Flexible Sensor for Invisible Respiratory Monitoring via Construction of a 2D Stacked Micronetwork

Jiajun Guo, Kailin Zhang, Ruixian Dai, Min Nie,* Yijun Li,* and Qi Wang

ABSTRACT: With the advent of 5G and the Internet of Things era, sensitive and stable sensors have begun to develop rapidly, which are important substantial fundaments of smart medical care. In this study, based on the positive temperature coefficient (PTC) in conductive polymer composites (CPC), a novel polyolefin elastomer (POE)/carbon fiber (CF) composite was prepared. By regulating the rheological behavior of the polymer matrix, we realized its controllable thermal expansion in the temperature field and finally realized the reversible construction—destruction of the conductive CF network. Under optimal molecular weight conditions, the POE/CF PTC sensor showed a high sensitivity of $0.11 \, ^\circ \text{C}^{-1}$ and stability. It was also demonstrated that the heat transfer efficiency of the composite material played an essential role in the sensitivity of the as-prepared PTC sensor. Most impressively, we have assembled an invisible respiratory monitoring device based on the POE/CF composite to achieve real-time monitoring of human breathing, which displayed wide potential prospects in thermal monitoring and provided good prospects for micron-scale functional composites.

1. INTRODUCTION

Since minor changes in human physiological characteristics such as variation of body temperature are often considered symptoms of diseases, great attention has been called on precisely monitoring for the real-time physical signs through sensing devices.1−3 Numerous studies have focused on the design and manufacture of various sensor components, which are able to detect the diversified physical signals of the human body and then convert signals into digital data as per the changes in resistance or voltage.4,5 As a result, biometric information can be easily collected and help to predict the illness in time. However, many of the prepared sensors are not exquisite-looking or exquisite.6,7

Meantime, the systematic research in the sensing mechanism at real-time physical signs has also been insufficient.8 Composites featuring a combination of various fillers and matrices can easily achieve a wide range of adjustable properties, which satisfy most kinds of applications and are used in every corner of our lives such as medical devices, vehicles, and intelligent robots.9,10 But most of the recent works concentrate on the preparation of nanocomposites with nanofillers (metal nanowire and graphene) and complex nanoscale network structures to implement certain functions.11−13 In general, high-cost nanofillers are always preprocessed with multiple steps such as dissolution and ultrasonic dispersion, and the obtained nanostructures are always difficult to control because of the weak interaction between fillers and matrices as well as the agglomeration tendency of nanofillers.14,15 Therefore, the elaborately prepared nanonetwork does not reflect its value or show its superiority under certain conditions. By contrast, the facile micron-level network with special stability and controllability is considerable.

On the other hand, the majority of polymer composites exhibit specific performance, which is benefited from the studies in terms of filler types, network configurations, and processing methods.16,17 As a result, the influence of the matrix property is easily despised and often overlooked in to the case of sensing composites. A few previous works have proved that the matrix nature is also crucial to the integrated performance of composites, such as the positive effects of multiphase matrices with different melting points on sensing sensitivity.18,19 Consequently, the lack of a systematic study about the impact of the matrix nature on sensing performance has greatly impeded the efficient design and manufacture of functional composites, especially for composites with micron-scale networks.
Carbon fiber (CF) is a one-dimensional fibrous material with high electrical conductivity. Especially, different from other nanoscale carbon-based materials, such as graphene and carbon nanotubes, micron-sized CF has the ability to disperse well during processing thanks to its superior rigidity. Its unique structure with high aspect ratio also makes it an ideal material to trace fluid flow. Therefore, a CF network can be induced to form an oriented arrangement under a simple flow field of a polymer melt and can be transformed into a wide variety of configurations. Most importantly, CPCs feature a unique positive temperature coefficient (PTC) effect. Since the thermal expansion coefficient of the conductive filler is much lower than that of the polymer matrix, when the temperature rises, the polymer expands, while the conductive filler remains in the initial position, resulting in the loose packing of the filler and the decreased density of the conductive pathways. It is rational to deduce that if the expansion rate of the polymer matrix can be precisely controlled, it is promising to prepare PTC sensors with a rapid and stable response.

