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Interactively mechanochromic electronic textile sensor with rapid and durable electrical/optical response for visualized stretchable electronics

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ABSTRACT

Textile-based electronic devices are significant components of emerging wearable electronics. However, the current textile-based electronics are limited by single electrical signal input/output, and unable to be visualized directly by human eyes, restricting their diverse applications in visualized human-interactive electronics. Here, we develop a series of novel interactively mechanochromic electronic textile (MET) sensors for visualized stretchable electronics. The MET sensors are based on the ingenious coupling of new supramolecular photonic elastomers (PEs) with hierarchical-fiber-structured conductive polyester textiles (CPTs). Benefiting from their semi-embedded structures, the MET sensors demonstrate not only distinct negative electrical response but also simultaneous mechanochromic capability upon stretching, via reconstructing conductive paths and adjusting the lattice spacing of the photonic crystals. Notably, the MET sensors exhibit bright structural colors, enhanced toughness (35.6 kJ m⁻³), excellent mechanical resilience, fast optical/electrical response (\approx 0.30 s) and recovery speed (≈ 0.22 s). Most importantly, the structural colors and electrical responses of the MET sensors remain constant even after 30,000 stretching/releasing cycles, showing outstanding mechanical stability, reliability, and excellent durability. Based on these merits, the MET sensors are used as visually interactive wearable devices for monitoring human joint movements in real time. This research provides a general platform for achieving visualized interaction of the electronic textiles, which shows great prospects in wearable devices, human-machine interfaces, and soft robotics.

1. Introduction

The ever-increasing demand for higher quality of life has motivated the rapid evolution of wearable and stretchable electronic devices that can be comfortably mounted on the human body. They aim to convert various personal and environmental stimuli into measurable electrical signals (e.g., resistance and capacitance) [1–7]. As an important component of wearable electronics, flexible electronic materials have shown great potential for numerous applications, including human health monitoring [8,9], electronic skin [10–12], soft robotics [13,14] and human–machine interactions [15,16]. Great efforts have been focused on the development of different types of flexible electronic materials. Among them, textiles that are indispensable in our daily life have been regarded as appealing candidates, mainly due to their lightweight, flexibility, biocompatibility, washability, and human tissue-like adaptability [17–21]. By incorporating conductive fillers (e.g., metallic nanoparticles and carbon materials) [22-24] or transforming into carbon textiles through thermal treatment [25-27], these textiles can be endowed with improved electrical performance. Based on the conductivity, various textile-based sensors have been demonstrated [3,18]. However, most of them are limited by single electrical signal input/ output, which requires connection to external electrical instruments and subsequent signal processing for data acquisition [12]. In addition, the electrical signals are unable to be directly visualized by human eyes, thereby restricting their applications in visualized intelligent human--machine interaction. Furthermore, the electrical signals can fluctuate irregularly under interference (i.e., temperature, humidity) and thus cause measurement error in monitoring human activities due to their sensitive responsiveness [28,29]. To this end, the introduction of additional visible signal output (i.e., color change) that can be easily and accurately perceived by the human eye would be an effective strategy to address these issues.

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Structural colors, arising from light interaction with periodically ordered nanostructures that are widely known as photonic crystals (PCs), are widespread in nature and play a crucial role in the survival of many creatures [30-34]. As a representative example, chameleons can rapidly change skin color for camouflage via actively tuning the lattice spacing of guanine nanocrystal arrays within their iridophore cells in response to environmental changes [30,35]. Inspired by this fascinating phenomenon, mechanochromic PC materials have attracted increasing attention and shown great promise in visualized strain sensing, owing to their unique capability of producing instantaneous and intuitive visual response to the user [36-39]. Generally, these materials are constructed by combining colloidal PCs with different elastic matrixes, such as hydrogels [40-43] and polymeric elastomers [35,44-48]. Although photonic hydrogels show sensitive color-changing ability and excellent biocompatibility, they usually suffer from low mechanical strength and water evaporation-induced environmental instability, limiting their long-term practical uses [35]. Alternatively, combining colloidal PCs with elastomers (i.e., photonic elastomers, PEs) offers an effective approach to tackle the aforementioned issues [45]. Such PEs possess the unique advantages of improved environmental stability and robustness, thus facilitating their potential applications in visualized stretchable devices [44,47]. However, current PEs only convert mechanical stimuli into eye-perceptible color changes. These optical signals are difficult to be coupled with the majority of wearable devices that adopt electrical transducing modes to feedback information [12,49]. Thus, subsequent conversion and processing of these optical signals are required for PEs in wearable electronics [29,50,51]. The ideal PE-based material should possess the sensing and visualization of the stimuli through electrical and optical dual-signal simultaneously. Nevertheless, fabrication of the above-mentioned materials with such abilities remains challenging and is largely unexplored so far.

Herein, for the first time, we present the rational design and fabrication of a series of novel interactively mechanochromic electronic textile (MET) sensors with rapid and durable electrical/optical response for visualized stretchable electronics. As shown in Fig. 1, the MET sensor is prepared by incorporating a new mechanochromic supramolecular PE into a hierarchically-structured conductive polyester textile (CPT). The supramolecular PE is composed of non-close-packed PS@SiO₂ photonic arrays within the elastomeric matrix, offering the capability of visualizing external strain through lattice spacing regulation. On the other hand, CPT was obtained by chemically and coaxially growing conductive and transparent Al-doped ZnO (AZO) films onto the fiber surface via atomic layer deposition (ALD), providing resistance variation via conductive paths reconstruction. The MET sensor possesses not only instantaneous and reversible color-changing capability (i.e., mechanochromism) but also simultaneous resistance changes while stretching. Importantly, it shows bright structural color, high toughness, excellent mechanical resilience, fast optical/electrical response and recovery speed, as well as stable optical/electrical response. To the best of our knowledge, such textiles exhibiting both excellent mechanochromism and electrical response simultaneously have not yet been reported. As a proof-of-concept, we further demonstrate the versatile capabilities of the MET sensor as a visualized interactive device for detecting human joint movements, showing numerous potential applications in wearable electronics, intelligent robotics and so forth.

2. Materials and methods

2.1. Materials

Polytetramethylene ether glycol (PTMG, $M_n = 2000$), isophorone diisocyanate (IPDI, 99%), 1,5-pentanediol (1,5-PDO, 97%), dibutyltin dilaurate (DBTDL, 95%) and vinyltriethoxysilane (VTES) were purchased from Aladdin (China). Styrene (St), sodium dodecyl sulfate (SDS), and potassium peroxydisulfate (KPS) were provided by Sinopharm Chemical Agent Company (Shanghai, China). St was alternately washed in a separatory funnel three times with 0.1 M NaOH to eliminate polymerization inhibitors and then washed with deionized water until neutrality. Absolute ethanol and ammonia solution (25 wt%) were obtained from Fuyu Fine Chemical Co. Ltd (Tianjin, China). Trimethyl aluminum (TMA, 99.999%) and diethyl zinc (DEZ, 99.999%) were purchased from AiMouYuan Scientific Equipment Co., Ltd (Nanjing, China). Deionized water and high-purity nitrogen (99.999%) were produced by Dalian University of Technology and Dalian Guangming Special Gas Co., Ltd., respectively. Commercially available polyester fabrics were supplied by Changxing DAXIN Textile Co., Ltd (Zhejiang, China).

