Solar heat gain coefficient and heat transmission coefficient of Al-doped ZnO thin-film coated low-emissivity glass

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In this study, we investigated the solar heat gain coefficient of aluminum-doped ZnO (AZO) low-emissivity (low-e) glass. Silver-based low-e glass has excellent solar heat shading performance. However, this is undesirable in winter. When the outside temperature is low, low-e glass should allow the passage of solar heat into a building. Transparent conductive oxides can be applied for this purpose, as they transmit near-infrared light. Indium tin oxide (ITO) has already been used in this way. However, ITO is toxic, and the availability of indium is limited, making it expensive. As an alternative material, AZO is promising. We fabricated AZO films on glass substrates and measured their transmittance, specular reflectance and absorbance. The solar heat gain coefficient and the heat transmission coefficient were then calculated.

Low emissivity (low-e) glass was developed in Europe and North America and spread widely in the 1970s and 1980s. There are two kinds of low-e glass. One uses a silver-based low-e film coating on the glass and is fabricated by sputtering, and the other uses a indium-based film applied using a pyrolytic process. Silver-based low-e glass has excellent performance in infrared shading and thermal insulation.¹

Silver-based low-e films with a single silver layer (ZnO/Ag/ZnO), two silver layers (ZnO/Ag/ZnO/Ag/ZnO) and three silvers layers (ZnO/Ag/ZnO/Ag/ZnO/Ag/ZnO) have been developed. These low-e films shut out infrared and ultraviolet wavelengths and only allow visible light to be transmitted. Increasing the number of silver layers improves the efficiency of near-infrared filtering. Because silver-based low-e films block near- and far-infrared radiation, they can significantly reduce the need for air conditioning. This makes the application of these films particularly beneficial for housing in regions with a tropical climate.

Temperate climates are hot in summer and cold in winter. In the summer, as in tropical regions, silver-based low-e films perform well as an infrared shutter. This kind of low-e film is called the solar-heat-shading type. In the winter of temperate regions, the need for heating is inevitable; however, silver-based low-e films continue to block near-infrared light even when allowing it through would be beneficial, and this increases the amount of energy required for heating. Furthermore, the physical feeling of warmth from sunlight is lost by blocking near-infrared radiation. Conversely, at night, silver-based low-e films reflect far-infrared radiation back into the building, which is advantageous.

More attention needs to be paid to silver-based low-e films, which block near-infrared in winter. In temperate regions, silver-based low-e films should be replaced by an alternative film that allows near-infrared light to enter a building through the windows in the winter.

Transparent conductive oxides (TCOs), such as indium tin oxide (ITO), aluminum-doped ZnO (AZO) and gallium-doped ZnO, do not block near-infrared light. ITO is the most widely used TCO. Although the toxicity of indium was not recognized until the 1990s, recent studies have reported that the inhalation of indium powder can cause interstitial pneumonia.²,³ Moreover, indium resources are limited, and its cost is rising steeply. As a replacement TCO in optical applications, AZO is a realistic alternative, because ZnO is abundant and non-toxic. Indeed, because ZnO has an astringent effect, it is also used for the treatment of wounds.

In order to allow near-infrared light into housing in the winter, there is a precedent for ITO film being used instead of silver-based low-e film. However, the price of ITO low-e glass is relatively high, and the use of low-e ITO glass has therefore been relatively limited.

In the area of low-e glass, ITO thin films are still used for solar-heat-gain-type low-e glass. The development of

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Key-words : AZO thin film, DC sputtering, Low-e glass, Solar heat gain coefficient, Heat transmission coefficient

[Received November 25, 2019; Accepted January 23, 2020]
heat-gain-type low-e glasses using AZO films is a matter of great urgency. As noted, ZnO is abundantly available and non-toxic, as illustrated by its use as a medicine. As such, zinc oxide is a safe material. Aluminum is the third-most abundant element in the earth’s crust, after oxygen and silicon. Therefore, ITO films should be replaced by AZO films as soon as possible.

In this study, we fabricated AZO films on glass substrates and calculated their solar heat gain coefficient and heat transmission coefficient. We then investigated this AZO low-e glass for its practical applications.

