



Electroactive shape memory composites with TiO₂ whiskers for switching an electrical circuit

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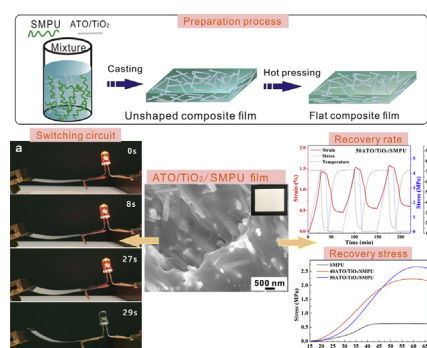
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HIGHLIGHTS

- The conductive shape memory composites with ATO/TiO₂ whiskers are developed and employed to switch an electrical circuit.
- The nearly white conductive films, which are free of the restriction of color, have general applicability.
- The ATO/TiO₂ composites show the improvements in moduli (at least 390%) and recovery stress (more than 250%).
- The switch made of ATO/TiO₂ composite is able to turn off the electric circuit within 30 s when the applied voltage is 70 V.

GRAPHICAL ABSTRACT



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ABSTRACT

In this paper, electrically actuated shape memory composites were prepared by compounding shape memory polyurethane (SMPU) with conductive antimony-doped tin oxide/TiO₂ (ATO/TiO₂) whiskers. The resultant composites, ATO/TiO₂/SMPU, can be activated by electric voltages because of heating Joule and enhancement of heating efficiency by the conductive network resulting from the overlaps of whiskers. In addition to conductivity, ATO/TiO₂/SMPU composites featured lighter color than most electroactive shape memory composites, which exhibit black color due to the addition of carbon materials. The composites exhibited uniform electrical resistance and rapid heat transfer performances. When the composites with 50 wt% ATO/TiO₂ whiskers were used in a switch electric circuit as the switch, the circuit can turn off within 30 s. ATO/TiO₂ whiskers improved Young's moduli by at least 390% and recovery stresses by more than 250% compared with pristine SMPU. Although the recovery rates were unsatisfactory in the first test cycle, composites with 40 wt% ATO/TiO₂ whiskers and 50 wt % ATO/TiO₂ whiskers still showed recovery rates higher than 96% and 94% in the third cycle, respectively.

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1. Introduction

Shape memory polymers, with unique shape memory behavior triggered by an external stimulus, are regarded as an important category of multifunctional smart materials for various engineering applications,

such as aerospace field [1], various actuators [2,3], biomedical applications [4,5], 4D printing [6,7], energy-harvesting devices [8,9], self-healing systems [10,11], functional fabrics [12–14], and circuit control [15]. Shape memory polymers can be deformed and fixed into any temporary shape. After exposure to stimulus (e.g., heat, electricity, light, moisture, magnetism, and solvent), these polymers will recover to their permanent shapes automatically [16,17]. As a kind of thermo-responsive shape memory polymer, shape memory polyurethane

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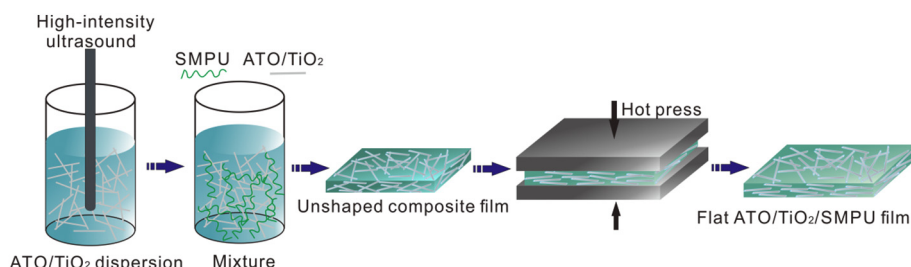


Fig. 1. Schematic of the manufacturing process of ATO/TiO₂/SMPU composite films.

(SMPU) is explored extensively owing to its low cost, excellent biocompatibility, easy processing, and high shape deformability [18]. The shape memory effect stems from the dual-segment system of SMPU, that is, the cross-linked segments that “remember” permanent shapes and soft segments with specific switching transition temperature that achieve shape deformation and fixation [19,20].