2.2. Preparation of monodispersed $PS@SiO_2$ core-shell nanospheres and $PS@SiO_2$ opal templates

Monodispersed polystyrene (PS) nanospheres were prepared by emulsion polymerization as reported previously [52]. PS@SiO₂ core-shell nanospheres were prepared using a modified Stöber method involving the hydrolysis of vinyltriethoxysilane (VTES) in aqueous solution of dispersed PS nanospheres [53]. Typically, 3 mL of the PS nanosphere dispersion (10 wt%) was mixed with 57 mL of deionized water under magnetic stirring at room temperature. Then, 4 mL of ammonia solution (NH₃·H₂O, 25 wt%) and 27 mL of water were added into the PS solution, and the mixture was stirred at a speed of 300 rpm for 15 min. Subsequently, 1 mL of VTES was added into the reactor with a syringe pump under continuous stirring for 2 h. The hydrolysis



Fig. 1. Schematic illustration showing the structure and application of the MET sensor for flexible electronics.

reaction was carried out at 20 °C for 6 h. Finally, the suspension was rinsed three times with ethanol, and the resultant $PS@SiO_2$ core–shell nanospheres was dispersed in 25 mL of ethanol for further use. $PS@SiO_2$ opal templates were prepared via the evaporation-induced self-assembly. The polytetrafluoroethylene (Teflon) molds were washed carefully with ethanol before use. $PS@SiO_2$ opal templates were obtained on the surface of Teflon molds at 40 °C for 5 min by self-assembly during solvent evaporation.

2.3. Preparation of the CPT

The CPT were prepared according to our previously reported work [54]. The polyester textiles with the weft-knitted structure were utilized as raw materials. First, the textiles were cleaned using water and ethanol alternately with ultrasonication to remove impurity. After being dried in air, the surfaces of the textile were activated by plasmonic treatment in a plasma cleaner using air as the plasma source (15 Pa, 25 °C, 10.2 W, Shenyang Kejing AutoInstrument Co., Ltd) for 10 min, in which many highly active hydrophilic groups were introduced onto the surface, thereby facilitating the following deposition process [55,56]. Next, transparent and conductive AZO films were deposited onto the textile surface at 150 °C via ALD technique. In AZO deposition process, diethyl zinc [DEZ; Zn(C₂H₅)₂], trimethyl aluminum [TMA; Al(CH₃)₃] and deionized water (DI; H₂O) were used as precursors for Zn, Al and O, respectively. High-purity nitrogen (N₂) was used as the carrier gas at a flow rate of 20 sccm (standard cubic centimeters per minute). During the deposition process, the textiles were placed in the ALD reaction chamber (LabNanoTM, Beijing Ensure Nanotech Co., Ltd) and the temperature was raised to 150 °C in vacuum. Subsequently, DEZ and H₂O were alternatively pulsed into the ALD reactor to deposit ZnO layer according to the following sequence: DEZ (0.02 s)/N₂ (20 s)/ H_2O (0.02 s)/N₂ (20 s) (Scheme S1-2), which consists of a complete ZnO cycle. For Al doping into the ZnO films, a single Al₂O₃ cycle, conforming to the sequence: TMA (0.02 s)/N₂ (20 s)/H₂O (0.02 s)/N₂ (20 s), was inserted after 20 cycles of ZnO layers (Scheme S3-4). Then, 20 ZnO cycles and one Al₂O₃ cycle were repeatedly deposited for AZO films, and the total supercycle was set to 30 (Al doping level 3.61%), as schematically shown in Fig. S1. Therefore, AZO films, in which the ratio of ZnO:Al₂O₃ ALD cycles was fixed to 20 invariably, were deposited onto the textile surfaces. Finally, the as-fabricated CPT were cooled to room temperature naturally. Fig. S2 shows the photographs and SEM images of polyester textile before and after AZO deposition, proving its intrinsic morphology and structure were not be affected during this process.

2.4. Preparation of PTIP, PE and MET sensor

Typical polymerization procedures for PTIP supramolecular elastomer are described as follows: PTMG (5 g, 2.5 mmol) was stirred under vacuum and heated at 120 °C for 2 h to remove moisture residue and then cooled to room temperature prior to use. IPDI (1048 µL, 5.0 mmol), 1,5-PDO (0.26 g, 2.5 mmol) and catalyst DBTDL (one drop) was then added into the melted PTMG at room temperature under nitrogen atmosphere and further stirred for 15 min. The resultant mixture was poured into Teflon mold and placed in the oven under 80 °C for 2 h to complete the polymerization reaction. Finally, the obtained PTIP elastomer was peeled off from mold and cut into certain dimensions for further use. PTI elastomer was also prepared in the same way except for not adding the chain extender 1,5-PDO. In all cases, the stoichiometric ratio of [NCO]/[OH] was kept at 1:1. For the preparation of PE, PS@SiO2 opal template was firstly self-assembled on the Teflon molds by using PS@SiO₂ nanospheres as building blocks (the reasons for the selection of PS@SiO2 nanospheres are presented in Supporting Information). Then, the above-mentioned mixture was poured onto PS@SiO2 opal templates and permeated into the interstices between the PS@SiO2 nanospheres by utilizing strong capillary force. The redshift in structural color of the opal template indicated that the elastomer precursor had

completely filled the gaps between them. The resulting PE was finally obtained by polymerizing at 80 °C for 2 h, followed by peeling off from the Teflon molds. For the preparation of MET sensor, the CPT was firstly cut into rectangular shape with certain dimensions. The mixture of elastomer precursors was coated on the Teflon mold with PS@SiO₂ opal templates, followed by covering and gently pressing CPT on top with the assistance of microslide in order to ensure the close contact between CPT and PE. During this time, elastomer precursor would partially penetrate into the structure of CPT. This is the key to forming the semi-embedding structure. In the end, it was prepared after completing polycondensation reaction between these elastomer precursors at 80 °C for 2 h.

