The influence of sealing temperature on the thermal and light properties of Low-E coating vacuum glazing

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The lateral sealing method of vacuum sealing used to fabricate Low-E vacuum glazing. The optical performance and surface morphology of the film at each sealing temperature were tested. The heat transfer rate to the central vacuum glazing was also tested. High-temperature sealing treatment may increase the density of the Low-E film surface. The visible spectral transmittance of the film and the far infrared spectral reflectance were significantly increased. Whereas the near-infrared spectral absorption ratio decreased slightly. The lowest rate of central heat transfer of vacuum glazing was 0.9 W/(m²·K) when the sealing temperature was 500°C. However, above 500°C, after the Ag film breakage and reunion to form a polymer film, the film performance degrades.

Key-words : Vacuum glazing, Low-emittance coating, Microstructure, Thermal properties, Optical properties

1. Introduction

Vacuum glazing is a new type of energy-saving transparent thermal-insulation constructive material with low heat loss and high visible transmittance.5,6 These properties give it good development potential and prospects, enabling it to become a research focus of glass processing technology.5,6,7,8 Collins and Robinson (1991) reported the fabrication of the first successful vacuum-glazing sample by using a low-melt glass that formed a hermetic edge seal at Sydney University.4) The use of a low-emittance coating on one or two glass surfaces within the evacuated gap of the vacuum glazing (Collins and Simko, 1998) increases thermal resistance.23) However, at a processing temperature above 400°C, many types of soft Low-E coatings degrade.6) Only hard Low-E coatings have been successfully employed in vacuum glazing.7,8,9 Griffiths produced a vacuum glazing using a low-temperature sealing method (less than 200°C) (Griffiths et al., 1998; Hyde et al., 2000), allowing a more functional film layer to be applied to the fabrication of vacuum glazing.9,10) The thermal properties of such glazing were studied by Fang (2006), and its heat transfer coefficient is less than 1 W/(m²·K).11,12) The low-E glass film properties, including the overall vacuum heat transfer coefficient and solar heat gain of the glass, were studied using finite element simulation and experimental methods.13,14) However, this method making employs In or In alloy as a sealing material, and due to the high prices of In, vacuum glazing of this type has not been promoted in the market.15) Therefore, to find an ideal Low-E coating both suitable and cost-efficient for the sealing process is of great significance in promoting the commercialization of vacuum glazing.

In this paper, for double silver Low-E coated glass vacuum glass preparation, the sealing temperature of the film, its transmittance in the visible spectrum, near infrared spectroscopy and far infrared absorption spectra, its spectral reflectance performance, the film surface morphology at different sealing temperatures, and its heat transfer coefficient were examined to analyse the impact of changes in temperature sealing process on the film vacuum glass optical properties and thermal properties.

2. Testing material and method

2.1 Testing material

In a Germany Leybold room, twenty-five groups of five double-cathode vacuum magnetron sputtering systems were used in the preparation of Low-E vacuum glazing with Low-E glass sheets. The vacuum chamber is controlled to less than 5.0 × 10⁻¹⁰ Pa. The working gas is argon of 99.99% purity; chromium, silver and other targets are used with purity of 99.99%; a high-purity target is used; and the spacing between target and substrate is maintained at approximately 15 cm. The substrate temperature (deposition temperature) is maintained at 400°C, and the working pressure is 0.5–0.75 Pa.

Vacuum glazing is performed with two sheets of Low-E coated soda-lime glass (800 × 600 mm, thickness of 4 mm) which includes seven chemical films and the structures are glass matrix/medium/Ag/protective shield/medium/Ag/protective shield/medium. The glass has an electric modulus of 72.45 MPa, Poisson’s ratio of 0.22, a tensile strength of 40 MPa, a compressive strength of 880 MPa (tensile, compressive strength according to limit selection), and softening temperature of near 580°C. The spacing distance between the support pillars is 40 mm; the diameter of the support pillars is 0.6 mm; the height of the support pillars is 0.2 mm; and the elastic modulus of the support pillars is 55 GPa, with a Poisson’s ratio of 0.25 and a density of 2500 kg/m³. The PbO–TiO₂–SiO₂–RxOy powder is selected as sealing solder and the sealing temperature range is from 420 to 520°C.

