

Optics Letters

Electrically tunable window device

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A device for controlling the transmittance of light over large areas, such as windows, is described. It is based on electrostatically induced surface deformation of soft dielectric elastomer sheets produced when a voltage is applied between two networks of electrically conducting nanowires on either side of the elastomer. Variations in the surface curvature are produced by the applied voltage refract light, decreasing the optical transmittance at all wavelengths. As the device relies on changes in the geometric propagation of light, rather than on chemical changes, it is color neutral. © 2016 Optical Society of America

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In contrast to the large number of devices that have been developed for modulating laser beams, there are few that can be used to modulate the optical transmittance over large areas, such as windows in buildings and transportation vehicles. The best known are electrically controlled polymer-dispersed liquid crystal films (PDLC) and electrochromic devices [1]. PDLC films consist of droplets of liquid crystals suspended in a polymer matrix that is sandwiched between two electrodes polymer films. In the absence of an electric field, the crystals within the droplets are randomly oriented which scatters incoming light. When an electric field is applied to the electrode, the crystals are aligned to the direction of the field, reducing the scatter and resulting in transparency [2,3]. Such control of particle alignments under an electric field is generalized in suspended particle devices that use anisotropic particles, such as rods suspended in a fluid layer [4–6]. Electrochromic devices consist of layers of electrodes and active materials sandwiched between two glass panes. The composition of the active materials includes a host transition metal oxide and mobile ionic species, such as tungsten oxide and hydrogen ions, respectively. When a voltage is applied, current flows, resulting in an electrochemical reaction which changes the oxidation states of the active materials. Since the oxidation states of the active materials have different optical properties, for instance, opacity, the electrical current consequently changes the optical transmittance from clear to dark, or vice versa. There is usually a

corresponding color change, a bluish tint for the tungsten oxide windows. In a variant of these devices, using transition metal hydrides, the window can be switched from a clear to a reflective state, rather than being absorbing [1].

In this Letter, we describe the use of random networks of nanowires to create local surface deformations of a soft elastomer film which, in turn, can be used to control light transmission through the elastomer. The underlying concept is similar to the principle of operation of dielectric elastomer actuators. In these, a voltage is applied across the thickness of a dielectric, and the Coulombic attraction between the opposing charges on flat electrodes leads to a spatially uniform Maxwell stress that compresses the dielectric. The magnitude of this stress is $2\sigma = \epsilon_r \epsilon_0 E^2$, where E is the electric field, and ϵ_r is the relative permittivity of the dielectric. In most dielectrics, the resulting deformation strains are very small (ppb to few ppm) since their elastic moduli are high, typically ~ 100 GPa for oxides in capacitors and piezoelectrics. However, the elastic moduli of elastomers are commonly 10–1000 kPa; thus, the displacements produced are correspondingly orders of magnitude greater. When a percolative electrical network of conducting nanowires is used in place of a flat, continuous plate electrode, there is both a macroscopic compression of the elastomer arising from the average charge distribution over the conducting nanowire network, together with spatially varying local displacements associated with electrostatic force on individual nanowires. As will be shown, the resulting voltage-induced surface displacements alter the optical transmittance and can be used as a voltage controlled light modulator.

The simplest device structure consists of a thin nanowire mat, such as a network of silver nanowires, on a uniform thickness elastomer film attached to a transparent, stiff substrate, such as an ITO-coated glass sheet. The substrate is not only transparent, but also much stiffer than the elastomer. Consequently, the substrate constrains the lateral expansion of the elastomer film when the voltage is applied, ensuring that the Coulombic forces produce displacements primarily in the thickness direction and altering the surface morphology. This is illustrated by comparing the bright field microscope images shown in Figs. 1(a) before and 1(b) with a voltage applied. With no voltage applied, the elastomer appears uniformly gray but, when a voltage is applied, there are variations in contrast in the optical image, mostly in the immediate vicinity of the nanowires.

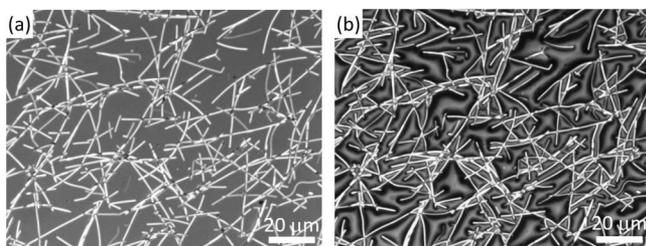


Fig. 1. (a) Bright field optical microscope images illustrating the random arrangement of silver nanowires (90 nm average diameter, 20–60 μm length, SLW-NW-90, Blue Nano, Charlotte, N.C.) dispersed on the surface of an elastomer sheet. (b) Same area and imaging conditions when a voltage is applied with respect to a flat electrode underneath. The nominal electric field is 110 V/ μm . The variations in contrast in the vicinity of the nanowires are due to variations in local curvature associated with the nanowires being displaced downward relative to the average surface. Bulging of the surface between the nanowires is most pronounced where the fibers are well separated.

