Breathable and Flexible Polymer Membranes with Mechanoresponsive Electric Resistance

Qiang Gao, Bernd A. F. Kopera, Jian Zhu, Xiaojian Liao, Chao Gao, Markus Retsch,* Seema Agarwal, and Andreas Greiner*

Flexible low-resistance membranes play an important role in soft electronics as sensors for robotics, body movement monitoring, nanogenerators to collect kinetic energy from body movements, and flexible batteries. Despite great efforts, low-resistance, mechanically stable large-dimensional membranes that tolerate very high deformability without sacrificing resistance produce low joule heating and allow passage of gases for human comfort are still being sought. Here, one of the solutions is provided by sandwiching a network of silver nanowires (AgNWs) between two highly porous electrospun thermoplastic polyurethane (TPU) membranes. The membranes are mechanically robust (both for bending and stretching) with a strong interface and large strain before breakage (more than 700%). The sheet resistance is as low as $<0.1\ (\pm 0.01)\ \Omega/\text{sq}$, and changed to only $1.6\ (\pm 0.43)\ \Omega/\text{sq}$ upon stretching to 100% strain. The combination of polymer elasticity and the AgNW network structure provides a reversible change in resistance beyond 100% strain. A detailed thermographic analysis is employed to in situ image and characterize the AgNW network morphology during various stretched conditions. It is believed that this flexible, sandwich-like, electrically conductive membrane is a good candidate for smart wearable devices and soft robots.

1. Introduction

Traditional electronics are typically composed of intrinsically heavy and rigid materials, like silicon, metals, and glass, that have extremely limited flexibility, stretchability, bendability, twistability, and impact resistance. There is great need for flexible and deformable circuits and electrodes with low electric resistance that maintain performance even at strained state because of the current rapid development of lightweight, wearable, and flexible electronic devices. Efforts to construct flexible circuits and electrodes with low resistance focused on elastomeric electrically conductive materials. Examples include electrically conductive sponges, hydrogels, nonwovens, films, and fibers. These materials are promising for energy production and storage, actuators, sensors, electronic skin, and soft robotics. However, obtaining flexible conductors with both high strain properties and low resistance is a challenge for design and production of flexible electronic devices. Further, porosity is an important requirement in case electronic devices are meant for contact with skin. Air permeability is very important for the comfort of wearable devices. Achieving a combination of flexibility, elasticity, high conductivity, in porous membranes with good mechanical integrity is a continued search.

Presently, typical flexible electrodes are composed of an elastic substrate as the elastomer to support bending, stretching, and twisting. The most commonly used materials...
are: polydimethylsiloxane (PDMS), thermoplastic polyurethane (TPU), electrically conductive polymers, carbon materials, metal nanoparticles, and metal nanowires. Technically, low-resistance electrodes provide the advantages of less energy loss and joule heating. Silver is an excellent material for electrodes and is often used for flexible electrodes with low resistance. In our previous work, we prepared electrospun polymer membranes with a metal-like conductivity of $7.5 \times 10^5$ S m$^{-1}$ using a very low content of silver nanowires (AgNWs: 3.35 vol%) as an additive. However, this kind of electrically conductive membrane is bendable but not sufficiently stretchable.

To prepare flexible membranes, printing fabrication techniques have been developed by transferring electrically conductive materials from a working substrate to another target substrate, such as PDMS, TPU, and polyethylene terephthalate (PET). Additionally, printing fabrication techniques provide a simple and versatile way to design and print patterns on substrate materials using computer control. In addition, microfluidic processing, microchannel wetting patterning, direct laser patterning, and photolithographic methods have also been developed to fabricate flexible conductors. Other methods, such as filling of a AgNW network prepared by spray drying of a AgNW dispersion with PDMS was also used for making stretchable, conductive electrodes. Nevertheless, the stretchable conductors prepared with these methods are all airtight due to their nonporous elastic substrate.

Alternatively, electrospinning is a promising technique for fabricating porous substrates with high conductivity, flexibility, and air permeability. The good electrospinnability of many different polymer types also provides the advantage of tuning the membrane mechanical characteristics. For example, electrospun polyamide nanofiber nonwovens embedded with AgNW networks showed 50% stretchability and a sheet resistance of 8.2 \ \Omega \ \text{sq}^{-1}. Electrospun polyurethane nonwoven porous substrate coated with AgNW provided highly stretchable membrane (more than 300%) with high conductivity.

