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Controlled Micro-wrinkling of Ultrathin Indium-Tin-Oxide Films for Transparency Tuning

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Abstract

Smart windows can electrically switch between clear and opaque states. Current smart windows based on polymer dispersed liquid crystal are expensive and they have moderate range of transparency tuning. Elastomeric tunable window devices are being researched as the low-cost alternates. They consist of a transparent elastomer substrate with surface electrodes that provide electrically controlled micro-wrinkling. They diffusely scatter the transmitted light and thus appear opaque when the surfaces are micro-wrinkled. On electrical activation the wrinkles are flattened, thus making the windows transparent like window blinds. However, the initial prototypes of these electrically tunable window devices showed limited transparency tuning because their transparent electrodes cannot be completely flattened. For example, the brownish e-beam evaporated indium-tin-oxide thin films (50 nm thick) remains mildly wrinkled (with 52.08% transmittance) even when subjected to 37% areal expansion, while its opaque state allows 39.14% transmittance. There is a need for more transparent thin-film electrode with better controllability of surface micro-wrinkling. This work reports a greatly improved tunable window device with enlarged range of transmittance tuning: a clear state of 71.5% transmittance and an opaque state of 2% transmittance. This new device made use of ultra-thin (6 nm) ITO thin films as the transparent compliant electrodes, which were initially wrinkled and can be flattened by 12.2% voltage-induced areal expansion. These ultra-thin ITO thin films are clearer with fewer thermally-induced wrinkles on the flat elastomer substrate (VHB 4905) as they were deposited at a lower surface growth temperature using the RF magnetron sputtering technique. In addition, they make compliant electrodes of higher electrical conductivity and can electrically unfold the mechanically induced micro-wrinkles by a small voltage-induced areal expansion (~12.2%). With the greatly enhanced performance, this electrically tunable window device is promising approach for low-cost smart windows.

Key words: smart window, dielectric elastomer actuator, wrinkling, transparency tuning, transparent compliant electrodes

1 Introduction

Smart windows, which can adjust the light transmittance between opaque and transparent mode by responding to external stimuli such as light, heat, and voltage, have attracted significant attentions due to their wide potential applications including energy savings and personal privacy demands for architectural or vehicle glass windows, skylights, doors, and internal partitions; light harvesting for solar cells; light diffusion for light emitting diode (LED) devices and backlight units of liquid crystal displays

(LCDs)^{1,2,3,4,5}. To date, the solutions of optical modulation in windows or coating layers are achieved by external-stimuli-triggered change in morphology and/or chemistry to result in the switchable optical properties, such as suspended particle devices (SPD)⁶, electrochromic (EC) smart glasses⁷, polymer dispersed liquid crystal displays (PDLC)⁸, and chromogenic materials, triggered by electrical field⁹, temperature¹⁰ and light². However, these solutions still have known issues. The first issue is the low off-state transmittance, for example, the highest transmission of SPD being not more than 60%. The second issue is the life time stability. Many of these active materials in smart windows are chemically unstable during switching process. The third issue is the need for complicated manufacturing processes for the complex device assembly. The fourth issue is the high cost of existing smart windows due to the use of expensive chemicals and high manufacturing overheads. Up to 2014, the electrochromic smart window costs high for \$58/ft².¹¹ Therefore, a facile and low-cost material handling and fabrication process is highly desired to make smart window practical for large-area durable transparency switching and rapid response.

Recently, the tunable window devices based on elastomers are developed as the low-cost alternatives of smart windows.^{12, 13, 14, 15} These devices are Dielectric Elastomer Actuators (DEAs) in a capacitor configuration which is composed of the elastomer membrane sandwiched between a pair of thin compliant (highly stretchable and soft) electrodes. These smart windows are opaque when their initial surfaces are micro-wrinkled to scatter transmitted light; they appear clear when the surfaces are flattened upon electrical activation by a compressive Maxwell force. Therefore, this device can tune transparency by scattering light through rough surface.

