



# Low-voltage-driven, flexible and durable paraffin–polydimethylsiloxane-based composite film with switchable transparency



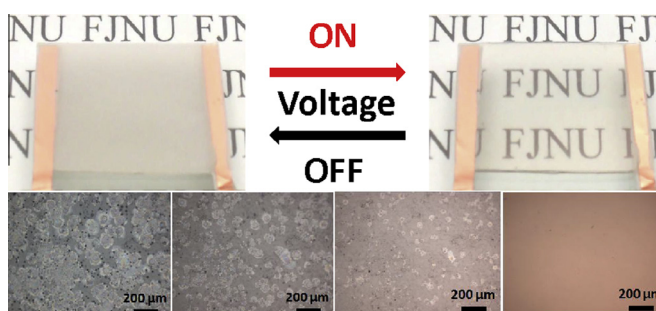
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## HIGHLIGHTS

- The paraffin–polydimethylsiloxane-based composite film with switchable transparency is successfully fabricated.
- The composite film is flexible and can be driven by low voltages.
- The switching speed from opaque state to transparent state is fast.
- The composite film has potential applications in transparency-switchable devices.
- The fabrication method is easy to scale up for commercial productions.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Recently, transparency-switchable devices were widely used in the fields of solar control, privacy, tactical display and flexible auto windows. In this work, we successfully fabricate a new type of electrical-driven composite material with switchable transparency. The composite is a trilayer structure, composing of paraffin wax and polydimethylsiloxane composite, indium tin oxide and polyethylene terephthalate. When a DC voltage of 10 V is applied to the composite film, it shows a quick switching from opaque state to transparent state in less than 8 s. The transmittance changes from 2% to 75%. After the voltage is turned off, the composite film reverses back to opaque state. Moreover, the composite film shows a stable and reversible optical performance after 200 times cycling of opaque–transparent switching. These features indicate that this new type of composite, which possesses the properties of excellent flexibility, rapid response and durable optical switching, will be widely used in the fields including optical switches, biomimetic camouflages and so on.

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## 1. Introduction

Nowadays transparency-switchable devices with switchable optical properties attract great interests due to their applications in the field of energy saving. In recent years, energy saving has become an important issue due to the ever increasing energy crisis of the world following the rapid industrial and technological advancement. Motivated by the potential for energy saving,

scientists have paid more attention to developing smart windows that can intelligently control the amount of transmitted light and heat [1–6]. Due to their switchable optical properties, smart windows have the potential to be used in privacy windows, low-consumption displays, and solar control [7–13]. The applications include electrical shutters, projection screen, intelligent displays, and band-pass filters. To date, there are mainly two categories of achieving switchable transparency. The first category utilizes a change in molecular arrangements, such as suspended-particle devices and polymer dispersed liquid crystals [8,14–16]. The complex assembly method and special equipment limit their applications in a wide range. The second category mainly utilizes chromogenic materials through external stimulus, such as light, heat, gas, and electrical voltage [17–29]. During switching, however, many chromogenic materials are chemically unstable, such as viologen derivatives and certain transition metal oxides [7,20,30,31]. Therefore, chemically stable materials together with simple assembled methods are needed for the fabrication of large-scale products with durable transparency switching and rapid response.

Several approaches have been explored in recent years. For example, hierarchically structured polydimethylsiloxane (PDMS) and a silica nano-particles/PDMS composite film both had switchable optical properties by mechanical extension and release [10,32]. Gao et al. made some progress in VO<sub>2</sub> based thermochromics composite film [27–29]. The prepared VO<sub>2</sub>-ZrV<sub>2</sub>O<sub>7</sub> composite films on silica glass substrate significantly enhanced luminous transmittances with increasing Zr/V ratios [29]. A composite film fabricated by sandwiching a carbon nanotube sheet within polyurethane films could switch from opaque to transparent with voltages higher than 30 V (2.1 V/mm) [33]. However, the driving voltage need to be further reduced for the application in commercial filed. In the past few years, scientists found that composites of paraffin wax (PW) and PDMS showed an obvious switching between an opaque state at low temperatures and a transparent state at high temperatures [34,35]. Nevertheless, PW–PDMS composites rely on external thermal stimuli to achieve switching transparency. This drawback limits their application in which the transparency could switch according to users' request. Hence, there is an urgent need to develop high-performance, low-voltage-driven, rapid-response materials with switchable transparency. Recently, a PDMS–paraffin/graphene laminated film reported by Kim et al. was driven by a DC voltage of 18 V and had a long switching time of 60 s [26]. However, the driving voltage and switching time could be further reduced. What's more, the complex fabrication process with a few steps of transfer and removal procedures may not be easy for scale-up.