Herein, polyolefin elastomer (POE)/CF composite films with micron-sized orientation networks based on CF for better processing and stability have been prepared. First, we studied the relationship between the molecular chain structure of the matrix and the macro expansion of composite films under the temperature field, and the consequent resistance changes with adjustable PTC have been achieved. In consideration of the timeliness of heat transfer, the sensing sensitivity under the fluctuant temperature field was also adjusted by regulating the thickness of the films. Furthermore, the samples were set into the nasal cavity and the sensing performance of the sample was investigated for human respiratory monitoring, demonstrating excellent sensitivity and stability. Overall, a valuable demonstration for efficient and stable invisible respiratory monitoring based on the proposed micro-scale orientation network has been put forward, which opens an opportunity for other flexible devices and functional composites.

### 2. RESULTS AND DISCUSSION

The mobility of molecular chains in a polymer is directly related to its thermal expansion coefficient. When the movement of the molecular chain is sensitive to temperature, due to the increase in entropy, the oriented molecular chains are keen to shrink into a random coil, which exhibits a large volume expansion in the macroscopic view. POE, a typical thermoplastic elastomer, is composed of ethylene and octene segments. The former can crystalize as a physical cross-linking point, while the latter weakens the regularity of the ethylene segment to form an amorphous region with hyperelastic. As shown in Figure 1a,b, the molecular weight distribution curves of L-POE and H-POE presented single and narrow peaks as well as narrow molecular weight distributions. The number average molecular weight ($M_n$) of L-POE was 73458, which was 1.6 times that of H-POE. Obviously, L-POE displayed more long-chain branched octene chain blocks (Table 1), which will hinder the crystallization of ethylene segments.

![Figure 1](https://dx.doi.org/10.1021/acsomega.0c05367)

**Table 1. Product Information and Number Average Molecular Weight of Two Grades of POE**

| trade name | $M_n$ | comonomer content (wt %) |
|------------|-------|--------------------------|
| Engage 8400 | 73458 | 40                       |
| Engage 8450 | 46220 | 20                       |
more. Since the ethylene segment that is not constrained by the crystal lattice presents an amorphous shape, the thermal motion of the molecule is much stronger than that of a crystalline ethylene chain. Unambiguously, as the temperature increases, the molecular chain movement of L-POE is more sensitive than that of H-POE.

The thermodynamic behavior of the material involves the characteristic glass-transition temperature, melting point, etc.; to understand the performance of POE with different segment structures, we have plotted the melting curves of specimens, as shown in Figure 1c. All curves show a step at 42 °C, which suggests the glass-transition temperature of the polymer. One can notice that L-POE and H-POE also showed complete melting peaks at 60 and 102 °C, indicating the lower melting temperature of L-POE. It can be attributed to the fact that on the one hand, the molecular chain containing more octene blocks is imparted with better flexibility; on the other hand, the branch leads to more crystal defects and lower melting temperatures. Accordingly, it can be concluded with confidence that L-POE has fewer crystalline ethylene segments as physical cross-linking points and more octene segments with side chains, so the molecules are more flexible and the diffusion movement of the segments requires lower energy. Moreover, it can be predicted accordingly that macroscopic thermal expansion of L-POE is more obvious.

Linear expansion is a critical indicator of the thermal expansion of a material, meaning the expansion ratio of the material is in one direction, which can be measured by the TMA test (Figure 1d). Although the addition of CFs enhanced the stiffness of the material and hindered its expansion by steric hindrance, the expansion ratio of the L-POE/CF film remained at a fairly high level of 5.7% when the samples were heated from 20 to 42 °C, which is 3 times that of H-POE/CF, confirming the excellent temperature sensitivity of the movement of the flexible L-POE molecular segment. It is worth noting that the thermal expansion coefficient of L-POE is calculated to be $909 \times 10^{-6}$ m/m·k based on the equation $\sigma = \Delta L/(L \times \Delta T)$, which is significantly higher than that of conventional plastics such as PE with a thermal expansion coefficient of $200 \times 10^{-6}$ m/m·k.

To understand the micron-scale CF network structure of the composite films intuitively, scanning electron microscopy (SEM) images of the frozen fracture surface of the composite films are shown in Figure 2a,b. One can obviously observe that due to the advection of the unconfined composite melt under pressure during compression molding, the CF crossover network exhibited a lamellar stack structure in the film, and there was almost no carbon fiber penetrating between the layers, presenting a two-dimensional network configuration. Although the fibers in each layer of the network are interconnected with each other stably, the connection between the layers is weak and easy to be misplaced and detached.

