ to prevent heat loss from the inside of the box. The test glass plate was placed at the opened window of the box. On the back wall also has an opened hole (ϕ = 5 cm) and instabled with a data logger (Georesearch Volcanedo, DK 323, Germany) with a dimension (diameter x height) of 5 x 3 cm^2. On the top surface of data logger, there are three sensors : temperature, humidity and illuminance sensors.

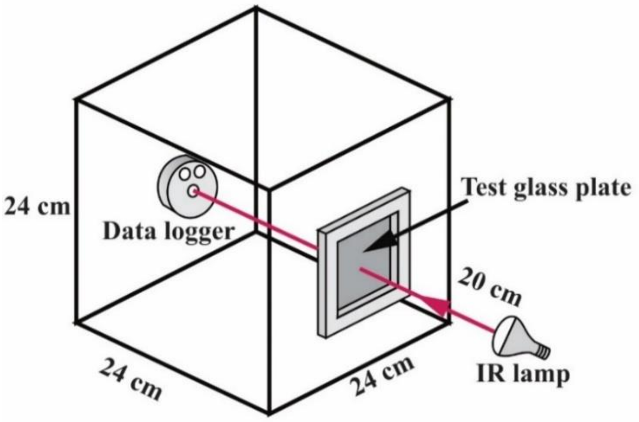

Figure 7 Experimental setup for infrared protection testing.

In the experiment, an infrared (IR) lamp, 8 A and 250 W (Philips, BR 125 IR 250) was used as an IR source. It was placed at a distance 20 cm in front of the test glass plate. The experiments were carried out on the following samples: (1) glass plate deposited with a multilayer stack of AlN 40 min/Ag 15 s/ AlN 40 min, (2) commercial glass (AGC Glass Company North America, ES 25 Pure Grey), (3) glass without deposited films and (3) no glass. For each tested sample, the IR lamp was turn on for 2 h and the temperature in the box was measured.

Figure 8 shows the variations of temperature in the box as a function of time for various tested samples. It is seen that, without glass protection (no glass) placed in front of the opened window of the box, the temperature in the box increased from room temperature (25°C) to about 60°C within 60 min. For glass plate without deposited film, the temperature in the box increased to about 45° within 60 min. For the glass deposited with AlN 40 min/Ag 15 s/AlN 40 min film stack, the temperature in the box increased to a maximum of 40°C within about 75 min and remained constant Thereafter. According to the specification data of AGC Glass Company North America on the Low-E Glass, product no. ES25 Pure Grey which was used for comparison, the transmittance in visible range and solar range are 36 and 18%, respectively (AGC Glass Company North America, 2021). In comparison with the results obtained in this work, the average solar transmittances in the visible range and in the near infrared range were found to be 48.05 and 15.17 %, respectively.

The results show that the solar transmittance in the visible range obtained in this work is higher than that of commercial glass. While in the infrared range, it is lower. It indicated that the glass plate deposited with multilayer stack of AlN 40 min/Ag 15 s/AlN 40 min prepared in this work has higher reflectance in the infrared range than that of commercial glass.

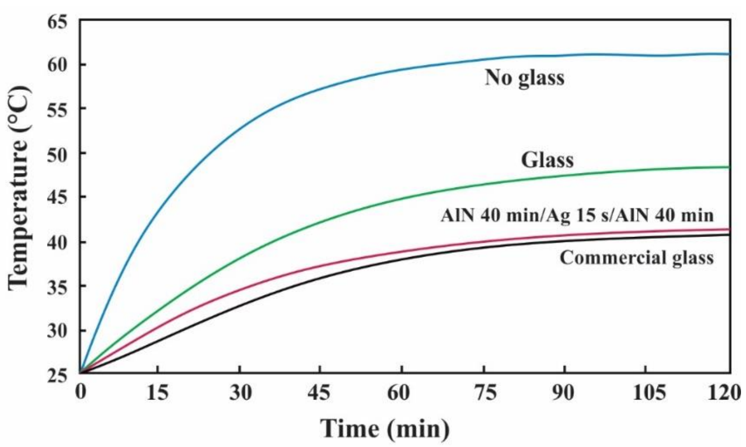

Figure 8 Variations of temperature as a function of time for various tested samples.

5. Conclusion

A home-built DC magnetron co-sputtering was used to deposit AlN and Ag films independently on Si wafers and glass slides at different deposition times of 20, 30 and 40 min, and 5, 10 and 15 s, respectively. It was found that the optimum deposition times for coating AlN and Ag films in the multilayer AlN/Ag/AlN film stack were 40 min and 15 s, respectively. Then, the multilayer AlN/Ag/AlN film stack was deposited with the above deposition times on the glass slide for transmittance measurement and a test glass plate with a size of 10 cm ×10 cm for infrared protection testing. The results showed that, for the glass slide deposited with AlN 40 min/Ag 15 s/AlN 40 min film, the average solar transmittance in the visible range and in the near infrared range were 48.05 and 15.17%, respectively. In comparison, the transmittances of deposited glass obtained in this work are comparable with those of obtained from a commercial glass.

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