2.2 Testing

The vacuum glazing process is to put one clean glass pane above another clean glass pane with a pillar array and then coat around the two glass panes with welding solder, heat them in a 420–520°C vacuum environment for 15–60 min, seal around the two glass panes with welding solder while removing the water and organic materials on the glass, and form an integral unit. The
The guarded hot box calorimeter is designed as shown in Fig. 1. The guarded hot box calorimeter consists of a closed system with a test specimen, and access to the interior is provided by cold XMT-K-type thermocouple is arranged on the surface of the test specimen, and access to the interior is provided by cold XMT-3000 industrial junction compensation member controls regulators, according to the output voltage through conversion. A sample can be tested under simulated conditions of heat transfer coefficient, and the specific parameters are as follows: the temperature of the hot air case is 20°C, the cold box air temperature is -20°C, the thermal conductivity of the filler is 0.72 W/(m²·K), the cold box air speed is 3 m/s, and the test specimen size is 800 mm × 600 mm × 8.2 mm.

3. Testing result and analysis

3.1 Impact analysis of the heat transfer performance

The heat transfer of vacuum glazing includes three forms: conduction, radiation and convection. The overall heat transfer rate can be written as follows:

$$Q_{\text{overall}} = Q_{\text{conduction}} + Q_{\text{radiation}} + Q_{\text{convection}}$$

where $Q_{\text{conduction}}$, $Q_{\text{radiation}}$, and $Q_{\text{convection}}$ are the conduction, radiation, and convection heat transfer rates, respectively. The overall heat transfer rate can ultimately be expressed as the relationship between $T_1$ and $T_2$:

$$Q_{\text{radiation}} = E_{\text{effective}} A (T_1^4 - T_2^4)$$

where $\sigma$ is the Stefan-Boltzmann constant, $E_{\text{effective}}$ is subject to effective emissivity, and wavelength affects the whole area of the two surfaces. Approximate calculation of the effective emissivity of a preferred vacuum glazing sample, the conducting vacuum layer and the effective emissivity, and wavelength affects the whole area of the two surfaces. Approximate calculation of the effective emissivity can be expressed as:

$$E_{\text{effective}} = (\varepsilon_{1,\text{hemispherical}} + \varepsilon_{2,\text{hemispherical}} - 1)^{-1}$$

Due to slight differences in the surface temperature, the radiation conductivity can be expressed as:

$$C_{\text{radiation}} = 4E_{\text{effective}}\sigma T^3$$

where $T$ is the average temperature of the surface. The heat conduction of the vacuum glazing’s entire support pillar array is as follows:

$$C_{\text{glass-glass, center-of-glazing}} = k_{\text{glass}} A / p^2$$

where $p$ is the distance of an array of support pillars.

The radiation emission rate is considered to be unrelated to the wavelength of the inner radiation and surface temperatures. Without considering the impact of these factors, for vacuum glazing without Low-E coatings, the maximum error is 4%. In a preferred vacuum glazing sample, the conducting vacuum layer has several effects on radiation heat transfer, and the overall heat transfer compared to the level of heat transfer of the glass can be negligible. Therefore, the total heat transfer rate can ultimately be expressed by Collins as follows:

$$C_{\text{glass-glass, center-of-glazing}} = 0.8 P_{\text{r}} + 4E_{\text{effective}}\sigma T_{\text{average}}^3 + ak_{\text{glass}} A / p^2$$