To demonstrate more clearly the effect of this expansion on the isolation of two-dimensional CF networks, SAXS measurements have been performed to characterize the composite film assembly structure at different temperatures, and the result is shown in Figure 2d1−d3. It is well documented that the scattering intensity after being fitted by the power law is a key indicator for fiber intersections and exhibits $q \sim n$ dependence at a low-q regime, and the value of $n$ is considered as a normal to evaluate the perfection of the filler network. Moreover, a more perfect network generally accompanies a larger $n$ value.
When the samples were heated to 40 °C, the $n$ value declines from 3.368 to 3.189, with a large decreasing value of $\sim 0.17$, while for other POE composites, only a value of $\sim 0.04$ can be observed in Figure 2e. It can be inferred that the expansion of the matrix inserted in the CF network contributed to the separation and slippage of CF between each other. The biggest expansion ratio of L-POE/CF indicated the farthest distance of CF separation and the most noticeable loosening of the network structure; consequently, an obvious variation in the L-POE/CF $n$ value has been found. In addition, the structural distortion of the micron-scale network is also the inducement of the apparent resistance change of the composite, so it is foreseeable that the change in the composite film resistance under temperature fluctuation is inevitably related to the nature of the matrix. Thanks to this two-dimensional network structure, the expansion of the polymer in the gap can easily increase the distance between the network layers, as illustrated in Figure 2f.

The electrical resistance of the composite films was positively correlated with temperature and exhibited continuous temperature dependence, as shown in Figure 3a. The resistance of the L-POE/CF composite film increased to almost 3.4 times that of the initial, while the resistance of the H-POE/CF one only became less than 1.4 times the starting value when the temperature rose to 45 °C. Combined with preceding conclusions, such remarkable changes can mainly be attributed to the significant expansion of L-POE inserted between CF networks on account of the large degree of molecular segment motion. To make the resistance change more intuitive, the TCR coefficient that represents the temperature-dependent sensitivity has been calculated to quantify the level of change in resistance through fitting the resistance-temperature curves according to the following equation:

$$\text{TCR} = \left(\frac{1}{R_0}\right) \times \left(\frac{dR}{dT}\right)$$  \hspace{1cm} (1)

where $R_0$ represents the sample resistance at the initial temperature (23 °C) and $R$ represents the sample resistance at temperature $T$. Besides, the conductivities of the composite films at room temperature were also calculated and listed with the TCR coefficient together in the inset of Figure 3a. Although the initial conductivity of the three samples is almost the same, L-POE showed an unexpectedly high TCR coefficient of 0.11 °C$^{-1}$, which is 4.4 times the value of B-POE and 6.5 times that of H-POE. The adjustable sensitivity of POE/CF films in the field of temperature sensing has been initially demonstrated.

The conductive mechanism of POE/CF films was investigated by the voltammetric test, where the L-POE/CF film was tested under different temperatures from 23 to 45 °C (Figure 3b). Obviously, the volt-ampere characteristic curves at all temperatures were almost strictly linear, which was in accordance with Ohm’s law, demonstrating a direct contact conduction mechanism of the filler. Although it is easy to achieve nanoscale separation of CFs due to the expansion of the matrix inserted between the oriented CF network, the matrix expansion is not strong enough to change the tight linkage of most parallel-crossed carbon fiber networks, which is considered to be one of the main factors inducing electron tunneling. Besides, the integrated conductive network is also experiencedly regarded as a parallel circuit of all conductive paths. As a result, the composite films involving micron-sized...
orientation networks follow the contact conduction mechanism under increasing temperature.

To study the sensing sensitivity more systematically, we measured the resistance change of L-POE/CF films under gradient temperature fluctuation with steps of 2°C, and the result is shown in Figure 3c. The resistance displayed a simultaneous stepped upward trend accompanied by increasing temperature. It was notable that the resistance remained invariant at the interval. Therefore, the favorable sensitivity of composite films for monitoring temperature fluctuation can be inferred. Furthermore, the heat exchange efficiency between the composite film and the environment is also a key factor for the sensing performance of sensors in the changing temperature field. The thermal conductivity, which is the major indicator to evaluate the thermal transmission capacity of the material, was measured as shown in Figure 3d. The thermal conductivity of all samples was maintained at a relatively low level, not more than 0.6 W/(m·K), so the thermal energy is difficult to transfer to the middle layer of the film and the corresponding thermal expansion should be weak. However, it provides an opportunity to adjust the sensing sensitivity of the film through regulating the thickness of the film because the ratio of the effective volume of thermal expansion to the total volume is moderated by changing the film thickness. It should also be noted that the thermal conductivity of the H-POE/CF film is higher than that of L-POE derived from the enlarged ethylene crystallization zone, where better steric regularity is conducive to faster thermal transmission.

