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Polymer films with multilayer low-E coatings

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ABSTRACT

The paper presents the experimental results on depositing a multilayer low-emissivity (low-E) coating with oxide–metal–oxide structure on polyethylene terephthalate (PET) and polyethylene (PE) films by magnetron sputtering. The TiO_2/ZnO:Ga/Ag/ZnO:Ga/TiO_2 coating on the PET film with high water-resistance and capability to be used outside of sealed double-glazed panes was proposed. The optimal thickness of coating layers was experimentally determined. The coating with the optimal structure has 82% transmittance over the visible spectrum and 91% reflection over the infrared spectrum. The window with a PET film and low-E coating was investigated in terms of heat engineering. It was revealed that heat transfer resistance increased up to 0.73 $m^2\,°C\,W^{-1}$ for the windows with a PET film and low-E coating. Heat transfer resistance of the windows without a polymer film was 0.38 $m^2\,°C\,W^{-1}$. The water-resistant ZnO:Ga/Ag/ZnO:Ga/SiO_2 coating on a PE film with 77% transmittance and 91–92% reflection in the IR range was proposed to be used as greenhouse covering material. The possibility of using the PE film with a low-E coating to reduce heat loss in greenhouses and enhance yielding capacity was demonstrated.

1. Introduction

Nowadays high and rising energy prices, limitations in energy supply, and growing concerns about climate changes are daunting challenges. Thin film technologies are of great importance in dealing with these issues. In particular, one of the energy saving technologies is the deposition of spectral-selective coatings. It is known that about 40% of heat losses in residential buildings and industrial facilities occur as a result of leakage through the transparent constructions (windows, balconies, etc.). In addition, approximately 2/3 of such heat loss is due to radiation, and the rest part is the result of heat conduction. At the same time, architectural windows and glass facades are primarily designed to provide apparent visual contact between indoors and outdoors, together with the day-lighting which must be kept at the appropriate level. Therefore, window areas are not made too small. The modern architectural trends tend to enlarge the window area, so the energy issue may become even more pressing in the future.

To prevent outward heat loss from buildings, multilayer coatings with low infrared emissivity (low-E) are widely used for architectural glazing [1]. Low-E coatings are typically deposited on glasses for architectural applications to the effect of high transmittance in visible range and, at the same time, high near-infrared reflectivity. Deposition is mostly performed by magnetron sputtering of multilayer coating, generally comprising at least one conductive metallic layer (typically silver) with a thickness of about 8–12 nm. Other highly conductive metals, such as Au and Al could be used as metal layers in low-E coatings. However, Ag has the lowest absorption coefficient (5%), as compared to that of 8 and 30% for Au and Al, respectively [2]. Heat-reflecting properties of a multilayer coating can be improved by

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increasing the number of Ag layers, as in the TiO_2/Ag/TiO_2/Ag/TiO_2 structure [3].

One of the major issues in silver application is poor atmospheric stability of a pure thin Ag layer [4]. Moisture was found to intensify the Ag atom migration in the film resulting in their agglomeration. This is one of the reasons why the silver layer is usually covered by dielectric protective layers such as oxides of zinc, tin or titanium with a thickness below 40 nm and a refractive index preferably more than 2 [5]. Dielectric layers may perform antireflective functions in the visible spectral range and/or protect the metallic layer from chemical and mechanical damage. In addition, they act as adhesion layers, nucleation templates, and diffusion barriers. Further components such as titanium, nickel, and chromium alloys are applied as sacrificial layers with a thickness of <5 nm to protect the metallic layer from oxidation during deposition of dielectric layers in oxygen-containing plasma.

Ag-based thin films for energy-efficient glazing are now highly optimized, and a great number of products with specified thermal and solar properties are available in the market. The aim of this work was to produce a multilayer low-E coating on polymer films with sufficient environmental durability. Transparent polymer films are very important substrates for low-E coatings, and basically, can substitute glass. Polyethylene terephthalate (PET) film excels in having an exceptional combination of optical, physical, mechanical, thermal, and chemical properties. Therefore, they are often used as a substrate for a variety of processing technologies, such as vacuum metalizing, coating and laminating. PET films exhibit dimensional stability and superior optical properties: excellent light transmittance (92%) with low haze ($<0.1\%$). Polyethylene terephthalate film can be suspended between glass panes and used in practical window constructions.