An alternative form of the window device is shown schematically in Fig. 2, where light has to pass through two elastomer surfaces, each with a nanowire electrode. In this structure, the continuous ITO electrode on glass is replaced with a second nanowire network on the opposite side of an intervening transparent sheet, such as a stiff polymer or a thin glass sheet. To create this laminated structure, two thin polyacrylate elastomer films (3M VHB F9460PC, nominal thickness of 50 μm) were biaxially pre-stretched to 300% linear strain, resulting in a thickness of 3.1 μm and then laminated on both sides of a Mylar (polyethylene terephthalate) sheet (13 μm thickness). Light passing through this device structure is incoherently refracted twice, once by the front deformable surface and then again by the back deformable surface.

Dramatic changes in the light transmittance upon application of an electric field are shown in Figs. 2(c)–2(f). These images, recorded at successively higher applied voltages, are of a logo and letters placed 15 cm behind a device mounted on a circular ring. Initially visible when no voltage is applied, they become progressively less visible as the voltage was increased to 2.2 kV at 3 Hz. (The reason for using an alternating voltage is discussed in a later paragraph.) The transition from transparent to nearly opaque occurs in <1 s and is reversible, as shown in Visualization 1.

The optical transmittance data are shown in Fig. 3(a). Without the nanowire electrodes, the three dielectric layers have a combined in-line transmittance of 90%, and most of the light lost is attributed to the surface reflectance at the two interfaces between the elastomer film (polyacrylate, refractive index 1.48–1.50 at 550 nm) and the air. Additional losses may be attributed to several factors, including interface losses due to the slight difference in the refractive index between the polyacrylate and Mylar ($n = 1.65$, at 550 nm) [7,8] and slight light absorption by the polyacrylate. The addition of two layers of silver nanowire electrodes having a density of, in this case, 78 mg/ m^2 , decreases the transmittance to 62%. The decrease is mainly caused by scattering from the metal nanowires, rather than any optical absorption. In this device, the in-line transmittance drops to 8% at a voltage of 2.4 kV. The transmission is relatively flat across the visible wavelength, indicating that

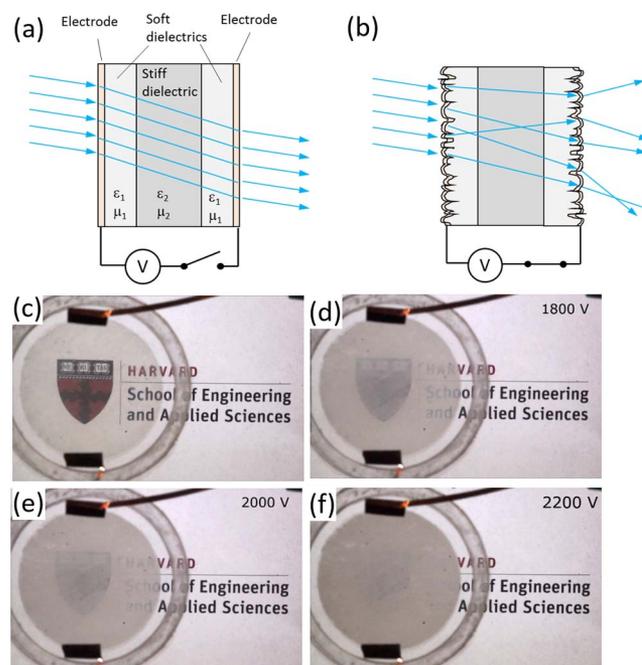


Fig. 2. (a) Schematic cross-sectional diagram of the device. In the absence of an electric field, the surfaces are relatively smooth, and light passes through without being scattered or refracted. (b) When connected to a high-voltage source, the nanowires locally compress the elastomer, deforming its surface. Since the nanowires are randomly oriented, the resulting deformation diffuses light passing through the device. (c–f) Changes of opacity at the indicated actuation voltage from 0 to 2.2 kV, demonstrating control over the in-line transmittance using electrical potential. The logo and text are located 15 cm behind the circular film. The black rectangles are electrical contacts to the two sides of nanowire electrodes. The real-time video switching from 0 to 2200 V is provided in Visualization 1.