To explore the potential sensing application of composite films, the samples were set into the nasal cavity and linked to a Keithley 6487 apparatus. The samples were fixed in a U-shaped nose clip with a cross-clip cavity, as shown in Figure 4a, and the corresponding resistance changes were calculated as $\Delta R/R$ and are shown in Figure 4b. One can notice that the resistance increased when the participant exhaled, whereas it declined when the participant inhaled. This can be attributed to the fact that the heat inside the body carried by the exhaled gas warms up the sample, while the ambient air that is inhaled cools the sample. Consequently, the sample repeatedly expands and shrinks under the temperature fluctuation field in the nasal cavity, causing the periodically varying resistance of samples. We recorded the resistance in 500 s, and the stability of the sensing behavior was confirmed. Among them, L-POE/CF films exhibited the maximum amplitude of periodical variation, while the change of H-POE/CF was faint, agreeing well with the fact that the most sensitive response was of L-POE/CF films. In addition, to prove the influence of the film thickness on the sensing performance, L-POE/CF films of different thicknesses were steeped in 42 °C warm water and then were taken out. The changes in resistance were measured and are plotted in Figure 4c. The thinner the film, the more dramatic the change in the resistance. The $\Delta R/R$ value became $\sim 4.8$ times higher than that of 1 mm thickness. It may be caused by different sample heating modes: in water, the sample was omnidirectionally heated, while on a hot stage, the sample was heated only through the contact surface. Consequently, larger expansion areas contributed to more remarkable changes in the resistance. The curves were also fitted according to the attenuation formula as follows to calculate the response time and the recovery time.

Figure 4. (a) Schematic diagram of an invisible respiratory monitoring device. (b) Curves of the $\Delta R/R$ value of three composite films over time when the films were set into the nasal cavity and (b1) locally amplified curves. (c) $\Delta R/R$ values of L-POE films with different thicknesses as a function of time when the films were steeped in 42 °C warm water and then were taken out; the response time and the recovery time fitted according to the attenuation formula are listed in the inset table.
\[ I(t) = I_0 + A \exp \left[ -(t - B)/T \right] \]  

(2)

where \( I_0 \) is the current at the initial temperature, \( A \) is the amplitude, \( B \) is a constant, and \( r \) is the recovery/response time. Here, the response time is defined as the time from the initial state \( (A) \) to the value of \( A/e \) (not to the stable value). The response time and the recovery time as the two indicators of sensing sensitivity were linked with the thickness of the film. When the thickness reduced from 2 to 0.5 mm, both the response time and the recovery time were shortened to 1/2 or less. Especially, the response time of a 0.5 mm film is extremely short, just \( \sim 0.7 \) s, which is almost negligible in respiratory monitoring.

For further understanding the sensing properties of the films with different matrices and forms, all kinds of composite films with different thicknesses were tested for respiratory sensing. As shown in Figure 5, a 0.5 mm L-POE/CF film showed the best sensitivity and excellent stability, which has a 44.31% peak value of \( \Delta R/R \) with periodic reproducibility within 5 min. In the end, a respiratory sensor with a dual mediation mechanism is simply processed.

3. CONCLUSIONS

In this study, we prepared POE/CF composite films with thermal sensing capability by constructing a micron-scale oriented network. It was demonstrated that the molecular segment structure played an essential role in adjusting the thermal expansion, which resulted in the resistance change of the composites under the temperature field. Since the polymer layer in the middle of the conductive network is a heat-resistant conductor, a thinner thickness was not only more beneficial for the sensitivity of the composite but also favorable for reproducible resistance variation. Impressively, the respiratory sensing application of an adjustable temperature-sensitive sensor has also been successfully implemented. We believed that the facile POE/CF composite films displayed wide potential prospects in thermal monitoring with slight changes, providing good prospects for micron-scale functional composites.