Vacuum glazing overall heat transfer coefficient can be measured using a guarded hot box calorimeter, as described by Fang et al. (2006):

$$h_{\text{out}}(X_m) = 0.029 P_{\text{r}} 0.43 \left(\frac{H}{W}\right) 0.8k$$

Figure 3 shows the test results for the Low-E glass vacuum heat transfer coefficient at different sealing temperatures. When the sealing temperature is below 400°C, the heat transfer
The coefficient of the Low-E coated vacuum glazing is 1.6 W/(m²·K). Due to the edge solder used at temperatures above 400°C, high-temperature sealing on Low-E film must be considered. In fact, the heat transfer coefficient, with increasing temperature, dropped and then experienced a sharp increase during the process, rather than increasing directly as a result of failure of the coating. For a heat sealing temperature of 500°C, the thermal transmittance value of the minimum vacuum glass was 0.92 W/(m²·K).

3.2 Effect of the optical performance

3.2.1 Impact on the performance of the visible spectral transmittance

Figure 4 shows the visible spectral (380–780 nm) transmittance curve of Low-E vacuum glazing at different sealing temperatures. The sealing temperature has a significant influence on the Low-E vacuum glazing visible light transmittance, and its trends are essentially the same with the change of wavelengths. Compared with non-high-temperature sealing of Low-E glass of the original film, Low-E vacuum glazing transmitted a significantly increased range of light wavelength. The maximum under different sealing temperatures all occurs at a wavelength of visible light transmittance of approximately 500 nm and can even reaches 62%. It can also been seen that Low-E vacuum glazing shows improved visible light transmittance within the wavelengths range of 380–600 nm. The visible spectral transmittance of the Low-E vacuum glazing at wavelengths of 500 nm or less first increased and then decreased with increasing sealing temperature. The sealing temperature with the maximum transmittance of the visible spectrum was 500°C. For wavelengths greater than 680 nm, Low-E vacuum plate glass transmittance in the visible spectrum was also reduced after the initial increasing trend with higher sealing temperatures. The sealing temperature of 500°C minimized the overall visible spectral transmittance. Thus, the transmittance of Low-E vacuum plate glass to the whole band of visible light is most sensitive at a sealing temperature of approximately 500°C.

3.2.2 Impact on the near-infrared spectral absorption rate

Figure 5 shows the near-infrared absorption spectrum (780–2500 nm) graph of Low-E vacuum glazing at different sealing temperatures. The figure shows the effect of the sealing temperature of the Low-E vacuum glazing on near infrared absorption. The same trend appears throughout but becomes more pronounced with increasing wavelength. The infrared spectrum in the wavelength range of 780–1600 nm absorption wavelength varies linearly, and the growth rate slows in the 1600–2500 nm wavelength range. Compared with non-high-temperature sealing of Low-E glass of the original film, high-temperature Low-E vacuum glazing decreased the near infrared absorption rate at different wavelengths. In the wavelength range of 780–1750 nm, the influence of the sealing temperature on Low-E plate vacuum glazing transmittance of the visible spectrum is not clear with the increase of wavelength. In the 1750–2500 nm wavelength range, the Low-E vacuum glazing spectral transmittance was significantly decreased only while the sealing temperature kept around 440 or 480°C, and spectral transmittance appeared fluctuation phenomenon to a certain extent at the wavelength of 2300 nm. The observed effect of the sealing temperature within the rest of the range of variation on Low-E vacuum glazing spectral transmittance is not obvious.