However, these electrically tunable prototype window devices still face the challenges of the limited transparency tuning range. The electrodes of these devices are often made of thin metal such as gold¹⁶ or other transparent materials such as silver nanowire¹⁷ or carbon nanotube network.¹⁸ The highest available transmittance in these devices is <60% due to the high reflection of the metal layer or the high light scattering due to the as-grown haze in nanowire network. Transparent conductive oxides (TCO) thin films such as Indium tin oxide (ITO) and Aluminum doped Zinc Oxide (AZO) can be the better alternatives of stretchable electrodes on elastomer substrate. TCO can overcome the low transmittance issue owing to the wide optical bandgap (> 3 eV)¹⁹. ITO is one of the most common TCO materials and is widely used in optoelectronic devices due to its low resistivity ($\sim 10^{-5} \Omega\cdot\text{cm}$) and high transparency in visible light range ($\sim 90\%$) due to its wide bandgap (~ 4 eV)¹⁹. ITO has also been used in the optically switchable smart window devices in our previous work¹². However, brittle ITO thin film has high risk to form cracks under a large compressive stain. As ITO film cracks, the electrode resistance dramatically increases and the micro-wrinkles are released from the stress relief. This has a negative impact on the reversible electrical tuning of the device transparency.

In our previous work, smart window device was developed with 50-nm-thick compliant ITO film coated on adhesive poly-acrylate dielectric elastomer (3M VHB4905) substrate by electron-beam (e-beam) evaporation method. The electrical unfolding converts the translucent state of DEA-based smart window, with the $14.2\% \times 14.2\%$ initially crumpled ITO thin-film electrodes, into transparent state, with transmittance increased from 39.14% to 52.08%¹². No cracks are observed in the wrinkled ITO film within 25% bi-axial compressive stain in the wrinkled ITO coating, which is attributed to nanometric film thickness and the very high bond with the VHB substrate. However, the present smart window still faces the following challenges. (1) The major issue is the limited transparency tuning range because the electrodes cannot be completely flattened. This is attributed to the mild wrinkles formed by the radiation heating of thermal sources in the e-beam evaporation.²⁰ (2) The ITO electrode on VHB membrane exhibits the extraordinarily high sheet resistance of $2.15 \text{ M}\Omega/\square$, which may affect the electrical activation of the device. (3) The 37% areal expansion for the mediate transparency tuning is too large and impractical for a real window of constant area. Therefore, the alternative method for ITO deposition is highly required. RF magnetron sputtering is the most common technique to deposit the highly conductive and transparent ITO thin films at low substrate temperature¹⁹. Furthermore, RF magnetron sputtering also features the low self-heating effect on the substrate and low damage on the substrate surface. This work reports the greatly improved transparency turning range (2%~71.5%) of window device under 12.2% voltage-induced areal expansion when ultra-thin (6 nm) ITO thin-film transparent and compliant electrodes are deposited by RF magnetron sputtering.

2 Experimental Section

Figure 1 illustrates the process flow of the fabrication of DEA-based smart window device. The process flow is divided into the follow steps^{19, 23, 24}. (1) A membrane of polyacrylate elastomer (3M, VHB 4905) is attached and fixed on mechanical radial stretcher. (2) The VHB substrate is equi-biaxially pre-stretched by 1.78×1.78 times by the stretcher. (3) The double surfaces of pre-stretched elastomer membrane are coated with 6-nm-thick ITO thin films by 13.56 MHz RF magnetron sputtering. A RF magnetron sputtering source is used to sputter 4-inch ITO target with a composition of 90% In_2O_3 and 10% SnO_2 by weight and deposit ITO thin film on the pre-stretched VHB substrate at room temperature. The ITO electrode is fabricated under a RF power of 50 W, a process pressure of 0.6 Pa, and the Argon mass flow of 20 sccm while the target-to-substrate distance is kept around 10 cm. (4) the ITO-coated elastomer substrate is relaxed from the initially pre-stretched state by using the mechanical radial stretcher to generate wrinkles on both surface of the device. The absolute radical compression strain (c), which is defined as $c=(1-D_2/D_1)$, of DEA-based window device is used to estimate the compression state of device compared with its initially planar state without the intentional crumpling. (5) ITO thin-film electrodes of