Aluminum zinc oxide films were fabricated using a DC magnetron sputterer (Anelva SPF-210H) with a four-inch target (Al2O3 2 wt%) purchased from Furuuchi Chemical Corp. The sputtering power was 400 W, and deposition was carried out for 20 min. The distance between the substrate and the target was 65 mm. A quartz glass substrate (USD-300, Daiko Seisakusho Inc.) with dimensions of 20 × 20 × 1 mm was used. The surface of USD-300 quartz is hydrophobic; therefore, the substrates were immersed in 1 M NaOH solution for 30 min to make them hydrophilic. Before sputtering, the substrates were treated in acetone for 10 min using an ultrasonic cleaner, rinsed with ion-exchanged water and dried at 120 °C. In order to avoid the incidence of oxygen anions created at the target surface, the substrates were held in the off-axis position. The crystalline phases were examined using a thin-film X-ray diffractometer (Rigaku ATX-G) operated at 40 kV and 300 mA with Cu Kα (λ = 0.154056 nm) from a rotor radiation source. The Raman spectrum was obtained using a Renishaw RE04. The transmittance and the specular reflectance were determined using a spectrophotometer (U4100 Hitachi Inc.) in the wavelength range between 200 and 2500 nm. The thermal emission between 5.5 and 50 μm (between 2000 and 200 cm⁻¹) was obtained using a FT/IR-6700FV all-vacuum system fourier transform infrared spectroscopy spectrophotometer with a specular reflectance unit attached (JASCO Inc.). The reference was an Au mirror manufactured by vapor deposition. The resolution was 2 cm⁻¹ and the spectrum scan between 200 and 2000 cm⁻¹ was integrated 256 times. The detector was made from triglycine sulfate.

**Figure 1** shows the thin-film X-ray diffraction (XRD) pattern. The diffraction peak of ZnO (002) was observed and was the only diffraction peak. The AZO thin film was highly c-axis oriented.

**Figure 2** shows the Raman spectrum of the AZO thin film as deposited. This spectrum is quite similar to that in the study of Charpentier et al. The A1(LO) vibrations (570 cm⁻¹) were clearly observed. E2(high) vibrations were also observed as a weak shoulder at about 440 cm⁻¹. As described above, the vibrations that characterize the AZO crystal structure were observed.

As shown in our previous report, the thickness of an AZO film fabricated at 400 W for 20 min varies from 1000 to 1600 nm. The sputterer used was not equipped with a substrate rotation apparatus, either in this or our previous study. Accordingly, some unevenness in the thickness is inevitable. The fabrication conditions were the same as in our previous report, and the thickness of the film in these experiments will thus be similar.

Transmission in the visible light region was around 80% and began to decline at wavelengths above 1000 nm. At 2500 nm, the transmission had decreased to 10%.

The solar heat gain coefficient through the AZO film was calculated from the transmittance, the specular reflectance at wavelengths between 300 and 2100 nm and the specular reflectance at far-infrared wavelengths, between 5 and 50 μm. The procedure to calculate the solar heat gain coefficient is described in Japanese Industrial Standard (JIS) R 3106. In JIS R 3106, the calculation procedures for double- and triple-pane glasses are also described. In this study, the solar heat gain coefficient of a single pane of glass was obtained. In such a case, the calculating procedure is simple, and is described below. The heat transmission coefficient also has to be obtained. The procedures to calculate the heat transmission coefficient is described in JIS R 3107.
The following equations describe the solar radiation transmission ($\tau_e$), the specular reflectance ($\rho_e$) and the solar radiation absorbance ($\alpha_e$) of a single pane of glass.

$$\tau_e = \frac{\sum \lambda E \lambda \cdot \tau_{1,1}(\lambda)}{\sum \lambda E \lambda \cdot \Delta \lambda}$$ (1)

$$\rho_e = \frac{\sum \lambda E \lambda \cdot \Delta \lambda \cdot \rho_{1,1}(\lambda)}{\sum \lambda E \lambda \cdot \Delta \lambda}$$ (2)

$$\alpha_e = \frac{\sum \lambda E \lambda \cdot \Delta \lambda \cdot \alpha_{1}(\lambda)}{\sum \lambda E \lambda \cdot \Delta \lambda}$$ (3)