Vacuum coating of polymer films in roll-to-roll coaters became highly instrumental long ago. This is clearly evident from the annual capacity of vacuum web coating, which amounts to 10^{10} m^2 [6]. The advantage of using polymer films as a substrate for multilayer coatings results from their flexibility, low weight, and impact strength. However, it should be noted that the initial film growth on a polymer surface tends to be different from that on glass surface. For example, the In_2O_3:Sn film deposited onto the acrylic-coated PC became conducting when the film thickness exceeded 14 nm, whereas the same value for the In_2O_3:Sn film on glass was 4 nm [7]. Despite a number of difficulties concerning the fabrication of low-E coatings on polymers (difference in mechanical properties between films and polymer substrates, adherence of a film to a polymer substrate), deposition at low substrate temperatures onto PET is feasible. The CdO and ZnO bilayers, SnO_x, $(In_2O_3)_{1-x}(ZnO)_x$, and In_2O_3–ZnO–SnO_2 films were deposited on PET substrates [8–11].

In the context of the study, the TiO_2/ZnO:Ga/Ag/ZnO:Ga/TiO_2 and ZnO:Ga/Ag/ZnO:Ga/SiO_2 sandwich structures were chosen as a low-E coating on PET and PE substrates, respectively (Fig. 1). Titanium dioxide has a function of anti-reflection layer, due to the high refractive index of 2.5. The authors propose using the gallium doped zinc oxide (ZnO:Ga) thin films instead of metal barrier layers to protect an Ag layer during magnetron sputtering of TiO_2. Even the sub-nanometer-thick metal barrier layers noticeably decrease the coating transmittance. The ZnO:Ga films do not reduce transmittance so significantly and they could be deposited by magnetron sputtering of the ZnO:Ga_2O_3 cathode in an Ar atmosphere, which prevents Ag oxidation during the sputtering process. Moreover, ZnO-based films are known to be sensitive to humidity arising from reactions of water molecules with oxygen vacancies [12]. SiO_2 was selected as a protective water-resistant layer for the films on PE substrates.

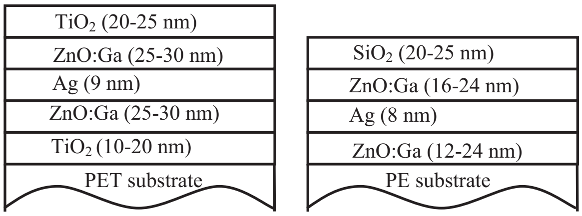

Fig. 1. Scheme of low-E coatings structure on PET (a) and PE (b) substrates.

2. Material and methods

The vacuum web coater consisted of a cylinder vacuum chamber, 2.2 m long and 1.2 m in diameter, three extended magnetron sputtering systems, an ion source with a closed electron drift and a winding system. Magnetrons with cylindrical rotating cathodes (1600 mm long and 89 mm in diameter) were used for TiO_2 and Ag deposition. Such magnetrons have a high efficiency of target utilization (up to 80%). For the ZnO:Ga film deposition a planar magnetron was used, since the fabrication of a tubular ceramic cathode is economically inefficient. A planar ZnO:Ga_2O_3 (3.5 at%) target has an area of 1600×140 mm^2. For the SiO_2 layer deposition the cylindrical magnetron with a Ti target was replaced with a planar one with a Si target. The thickness non-uniformity in the deposited coating over a length of 1200 mm was ±2–5%. The coatings were deposited at room temperature on a 100 μm-thick PET film (Tasma Ltd, Russia) and on a 100 μm-thick polyethylene (PE) film, traditionally used in greenhouses. Samples sized 400×200 mm^2 were used to find the optimal structure and investigate the low-E coating characteristics. The low-E coating samples sized up to 1.2×40 m^2 were fabricated for full-scale testing in transparent constructions of buildings and greenhouses.

Prior to the coating deposition, the substrate was cleaned by an ion beam at a discharge voltage of 1.8 kV and discharge current of 200 mA. The polymer film was moved at a speed of 2.7 m/min. The TiO_2 layer was deposited in the atmosphere of Ar and O_2 at a pressure of 0.3 Pa, in pulsed regime with a frequency of 1 kHz and at the average discharge power of 10 kW. The thickness of TiO_2 layers was varied in the range of 6–50 nm. The ZnO:Ga layer was deposited in a DC regime with a discharge power of 2 kW at the Ar pressure of 0.13 Pa. The discharge power was limited by the poor heat removal from the ceramic target. The thickness of these layers was varied from 12 to 60 nm. The Ag layer was also deposited in an Ar atmosphere in pulsed regime at a frequency of 5 kHz with the average discharge power about 2 kW. Our previous studies of the ultra-fine Ag films deposition on architectural glass revealed that the films deposited by magnetron