EGaIn Fiber Enabled Highly Flexible Supercapacitors

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ABSTRACT: Attributed to their soft and stretchable feature, flexible supercapacitors have attracted increasing attention in areas of soft electronics, wearable devices, and energy storage systems. However, it is a challenge to manufacture all-soft supercapacitors with highly flexible properties and excellent electrochemical performance. Here, an EGaIn-based fibrous supercapacitor, which is composed of two paralleled stretchable fibers, is designed and demonstrated first with flexible and stretchable properties. EGaIn coated on the surface of polyurethane (PU)@polymethacrylate (PMA) fibers can serve as a current collector. The prepared supercapacitor is measured with an areal specific capacitance of 26.71 mF·cm⁻² by mixing Fe₃O₄ microparticles with EGaIn. This value can increase up to 61.34 mF·cm⁻² after vacuum pumping, the mechanism of which is further revealed to be related with the coarser surface and airhole formation on the fibers. The supercapacitor maintains an excellent electrochemical performance when stretched to 120% strain and exhibits a long cycling life through a charge−discharge cycle of over 1000 times. Finally, the supercapacitors are adopted to light the LED, demonstrating that those supercapacitors can work successfully. All these characteristics indicate the huge potential of EGaIn-based supercapacitors in the field of flexible electronics and wearable devices.

INTRODUCTION

The development of soft electronics and wearable devices are attracting more and more interest due to their potential applications on health monitoring and motion tracking.¹−⁵ Stretchable and flexible sensors, circuits, and soft energy storage devices are indispensable to the performance of wearable devices.⁶−⁹ The production of high-performance soft energy storage devices are considered as the challenging part in wearables.¹⁰ Flexible supercapacitors that have excellent energy storage performance can be used as soft energy storage devices. In fact, supercapacitors, which were first introduced in 1957, have been widely applied as energy storage devices.¹¹ They are distinguished for their fast charging and discharging processes, great capacitance, and ultralong life span.¹²,¹³ Higher power density and energy density are also obtained simultaneously.¹⁴,¹⁵ These excellent electrical properties make supercapacitors stand out from diverse energy storage devices. What is more, supercapacitors are considered to be a potential alternative to conventional lithium ion batteries to solve the limitations of large sizes and heavy weights.¹⁶

In order to improve the performance of flexible supercapacitors, various methods have been explored, including optimizations on the designs and materials. Generally, flexible supercapacitors include fibrous supercapacitors, planar supercapacitors, etc.¹⁷−²¹ The electrodes of fibrous supercapacitors can be placed in the form of parallel, spiral or coaxial structures. Compared to planar supercapacitors, fibrous supercapacitors show better flexibility and usually exhibit high stretchability for wearables.¹⁸ Besides, the electrode materials are influential as well. Carbon-based materials have been widely used, like carbon nanofibers and graphene.¹⁷−²² Advantages of carbon nanomaterials are that they have high specific surface area and good flexibility. However, the conductivity of carbon-based materials is inferior to metallic
materials. And the fabrication of supercapacitors should be further simplified and standardized. Finally, Metal oxide microparticles, such as MnO₂ and Fe₃O₄, which can increase effective facing area of supercapacitors and are also verified pseudo-capacitive materials, have been commonly used to improve the specific capacitance of supercapacitors.¹⁷,¹⁸

Recently, liquid metal, a new emerging material for soft electronics, is attracting increasing interest because it can provide not only high conductivity but also perfect flexibility.²³,²⁴ Studies on flexible electronics based on liquid metals include flexible conductors, antennas, transistors, and sensors, as well as soft supercapacitors and batteries.²⁵⁻³⁰ For instance, Park et al. fabricated a stretchable supercapacitor and integrated it with a strain sensor using liquid metal.³⁰ This system was proven to work well for real-time health monitoring. Furthermore, Kim et al. designed a planar supercapacitor with liquid metal electrodes.³¹ Carbon nanotubes were uniformly deposited on the surface of liquid metal electrodes to improve the capacitance. The highest areal capacitance of supercapacitors fabricated by this method was about 12.4 mF·cm⁻². At this stage, the supercapacitors could only remain stable under 30% strain. Overall, despite the advances, difficulty remains in designing supercapacitors with high capacitance, stability, and flexibility.