2.5. Characterization

SEM images were obtained by a scanning electron microscope (Nova Nano SEM 450). Energy-dispersive X-ray spectroscopy (EDS) mapping analysis was performed using an inbuilt EDS detector. The normalincidence static reflection spectra and the dynamic reflection spectra in response test were obtained using a spectrophotometer (Hitachi U-4100, Japan) and a Fiber Optic Spectrometer (EQ 2000). DSC analysis was carried out in a nitrogen atmosphere by using an American TA Instruments 910S DSC thermal analyzer over a temperature range from -100 to 0 °C at a heating rate of 10 °C min⁻¹. FT-IR spectra were recorded using a Thermo Fisher iN 10 spectrometer. Digital photos of all samples were captured with a smartphone. Mechanical tensile test was carried out by a universe testing machine (PT-305, Dongguan Precise-Test Equipment Co., Ltd.). The tensile rate was 10 mm min^{-1} , and the tested samples of PU, PE and MET sensor with a length of 20 mm, a width of 5 mm, and a thickness of 0.6 mm was prepared in a mold. XPS (Thermo Scientific Escalab Xi⁺) measurements were taken using Al K α radiation source. The resistance test was performed using a Digital Multimeter (Keysight 3440A/11A Digital Multimeter (Agilent)) in conjunction with the computer-controlled universe testing machine. The repeated stretching and releasing behavior of the sensor was controlled by a homemade cyclic tensile tester with adjustable frequency. Rubbing resistance measurement was performed on a YB571-II crockmeter (Wenzhou Darong Textile Standard Instrument Factory, China) according to the ISO 105 \times 12 standard. Both sides of the CPT were first fixed to the test platform, and the CPT was subjected to reciprocating rubbing for 10 cycles using a loaded head (with a pressure of 50 kPa).

3. Results and discussion

3.1. Preparation and characterization of the MET sensor

Fig. 2 shows the preparation and characterization of the MET sensor. Firstly, a new supramolecular elastomer (namely PTMG-IPDI-PDO, simplified as PTIP) was synthesized through a polycondensation reaction of polytetramethylene ether glycol (PTMG), isophorone diisocyanate (IPDI) and 1,5-pentanediol (PDO) in the presence of dibutyltin dilaurate (DBTDL, catalyst), as shown in Fig. 2a. It can be seen from the Fourier transform infrared (FTIR) spectra in Fig. 2c that as-prepared PTIP shows the typical absorption bands at 3327 and 1530 cm^{-1} , attributed to the stretching and bending vibration of N-H bond, respectively. The absence of characteristic peaks of -OH and -NCO in the spectrum of PTIP confirms that -NCO in IPDI has reacted completely with —OH in PTMG. The absorption peak at 1716 cm^{-1} is attributed to the stretching vibration region of C=O groups, demonstrating the formation of carbamate. Besides, the C=O groups is split into two peaks at 1720 and 1700 cm^{-1} ascribing to free C=O and hydrogen-bonded C=O groups, respectively (Fig. S3), indicating the formation of a supramolecular crosslinking network through hydrogen bonding in PTIP [57-59]. High-resolution X-ray photoelectron spectroscopy (XPS) spectrum of C 1 s can be resolved in four main components including C-C/C-H, C-N, C-O-C, and -COOR bonds corresponding to the peaks at 284.6, 285.1, 286.0, and 288.5 eV,



Fig. 2. (a) Synthesis and chemical structure of PTIP supramolecular elastomer, with a proposed ideal microphase-separated network structure of PTIP dynamically cross-linked through hydrogen bonds between urethane groups. (b) Schematic illustration of the fabrication process of the MET sensor. (c) FT-IR spectra of PTMG, PDO, IPDI and PTIP, respectively. (d) Photographs of (d_1) CPT, (d_2) PE and (d_3) MET sensor, as well as the twisted and bended configurations of MET sensor, respectively. Scale bar is 0.5 cm. (e) Reflectance spectra of green PS@SiO₂ PCs and orange MET sensor, respectively. (f) Cross-sectional SEM image of the MET sensor. (g) Enlarged cross-sectional SEM images showing the PE containing a non-close-packed array of PS@SiO₂ nanospheres. (h) SEM image and the corresponding EDS mapping images of the AZO layer deposited on a single polyester fiber: red, green, blue and yellow represent the C, O, Al and Zn element mapping results, respectively. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

respectively, further implying the formation of PTIP (Fig. S4) [60,61]. Therefore, the above results indicate the successful synthesis of PTIP supramolecular elastomers cross-linked through hydrogen bonding.

In this supramolecular system, the PTMG parts of the PTIP were used as the soft segment, endowing the elastomer with excellent deformability, whereas IPDI and PDO components together acted as the hard segment, which could form the hard domains through hydrogen bonding between urethane and urea groups [62-64], as schematically shown in Fig. 2a. The hard domains play an important role in increasing the toughness for the elastomer [65]. To verify this, a reference elastomer (namely PTMG-IPDI, simplified as PTI) was also prepared through a similar polycondensation reaction between PTMG and IPDI without adding the chain extender PDO (Fig. S5). Differential scanning calorimetry (DSC) analysis reveals that glass transition temperature (T_g) of PTIP and PTI are nearly the same value of -75 °C, implying that these elastomers are in the highly elastic state at room temperature (Fig. S6). It is observed from the stress-strain curves that the tensile strength and the elongation at break of PTIP are 2.0 MPa and 1270%, respectively. In contrast, these are reduced to 1.4 MPa and 970% for PTI, respectively, demonstrating that the PDO acts as part of the hard segment and greatly enhances the mechanical toughness (from 8.5 to 17.6 kJ m^{-3}) of the elastomer (Fig. S7).

Taking advantage of the merits of PTIP supramolecular elastomer, PE was then prepared by incorporating the PS@SiO2 PCs within PTIP. CPT was obtained by the chemical and coaxial growth of conductive and transparent AZO films onto the fiber surface via ALD. Thus, the resulting CPT showed an electrical conductivity of 0.012 S/m and stable mechanical property due to the formation of chemical interactions between textiles and AZO films (Figs. S8 and S9). The MET sensor was finally obtained by integrating mechanochromic PE with hierarchical-fiberstructured CPT through partially embedding PE into CPT, as depicted in Fig. 2b (see the Experimental Section for details). As displayed in Fig. 2d, the MET sensor exhibits more pronounced structural color (orange) in comparison with the pure PE. This is because the black-colored CPT absorbs the background light and decreases incoherent scattering, thus enhancing color saturation [66] (Figs. S10). The sensor also possesses robust mechanical properties and flexibility that can undertake various deformations including stretching (discussed below), twisting and bending, making this an ideal candidate for flexible electronics. The wavelength of PS@SiO2 PCs shifted from 527 to 604 nm after infiltrating PTIP elastomer (Fig. 2e). This is because PTIP fills the interstices between PC template, which increases the effective refractive index. What is more, elastomer infiltration increases the lattice spacing of the PC template, thus forming a non-close-packed hexagonal structure (Fig. 2g), which facilitates mechanochromism [35,46]. In addition, the cross-sectional scanning electron microscopy (SEM) image in Fig. 2f clearly shows the hierarchically semi-embedded structure of the MET sensor, including the top layer of PE, the intermediate auxiliary layer of PTIP and the bottom layer of CPT. It can be seen that the CPT was partially embedded in PTIP elastomer and therefore, the two binds tightly together, ensuring their simultaneous movements during stretching. Fig. S11 presents the SEM images of the AZO films deposited on a single polyester fiber. As expected, they were wrapped around the fiber surface tightly and uniformly, indicating the conformal and uniform growth via ALD. This result was further confirmed by energydispersive X-ray spectroscopy (EDS) shown in Fig. 2h and S12. It is observed that distributions of four elements (C, O, Al and Zn) on single fiber were uniform, implying high uniformity and conformability.