the laminate is color neutral in contrast to the “bluish” tint normally seen in many electrochromic devices. The transmittance can be tuned continuously between the maximum and minimum transmittance values by controlling the actuation voltage, as shown in Fig. 3(b). The transmittance curve as a function of voltage has a nonlinear shape, particularly near the low and high voltages, but is approximately linear between these two extremes. The nonlinearity at the low end limits is a result of the quadratic dependence of Maxwell force on the electric field whereas, at the high limit, the nonlinearity is caused by approaching a saturation in the amount of light that can be scattered. The transmittance response as the voltage is increased to successively larger values and turned off between is shown in Fig. 3(c). At each change in voltage, there is a rapid change (~ 1 s) in transmittance of about 90% followed by a slower change. Concurrently, as the applied voltage is increased, the transmitted beam broadens. An example, recorded at a wavelength of 632 nm, using a 2D CMOS detector, is shown in Fig. 3(d).

The effect of the viewing angle is shown in Fig. 4. The in-line transmittance as a function of the illumination angle is shown in Fig. 4(a). Without any voltage applied, the direct transmittance is relatively flat between $\pm 45^\circ$ and diminishes quickly above about 50° . This is close to the Brewster angle, 56° , calculated

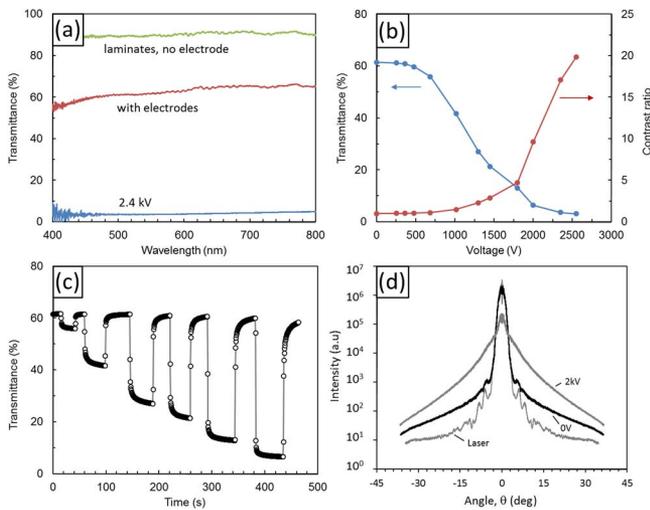


Fig. 3. (a) A significant decrease in the in-line transmittance is observed at all wavelengths with the applications of 2.4 kV. (b) Optical transmittance at a wavelength of 550 nm and corresponding contrast ratio (T_{\max}/T) as a function of electrical voltage. (The lines are guides to the eye.) (c) Response time of the device upon application of increasing actuation voltage from 700 to 2000 V. The sampling period is 0.5 s. (d) Intensity profile of the transmitted light as a function of the angle for the indicated applied voltages. This profile was measured by shining an He-Ne laser (632 nm, 1 mm beam diameter) normal to the device and by recording the spread of the exit beam using a 2D CMOS sensor (15.6 × 23.5 mm, model NEX-3N, Sony Corp.) placed on a translation stage, at a distance of 22 mm away from the device.

at flat interface between air and polyacrylate. When a voltage is applied, the transmittance at a normal angle decreases, and its variation with angle increases. This effect is typically caused by rough surfaces that increase the diffuse scattering, consistent with the smoother roll-off in transmittance with the viewing angle at the higher voltages in Fig. 4(a). It is also consistent with the broadening at a single wavelength shown in Fig. 3(d). The haze, defined by $I_{\text{scatter}}/I_{\text{total}} \times 100\%$, where I_{total} is the total intensity of incoming light, and I_{scatter} is the integrated intensity of the spread beam surrounding the original transmitted beam, as a function of voltage, as shown in Fig. 4(b). In the absence of any voltage, the device has an intrinsic haze of 8%. This value depends on the density of the silver nanowire mats. Note that because of technical difficulties, the maximum voltage used for haze measurement was limited to 1700 V. It is anticipated that the haze will increase with still higher voltages.