4. EXPERIMENTAL SECTION

4.1. Materials. POE (Engage 8450) with a density of 0.902 g/cm\(^3\) and a melt flow index of 3 g/10 min (190 °C/2.16 kg) and POE (Engage 8400), with a density of 0.870 g/cm\(^3\) and a melt flow index of 30 g/10 min (190 °C/2.16 kg) were supplied by Dow Chemical Company. Shortcut CFs (T700) were purchased by Sino Fibers Technology Developing Co., Ltd., China, with a density of 1.76 g/cm\(^3\), a diameter of 7 μm, and an electric conductivity of \( \sim 1 \) S/cm.

4.2. Sample Preparation. For the sake of convenience, we referred to 8450 and 8400 POE as H-POE and L-POE. First, the granular H-POE and L-POE were mixed by a torque rheometer at a ratio of 1:1 at 190 °C; the blend was granulated and named as B-POE. The three POE granules were then thoroughly mixed with 20 wt % CFs by a torque rheometer at 190 °C, separately. Finally, the three composites prepared were
formed into films with thicknesses of 0.5, 1, and 2 mm by compress molding at 200 °C.

4.3. Characterization. 4.3.1. Gel Permeation Chromatography (GPC) Analysis. Gel permeation chromatography (GPC) was used to measure the molecular weight distribution and polydispersity index (PDI) of the two kinds of POE. The samples were tested at 158 °C with 1,2,4-trichlorobenzene (TCB) as a solvent and polystyrene standard samples with a narrow molecular weight distribution as references. The measurements were performed on a GPC analyzer (PLgel 3 ln, U.K.) with MIXED-B LS 300 3 7.5 mm columns and a refractive index detector.

4.3.2. Scanning Electron Microscope (SEM) Observation. The frozen fracture cross sections of composite films were gold-sputtered and observed by an FEI Inspect F-SEM instrument with an acceleration voltage of 20 kV.

4.3.3. Differential Scanning Calorimetry (DSC) Analysis. The melting behaviors of the samples were investigated with a Q20 differential scanning calorimetry apparatus (TA) under nitrogen protection. The samples (~6 mg) were heated from −20 to 200 °C at a heating rate of 10 °C/min.

4.3.4. Thermal Mechanical Analyzer (TMA) Test. To observe the thermal expansion behavior of the composite, we tested the linear expansion of the samples from 20 to 40 °C with a Q400EM thermal mechanical analyzer (TA) at a heating rate of 3 °C/min.

4.3.5. Two-Dimensional Small-Angle X-Ray Scattering (2D-SAXS) Measurement. To unearth the change of the aggregate structure of the filler in the composite film with temperature, 2D-SAXS measurement operating under Xeucx 2.0 system (GeniX3D Cu ULD, Xenocs SA, France) was carried out. The wavelength of X-ray was 0.154 nm, and the distance from the sample to the detector was 2481 mm. The sample was placed on a hot stage (Linkam THMS600, Linkam Scientific Instruments Ltd., U.K.). The heating rate was 10 °C/min, and the samples were isothermal for 20 min at 20 and 40 °C. Then, the 2D-SAXS pattern was recorded at each temperature.

4.3.6. Thermal Conductivity Test. The thermal diffusivity (σ) of the composites was measured by a laser flash conductometer (NETZSCH LFA457 MicroFlash). The thickness of the specimens was 1 mm.

4.3.7. Electric Measurements. All specimens were cleaned with alcohol, and copper wires were attached at their two ends with conductive silver glue before performing the measurements to eliminate the contact resistance between the specimens and the electrodes. The conductivities of the specimens was 1 mm.

4.3.8. Respiratory Monitoring Test. The sheet-like film with a copper wire was fixed in a U-shaped nose clip with a clip cavity and invaded into the nasal cavity, and its resistance was measured by a Keithley 6487 apparatus and the signal was transmitted to the computer terminal in real time.