To date, plenty of researches on electrical conductive SMPU have been conducted to satisfy the demands of electro-activated shape recovery behavior [21–24]. Conductive fillers with excellent electrical conductivity are generally used to endow insulated SMPU with conductivity, examples of such fillers include carbon nanotubes, carbon nanofibers, silver nanowires, graphene, and metal fibers and nanoparticles [22,25–27]. Thereinto, 1D fillers can mutually overlap easily to provide continuous and steady conductive paths to significantly reduce electrical resistivity [28]. Surface modification of conductive fillers is used to improve not only the interface between polymer and filler but also the dispersion property of fillers [29,30]. 3D network structures and synergies with thermal conductive filler are employed to achieve more uniform and efficient electric heating [31,32]. Overall, conductive SMPUs that exhibit rapid shape recovery respond without an external thermal heater by incorporation with various conductive fillers. The present work emphasizes the fabrication of SMPU composites with high conductivity without prejudicing the shape recovery rate of SMPU by using as few fillers as possible. However, the drawbacks of low elastic moduli and recovery stresses were barely investigated.

Conductive components not only offer electro- and thermal-conductive pathways to enable uniform Joule heating and subsequently accelerate recovery but also influence the mechanical properties of SMPU. Studies have proposed that addition of conductive fillers (especially 1D fillers) will diminish the recovery rate of SMPU, whereas some deficiencies, such as low deformation stiffness and recovery stresses, will be improved [25,33–35]. Nano-sized, low-yield conductive fillers used in previous works are generally expensive. SMPU composites with high content of conductive fillers, especially for 1D fillers, are rarely reported due to the loss of shape recovery rate and high cost. Studies seldom investigate the comprehensive mechanic performance of this kind of composite and barely consider the color of electroactive shape memory composites.

In this study, we report a kind of 3D network conductive whisker-SMPU composite films with high filler content and free of color restriction due to the light-colored conductive antimony-doped tin oxide/TiO₂ (ATO/TiO₂). ATO, as a conductive shell, surrounds TiO₂, which was used as a whitening agent. Results showed that Joule heating can trigger the composite films to activate shape memory effect in a few seconds. Application of voltage and filler ratio profoundly affected heating rate and thermal equilibrium temperature. Thermodynamic performances and static mechanical behaviors of composite films with different filler contents were investigated systematically. Young's moduli and recovery stresses were significantly enhanced. Recovery behavior of composite films was unsatisfactory during the first cycle and improved with increasing cycle time, indicating that high shape recovery rate can be achieved after the pre-deformation cycle. The obtained films with uniform heating performances and good recovery rates can be used to switch an electrical circuit and showed rapid response. Compared with most conductive materials, ATO/TiO₂/SMPU film appeared white, which will be preferable for further applications, such as color-sensitive and luminescence applications.

2. Experimental method

2.1. Preparation of ATO/TiO₂/SMPU composite films

The composite films were fabricated by incorporating ATO/TiO₂ into the SMPU matrix. ATO/TiO₂ is a conductive filler with core-shell structure and was synthesized by following the process reported in Reference [36]. Specifically, TiO₂ whiskers were first prepared by hydrolysis reaction of tetraisopropyl orthotitanate in ethylene glycol medium under microwave heating and then decomposition reaction of hydrolysis products. Second, the conductive ATO layer was obtained after synchronous hydrolysis reaction of tin and antimony salts on TiO₂ whiskers and calcination treatment of precipitates. The average diameter of ATO/TiO₂ whisker approximated 250 nm. Fig. 1 illustrates and describes the concrete steps as follows. The laboratory-made ATO/TiO₂ with a resistivity of 210 Ω·cm was first added into tetrahydrofuran (THF, 99.5%, WAKO, Japan) at ratios of 40 and 50 wt% relative to the polymer matrix. Then, ultrasonically broken instrument (Sonifier 250,

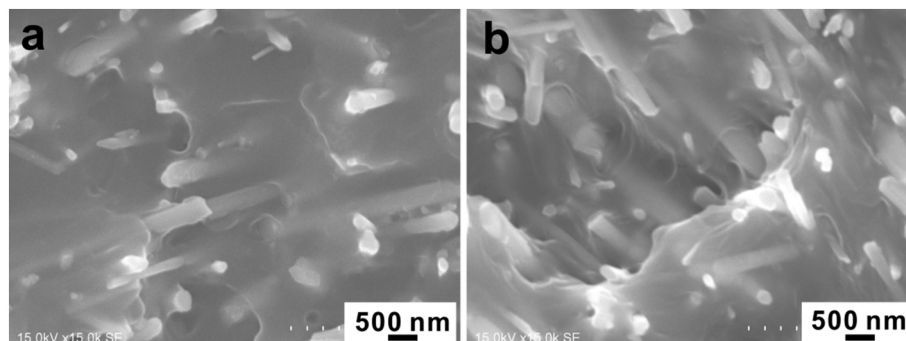


Fig. 2. SEM images of ATO/TiO₂/SMPU composite films with different whisker contents (a: 40ATO/TiO₂/SMPU film; b: 50ATO/TiO₂/SMPU film).

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