Here, we present a flexible and breathable polymer membrane with bending/stretching stability and very low electrical resistance as a new electrode material. Also, we provide important fundamental studies regarding analysis on island formation, evolution of anisotropy and hot-spots, and thermal transport in such structures. Our study shows 1) a simple preparation method for making such membranes which is applicable to large-dimension samples. We use a network of AgNWs as conductive layer sandwiched between two porous TPU fibrous nonwovens made by electrospinning. We improved the interface stability with polycaprolactone (PCL) hot glue in the form of short fibers dispersed in between the AgNW network. 2) Our membranes show a sheet resistance as low as <0.1 \ \Omega \ \text{sq}^{-1}. The resistance does not significantly increase upon deformation (twisting, bending and stretching to approximately 100% strain). 3) We also show a reversible change in the resistance upon stretching or bending with more than 100% strain for several cycles. The AgNW networks reversibly breaks and reforms during stretching and releasing, respectively. 4) Our membranes are breathable. This allows the exchange of gases, which is important for human comfort. 5) Our membranes are thermal insulators comparable to porous polymers, despite their electrical conductivity. We think that our membranes are a promising material for electrodes in smart textiles and other wearable devices.

2. Results and Discussion

The concept for the preparation of flexible and breathable electrically conductive membranes is illustrated in Figure 1. First, we prepared TPU nanofiber nonwovens via
We then used the porous TPU membranes as a filter for the filtration of AgNWs (17 mg mL$^{-1}$, 126 ± 10 nm in diameter and 18 ± 4 µm in length, Figure S1, Supporting Information) mixed with a PCL short-fiber dispersion (1 mg mL$^{-1}$, average aspect ratio ≈1000, Figure S2, Supporting Information). The TPU nonwovens consist of randomly oriented fibers (the 2D order parameter is around 0.1) with an average diameter of 1.6 ± 0.5 µm (Figure 2a,b). After filtration, the AgNWs and short PCL fibers generated a double network-type structure due to percolation (Figure 2c,d). Afterward, we covered the AgNW/PCL layer with another layer of TPU nonwoven by electrospinning. Finally, we hot pressed the three-layer stack at 75 °C for 30 min to melt the PCL and increase the adhesion of the layers (Figure 2e). The resulting three-layer membranes are designated as TPU-AgNW/PCL-TPU. We checked the presence of an AgNW layer sandwiched between the two TPU layers by EDX (Figure 2f). The cross-sectional SEM images of TPU-AgNW/PCL-TPU and TPU-AgNW-TPU (without the use of PCL short fibers for comparison) are shown in Figure 2g,h, respectively. The PCL short-fiber dispersion acts as a glue between the AgNW network and the TPU. The importance of providing a good adhesion between the fiber support and the conducting nanowires was also investigated by Jiang et. al.[28] Omitting the PCL left us with a loose interface between the layers. Figure S3, Supporting Information, shows the morphology of the middle layer composed only of PCL short fibers after hot pressing at 75 °C for 30 min. Patches of molten fibers can be observed.

The pore size distribution (Figure S4, Supporting Information) of the sandwich membrane without AgNWs is mainly distributed at 2.8 µm with 80% porosity. Upon formation of the AgNW network layer, the average pore size was 1.5–0.9 µm. Additionally, the thickness of the sandwich membrane increased slightly with increasing volume of the AgNW solution, from 0.18 ± 0.4 to 0.27 ± 0.02 mm, as shown in Figure S5, Supporting Information.

Different amounts of the AgNW dispersion were used to investigate the influence of the AgNW concentration on the air permeability and electrical properties. The relevant parameters are illustrated in Table 1. The sheet resistance was as high as 6.1 ± 0.3 × 10^8 Ω sq$^{-1}$ without an AgNW layer. The sheet resistance decreased to as low as 0.09 Ω sq$^{-1}$ upon increasing the amount of AgNWs because of the formation of a dense network of AgNWs. Even the use of a small amount of AgNWs...
The 2D order parameter \( S_{2D} \) measures how well the fibers are aligned. The order parameter is given by

\[
S_{2D} = \frac{2}{N} \left[ \sum_{i=1}^{N} \left( \cos(\alpha_i - \bar{\alpha}) \right) \right] - 1
\]