As mentioned earlier, the graphs presented as a function of voltage were recorded under AC conditions. This was because it was observed that under a constant DC voltage, the light transmission decreased rapidly in less than 0.5 s followed by a slow increase reverting toward its original transmittance. This behavior is shown in Fig. 5(a). Under AC conditions, this phenomenon was not observed. The drift suggests that there is a change of the electric field induced surface morphology with time. Two explanations are possible: a viscoelastic relaxation of the elastomer and a charge re-distribution. To learn if the intrinsic viscosity of the elastomer was responsible for the decay, a comparison was made between two devices, one made with the polyacrylate and the other with a silicone elastomer, an elastomer that is known to exhibit much less viscoelasticity. The low

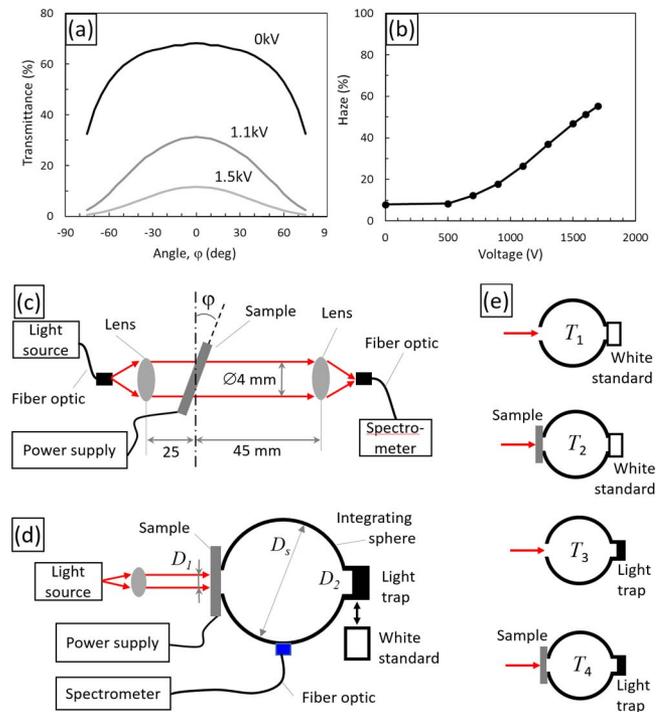


Fig. 4. (a) In-line transmittance as a function of the incident illumination angle at the indicated actuation voltages. (b) Haze as a function of actuation voltage. (c) Schematic of the in-line optical transmittance as a function of the illumination angle, ϕ . (d) Schematic of the total transmission and haze measurement. $D_1 = 4$ mm, $D_2 = 7.9$ mm, $D_s = 38.1$ mm. The haze was measured following the ASTM standard [9] using an integrating sphere (Model RSA-FO-150, Labsphere, Inc.) and a reference white standard (Spectralon SRS-99, Labsphere, Inc.). The standard specifies that a specular transmission has a maximum spread of 2.5°; anything larger is considered to be diffusively scattered. Because of the size of our collimator, we used a slightly different spread angle of 2.9°. (e) Schematic of the integrating sphere measurements used to determine the total transmission, $T_t = T_2/T_1$; the diffused transmission, $T_d = [T_4 - T_3(T_2/T_1)]/T_1$; and the haze = $(T_4/T_2 - T_3/T_1) \times 100\%$.

viscoelasticity of the silicone is evident from the faster response time compared to polyacrylate, which can be seen from the higher curvature when the voltage is initially held [see arrow “a” in Fig. 5(b)]. However, after prolonged times, as indicated by arrows “b” in Fig. 5(b), both curves become relatively linear with similar slopes. This is a strong indication that the same process governs the observed response in either material. Based on this comparison, it was concluded that viscoelastic relaxation is not responsible for the observed drift.

The other mechanism that might be responsible for the transmittance drift is migration of charge from the nanowire electrodes spreading over the surface of the elastomer. The effect of charge re-distribution would be to decrease the local electric field on the nanowires and, consequently, the electrostatic force on the nanowires locally deforming the elastomer. In turn, the effective roughness of the elastomer would decrease, leading to an increase in the in-line transmittance. In the limit, the charge would migrate until it is uniformly distributed over the surface, and the morphology would be the same