AUTHOR INFORMATION

Corresponding Authors
Min Nie — State Key Laboratory of Polymer Materials Engineering, Polymer Research Institute of Sichuan University, Chengdu 610065, China; orcid.org/0000-0001-8386-7547; Phone: +86-28-85405133; Email: poly.nie@gmail.com
Yijun Li — State Key Laboratory of Polymer Materials Engineering, Polymer Research Institute of Sichuan University, Chengdu 610065, China; orcid.org/0000-0002-6788-0730; Phone: +86-28-85405133; Email: ruddph@gmail.com

Authors
Jiajun Guo — State Key Laboratory of Polymer Materials Engineering, Polymer Research Institute of Sichuan University, Chengdu 610065, China
Kaolin Zhang — State Key Laboratory of Polymer Materials Engineering, Polymer Research Institute of Sichuan University, Chengdu 610065, China
Ruxian Dai — State Key Laboratory of Polymer Materials Engineering, Polymer Research Institute of Sichuan University, Chengdu 610065, China
Qi Wang — State Key Laboratory of Polymer Materials Engineering, Polymer Research Institute of Sichuan University, Chengdu 610065, China

Complete contact information is available at: https://pubs.acs.org/10.1021/acsomega.0c05367

Notes
The authors declare no competing financial interest.

ACKNOWLEDGMENTS

This work was financed by the Program of Innovative Research Team for Young Scientists of Sichuan Province (2016TD0010), the National Natural Science Foundation of China (52003174 and 51721091), and the State Key Laboratory of Polymer Materials Engineering (Grant Nos. sklpme2016-3-05 and sklpme2020-3-05).