(1)

where \( N \) is the number of stretched fiber orientations in the SEM image, \( \alpha_i \) is the angle between the fiber and the horizontal.
axis, and $\alpha$ is the average angle.\textsuperscript{[29]} The 2D order parameter of pure TPU remains constant at a value close to one after 100% strain (Figure 4d). For pure TPU the thermal diffusivity increases along the stretching direction and decreases perpendicular to it (Figure 4e,f). Since TPU is a thermal insulator the absolute thermal diffusivity remains at a low level. The evolution of anisotropy can be explained by the geometric changes, where the fibers start to align, forming an anisotropic fiber network (Figure S10). Heat travels along these oriented fibers faster than perpendicular to it, resulting in a high anisotropy ratio (Figure 4g). The geometric alignment can be accompanied by a molecular orientation of the polymer chains within the fibers, leading to an additional increase in the thermal anisotropy.\textsuperscript{[30]} The thermal diffusivity at 0% strain is higher for the AgNW-TPU sample (TPU-2.0AgNW/PCL-TPU) than for the pure TPU support structure. This can be understood by the additional thermal transport via electronic conduction through the AgNW network. The angular dispersion is rather shallow demonstrating an isotropic temperature spreading in the unstretched and disordered fiber network. Stretching of the fiber network immediately introduces anisotropy to the thermal diffusivity. For AgNW-TPU also some degree of anisotropy evolves (Figure 4h,i), but the thermal diffusivity overall decreases compared to the unstretched state (Figure S11, Supporting Information). This can be understood by a decrease in electronic thermal conductivity in the AgNW layer. The overall thermal diffusivity remains, nevertheless, higher compared to pure TPU. Simultaneously, also for AgNW-TPU samples, we find an evolution of anisotropic heat spreading (Figure 4g). Overall, stretching of the AgNW-TPU sample leads to a smaller amount of anisotropy compared to the pure TPU network structure (Figure 4g). This corroborates our previous findings that the AgNW network is torn into isolated island structures upon stretching. Thermal transport within these island-like structures happens in an isotropic fashion, which shows that the anisotropic orientation of the TPU fiber network is only partially transferred to the AgNW network and counteracted by the evolution of gaps between such islands parallel to the

Figure 3. a) Stress–strain curves of sandwich membranes with different amounts of AgNWs. b) Resistance of sandwich membranes with different amounts of AgNWs with different strains. c) Optical images of TPU-2.0AgNW/PCL under different stains. Note the appearance of white spots at 150% strain due to cracks in the AgNW layer. (scale bar: 500 µm) d) EDX-SEM image of TPU-2.0AgNW/PCL at 100% strain. (scale bar: 50 µm) Ag is indicated in purple. e) Thermography images of the TPU sandwich sample upon stretching. White lines indicate the film boundary. A constant current of 2 mA passes through the sample. Note the appearance and shift of hotspots due to local bottlenecks for the electric current created by the cracks in the AgNW layer (scale bars: 3 mm).
We corroborate this interpretation with additional orientation analysis of AgNW-TPU samples upon stretching, summarized in Figures S12 and S13, Supporting Information.

We now focus on the reversibility of the stretching/compression process and its implications for electromobility measurements. The membranes, even with very high amounts of AgNWs (TPU-2.0AgNW/PCL-TPU), showed excellent stretchability and bending stability. The samples were subjected to 100 cycles of stretching (Figure 5a) and bending (Figure 5b).

The RC of TPU-0.25AgNW/PCL-TPU with only 1.2 wt% of AgNWs significantly increased after only 60 stretching cycles, the RC was >20 000 under 50% strain. However, the same sample showed a much better bending stability (bending curvature from 0° to 150°) without a significant change in resistance. The RC of TPU-0.25AgNW/PCL-TPU after 100 bending cycles was merely 1.32. The sample TPU-2.0AgNW/PCL-TPU with a dense network of AgNWs showed both excellent bending and stretching stability. No significant change in the RC was observed for bending tests. During strain testing RC increased by only a factor of ≈20 after 100 cycles. It is important to note that our method of fabrication is particularly suitable to adjust the AgNW concentration in a simple manner. This can even be achieved locally by filtration through a mask. In Figure S14, Supporting Information, we demonstrate, that the local variation of the AgNW concentrations leads to locally confined heating powers. Hence the AgNW...
concentration, sheet resistance, and response to stretching or bending can be laterally adjusted and optimized for a given application.