Here, $\tau_{1,1}(\lambda)$ and $\rho_{1,1}(\lambda)$ are the measurements of transmittance and specular reflectance as shown in Fig. 3, and $\alpha_1 = 1 - \tau_{1,1}(\lambda) - \rho_{1,1}(\lambda)$. The solar radiation absorbance of a single pane of glass is given by Eq. (3) and is also indicated in Fig. 3. $E \lambda \cdot \Delta \lambda$ is the adjusting weighting factor given in JIS R 3106. This factor weights solar thermal radiation according to the actual solar radiation intensity that reaches the surface of the earth, as shown in Fig. 4. The values of $\tau_e$, $\rho_e$ and $\alpha_e$ were 0.742, 0.0889 and 0.169.

In order to calculate the solar heat gain coefficient, the vertical thermal emissivity has to be measured. The vertical emissivity of the surface of float glass is given as 0.896. The vertical emissivity on the surface of the AZO thin film was obtained using the procedure described below.

The vertical thermal emissivity at room temperature in the far-infrared region is obtained between 5.5 and 50.0 $\mu$m using Eq. (4).

$$\rho_n = \frac{1}{30} \sum_{i=1}^{30} \rho_n(\lambda_i)$$ (4)

The sampling wavelength is shown in Table 1. The vertical thermal emissivity $\rho_n(\lambda)$ is obtained from Fig. 5 using Eq. (4). The value of $\rho_n$ was 0.769.

The vertical emissivity on the AZO film is expressed as

$$\varepsilon_n = 1 - \rho_n,$$ (5)

and was found to be 0.231.

The surface heat transfers out of the room ($h_e$) and into the room ($h_i$) are then calculated using the following equations.

$$h_e = h_{re} \cdot \varepsilon_e + h_{ce}$$ (6)

$$h_i = h_{ri} \cdot \varepsilon_i + h_{ci}$$ (7)

![Fig. 3. Transmittance, specular reflectance and absorbance of the AZO thin film.](image)

![Fig. 4. Adjusting weighting factor at each wavelength.](image)

![Fig. 5. Thermal radiation at normal temperature.](image)

| No. | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 | 11 | 12 | 13 | 14 | 15 |
|-----|---|---|---|---|---|---|---|---|---|----|----|----|----|----|----|
| $\mu$m | 5.5 | 6.7 | 7.4 | 8.1 | 8.6 | 9.2 | 9.7 | 10.2 | 10.7 | 11.3 | 11.8 | 12.4 | 12.9 | 13.5 | 14.2 |
| No. | 16 | 17 | 18 | 19 | 20 | 21 | 22 | 23 | 24 | 25 | 26 | 27 | 28 | 29 | 30 |
| $\mu$m | 14.8 | 15.6 | 16.3 | 17.2 | 18.1 | 19.2 | 20.3 | 21.7 | 23.3 | 25.2 | 27.7 | 30.9 | 35.7 | 43.9 | 50.0 |
Table 2. Surface heat transfer coefficients outdoor and indoor (W/m²·K)

|        | Thermal emissivity | Convection | Thermal emissivity | Convection |
|--------|--------------------|------------|--------------------|------------|
| Outdoor| $h_{e0} = 6.5$     | $h_{i0} = 12.2$ | $h_{e0} = 5.1$     | $h_{i0} = 16.3$ |
| Indoor | $h_{e0} = 6.3$     | $h_{i0} = 3.9$  | $h_{e0} = 5.6$     | $h_{i0} = 3.3$  |

Here, $\varepsilon_e$ and $\varepsilon_i$ are the hemisphere thermal emissivity of the outside of the room and the inside of the room, respectively, and their values were found to be 0.837 and 0.251. These are modified thermal emissivity values and are obtained by multiplying the vertical thermal emissivity by the coefficients listed in Table 1 of JIS R 3107. The parameters $h_{e0}$, $h_{i0}$; $\varepsilon_e$; and $\varepsilon_i$ are the thermal emissivity components, and $h_{e0}$ and $h_{i0}$ are the convectional components.

The values of $h_{e0}$ and $h_{i0}$ were found to be 17.6 (outdoor) and 5.48 (indoor) for summer and 20.6 (outdoor) and 4.71 (indoor) for winter, respectively. These values are different in the summer and the winter and are given as shown in Table 2, from Table 1 in JIS R 3106.