By varying the diameter of the $PS@SiO_2$ nanospheres, two kinds of MET sensors with different structural colors (orange and green) (Fig. S13) were readily constructed. In general, the reflection peak positions for the non-close-packed face-centered-cubic (FCC) structure can be estimated by equation (1)

$$\lambda_{max} = 2dn_{eff} \left(\frac{\pi}{3\sqrt{2}\,\varphi}\right)^{\frac{1}{3}} \left(\frac{8}{3}\right)^{\frac{1}{2}} Dn_{eff} \tag{1}$$

where λ_{max} is the maximum reflection wavelength, *d* is the (111) plane spacing, *D* is the diameter of the nanospheres, φ is the volume fraction of the nanospheres. In addition, n_{eff} refers to the effective refractive index of the MET sensors as given by equation (2)

$$n_{eff} = \sqrt{n_{PS@SiO2}^2 \varphi + n_{PTIP}^2 (1-\varphi)}$$
⁽²⁾

where $n_{PS@SiO2}$ (calculated by using equation S3) and $n_{PTIP} \approx 1.47$ (measured using spectroscopic ellipsometry) are the refractive indices of the PS@SiO₂ nanospheres and the PTIP elastomer, respectively. For example, orange MET sensor with λ_{max} at 604 nm was fabricated using 222.2 nm PS@SiO₂ nanospheres as building blocks (Fig. S14). A value of $\varphi = 0.55$ is calculated from Eqs. (1) and (2). Furthermore, the λ_{max} of the PC films were roughly equal to the theoretical values calculated by equations S1-2 (Fig. S15 and Table S1) according to Bragg's law [67,68] (see the Supporting Information for details of calculation).

3.2. Mechanical and mechanochromic properties of the MET sensor

The MET sensor shows excellent mechanical and mechanochromic properties. As shown in Fig. 3a, after the introduction of PCs into PTIP, the tensile strength of PE was enhanced (3.3 MPa) while its elongation at break was slightly reduced (1126%) in comparison with that of pure PTIP. This is possibly due to chemical interactions between the PTIP network and hydroxyl groups on the surface of PS@SiO₂ nanospheres [44,48,69]. Remarkably, coupling PE with CPT exhibited a tremendous increase in strength (16.1 MPa) and toughness (35.6 kJ m⁻³), compared with pure PE (3.3 MPa, 20.7 kJ m⁻³) and CPT (10.3 MPa, 24.9 kJ m⁻³), thus providing unique advantages for the preparation of robust and stretchable electronic composites. This is because the synergistic combination of PE and CPT induced by part of the CPT fibers were cross-linked by the PTIP elastomers.

By varying the thickness of pure PTIP, a series of MET sensors with different mechanical properties were obtained (Fig. S16). Moreover, it displays good elastic performance as evidenced by the repeated cyclic tensile tests (Fig. 3b). It is observed that the hysteresis loop in the second cycle was much less than that of the first cycle, and the curves tended to be stable after four cycles, indicating that the energy dissipation during the rupture of the hydrogen bonds occurred mainly in the first cycle. After relaxing for 30 min at 25 °C, it showed a loading-unloading curve similar to that of the first cycle, indicating its good mechanical resilience. A further cyclic test at a strain of 100% for PTIP and MET sensor showed a similar trend (Fig. S17). Furthermore, the MET sensor also exhibits an outstanding elasticity during multistep cyclic stress-strain testing from 30% to 100%, as shown in Fig. 3c. After the first tensile cycle with a small strain (30%), most of the mechanical properties were restored by recovering partially broken hydrogen bonds and therefore, driving the PTIP networks almost back to their original state. This revealed good elasticity within strains up to 100%, feasible for many practical applications.

Notably, the MET sensor demonstrates good mechanochromic property and exhibits excellent reversibility. The structural color of orange-colored MET sensor underwent a gradual change from orange to green when longitudinally stretched from 0% to 20%, as shown in Fig. 3d and Movie S1. During its elongation from 0% to 51.6%, the reflection peak gradually and continuously shifted from 604 to 558 nm (Fig. 3f). A similar blueshift in structural color can also be intuitively observed in green-colored MET sensor (Fig. S18 and Movie S2). This color switching phenomenon can be explained by the following equation (3), as depicted schematically in Fig. 3e.

$$A_{max} = 2d(1 - v\varepsilon_x)n_{eff} \tag{3}$$

where ε_x is the tensile strain parallel to the stretching direction. During stretching, the thickness along observation direction (*z*) is decreased when assuming the total volume of the MET sensor remains unchanged.



Fig. 3. (a) Typical stress–strain curves of PTIP elastomer (black), PE (red), CPT (blue) and MET sensor (pink) at a tensile strain rate of 10 mm min⁻¹, respectively. Sample width, 5 mm; length, 20 mm. (b) Repeated cyclic tensile curves of MET sensor at 50% strain. There was no waiting time between two consecutive cyclic tensile (cycle 1-cycle 10). (c) Sequential cyclic tensile curves of the MET sensor at different strains without waiting time between two consecutive loadings. (d) Series of photographs of the MET sensor under different strains. Scale bar is 0.5 cm. (e) Mechanism illustration of the reversible changes of structural color during stretching and releasing. (f) Reflectance spectra of the MET sensor under various strains. (g) The position of reflection peak (λ_{max}) as a function of strain. (h) The reversible changes of λ_{max} at initial 10 stretching-releasing cycles, after 15,000 and 30,000 cycles of extension to 30% and relaxation to 0%. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

The tensile strain perpendicular to the stretching direction ε_z can be expressed in terms of ε_x and Poisson's ratio (ν): $\varepsilon_z = -\nu \varepsilon_x$. According to equation (3), when the MET sensor is stretched in the horizontal direction, the lattice spacing *d* is reduced from d_0 to d_1 in the vertical direction, thus resulting in a decrease in λ . In contrast, upon releasing, it returns rapidly to its original state, accompanied by the fully recovery of the structural color. This analysis is consistent with our experimental results.

As shown in Fig. 3g, there is a monotonic decrease in λ_{max} with strain up to 51.6%. Obviously, the curve can be divided into two linear regions with different slopes. When the strain $\varepsilon_x < 15.9\%$, λ_{max} changed rapidly with increasing strain because the interparticle distance is sufficient to avoid the contact between two nanospheres; the deformation of the MET sensor is governed by the elastic PTIP elastomer for small strain. When further increasing strain (15.9% < $\varepsilon_x < 51.6\%$), no significant shift in λ_{max} was observed due to the closed distance between neighboring PS@SiO₂ nanospheres [35,46]. Similar variations of λ_{max} as a function of strain can also be observed from the green-colored MET sensor shown in Fig. S19. Most importantly, the color shift generated by tensile strain is fully reversible and reproducible as shown in Fig. 3h. Owing to the hysteresis upon stretching to 30% (Fig. 3c), the variation of λ_{max} decreases slightly in the first 5 cycles and then tends to be stable with further increasing the number of cycles. Similar tendency can also be observed when it was tested with a strain of 50% (Fig. S20). These results demonstrate its excellent stability and durability during 30,000 continuous stretching/releasing cycles, which is beneficial for a visualized sensor in flexible electronics.