We also investigated the changes to the AgNW upon repeated stretching and un-stretching with thermography. This measurement was conducted using a sample of intermediate AgNW concentration (TPU-1.0AgNW/PCL-TPU, 3.5 wt%). Upon stretching (50% strain) we measured an increase in electrical resistance, which translated into hot spot areas.

Consistent with our previous analysis such islands of AgNW network (Figure 3d) almost completely disappear upon release of the strain. In the following stretching cycles these hot-spots reform and increase in number and evolved heat. With this intermediate AgNW concentration, a disconnection between the well conducting AgNW islands happens easier and the re-formation of the intact network becomes less complete, in contrast to the better performing samples with a higher amount of AgNWs.
An LED was utilized to demonstrate the flexibility of the prepared sandwich membrane under various mechanical loads during working conditions. TPU-0.25AgNW/PCL-TPU was connected to a circuit, as shown in Figure 5e, and was bent and twisted. The LED maintained almost the same brightness. However, after 50% strain stretching, the brightness of the LED could hardly be observed. This behavior corroborates the formation of a very high RC of TPU-0.25AgNW/PCL-TPU after stretching. When using a more highly concentrated AgNW network (TPU-2.0AgNW/PCL-TPU) twisting, bending, and stretching (50% strain) are tolerated and the brightness of the LED remained almost unchanged. In addition, detailed information is shown in Video S3, Supporting Information.

The use of TPU-2.0AgNW/PCL-TPU as resistive-type strain sensor in a preliminary test for monitoring finger and wrist bending and unbending movements for several times (Figure S15, Supporting Information) worked very well. The sensitivity and precision would be further investigated and other application areas will also be explored.

3. Conclusion

In conclusion, we proposed a new strategy to fabricate sandwich-like electrically conductive membranes with very low electric resistance. Our material consists of an AgNW network sandwiched between two porous electrospun TPU nonwovens. PCL short fibers act as glue and provide a strong interface between the three layers. Our membranes show good bending and stretching stability, high stretchability (breaking elongation less than 88% and 90%, respectively), and stretching stability, high stretchability (breaking elongation less than 88% and 90%, respectively), high mechanical strength, and good wound healing behavior, as well as high electrical conductivity and gas permeability. These characteristics enable the membranes to be used in various applications, including wearable devices to collect body movement signals using a wearable sensor. The combination of good mechanical and electrical properties makes our materials suitable for use in flame retardant materials and in the field of electronic skin, where they can be used to monitor various physiological signals.

4. Experimental Section

Material: In this study, TPU (Desmopann DP 2590, Bayer Materials Science, €Mw \(145\ 000\), M<sub>l</sub> 88 900, M<sub>l</sub> 145 000), PCL (Capa 6800, €Mw 120 000, Perstorp UK Ltd.), poly(vinylpyrrolidone) (PVP K30, €Mw 40 000, Sigma-Aldrich), ethylene glycol (p.a. >95%, Fluka), silver nitrate (AgNO<sub>3</sub>, p.a. 99.9999%, Sigma-Aldrich), iron chloride (FeCl<sub>3</sub>, p.a. 98.0%, Sigma-Aldrich), sodium chloride (NaCl, p.a. 99.0%), and dimethylformamide (DMF, p.a. 99.8%) were used as obtained. Other solvents were distilled before use. The AgNWs were synthesized using a previous method.

Electrospinning: A Zeiss LEO 1530 (Jena, Germany; Schottky field-emission cathode) was used for SEM characterization of the AgNWs and their corresponding networks. EDX was performed using a Zeiss Ultra Plus (Jena, Germany; acceleration voltage of 15 kV). TEM measurements were performed using an electron bright-field transmission electron microscope (TEEM, Zeiss 922 Omega) at a voltage of 200 kV. ImageJ software was used to determine the AgNW diameter. An average of 100 AgNWs was taken for the diameter calculation.

The AgNW content was determined from thermogravimetric analysis (TGA Libra F1, Netzsch, Selb, Germany). A heating rate of 20 °C min<sup>−1</sup> was used. The measurements were performed in synthetic air.

Pore size measurements were performed with a PSM 165/H (Dresden, Germany) to determine the pore size. Topor was used as the standard test liquid (surface tension = 16.0 mN m<sup>−1</sup>). The sample holder had a diameter of 11 mm, and a flow rate of up to 70 L min<sup>−1</sup> was applied. At least three measurements were carried out for each sample, and an average was taken.