3.2.3 Impact on the mid-infrared and far infrared spectral reflectance properties

Figure 6 shows the Low-E vacuum glazing mid-infrared and far infrared spectrum (2500–25000 nm) graph of the reflectance at various sealing temperatures. The figure shows that the sealing temperature had no effect on the mid-infrared and far infrared spectral reflectance of Low-E vacuum glazing. Compared with
The reflectivity curve shows basically the same trend. At wavelengths of 2500, 5000–7000, 15000 and 24000 nm, four reflection peaks appeared, with a particularly clear peak at 5000–7000 nm, which means that Low-E vacuum glazing has good reflectivity within these wavelengths. For all wavelengths of infrared light, the mid-infrared and far infrared spectral reflectance rate of the Low-E vacuum glazing has increased significantly. Especially, the reflection peak at 6000 nm was twice as high as the standard background infrared reflectivity, at 100%. Without high temperature sealing of the Low-E glass sheet, reflective double peaks were 94 and 95.2%, because high temperature processing improved the coating density. Although different sealing temperatures exert no obvious influence on the rate of increase, for the performance of a specific wavelength in the infrared reflection spectrum, some fluctuations in the reflectivity of wavelength between the 15000 and 16000 nm appeared, and the reflective double peak reached 99% at the temperature of 520°C. This is due to further refinement of the coating with the increase of sealing temperature and reflection effect of mid-infrared and far infrared enhanced for the sharp increased grain boundaries when the temperature reached 520°C.

3.3 Analysis of impact on film properties

Figure 7 shows SEM images of the Low-E vacuum glazing surface for different sealing temperatures. As shown in Fig. 7(a), the Low-E glass sheet surface without high temperature sealing exhibits an uneven coating particle distribution. Random-shaped grains and crystals also appears and the main sizes are between 0.2–0.5μm. The particles in the nearby area have grown significantly and show aggregation phenomenon. As the sealing temperature increases through the range of 420–500°C, the Low-

Fig. 6. Mid-infrared and far infrared reflectance curve of Low-E film at different sealing temperatures.

Fig. 7. Microstructure of the Low-E coating sealed at different temperatures.
E coating surface of the vacuum glazing undergoes dense refinement. The size becomes very consistent with each less than 500 nm, and few agglomerates of larger diameter appear. Figures 7(b) and 7(c) show that as the sealing temperature further increases, the Low-E vacuum glazing coating particles are further refined, and when it reaches 520°C, there appears to be certain heat-affected area, dark spots and many small surface cracks, as shown in Fig. 7(d).

Thus, through the Low-E vacuum glaze sealing process for preparing high-temperature glass surface coating, the particles can be refined to the same size, and the coating can be made more dense the coating to eliminate voids and gaps at junctions of different sized particles, thereby reducing the unit grain boundaries within the area, reducing the grain boundary scattering effect of the near-infrared spectrum, and improving the performance and visible spectral transmission properties along with far infrared spectral reflectance. In addition, refining the dense coating particles improves the coating strength and corrosion resistance of the Low-E vacuum glazing to extend its life. However, at excessively high sealing temperatures, the linear expansion coefficients of the glass and film are not the same, and excessive thinning makes the coating particles easy to crack during cooling, thus affecting the properties of Low-E vacuum glazing with regard to light and heat.

High-temperature sealing produces certain chemicals through the physical changes caused by proliferation and migration between the coating layer and the glass substrate, which can affect the thermal performance of Low-E vacuum glazing optical elements. The elements of the migration process, the proliferation and elements in the coating chemical and the physical reactions at the interface, generate new material and new chemical bonds that improve the thermal performance of the optical coatings to a certain extent. However, the excessive formation or migration of new substances will adversely affect performance. In the coating used for Low-E vacuum glazing of SnO$_2$-Si$_2$N$_4$-Ag, a glass substrate used for an ordinary soda lime glass, the main components are Si, Na, Ca and O. Si is common to both elements, shown in Fig. 8. When the sealing temperature is below 480°C, the change in the content of all elements is not substantial, and no significant diffusion and migration between the plated layer and the glass matrix elements occurs, resulting in high thermal stability resistance. When the sealing temperature exceeds 480°C, Si decreases, but the content of the other elements does not change substantially, indicating that Si has undergone significant diffusion and migration between the plated layer and the glass substrate. The migration of Si will cause failure of the coating performance, thus affecting the thermal performance of Low-E light vacuum glazing.