3.3. Electromechanical and interactive performance of the MET sensor

As discussed above, conductive AZO films were deposited onto the fiber surface to offer desired electrical property and mechanical stability; hence, the obtained CPT demonstrated excellent electromechanical properties (Fig. S21). More attractively, the MET sensor shows both the excellent electromechanical and interactive performance compared with CPT. Fig. 4a presents its relative resistance change ($\Delta R/R_0 = (R - R_0)/R_0$) curves as a function of applied strain, where R_0 and Rrepresent the initial and real-time resistances, respectively. Unexpectedly, the sensor exhibits a distinctive negative resistance variation with increasing strain in contrast to the vast majority of reported textilebased sensors [22,23,25–27]. Specifically, the curve could be divided into three linear strain regions, 0–32%, 32–98% and 98–200%, with a gauge factor (GF = $(\Delta R/R_0)/\varepsilon$, where ε denotes the strain) of –1.76, –0.36 and –0.04, respectively, demonstrating both its sensitivity and wide workable strain range. In addition, the impact of the semiembedding depth of CPT on the sensitivity of the MET sensor is investigated, as shown in Fig. S22. Moreover, it can be observed from Fig. 4b that the $\Delta R/R_0$ of the MET sensor was independent of the frequency



Fig. 4. (a) Relative resistance change of the MET sensor as a function of strain. (b) Relative resistance variation under cyclic stretching–releasing with a strain of 50% at frequency of 0.125, 0.25, 0.5 and 1.0 Hz. (c) Performance of the MET sensor under 30,000 stretching–releasing cycles (30% strain, 2 Hz). (d) The corresponding SEM images showing the morphology changes of the MET sensor at different strains. (e) Schematic illustration of the resistance model of an elementary unit in the weft-knitted structure showing the reversible resistance changes during stretching and releasing. (f) Changes of relative resistance and reflection peak wavelength of the sensor under different strains of 5%, 7%, 10%, 20%, 30%, and 40% at a frequency of 0.25 Hz. (g) Response and recovery time measurement for electrical and optical signals of the MET sensor applied strain from 0% to 5%.

within the tested frequency range (0.125–1.0 Hz), demonstrating its excellent frequency-independent electromechanical property. Importantly, the MET sensor displays stable and repeated electrical responses which can even withstand 30,000 stretching-releasing cycles (Fig. 4c and S23). The $\Delta R/R_0$ showed no evidence of hysteresis or degradation in performance during this process, exhibiting outstanding durability in comparison with most of hydrogel-based sensors, which is conducive to numerous practical applications.

To investigate the underlying mechanism of negative resistance variation, we track the morphology changes of the MET sensor under different strains (Fig. 4d). It can be seen that adjacent loop yarns (regarded as a basic repetitive unit) perpendicular to stress become more and more compact upon stretching, and the fibers in the same loop yarn become closer as well. Based on that, a resistance model which can be considered as a parallel circuit was proposed, and theoretical analysis was carried out correspondingly (Fig. 4e). The total original resistance *R* of a basic repetitive unit can thus be calculated by equation (4)

$$R = \frac{R_1 + R_2 + 2R_c}{2}$$
(4)

where R_1 and R_2 refer to the resistances in the horizontal and vertical

parts of one loop yarn, respectively. R_c is the contact resistance between two adjacent loop yarns in the vertical direction without any applied strain. Two instantaneous structural changes can lead to the decrease of R with increasing strain: 1) the increase of the contact areas at the junction, which is a critical factor determining the sensor resistance response; 2) the compactness of the fibers in the same loop yarn, which also reduces the resistance of the yarn itself (see the detailed analysis in Fig. S24 and Supporting Information). Therefore, the total resistance of the MET sensor decreases with increasing strain, indicating its negative resistance variation.

As expected, the electrical response and the reflection wavelength change synchronously, stably, continuously and proportionally as the strain increased (Fig. 4f), which was consistent with the results displayed in Fig. 3g and 4a. Such synchronous changes of both optical and electrical signals could be ascribed to their tight binding between CPT and PE as discussed above. Notably, the MET sensor exhibits synchronous and rapid response (≈ 0.30 s) and recovery speed (≈ 0.22 s) under 5% strain (Fig. 4g). Moreover, its response (≈ 0.34 s) and recovery speed (≈ 0.28 s) under 15% strain are also displayed in Fig. S25. These results indicate the good sensitivity and visualized sensing capabilities of the MET sensor, which is promising for visualized interactive devices and



Fig. 5. (a) Schematic illustration showing the application of the MET sensor as a visualized interactive device for detecting human joint movements. Corresponding time-dependent signals of relative resistance change and reflectance spectra when monitoring the different joint movements of the human body. (b) Finger bending with different frequencies in real time. (c) Wrist bending. (d) Elbow bending. (e) Knee bending. (f) Neck bending. The photographs show the structural color changes of the MET sensor during the same process.

smart human-machine interfaces.

3.4. Applications of the MET sensor as a visualized interactive device

The excellent synchronous dual-signal response of the MET sensor enables its uses as a visualized interactive device. The color variations can be directly perceived by human eyes and quantified by optical spectroscopy, while the electrical readout can be accurately recorded by electrical instruments, thereby achieving direct human-machine interactive visualization (Fig. 5a and Movie S3). As a proof-of-concept, it was designed as a visualized interactive device to monitor human joint motions by directly attaching it onto finger, wrist, elbow, knee and neck, respectively (Fig. 5b-f). In the case of finger motion, the relative resistance change decreased immediately when the index finger began to bend, accompanied by the variation of structural color from orange red to green. This was further confirmed by reflectance spectra with λ_{max} shifting from 604 to 571 nm. With the recovery of the finger, the relative resistance change and structural color returned to the original state. In addition, the electrical signal changes and the visual color transitions of the sensor were consistent, displaying high stability and reliability even suffering from continuous finger motions at different frequencies; the sensor's dual-signal response also showed a frequency-independent property. Stable dual-signal responses of the sensor were also displayed in wrist, elbow, knee and neck joint movements as shown in Fig. 5c-f. Compared with the single electrical response, the introduction of additional optical response can effectively avoid the measurement errors under interference (i.e., temperature, humidity), as shown in Fig. S26. These results demonstrate that the MET sensor exhibited interactive visualization of strain and stable dual-signal response for detecting human joint movements. Compared with other types of sensors, the MET sensor can be easily knitted, sewed on garments or attached on human skin directly for tracking human joint motions with dual-signal response, which can bring intriguing developments in emerging interactive electronics.