The gas permeability test was performed with a homemade unit as described in our previous work in reference 3. The membrane was fixed at the bottleneck with the cap. A rubber tube (airtight) was used to connect to another glass bottle containing solid carbon dioxide. The glass bottle fixing the nonwovens at the bottleneck was immersed in a phenolphthalein/limestone mixture. The carbon dioxide permeated through the membrane and discolored the aqueous basic solution containing phenolphthalein indicator.

Tensile tests were carried out (sample length 2 cm, width 2 mm) using a tensile tester (ZwickLine Z0.5; BT1-FR0.STN. D14; Zwick/Roell, Germany). The following parameters were used: clamping length 1 cm, crosshead speed 10 mm min<sup>−1</sup>, temperature 20 °C and pretension 0.01 N mm<sup>−1</sup>. The thickness of the samples was measured with a screw micrometer. The 100-cycle stretching test with 50% and 100% strain was also measured with the abovementioned measuring conditions.

Sheet resistance measurements (four-point measurements) were performed using a Keithley 2420 high-current source meter coupled with a Signatone SYS-301. The sheet resistance was measured ten times for each sample.

A tensile tester (ZwickLine Z0.5; BT1-FR0.STN. D14; Zwick/Roell, Germany) was employed to control the stretching distance and bending degree, and a digital multimeter (EMOS Multimeter EM391) connected to measure the sample with a bronze conductor was employed to measure the stretching resistance and bending resistance. The size of the sample and the measuring parameters used here were the same as those used in the tensile test.

Thermography imaging was performed with an Infratec VarioCAM HD research IR camera (7.5–14 μm). The spatial resolution is 29 μm at closest proximity. All measurements were performed in air.

The resistance was measured simultaneously with a Keithley 2400 Sourcemeter. The Sourcemeter acted as a constant current source during the four wire resistance measurements. A home built stretching apparatus was used to elongate the samples. The strain was determined from the length change.

Thermal diffusivity was measured with our own, self-built Lock-In thermography setup. Briefly, the films were mounted on a stretchable holder inside a vacuum chamber. An intensity modulated line laser is focused onto the back side of the sample. The intensity modulation creates temperature oscillations inside the thin film. We then detect the surface temperature with an Infratec VarioCAM HD research IR camera. The samples were coated with 25 nm of carbon on both sides to enhance laser absorption and IR emission. The IRBIS active online software from the IR Camera detects amplitude and phase of the temperature fluctuations on the sample surface. The thermal diffusivity is extracted from the material by fitting the linearized amplitude and phase signal, which spreads perpendicularly to the line laser excitation.

Optical microscopy was performed on a LEXT Olympus OLS 5000 confocal laser scanning microscope. A 100x magnification lens with a working distance of 300 μm was used. Image analysis was conducted with the Matlab code published by Persson et al.

Preparation of TPU Nonwoven and PCL Short Fibers: Electrospinning of 16 wt% TPU in DMF/THF (80/20 wt%) was carried out to obtain TPU nonwovens with randomly oriented fibers. A round 325 mesh stainless-steel sieve (70 mm in diameter) at an electrode distance of 15 cm was used as the collector. Other spinning conditions were as follows: positive voltage = 15 kV; temperature = 25 °C; relative humidity = 35%. For spinning PCL, a 15 wt% solution in a mixture of THF/DMF (70/30 wt%) was used. Other electrospinning conditions were the same as those used for TPU spinning. The dispersion (1 g L<sup>−1</sup>) of short PCL nanofibers was obtained as reported previously by us.

Preparation of Sandwich TPU-AgNW/PCL-TPU Membranes: The preparation procedure for the sandwich membranes is shown...
schematically in Figure 1. First, a TPU nonwoven prepared according to the procedure described above via electrospinning was used as a filter. Then, a dispersion of AgNWs and PCL short fibers was filtered. The AgNWs and PCL short fibers formed a conductive layer on top of the TPU nonwoven fibers. The fibers were dried under vacuum for 10 min at a pressure of 0.1 mbar. Later, another layer of TPU nanofibers was electrospun on top of the nonwoven with AgNWs and PCL short fibers to construct a flexible, sandwich-like, electrically conductive nonwoven. To induce thermal annealing, the achieved sandwich-like, electrically conductive nonwovens were pressed between two glass plates and heated at a temperature of 75 °C for 30 min to melt the PCL fibers and bond the two layers of TPU nonwovens and AgNWs together.