DEA-based smart window are connected to the high voltage power supply (Trek, 610E) via thin aluminum foils. A voltage ramp at a 1 kV voltage step is provided to activate DEA-based smart window. The relative areal expansion strain of electrode (s_A) of DEA-based window device is defined as $s_A = (1 - D_2/D_3)$, which is used to estimate the voltage-induced expansion of electrode area of DEA-based device compared with the initial un-active state. The surface microstructure of ITO thin film on elastomer is characterized by optical microscopy. The in-line transparency of smart window is measured by UV-Vis spectrometer (PerkinElmer, Lambda 950) in the wavelength range of 400-800 nm. The 6-mm-diameter head of cable connected to spectrometer is 14 cm far away from the surface of device to meet the standard specifies that a specular in-line transmission should have a maximum spread of 2.5° . The resistance of ITO thin film is measured by using a multi-meter (Agilent, 34410A). The photos of smart window devices are taken by digital single lens reflex camera (Canon EOS 550D).

Figure 1 (a) Process steps for the fabrication of smart window device; (b) The schematic diagrams of light paths in the component or complete structure of smart window device.

3 Results and Discussion

The poor transmittance of the as-deposited ITO electrode on VHB substrate is one of the main issues for the required large tuning range of smart window. As mentioned above, the low transmittance is attributed to the uneven surface with micro-wrinkling induced by thermal effect during the ITO deposition process. Figure 2 (a) displays that the highly transparent ITO electrode deposited by RF magnetron sputtering at room temperature. The smart window with double-side 6-nm-thick ITO electrodes exhibits a transmittance 72.5% at 550 nm wavelength, as shown in Figure 2 (c). Figure 2 (b)

shows that the surface morphology of the as-deposited smart window. The surface of device shows a relatively flat state with the slight micro-wrinkles, yielding its initially high transmittance. This relatively flat surface can be attributed to the low self-heating effect of ITO thin film deposition in RF magnetron sputtering system. It was found that the substrate temperature would rise slowly from 25 °C to 60 °C in 10 min (measured with inbuilt thermocouple) during the deposition process. The deposition time in the present work is 30s so that the increase in substrate temperature can be negligible. The radiation in RF magnetron sputtering, which is mainly from the Argon plasma whose spectrum is mainly in the range between UV and near infrared, is much weaker than the heating radiation in e-beam evaporation.²⁰ The secondary electrons generated in the plasma of RF magnetron sputtering are trapped in cycloidal orbits near the target area and thus avoid self-heating to the substrate, leading to the relatively low substrate temperature during the deposition process²¹.

Figure 2 The photo image (a), surface morphology (b), and in-line transmittance spectrum (c) of as-grown 6-nm-thick ITO thin-film electrode on double side of DEA-based smart window.

The electromechanical activation of DEA-based smart window device with the initially crumpled ITO thin-film electrodes doesn't occur at the low voltages below a threshold voltage of 5 kV, corresponding to electrical field of ~32 MV/m, is applied. Figure 3 shows the photo images of DEA-based smart window at different driving voltages. The in-active smart window with translucent appearance at initial crumpling shows a translucent state, as shown in Figure 3 (a). As the driving voltage increases, the areal expansion of VHB unfolds the initially crumpled ITO electrodes. Thus, the image

beneath smart window device can be seen clearly when the device is optically switched from translucent state to transparent state.

The absolute radical compression strain (ϵ) of smart window with electrical activation gradually decreases with the increasing voltage-induced areal expansion from the initial 5.45% at 0 V to the final value of -0.17%, which indicates the small tensile strain, at 7 kV, as shown in Figure 4. The 5.6% change of the absolute radical compression strain here is much smaller than the value of 14.2% in the previous work.

Figure 3 The electromechanical activation of DEA-based smart window devices with the crumpled ITO thin-film electrodes: (a) the photo of in-active state of smart window with translucent state at initial crumpling; (b)~(d) the photos of active states of smart window with areal expansions of ITO electrodes and increased optical transmittance by unfolding the crumpled ITO electrodes.