4. Conclusion

In summary, we have developed a series of novel interactively MET sensors with rapid and durable electrical/optical response, and demonstrate their applications in visualized stretchable electronics. Typically, the MET sensors are realized by the rational integration of mechanochromic supramolecular photonic PTIP elastomers and ALDdeposited conductive AZO polyester textiles. Taking advantage of their semi-embedded structures, the resultant sensors exhibit not only distinct negative resistance variations, but also dynamic and reversible color-switching capabilities upon stretching simultaneously. Attractively, the MET sensor possesses brilliant structural color, excellent mechanical resilience, fast optical/electrical response (≈ 0.30 s) and recovery speed (\approx 0.22 s). Notably, the synergistic combination of PE and CPT imparts it with higher toughness (35.6 kJ m^{-3}) in comparison with PE (20.7 kJ m^{-3}) or CPT (24.9 kJ m^{-3}) alone. Most importantly, the MET sensor can even withstand 30,000 stretching-releasing cycles and its dual-signal response still remains steady, demonstrating its superior mechanical stability and excellent durability. Based on the admirable performances, we have demonstrated its versatile capabilities as a visualized interactive device for tracking human joint movements (i.e., finger, elbow, wrist, neck and knee joint bending). It is predictable that these MET sensors could be useful in a broad range of applications concerning visualized sensors, electronic skins and intelligent robots.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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References

- T.Q. Trung, N.-E. Lee, Flexible and stretchable physical sensor integrated platforms for wearable human-activity monitoring and personal healthcare, Adv. Mater. 28 (22) (2016) 4338–4372.
- [2] W. Gao, H. Ota, D. Kiriya, K. Takei, A. Javey, Flexible electronics toward wearable sensing, Acc. Chem. Res. 52 (3) (2019) 523–533.
- [3] C. Wang, K. Xia, H. Wang, X. Liang, Z. Yin, Y. Zhang, Advanced carbon for flexible and wearable electronics, Adv. Mater. 31 (2019) 1801072.
- [4] C. Luo, B. Tian, Q. Liu, Y. Feng, W. Wu, One-step-printed, highly sensitive, textilebased, tunable performance strain sensors for human motion detection, Adv. Mater. Technol. 5 (2020) 1900925.
- [5] B. Tian, Q. Liu, C. Luo, Y. Feng, W. Wu, Multifunctional ultrastretchable printed soft electronic devices for wearable applications, Adv. Electron. Mater. 6 (2020) 1900922.
- [6] B. Tian, W. Yao, P. Zeng, X. Li, H. Wang, L.i. Liu, Y.u. Feng, C. Luo, W. Wu, Allprinted, low-cost, tunable sensing range strain sensors based on Ag nanodendrite conductive inks for wearable electronics, J. Mater. Chem. C 7 (4) (2019) 809–818.
- [7] W. Wu, Stretchable electronics: functional materials, fabrication strategies and applications, Sci. Technol. Adv. Mater. 20 (2019) 187–224.
 [8] N. W. X. Cheng, O. Zhong, L. Zhong, W. Li, P. G. Wang, R. Lin, L. Zhong, Collisil
- [8] N. Wu, X. Cheng, Q. Zhong, J. Zhong, W. Li, B.o. Wang, B. Hu, J. Zhou, Cellular polypropylene piezoelectret for human body energy harvesting and health monitoring, Adv. Funct. Mater. 25 (30) (2015) 4788–4794.
- [9] Y. Zhang, P. He, M. Luo, X. Xu, G. Dai, J. Yang, Highly stretchable polymer/silver nanowires composite sensor for human health monitoring, Nano Res. 13 (4) (2020) 919–926.
- [10] Y. Liu, H. Zheng, L. Zhao, S. Liu, K. Yao, D. Li, C. Yiu, S. Gao, R. Avila, C. Pakpong, L. Chang, Z. Wang, X. Huang, Z. Xie, Z. Yang, X. Yu, Electronic skin from highthroughput fabrication of intrinsically stretchable lead zirconate titanate elastomer, Research 2020 (2020) 1085417.
- [11] J. Yang, J. Mun, S.Y. Kwon, S. Park, Z. Bao, S. Park, Electronic skin: recent progress and future prospects for skin-attachable devices for health monitoring, robotics, and prosthetics, Adv. Mater. 31 (2019) 1904765.
- [12] Y. Lee, J. Park, A. Choe, S. Cho, J. Kim, H. Ko, Mimicking human and biological skins for multifunctional skin electronics, Adv. Funct. Mater. 30 (2020) 1904523.
- [13] Z. Liu, Y. Wang, Y. Ren, G. Jin, C. Zhang, W. Chen, F. Yan, Poly(ionic liquid) hydrogel-based anti-freezing ionic skin for a soft robotic gripper, Mater. Horiz. 7 (3) (2020) 919–927.
- [14] Z. Lei, Q. Wang, S. Sun, W. Zhu, P. Wu, A bioinspired mineral hydrogel as a selfhealable, mechanically adaptable ionic skin for highly sensitive pressure sensing, Adv. Mater. 29 (2017) 1700321.
- [15] J. Zhong, Y. Ma, Y.u. Song, Q. Zhong, Y. Chu, I. Karakurt, D.B. Bogy, L. Lin, A flexible piezoelectret actuator/sensor patch for mechanical human-machine interfaces, ACS Nano 13 (6) (2019) 7107–7116.
- [16] L. Wang, W. Liu, Z. Yan, F. Wang, X. Wang, Stretchable and shape-adaptable triboelectric nanogenerator based on biocompatible liquid electrolyte for biomechanical energy harvesting and wearable human-machine interaction, Adv. Funct. Mater. 31 (2020) 2007221.
- [17] G. Chen, Y. Li, M. Bick, J. Chen, Smart textiles for electricity generation, Chem. Rev. 120 (8) (2020) 3668–3720.
- [18] S. Seyedin, P. Zhang, M. Naebe, S. Qin, J. Chen, X. Wang, J.M. Razal, Textile strain sensors: a review of the fabrication technologies, performance evaluation and applications, Mater. Horiz. 6 (2019) 219–249.
- [19] M. Zhang, M. Zhao, M. Jian, C. Wang, A. Yu, Z. Yin, X. Liang, H. Wang, K. Xia, X. Liang, J. Zhai, Y. Zhang, Printable smart pattern for multifunctional energymanagement e-textile, Matter 1 (1) (2019) 168–179.
- [20] T.Q. Trung, T.M.L. Dang, S. Ramasundaram, P.T. Toi, S.Y. Park, N.-E. Lee, A stretchable strain-insensitive temperature sensor based on free-standing elastomeric composite fibers for on-body monitoring of skin temperature, ACS Appl. Mater. Interfaces 11 (2) (2019) 2317–2327.
- [21] W. He, C. Wang, H. Wang, M. Jian, W. Lu, X. Liang, X. Zhang, F. Yang, Y. Zhang, Integrated textile sensor patch for real-time and multiplex sweat analysis, Sci. Adv. 5 (11) (2019) eaax0649, https://doi.org/10.1126/sciadv.aax0649.
- [22] Y. Li, Y. Li, M. Su, W. Li, Y. Li, H. Li, X. Qian, X. Zhang, F. Li, Y. Song, Electronic textile by dyeing method for multiresolution physical kineses monitoring, Adv. Electron. Mater. 3 (2017) 1700253.
- [23] S.J. Kim, W. Song, Y. Yi, B.K. Min, S. Mondal, K.-S. An, C.-G. Choi, High durability and waterproofing rgo/swcnt-fabric-based multifunctional sensors for humanmotion detection, ACS Appl. Mater. Interfaces 10 (4) (2018) 3921–3928.