REFERENCES

(1) Yamamoto, Y.; Yamamoto, D.; Takada, M.; Naito, H.; Arie, T.; Akita, S.; Takei, K. Efficient skin temperature sensor and stable gel-less sticky ECG sensor for a wearable flexible healthcare patch. Adv. Healthcare Mater. 2017, 6, No. 1700495.
(2) Nakata, S.; Arie, T.; Akita, S.; Takei, K. Wearable, flexible, and multifunctional healthcare device with an ISFET chemical sensor for simultaneous sweat pH and skin temperature monitoring. ACS Sens. 2017, 2, 443–448.
(3) Oh, J. H.; Hong, S. Y.; Park, H.; Jin, S. W.; Jeong, Y. R.; Oh, S. Y.; Yun, J.; Lee, H.; Kim, J. W.; Ha, J. S. Fabrication of high-sensitivity skin-attachable temperature sensors with bioinspired microstructured adhesive. ACS Appl. Mater. Interfaces 2018, 10, 7263–7270.
(4) Lu, Y.; Jiang, K.; Chen, D.; Shen, G. Wearable sweat monitoring system with integrated micro-supercapacitors. Nano Energy 2019, 58, 624–632.
(5) Gao, Y.; Yu, L.; Yeo, J. C.; Lim, C. T. Flexible hybrid sensors for health monitoring: materials and mechanisms to render wearability. Adv. Mater. 2020, 32, No. 1902133.
(6) Yang, J.; Li, Y.; Yu, X.; Sun, X.; Zhu, L.; Qin, G.; Dai, Y.; Chen, Q. Tough and conductive dual physically cross-linked hydrogels for wearable sensors. Ind. Eng. Chem. Res. 2019, 58, 17001–17009.
(7) Ge, G.; Lu, Y.; Qu, X.; Zhao, W.; Ren, Y.; Wang, W.; Wang, Q.; Huang, W.; Dong, X. Muscle-Inspired Self-Healing Hydrogels for Strain and Temperature Sensor. ACS Nano 2020, 14, 218−228.
(8) Nakata, S.; Shiomi, M.; Fujita, Y.; Arii, T.; Akita, S.; Takei, K. A wearable pH sensor with high sensitivity based on a flexible charge-coupled device. Nat. Electron. 2018, 1, 596−603.
(9) Li, Y.; Zhang, K.; Nie, M.; Wang, Q. Tubular Sensor with Multi-Axial Strain Sensibility and Heating Capability Based on Bio-Mimic Helical Networks. Ind. Eng. Chem. Res. 2019, 58, 22273−22282.
(10) Zhao, S. F.; Li, J. H.; Cao, D. X.; Zhang, G. P.; Li, J.; Li, K.; Yang, Y.; Wang, W.; Jin, Y. F.; Sun, R.; Wong, C. P. Recent Advancements in Flexible and Stretchable Electrodes for Electro-mechanical Sensors: Strategies, Materials, and Features. ACS Appl. Mater. Interfaces 2017, 9, 12147−12164.
(11) Ke, K.; Potschke, P.; Wiegand, N.; Krause, B.; Voit, B. Tuning the Network Structure in Poly(vinylidene fluoride)/Carbon Nanotube Nanocomposites Using Carbon Black: Toward Improvements of Conductivity and Piezoresistive Sensitivity. ACS Appl. Mater. Interfaces 2016, 8, 14190−14199.
(12) Liu, H.; Li, Q.; Zhang, S.; Yin, R.; Liu, X.; He, Y.; Dai, K.; Shan, C.-X.; Guo, J.; Liu, C.; Shen, C.; Wang, X.; Wang, N.; Wang, Z.; Wei, R.; Guo, Z. Electrically Conductive Polymer Composites for Flexible Sensor: A Critical Review. J. Mater. Chem. C 2018, 6, 12121−12141.
(13) Tian, M.; Lu, Y.; Qu, L.; Zhu, S.; Zhang, X.; Chen, S. A Pillow-Shaped 3D Hierarchical Piezoresistive Pressure Sensor Based on Conductive Silver Components-Coated Fabric and Random Fibers Assembly. Ind. Eng. Chem. Res. 2019, 58, 5737−5742.
(14) Yu, Y.; Peng, S.; Blanloeil, P.; Wu, S.; Wang, C. H. Wearable Temperature Sensors with Enhanced Sensitivity by Engineering Microcrack Morphology in PEDOT: PSS−PDMS Composites. ACS Appl. Mater. Interfaces 2020, 12, 36578−36588.
(15) Bang, J.; Lee, W. S.; Park, B.; Joh, H.; Woo, H. K.; Jeon, S.; Ahn, J.; Jeong, C.; Kim, T.; Oh, S. J. Highly Sensitive Temperature Sensor: Ligand-Treated Ag Nanocrystal Thin Films on PDMS with Thermal Expansion Strategy. Adv. Funct. Mater. 2019, 29, No. 1903047.
(16) Sun, T.; Luo, W.; Luo, Y.; Wang, Y.; Zou, S.; Liang, M.; Chen, Y.; Zou, H. Self-reinforced Polypropylene/Graphene composite with segregated structure to achieve balanced electrical and mechanical properties. Ind. Eng. Chem. Res. 2020, 59, 11206−11218.
(17) Zhao, H.; Cheng, Y.; Liang, X.; Du, Y.; Ji, G. Constructing large interconnect conductive networks: an effective approach for excellent electromagnetic wave absorption at gigahertz. Ind. Eng. Chem. Res. 2018, 57, 2155−2164.
(18) Deng, H.; Skipa, T.; Zhang, R.; Lellinger, D.; Bilotti, E.; Alig, I.; Peijs, T. Effect of melting and crystallization on the conductive network in conductive polymer composites. Polymer 2009, 50, 3747−3754.
(19) Canales, J.; Muñoz, M. E.; Fernández, M.; Santamaría, A. Rheology, electrical conductivity and crystallinity of a polyeurethane/graphene composite: Implications for its use as a hot-melt adhesive. Composites, Part A 2016, 84, 9−16.
(20) Pu, J. H.; Zha, X. J.; Zhao, M.; Li, S. Y.; Bao, R. Y.; Liu, Z. Y.; Xie, B. H.; Yang, M. B.; Guo, Z. H.; Yang, W. 2D end-to-end carbon nanotube conductive networks in polymer nanocomposites: a conceptual design to dramatically enhance the sensitivities of strain sensors. Nanoscale 2018, 10, 2191−2198.
(21) Haghgo, M.; Ansari, R.; Hassanzadeh-Aghdam, M. Prediction of electrical conductivity of carbon fiber-carbon nanotube-reinforced polymer hybrid composites. Composites, Part B 2019, 167, 728−735.
(22) Tseng, H.-C.; Chang, R.-Y.; Hsu, C.-H. Numerical prediction of fiber orientation and mechanical performance for short/long glass and carbon fiber-reinforced composites. Compos. Sci. Technol. 2017, 144, 51−56.
(23) Li, Y.; Nie, M.; Wang, Q. Facile Fabrication of Electrically Conductive Low-Density Polyethylene/Carbon Fiber Tubes for Novel Smart Materials via Multiaxial Orientation. ACS Appl. Mater. Interfaces 2018, 10, 1005−1016.