4. Conclusion

Low-E vacuum glazing samples were prepared via the side vacuum sealing method at different temperatures. The optical performance at each sealing temperature and the surface morphology of the film coatings were tested. The results showed that reasonably high-temperature sealing treatment increased the density of the surface of the Low-E film and the visible spectral transmittance of the film and significantly increased the far infrared spectral reflectance. Whereas the near-infrared spectral absorption ratio decreases slightly as the sealing temperature increases, the Low-E vacuum glazing surface coating undergoes thinning of dense particles: the size becomes very consistent, improving the optical performance of the Low-E glazing. The transmitted light wavelength range increased significantly, improving in the wavelength region of 600 nm or less. The maximum wavelength of visible light transmittance appears in the region of 500 nm, and visible light transmittance is the most sensitive for a sealing temperature of approximately 500°C. However, at temperatures above 500°C, the Ag film underwent breakage and reunion to form a polymer film, and the film performance decreased. The central heat transfer rate from the vacuum glazing also exhibited a sharp decline and then rise in the process. The heat transfer coefficient reached a minimum of 0.92 W/(m$^2$·K) at a sealing temperature 500°C. Thus, low-E vacuum glazing in the sealing process using a suitable sealing temperature can improve the thermal performance of the optical functional film to a certain extent.
Acknowledgements The authors acknowledge the support from the National Natural Science Foundation of China (No. 51372216, 51302242), the Natural Science Foundation (No. BK2012530) of Jiangsu Province, and Science and Technology Plan Project (No. 2014-10) of Yangzhou.

References
1) R. E. Collins, G. M. Turner, A. C. Fischer-Cripps, J.-Z. Tang, T. M. Simko, C. J. Dey, D. A. Clugston, Q.-C. Zhang and J. D. Garrison, Build. Environ., 30, 459–492 (1995).
2) Y. Fang, T. J. Hyde, F. Ayra, N. Hewitt, P. C. Eames, B. Norton and S. Miller, Renew. Sust. Energ. Rev., 37, 480–501 (2014).
3) R. Zhang, Beijing: China Agricultural University (2005).
4) R. E. Collins and S. J. Robinson, Sol. Energy, 47, 27–38 (1991).
5) R. E. Collins and T. M. Simko, Sol. Energy, 62, 189–213 (1998).
6) B. Lu, X. Xu, X. Yu and Z. Fan, Acta Optica Sin., 30, 283–286 (2010).
7) D. Zhang, M. Si, R. Liu and Z. Liu, Spectros. Spect. Anal., 32, 2438–2441 (2012).
8) S. Zhang, Y. Tan and K. Liang, Acta Metall. Sin., 47, 1147–1152 (2011).
9) P. W. Griffiths, M. Di Leo, P. Cartwright, P. C. Eames, P. Yianoulis, G. Leftheriotis and B. Norton, Sol. Energy, 63, 243–249 (1998).
10) T. J. Hyde, P. W. Griffiths, P. C. Eames and B. Norton, Proceedings World Renewable Energy Congress VI (WREC2000), Brighton, UK, 271–274 (2000).
11) Y. Fang, P. C. Eames, B. Norton and T. J. Hyde, Sol. Energy, 60, 564–577 (2006).
12) Y. Fang, P. C. Eames, B. Norton, T. Hyde, J. Zhao, J. Wang and Y. Huang, Sol. Energy, 81, 8–12 (2007).
13) N. Ng, R. E. Collins and L. So, Int. J. Heat Mass Tran., 49, 4877–4885 (2006).
14) N. Ng, R. E. Collins and L. So, Mater. Sci. Eng., 138, 128–134 (2007).
15) J. Zhou, Y. Ren, Z. Fu and C. Wang, J. Build. Struct., 28, 104–108 (2007).
16) Q.-C. Zhang, T. M. Simko, C. J. Dey, R. E. Collins, G. M. Turner, M. Brunotte and A. Gombert, Int. J. Heat Mass Tran., 40, 61–71 (1997).