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

Acknowledgements
Qiang Gao would like to thank the China Scholarship Council for awarding a fellowship for carrying out Ph.D. research in Germany in the lab of Prof. Andreas Greiner. Bernd A. F. Kopera acknowledges support from the Elite Network of Bavaria. Financial support by DFG (project GR 972/46-2) and Volkswagen Foundation is acknowledged.

Conflict of Interest
The authors declare no conflict of interest.

Keywords
electrospun membranes, flexible conductors, low resistance, thermal anisotropy

Received: September 12, 2019
Revised: December 3, 2019
Published online: January 9, 2020

[1] a) E. J. Markvicka, M. D. Bartlett, X. N. Huang, C. Majidi, Nat. Mater. 2018, 17, 618; b) Y. J. Hong, H. Jeong, K. W. Cho, N. Lu, D. H. Kim, Adv. Funct. Mater. 2019, 29, 1808247; c) S. Y. Huang, Y. Liu, Y. Zhao, Z. F. Ren, C. F. Guo, Adv. Mater. Interfaces 2019, 29, 1805924.
[2] S. H. Jiang, S. Reich, B. Uch, P. Hu, S. Agarwal, A. Greiner, ACS Appl. Mater. Interfaces 2017, 9, 34286.
[3] Y. Zhou, C. J. Wan, Y. S. Yang, H. Yang, S. C. Wang, Z. D. Dai, K. J. Ji, H. Jiang, X. D. Chen, Y. Long, Adv. Funct. Mater. 2019, 29, 1806220.
[4] S. Reich, M. Burgard, M. Langner, S. Jiang, X. Wang, S. Agarwal, B. Ding, J. Yu, A. Greiner, npj Flexible Electron. 2018, 2, 5.
[5] Y. Q. Zeng, T. Li, Y. G. Yao, T. Y. Li, L. B. Hu, A. Marconnet, Adv. Funct. Mater. 2019, 29, 1901388.
[6] a) W. Weng, P. N. Chen, S. S. He, X. M. Sun, H. S. Peng, Angew. Chem., Int. Ed. 2016, 55, 6140; b) S. Lee, S. Shin, S. Lee, J. Seo, J. Lee, S. Son, H. J. Cho, H. Algadi, S. Al-Sayari, D. E. Kim, T. Lee, Adv. Funct. Mater. 2015, 25, 3114; c) J. Lee, B. L. Zambrano, J. Woo, K. Yoon, T. Lee, Adv. Mater. 2019, 1902532.
[7] R. Cao, X. J. Pu, X. Y. Du, W. Yang, J. N. Wang, H. Y. Guo, S. Y. Zhao, Z. Q. Yuan, C. Zhang, C. J. Li, Z. L. Wang, ACS Nano 2018, 12, 5190.
[8] M. Amjadi, M. Sitti, Adv. Sci. 2018, 5, 1800239.
[9] R. P. Tong, G. X. Chen, D. H. Pan, H. S. Qi, R. A. Li, J. F. Tian, F. C. Lu, M. H. He, Biomacromolecules 2020, 20, 2096.
[10] a) A. Chortos, J. Liu, Z. Bao, Nat. Mater. 2016, 15, 937; b) S. H. Wang, Y. J. Oh, J. Xu, H. Tran, Z. Bao, Acc. Chem. Res. 2018, 51, 1033; c) Y. Lee, J. Park, A. Cho, S. Cho, J. Kim, H. Ko, Adv. Funct. Mater. 2019, 1904523.
[11] C. Majidi, Adv. Mater. Technol. 2019, 4, 1800477.
[12] G. Chen, N. Matsuhashi, Z. Y. Liu, D. P. Qi, P. Q. Cai, Y. Jiang, C. J. Wan, Y. J. Cui, W. R. Leow, Z. J. Liu, S. X. Gong, K. Q. Zhang, Y. Cheng, X. D. Chen, Adv. Mater. 2018, 30, 1801012.