- [24] Z. Yang, Y. Pang, X.-L. Han, Y. Yang, J. Ling, M. Jian, Y. Zhang, Y. Yang, T.-L. Ren, Graphene textile strain sensor with negative resistance variation for human motion detection, ACS Nano 12 (9) (2018) 9134–9141.
- [25] C. Wang, X. Li, E. Gao, M. Jian, K. Xia, Q.i. Wang, Z. Xu, T. Ren, Y. Zhang, Carbonized silk fabric for ultrastretchable, highly sensitive, and wearable strain sensors, Adv. Mater. 28 (31) (2016) 6640–6648.
- [26] M. Zhang, C. Wang, H. Wang, M. Jian, X. Hao, Y. Zhang, Carbonized cotton fabric for high-performance wearable strain sensors, Adv. Funct. Mater. 27 (2017) 1604795.
- [27] C. Wang, K. Xia, M. Jian, H. Wang, M. Zhang, Y. Zhang, Carbonized silk georgette as an ultrasensitive wearable strain sensor for full-range human activity monitoring, J. Mater. Chem. C 5 (30) (2017) 7604–7611.
- [28] S. Wang, Y. Fang, H. He, L. Zhang, C. Li, J. Ouyang, Wearable stretchable dry and self-adhesive strain sensors with conformal contact to skin for high-quality motion monitoring, Adv. Funct. Mater. 31 (2020) 2007495.
- [29] Z. Zhang, Z. Chen, Y.u. Wang, Y. Zhao, Bioinspired conductive cellulose liquidcrystal hydrogels as multifunctional electrical skins, Proc. Natl. Acad. Sci. U. S. A. 117 (31) (2020) 18310–18316.
- [30] J. Teyssier, S.V. Saenko, D. van der Marel, M.C. Milinkovitch, Photonic crystals cause active colour change in chameleons, Nat. Commun. 6 (2015) 6368.
- [31] S.A. Morin, R.F. Shepherd, S.W. Kwok, A.A. Stokes, A. Nemiroski, G.M. Whitesides, Camouflage and display for soft machines, Science 337 (6096) (2012) 828–832.
- [32] F. Liu, B.Q. Dong, X.H. Liu, Y.M. Zheng, J. Zi, Structural color change in longhorn beetles Tmesisternus isabellae, Opt. Express 17 (18) (2009) 16183, https://doi. org/10.1364/OE.17.016183.
- [33] D. Gur, B.A. Palmer, B. Leshem, D. Oron, P. Fratzl, S. Weiner, L. Addadi, The mechanism of color change in the neon tetra fish: a light-induced tunable photonic crystal array, Angew. Chem., Int. Ed. 54 (2015) 12426–12430.
- [34] P. Wu, J. Wang, L. Jiang, Bio-inspired photonic crystal patterns, Mater. Horiz. 7 (2) (2020) 338–365.
- [35] G.H. Lee, T.M. Choi, B. Kim, S.H. Han, J.M. Lee, S.-H. Kim, Chameleon-inspired mechanochromic photonic films composed of non-close-packed colloidal arrays, ACS Nano 11 (11) (2017) 11350–11357.
- [36] M. Vatankhah-Varnosfaderani, A.N. Keith, Y. Cong, H. Liang, M. Rosenthal, M. Sztucki, C. Clair, S. Magonov, D.A. Ivanov, A.V. Dobrynin, S.S. Sheiko, Chameleon-like elastomers with molecularly encoded strain-adaptive stiffening and coloration, Science 359 (6383) (2018) 1509–1513.
- [37] H. Yi, S.H. Lee, H. Ko, D. Lee, W.G. Bae, T.I. Kim, D.S. Hwang, H.E. Jeong, Ultraadaptable and wearable photonic skin based on a shape-memory, responsive cellulose derivative, Adv. Funct. Mater. 29 (2019) 1902720.
- [38] P. Wu, X. Shen, C.G. Schäfer, J. Pan, J. Guo, C. Wang, Mechanochromic and thermochromic shape memory photonic crystal films based on core/shell nanoparticles for smart monitoring, Nanoscale 11 (42) (2019) 20015–20023.
- [39] P. Zhao, B. Li, Z. Tang, Y. Gao, H. Tian, H. Chen, Stretchable photonic crystals with periodic cylinder shaped air holes for improving mechanochromic performance, Smart Mater. Struct. 28 (2019), 075037.
- [40] J. Chen, L. Xu, M. Yang, X. Chen, X. Chen, W. Hong, Highly stretchable photonic crystal hydrogels for a sensitive mechanochromic sensor and direct ink writing, Chem. Mater. 31 (21) (2019) 8918–8926.
- [41] Y. Yue, T. Kurokawa, M.A. Haque, T. Nakajima, T. Nonoyama, X. Li, I. Kajiwara, J. Gong, Mechano-actuated ultrafast full-colour switching in layered photonic hydrogels, Nat. Commun. 5 (2014) 4659.
- [42] X.-Q. Wang, C.-F. Wang, Z.-F. Zhou, S.u. Chen, Robust mechanochromic elastic one-dimensional photonic hydrogels for touch sensing and flexible displays, Adv. Opt. Mater. 2 (7) (2014) 652–662.
- [43] X. Jia, J. Wang, K.e. Wang, J. Zhu, Highly sensitive mechanochromic photonic hydrogels with fast reversibility and mechanical stability, Langmuir 31 (31) (2015) 8732–8737.
- [44] H. Tan, Q. Lyu, Z. Xie, M. Li, K. Wang, K. Wang, B. Xiong, L. Zhang, J. Zhu, Metallosupramolecular photonic elastomers with self-healing capability and angleindependent color, Adv. Mater. 31 (2019) 1805496.
- [45] M. Li, H. Tan, L. Jia, R. Zhong, B. Peng, J. Zhou, J. Xu, B. Xiong, L. Zhang, J. Zhu, Supramolecular photonic elastomers with brilliant structural colors and broadspectrum responsiveness, Adv. Funct. Mater. 30 (2020) 2000008.
- [46] T. Ito, C. Katsura, H. Sugimoto, E. Nakanishi, K. Inomata, Strain-responsive structural colored elastomers by fixing colloidal crystal assembly, Langmuir 29 (45) (2013) 13951–13957.
- [47] G.H. Lee, S.H. Han, J.B. Kim, J.H. Kim, J.M. Lee, S.-H. Kim, Colloidal photonic inks for mechanochromic films and patterns with structural colors of high saturation, Chem. Mater. 31 (19) (2019) 8154–8162.