Figure 4 shows that the relative areal expansion strain of electrode slowly increases with the increase in driving voltage. When the voltage is beyond the threshold voltage, this areal strain dramatically increases up to 12.2% at 7 kV. To our knowledge, this areal strain required for the transparency switching is much smaller than that reported 37%~100% for the pervious devices based on carbon nanotubes¹⁸, graphene²², and the e-beam evaporated ITO thin film.¹²

Figure 5 displays the surface topographies of the initially crumpled smart window at different driving voltages. As seen in insert of Figure 5, the initially crumpled surface with the 5.45% absolute radical compression strain exhibits the micrometer-scale random wrinkles. With the increases in voltage-induced electrical activation, the wrinkles are unfolded gradually and the density of wrinkles decreases, as shown in Figure 5. However, the wrinkles on VHB substrates have the memory effect of mechanically

induced wrinkling and cannot completely unfold the initially crumpled surface into a flat one. In addition, no cracks or other fractures are observed within the 0~5.45% radical compression strain and even the 0.17% radical tensile strain at 7 kV.

Figure 4 The dependence of relative areal expansion strain of electrode (s_A) and absolute radical compression strain (c), which are defined as $s_A=(1-D_2/D_3)$ and $c=(1-D_2/D_1)$, respectively, of DEA-based window device on the driving voltage. Relative areal expansion strain of electrode (s_A) is used to estimate the voltage-induced electrode expansion of DEA-based device compared with the initial un-active state, while the absolute radical compression strain is to estimate the compression state of device compared with the initially planar state without the intentional crumpling. These two strains versus the driving voltage are well fitted by quadratic curves.

Figure 5 Effect of DEA on surface topographies of the initially crumpled DEA-based smart window at different driving voltages. The insert in (a), taken with magnification times of 50, is the enlarged details of (a) by 2.5-times. (a)~(d) illustrate that the initial wrinkles are gradually unfolded with the increasing voltage-induced biaxial tension, without observation of thin-film fracture. Electrical activation cannot completely unfold the initial crumpling into a flatten state. The VHB-based smart window has the memory effect of mechanical micro-wrinkling.

Electrical-activated unfolding of micro-winkles results in the switching of our smart window from translucent to transparent state, as shown in the in-line transmittance spectra in Figure 6 (a). The in-line transmittance at 550 nm wavelength greatly increases from 2% at 0 V, with the initial 5.45% absolute radical compression strain, to 71.5% at 7 kV, corresponding 12.2% of the relative areal expansion strain of electrode and 0.17% of the absolute radical tensile stain. The tunable range of 2%~71.5% is the much wider than the tunable ranges (<20%) of all previously reported DEA-based smart window,^{13,14,15} within the smallest operation strain range (0-5.45%) mentioned above.

Figure 6 (a) The in-line transmittance spectra of smart window with 5.45% initially crumpled ITO electrodes at different driving voltages; (b) the correlation between in-line transmittance at 550 nm wavelength and the driving voltage of smart window.

4 Conclusion

In this work, the nanometric-thick ultra-thin ITO electrodes of DEA-based elastomer smart window are deposited by RF magnetron sputtering technique at room temperature. A relatively flat surface of ITO is achieved, and is attributed to the low thermal energy transferred to the substrate due to the reduced secondary electron energy after multiple collisions in plasma. The transparent, conductive, and compliant ultra-thin ITO electrodes of smart window are achieved, with low sheet resistance (several kilo Ohms), high in-line transmittance (>70%), and crack-free surface morphology. The greatly improved tunable window device with the enlarged range of transmittance tuning (a clear state of 71.5% and an opaque state of 2%), is achieved within the very small (12.2%) voltage-induced relative areal expansion strain of electrode. The tuning range of transmittance is wider with lower relative areal expansion strain of electrode compared with all the previously reported results. This electrical tunability of transparency makes elastomer-based window device with ITO electrodes a promising candidate for low-cost smart window.

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