Here, we propose a new type of composite with switchable transparency based on a trilayer structure, composing of PW–PDMS composite, indium tin oxide (ITO) and polyethylene terephthalate (PET). The speed of transparency switching is rapid. The PW–PDMS/ITO/PET composite film becomes transparent in 8 s with a DC voltage of 10 V (0.3 V/mm), and reverses back to opaque after voltage turning off. The switch between opaque state and transparent state can be repeated for at least 200 times without altering the optical properties. Furthermore, chemically stable materials and simple synthesis method make the PW–PDMS/ITO/PET composite developed here easily scale up for commercial production.

## 2. Experimental section

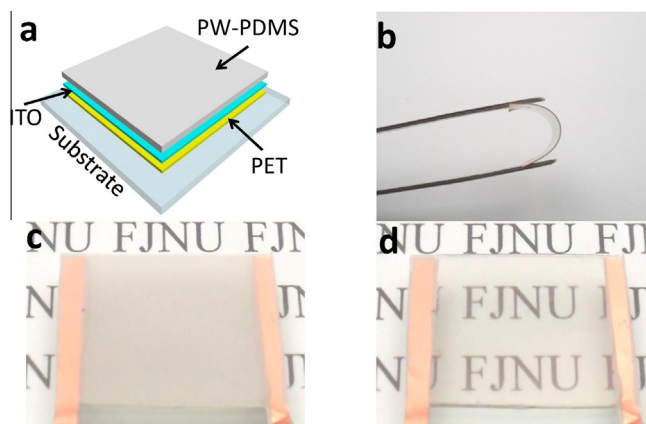
### 2.1. Materials

Dimethyl siloxane (DMS) and tetraethyl orthosilicate cross-link reagent, which can be mixed together to fabricate PDMS rubber

(GF-T2 Silicone Elastomer), were purchased from Beijing Hangtongzhou Technology Co., Ltd., China. The cured PDMS is a transparent and flexible polymer. Meanwhile it is also an electrical insulator with resistivity  $>10^8 \Omega\cdot\text{m}$ . PW with a melting point of 55 °C is supplied by Shanghai Huashen Rehabilitation Equipment Factory, China. PW undergoes an opaque–transparent switching when it melts during PW phase transition. This property results in PW–PDMS composites becoming transparent when the temperature is higher than the melting point of PW. ITO/PET films are purchased from Shenzhen South-China Xiang Science & Technology Co., Ltd., China. Pure PET film in ITO/PET film has a transmittance of 89% at the wavelength of 550 nm with a thickness of  $0.175 \pm 0.05 \text{ mm}$ , while the ITO/PET film has a lower transmittance of 81% due to plated ITO film with a thickness of  $185 \pm 5 \text{ nm}$ . ITO is a common component, which is used in the fabrication of transparency-switchable devices due to its transparency and conductivity. Because ITO is traditionally plated on glass, the ITO/glass substrate is rigid and hard to bend. In our work, the ITO/PET film employs PET as a substrate, which makes the ITO/PET film flexible and conductive with a resistance of  $\sim 6 \Omega/\text{square}$ . The features described above enable the ITO/PET film to be a transparent, flexible and conductive material.

### 2.2. Fabrication of the PW–PDMS/ITO/PET composite film

Fig. 1(a) schematically shows the trilayer structure of the PW–PDMS/ITO/PET composite film, which is constructed through a heat curing approach. ITO/PET film was fixed on a polystyrene substrate with the plated ITO side upwards. Copper electrodes (not shown in Fig. 1(a)) were at two ends of the ITO/PET film, so that the electrical current can pass through the ITO film in the length direction and the total electrical resistance of the composite film is proportional to the length of ITO/PET film. The PW–PDMS composites with various PW contents were generated by mixing DMS and cross-link reagent with various contents of PW. First, the DMS was mixed with PW at room temperature (29 °C), which forms an opaque suspension, but became a clear gluey liquid after stirring at 80 °C for 5 min. Second, the prepared mixture was mixed by tetraethyl orthosilicate cross-link reagent with the weight of 1/9 DMS. Third, the mixture was stirred for 10 min at 80 °C to make the cross-link reagent fully dispersed. Finally, the mixed liquid was cast onto the ITO/PET film. The thickness was controlled to be less than 0.6 mm. After curing in an oven at 80 °C for 12 h, a solid-state PW–PDMS/ITO/PET composite film was achieved.



**Fig. 1.** (a) Trilayer structure of PW–PDMS/ITO/PET composite film; (b) optical photo showing the flexibility of PW–PDMS/ITO/PET composite film; (c) an opaque state of PW–PDMS/ITO/PET composite film at 29 °C; (d) a transparent state of PW–PDMS/ITO/PET composite film at 65 °C.

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