[13] a) T. H. Kim, C. S. Lee, S. Kim, J. Hur, S. Lee, K. W. Shin, Y. Z. Yoon, M. K. Choi, J. Yang, D. H. Kim, T. Hyeon, S. Park, S. Hwang, ACS Nano 2017, 11, 5992; b) H. S. Liu, B. C. Pan, G. S. Liou, Nanoscale 2017, 9, 2633.
[14] A. D. Valentine, T. A. Busbee, J. W. Boyle, J. R. Raney, A. Chortos, A. Kotzian, J. D. Bernring, M. F. Durstrock, J. A. Lewis, Adv. Mater. 2017, 29, 1703817.
[15] H. H. Shi, N. Khalili, T. Morrison, H. E. Naguib, ACS Appl. Mater. Interfaces 2018, 10, 19037.
[16] a) N. Karim, S. Afroj, S. R. Tan, P. He, A. Fernando, C. Carr, K. S. Novoselov, ACS Nano 2017, 11, 12266; b) C. Zhu, A. Chortos, Y. Wang, R. Pfattner, T. Lei, A. C. Hinckley, I. Pochorovski, X. Yan, J. W. F. To, J. Y. Oh, J. B. H. Tok, Z. Bao, B. Murmann, Nat. Electron. 2018, 1, 183.
[17] M. Tavakoli, M. H. Malakooti, H. Paisana, Y. Ohm, D. G. Marques, P. A. Lopes, A. P. Piedadé, A. T. de Almeida, C. Majidi, Adv. Mater. 2018, 30, 1801852.
[18] H. Lee, M. Kim, I. Kim, H. Lee, Adv. Mater. 2016, 28, 4541.
[19] P. C. Hsu, X. Liu, C. Liu, X. Xie, H. R. Lee, A. J. Welch, T. Zhao, Y. Cui, Nano Lett. 2015, 15, 365.
[20] S. J. Benight, C. Wang, J. B. H. Tok, Z. Bao, Prog. Polym. Sci. 2013, 38, 1961.
[21] K. Arapov, E. Rubิงingh, R. Abbel, J. Laven, G. de With, H. Friedrich, Adv. Funct. Mater. 2016, 26, 586.
[22] S. Kustra, H. Wu, S. Basu, G. K. Rohde, C. J. Bettinger, Small 2012, 8, 3746.
[23] B. R. Yang, W. Cao, G. S. Liu, H. J. Chen, Y. Y. Noh, T. Minari, H. C. Hsiao, C. Y. Lee, H. P. Shieh, C. Liu, ACS Appl. Mater. Interfaces 2015, 7, 21433.
[24] J. W. Yoon, W. S. Chang, S. H. Cho, Opt. Lasers Eng. 2015, 73, 40.
[25] F. Xu, Y. Zhu, Adv. Mater. 2012, 24, 5117.
[26] V. Martinez, F. Stauffer, M. O. Adagunodo, C. Forro, J. Voros, A. Larmagnac, ACS Appl. Mater. Interfaces 2015, 7, 13467.
[27] Y. J. Fan, X. Li, S. Y. Kuan, Y. Kuan, L. Zhang, Y. H. Chen, L. Liu, K. Zhang, S. W. Ma, F. Liang, T. Wu, Z. L. Wang, G. Zhu, ACS Nano 2018, 12, 9326.
[28] Z. Jiang, M. O. G. Nayee, K. Fukuda, S. Ding, H. Jin, T. Yokota, D. Inoue, D. Hashizume, T. Someya, Adv. Mater. 2019, 31, 1903446.
[29] N. E. Persson, M. A. McBride, M. A. Grover, E. Reichmanis, Chem. Mater. 2017, 29, 3.
[30] Z. Liu, R. Yang, Phys. Rev. B 2010, 81, 174122.
[31] a) J.-H. Kim, S.-R. Kim, H.-J. Kil, Y.-C. Kim, J.-W. Park, Nano Lett. 2018, 18, 4531; b) S. Chen, S. Zeng, S. Liu, H. Liu, R. Zheng, K. L. White, A. T. Smith, L. Liu, L. Sun, Chem. Mater. 2019, 31, 8708.
[32] a) A. Philipp, N. W. Pech-May, B. A. F. Kopera, A. M. Lechner, S. Rosenfeldt, M. Retisch, Anal. Chem. 2019, 91, 8476; b) H. M. Akram, M. Maqsood, H. Rashid, Rev. Sci. Instrum. 2009, 80, 075103.