Finally, the solar heat gain coefficient, $\eta$, of glass with an AZO thin film was calculated using the following equations.

$$\eta = \tau_e + N_i \cdot \alpha_e$$

$$N_i = R_e/(R_e + R_i)$$

$$R_e = 1/h_e$$

$$R_i = 1/h_i$$

As shown in Table 2, there are the two sets of surface heat transfer coefficients for summer and winter. The solar heat gain coefficient in summer was 0.782, and in winter it was 0.774. These values are similar and rather high.

Commercial transparent double glazing without low-e films has a solar heat gain coefficient of 0.752. This value is quite close to the solar heat gain coefficients found in this work. Both double glazing and AZO low-e glazing are able to take in solar radiation in winter. However, AZO low-e glazing has an advantage when compared to simple double glazing. In winter, indoor objects radiate far-infrared, and the AZO film reflects this radiation, which has a peak at 10 μm, back into the building. Conversely, simple glazing absorbs far-infrared radiation and radiates it back to the outside. Therefore, AZO low-e glazing is able to conserve energy both during the day and at night.

The solar heat gain coefficient of the silver-based solar-heat-shading-type low-e film is below 0.4 and the minimum value is 0.2. These low values are favorable for shading the solar heat in the summer.

Heat transmission coefficient is another important factor as well as solar heat gain coefficient. This coefficient indicates the amount of the heat flow from the inside to the outside of the housing. For single pane glass, heat transmission coefficient $U$ is given by Eq. (12).

$$1/U = 1/h_e + R + 1/h_i$$

Here, $R$ is the thermal resistance and given by Eq. (13).

$$R = d/\Lambda = 1 \times 10^{-3} \text{ K·m}^2/\text{W}$$

Here, $d$ is the thickness of the glass and $\Lambda$ is the thermal conductivity of the glass. $d = 1 \times 10^{-3} \text{ m}$ and $\Lambda = 1 \text{ W/(m·K)}$.

From $R$, $R_e$ and $R_i$, the heat transmission coefficients were obtained by the Eq. (12). In summer, $U$ is 4.2 and 3.8 W/(m²·K) in winter.

These $U$ values in this study are close to that of the simple double glazing [2.9 W/(m²·K)]. In the case of a single pane, $U$ is 6.0 W/(m²·K). Intermediate hollow layer of the double glazing contributes to the reduction of the $U$ value largely.

In this study, $U$ is about 4 W/(m²·K) and much lower than 6.0 W/(m²·K) of a single pane. When a double glazing was constructed with an AZO low-e film deposited single pane, $U$ should be less than 2.9 W/(m²·K). As described before, AZO low-e film reflects far-infrared back into the housing.

At present, it is impossible to decide the precise $U$ value of the double glazing with AZO low-e film, because we did not construct a double glazing with AZO low-e film. This is the subject left in the future.

Acknowledgment A part of this work was conducted at Nagoya University, supported by “Nanotechnology Platform Program” of the Ministry of Education, Culture, Sports, Science and Technology (MEXT), Japan, Grant Number JPMPX09F19NU0048.

References

1) E. Ando and M. Miyazaki, *Thin Solid Films*, 351, 308–312 (2008).
2) H. Nogami, T. Shimada, S. Shoji and S. Nishima, *Respir. Investig.*, 46, 60–64 (2008).
3) O. Taguchi and T. Chonan, *Respir. Investig.*, 44, 532–536 (2006).
4) T. Horiuchi and T. Sonoda, *J. Ceram. Soc. Jpn.*, 127, 173–179 (2019).
5) Y. Li, G. S. Tompa, S. Ling, C. Gorla, Y. Lu and J. Doyle, *J. Vac. Sci. Technol. A.*, 15, 1063–1068 (1997).
6) C. Charpentier, P. Prod’homme and P. Roca i Cabarrocas, *Thin Solid Films*, 531, 424–429 (2015).
7) E. P. Zaretskaya, V. F. Gremenok, A. V. Semchenko, V. V. Sidsky and R. L. Juskenas, *Semiconductors*, 49, 1253–1258 (2015).
8) Japanese Standards Association Group, *JIS R 3016* (2019).
9) Japanese Standards Association Group, *JIS R 3017* (2019).