- [48] M. Li, B. Zhou, Q. Lyu, L. Jia, H. Tan, Z. Xie, B. Xiong, Z. Xue, L. Zhang, J. Zhu, Selfhealing and recyclable photonic elastomers based on a water soluble supramolecular polymer, Mater. Chem. Front. 3 (12) (2019) 2707–2715.
- [49] Y. Chang, L. Wang, R. Li, Z. Zhang, Q. Wang, J. Yang, C. Guo, T. Pan, First decade of interfacial iontronic sensing: from droplet sensors to artificial skins, Adv. Mater. 33 (2020) 2003464.
- [50] P. Snapp, P. Kang, J. Leem, S. Nam, Colloidal photonic crystal strain sensor integrated with deformable graphene phototransducer, Adv. Funct. Mater. 29 (2019) 1902216.
- [51] Y. Wang, Y. Yu, J. Guo, Z. Zhang, X. Zhang, Y. Zhao, Bio-Inspired stretchable, adhesive, and conductive structural color film for visually flexible electronics, Adv. Funct. Mater. 30 (2020) 2000151.
- [52] B. Tang, X. Zheng, T. Lin, S. Zhang, Hydrophobic structural color films with bright color and tunable stop-bands, Dyes Pigments 104 (2014) 146–150.
- [53] S.-Z. Yu, W.-B. Niu, S.-L. Wu, W. Ma, S.-F. Zhang, Robust and flexible thermalplasticizing 3D shaped composite films with invariable and brilliant structural color, J. Mater. Chem. C 6 (47) (2018) 12814–12821.
- [54] K. Zhao, W. Niu, S. Zhang, Highly stretchable, breathable and negative resistance variation textile strain sensor with excellent mechanical stability for wearable electronics, J. Mater. Sci. 55 (6) (2020) 2439–2453.
- [55] F. Chen, H. Yang, K. Li, B. Deng, Q. Li, X. Liu, B. Dong, X. Xiao, D. Wang, Y. Qin, S.-M. Wang, K.-Q. Zhang, W. Xu, Facile and effective coloration of dye-inert carbon fiber fabrics with tunable colors and excellent laundering durability, ACS Nano 11 (10) (2017) 10330–10336.
- [56] W. Niu, L. Zhang, Y. Wang, Z. Wang, K. Zhao, S. Wu, S. Zhang, A.I.Y. Tok, Multicolored photonic crystal carbon fiber yarns and fabrics with mechanical robustness for thermal management, ACS Appl. Mater. Interfaces 11 (35) (2019) 32261–32268.
- [57] Y. Lai, X. Kuang, P. Zhu, M. Huang, X. Dong, D. Wang, Colorless, transparent, robust, and fast scratch-self-healing elastomers via a phase-locked dynamic bonds design, Adv. Mater. 30 (2018) 1802556.
- [58] W. Wang, H. Chen, Q. Dai, D. Zhao, Y. Zhou, L. Wang, D. Zeng, Thermally healable PTMG-based polyurethane elastomer with robust mechanical properties and high healing efficiency, Smart Mater. Struct. 28 (2019), 015008.
- [59] Y. Yao, Z. Xu, B. Liu, M. Xiao, J. Yang, W. Liu, Multiple h-bonding chain extenderbased ultrastiff thermoplastic polyurethanes with autonomous self-healability, solvent-free adhesiveness, and aie fluorescence, Adv. Funct. Mater. 31 (2020) 2006944.
- [60] H. Bi, Z. Ren, R. Guo, M. Xu, Y. Song, Fabrication of flexible wood flour/ thermoplastic polyurethane elastomer composites using fused deposition molding, Ind. Crop. Prod. 122 (2018) 76–84.
- [61] S. Zhao, Z. Wang, H. Pang, W. Zhang, S. Zhang, J. Li, Li, Li, Organic-inorganic nanohybrid polyurethane elastomer based on dopamine-mediated biomimetic codeposition thought toward multiple improved properties, Appl. Surf. Sci. 493 (2019) 1340–1349.
- [62] Y. Chen, A.M. Kushner, G.A. Williams, Z. Guan, Multiphase design of autonomic self-healing thermoplastic elastomers, Nat. Chem. 4 (6) (2012) 467–472.
- [63] D. Wang, J. Xu, J. Chen, P. Hu, Y. Wang, W. Jiang, J. Fu, Transparent, mechanically strong, extremely tough, self-recoverable, healable supramolecular elastomers facilely fabricated via dynamic hard domains design for multifunctional applications, Adv. Funct. Mater. 30 (2020) 1907109.
- [64] W.B. Ying, Z. Yu, D.H. Kim, K.J. Lee, H. Hu, Y. Liu, Z. Kong, K. Wang, J. Shang, R. Zhang, J. Zhu, R.-W. Li, Waterproof, highly tough, and fast self-healing polyurethane for durable electronic skin, ACS Appl. Mater. Interfaces 12 (9) (2020) 11072–11083.
- [65] J. Hu, R. Mo, X. Jiang, X. Sheng, X. Zhang, Towards mechanical robust yet selfhealing polyurethane elastomers via combination of dynamic main chain and dangling quadruple, Polymer 183 (2019), 121912.
- [66] M. Iwata, M. Teshima, T. Seki, S. Yoshioka, Y. Takeoka, Bio-Inspired bright structurally colored colloidal amorphous array enhanced by controlling thickness and black background, Adv. Mater. 29 (2017) 1605050.
- [67] C. Fenzl, T. Hirsch, O.S. Wolfbeis, Photonic crystals for chemical sensing and biosensing, Angew. Chem., Int. Ed. 53 (13) (2014) 3318–3335.
- [68] J.B. Kim, S.Y. Lee, J.M. Lee, S.-H. Kim, Designing structural-Color patterns composed of colloidal arrays, ACS Appl. Mater. Interfaces 11 (16) (2019) 14485–14509.
- [69] Y. Qiao, Y. Li, W. Li, J. Bao, Y. Zheng, L. Feng, Y. Ma, K. Yang, A. Wu, H. Bai, Y. Yang, Preparation and luminescence properties of core-shell structure composites SiO2@ANA-Si-Eu and SiO2@ANA-Si-Eu-L and core-shell-shell structure composites SiO2@ANA-Si-Eu@SiO2 and SiO2@ANA-Si-Eu-L@SiO2, New J. Chem. 44 (3) (2020) 1107–1116.