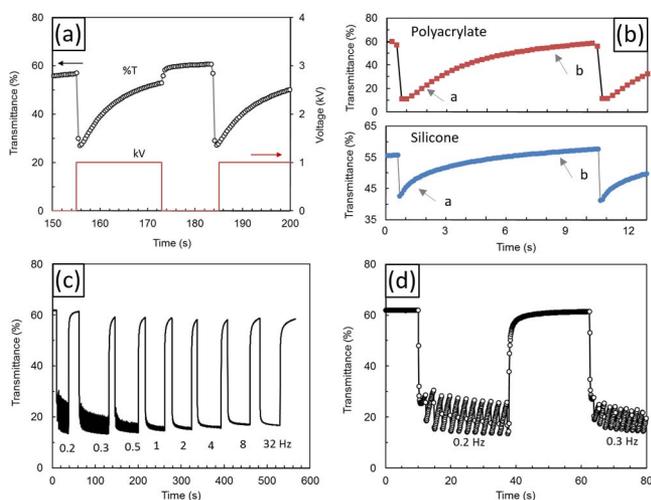


Fig. 5. (a) Optical transmittance response to a square DC voltage pulse. The sampling period between consecutive data points is 0.3 s. There is a fast (<0.5 s) initial decrease in transmittance followed by a slower increase in the transmittance. (b) Transient response of two different elastomers, a polyacrylate (VHB) and a silicone elastomer, are similar, suggesting that the response is not limited by a viscoelastic mechanical response of the materials themselves. In these measurements, the polarity of the voltage source was initially reversed at $t = 1$ s and then again after $t = 10$ s. (c) Dynamic response at different AC voltage frequencies. At switching frequencies of ~ 2 Hz and above, the drift is significantly suppressed. (d) Expanded region of figure (c) indicates a sawtooth profile that corresponds to the reverses in the polarity of the actuation voltage. The sampling period is 0.1 s.

as in the absence of the applied voltage. Based on images, such as Fig. 1(b), it is likely that the charge re-distribution would be most pronounced at the ends of individual nanowires, as these have a very small radius of curvature. This would be the surface analog of charge spraying from a pointed conductor as originally observed by Röntgen [10] over 100 years ago and employed in the Xerography process. More recently, charge spraying from a high-voltage needle has been used to spray charges to actuate dielectric elastomers without physical contact [11].

To limit the effects of charge migration, the voltage was reversed periodically so that the net voltage gradient driving surface charge migration would be zero. The effect of increasing the frequency of the voltage reversal on the in-line transmittance is shown in Fig. 5(c). As indicated, the drift decreases with increasing frequency and is almost completely suppressed at 2 Hz and above. Therefore, we used a square wave power source with a frequency of 3 Hz to ensure stable optical transmittance for analysis of the optical performance, such as for the time-evolution data shown in Fig. 3(c). Using AC power rather than DC effectively doubles the response time to a change in the DC value, but is still less than a second. The slower response time is attributed to the intrinsic viscoelasticity of the polyacrylate elastomer. Nevertheless, the overall response time is still significantly faster than that reported for electrochromic devices.

There are several intrinsic advantages of the light modulation device described in this Letter. These include the scalability to much larger areas, the suitability for roll-to-roll manufacture using commercially available materials, notably elastomers, and

Mylar, as well as the intrinsic light weight and flexibility of the materials. It is also possible that with some design modification of the laminate structure, the device can be retrofitted to existing windows. It is likely that the current materials can be further optimized in several aspects, including increasing the transparency of the elastomer, decreasing the actuation voltage using thinner and softer elastomer, optimizing the density of nanowires for maximum light scattering characteristics, increasing the conductivity of the nanowire network using post-processing, such as plasmonic welding and the incorporation of SWNT.

The use of a stiff dielectric inner layer is also important, regardless of whether it is Mylar or ITO-coated glass. First, it can provide support for the thin elastomer films, as well as the forces used to stretch the soft elastomer dielectric to decrease its thickness. The second reason for having a stiff intervening dielectric is to limit premature electrical breakdown due to electromechanical instability typically found for soft dielectrics [12]. Finally, although similar tunability in optical transmittance can be expected to occur using a regular geometrical electrode mesh, the periodicity of the mesh would likely lead to periodic variations in the transmitted beam. The randomness of the nanowire mat avoids any such preferred directionality in the transmitted light.

In conclusion, we have shown that it is possible to tune the in-line transparency of a transparent window by electrically modifying the surface morphology of a thin elastomer film. The underlying mechanism is the spatially nonuniform deformation produced in the elastomer surface when a voltage is applied to a percolative network of randomly oriented nanowires spread over the elastomer. The window devices operate under alternating voltage with a response time of the order of 1 s. The devices made using polymer and elastomer sheets are very lightweight (~ 25 g/m²) and flexible, making it easy for transportation and retrofitting into existing windows. It is anticipated that there will be several practical applications of the optical effects, including tunable privacy windows, smart glass, projector screen, displays, and camouflage.

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