In this work, we attempted to design all-soft fiber supercapacitors with high flexibility and capacitance based on eutectic gallium-indium alloy (EGaIn) and metal oxide microparticles. Fibers applied in this experiment were made of polyurethane (PU). EGaIn was coated onto the surface of polyurethane fibers with the help of polymethacrylate (PMA). The obtained PU@PMA@EGaIn (PPE) fibers were then placed parallel to each other to form supercapacitors. To obtain higher capacitance, metal oxide was introduced, including MnO₂ and Fe₃O₄ microparticles. In this article, EGaIn mixed with metal oxide microparticles was named as O-EGaIn. The results showed that both MnO₂ and Fe₃O₄ microparticles can greatly improve the capacitance, especially for Fe₃O₄ microparticles. The highest specific capacitive value we obtained is about 61.34 mF·cm⁻². The supercapacitors could tolerate tensile strain of up to 120%. It is expected that liquid metal will show more advantages in the development of next-generation flexible supercapacitors.

 RESULT AND DISCUSSION

Figure 1 illustrates the fabrication and the characterization of flexible PPE supercapacitors (PPE SCs). As shown in Figure 1a, the PPE SCs were composed of four layers, including PU fibers, PMA, O-EGaIn, and PVA electrolyte. The preparation of O-EGaIn is described in Figure S1. The fabrication process of PPE SCs is illustrated in Figure 1b. First, PU fibers were coated by PMA and O-EGaIn successively. The utilization of PMA can enhance the adhesion force between O-EGaIn and fibers, which was tested later. Afterward, two PPE fibers were placed parallelly and coated by NaCl-CaCl₂ PVA gel electrolyte. NaCl-CaCl₂ PVA gel electrolyte with different doping ratios were prepared and tested. Results are presented in Figure 2a. Obviously, with the increasing proportion of NaCl, the solution became opaque gradually. When the percentage of NaCl increased up to 100%, it failed to form a gel electrolyte. Thus, to obtain good mechanical properties and higher ionic conductivity, the most suitable proportion of NaCl is around 24%. For example, the NaCl doping ratio of 24% is used in this work to prepare the PVA gel electrolyte. Through repeated experiments, we found that EGaIn would react with acidic or alkaline PVA gel electrolyte. Thus, the neutral electrolyte was chosen to prepare the supercapacitor. Considering that the solubility of CaCl₂ is relatively higher than NaCl and the existence of CaCl₂ can avoid excessive water loss, we added both NaCl and CaCl₂ together to improve the ion concentration and mechanical performance of PVA gel electrolyte. NaCl-CaCl₂ PVA gel electrolyte with different doping ratios were prepared and tested. Results are presented in Figure 2a. It is observed that the increasing proportion of NaCl, the solution became opaque gradually. When the percentage of NaCl increased up to 100%, it failed to form uniform solution with the precipitation of some flocculent precipitates. On the other hand, when the percentage of CaCl₂ is 100%, the solution is too sticky to form a gel electrolyte film. Thus, to obtain good mechanical properties and higher ionic conductivity, the most suitable proportion of NaCl is around 24%.
40–60%. In our experiment, the mixing ratio between NaCl and CaCl₂ in PVA gel adopted was 2:3.

Guo et al. found that PMA caused high adhesion with EGaIn because of the hydrogen bonding formed between PMA and EGaIn liquid metal oxide. The function of PMA was tested by an adhesion experiment, as shown in Figure 2b. PU@PMA fibers were obtained by immersing PU fibers in PMA solution. The PU fiber and PU@PMA fiber were pressed onto the EGaIn droplet and then lifted up tardily. Obviously, EGaIn was more adhesive to the PU@PMA fiber than PU fiber. EGaIn could reach a higher position with the PU@PMA fiber. Moreover, when the fibers and EGaIn were separated, some EGaIn stuck to the PU@PMA fiber. However, almost no EGaIn remained on the PU fiber. Figure 2c depicts the critical height before EGaIn was separated from the fibers. The height of PU@PMA fiber was 4.68 mm, which was 3.7 times higher than that of PU fiber. Thus, it is concluded that PMA can greatly enhance the adhesion between EGaIn and fibers.

To test the electrochemical performance of the supercapacitor made of PPE, we first investigated the cyclic voltammetry (CV) response of the supercapacitor in a two-electrode electrochemical cell. The CV curves of PPE SC, PPE-MnO₂ SC, PPE-Fe₃O₄ SC, and PPE-Fe₃O₄ SC (after vacuum freeze-drying) were all measured for comparison as shown in Figure 3a. At a scan rate of 5 mV·s⁻¹, the areal specific capacitance of the PPE-Fe₃O₄ SC was 26.71 mF·cm⁻². However, the PPE-Fe₃O₄ SC (vacuum) showed an areal specific capacitance of 61.34 mF·cm⁻², which was 2.30 times that of the PPE-Fe₃O₄ SC, 5.30 times that of the PPE-MnO₂ SC, and 13.57 times that of the PPE SC. The nearly rectangular shape is actually a characteristic of the pseudocapacitance of MnO₂ and Fe₃O₄. To evaluate why the areal specific capacitances of PPE-Fe₃O₄ SCs (vacuum) are higher than PPE-Fe₃O₄ SCs, SEM images were observed, as shown in Figure S4. A coarser surface than PPE-Fe₃O₄ SC and many airholes were found on the surface of PPE-Fe₃O₄ fiber after PPE-Fe₃O₄ SCs were vacuum pumped. To calculate the capacitance of fibrous supercapacitors, we can treat them as a parallel plate capacitor approximately. Thus, the capacitance of PPE SCs can be calculated according to the following equation:

\[ C = \frac{\varepsilon S}{d} \]

where \( C \) is the capacitance and \( \varepsilon \) is the dielectric constant of the PVA gel. \( S \) and \( d \) are the effective facing area and distance of the PPE fibers. The coarser surface and airholes of PPE-Fe₃O₄ SCs (vacuum) led to larger \( S \); therefore, the capacitance was much higher. However, the capacitance of PPE-Fe₃O₄ SCs (vacuum) after being measured repeatedly was not steady, so the PPE-Fe₃O₄ SCs had better performance.
which was calculated from CV curves, differed regularly when the scan rate changed. As shown in Figure 3c, the areal specific capacitance of the PPE-Fe₃O₄ SC reduced gradually from 26.71 to 3.24 mF·cm⁻² when the scan rate varied from 5 to 200 mV·s⁻¹, with a reduction rate of 87.9%. In addition, because the upper and lower areas of the X axis were not the same, the integration result was divided to calculate the Coulomb efficiency (CE) during the charge and discharge processes. At a scan rate of 5 mV·s⁻¹, the Coulomb efficiency of the PPE-Fe₃O₄ SC was 99.66%, which indicated that leakage current of the PPE-Fe₃O₄ SC was low.

The galvanostatic charge–discharge curves of the PPE-Fe₃O₄ SC were studied to further evaluate the charge–discharge storage capacity of the PPE-Fe₃O₄ SC, as shown in Figure 3d. Consistent with the range of voltage in the CV curves in Figure 3b, the voltage of charge–discharge curves ranged from 0 to 0.6 V, with current densities ranging from 0.22 to 0.75 mA·cm⁻². The nearly symmetric curves indicated the good capacitive characteristics of the PPE-Fe₃O₄ SC.

Figure 3e shows the Nyquist plot of the PPE-Fe₃O₄ SC. Electrochemical impedance spectroscopy (EIS) was used to characterize the performance of the PPE-Fe₃O₄ SC at a frequency from 0.01 Hz to 100 kHz. The equivalent series resistance or internal resistance (ESR) of the PPE-Fe₃O₄ SC calculated from the high-frequency intercept of the semicircle on the real axis was about 6.42 Ω, which was contributed by the resistance of electrolyte, the current collector, and also the contact resistance at the interface of the PPE-Fe₃O₄ fiber. According to pre-experiment results, mixing oxide with EGaIn showed smaller ESR than that of attaching oxide on the surface of the PPE fibers. In addition, the PPE-Fe₃O₄ fiber not only were active materials but also replaced the metal plate as current collectors.

In addition, we measured the capacitance of the PPE-Fe₃O₄ SC over multiple charging and discharging cycles, as shown in Figure 3f, because stable performance of a supercapacitor is required in actual applications. The CV response was measured to gain the change over 1000 cycles between −0.3 to 0.3 V at a scan rate of 200 mV·s⁻¹. The areal specific capacitance reduced to 92.51%, and the CV curves remained symmetric after 1000 cycles, which indicated that the PPE-Fe₃O₄ SC had a long cycling life.

Figure 4 displays the electrochemical stability of the PPE-Fe₃O₄ SC. In a practical application, supercapacitors need to adapt to various working conditions; therefore, it is necessary to measure the electrochemical stability of supercapacitors. With an increasing strain of 120%, it can be seen that 97.08% of the specific capacitance was maintained, as shown in Figure 4a. In fact, there is no significant reduction with the increase in strain up to 120%. The optical photographs and CV curves are shown in Figure 4b and Figure S5c when the PPE-Fe₃O₄ SC was stretched to 200% and 150%, respectively. In addition, the CV curves of the PPE-Fe₃O₄ SC that was stretched under 50% strain and released for 100 cycles were measured. In Figure S5d we found that the CV curves are almost unchanged. As shown in Figure 4c, the specific capacitance decreased gradually when the bending angle increased, which retained 74.53% at least at the bending angle from 0 to 180°. The above study indicated that the PPE-Fe₃O₄ SC had superior electrochemical stability.

To demonstrate the power supply ability of the PPE-Fe₃O₄ SC, a charge–discharge experiment was conducted. As Figure 4d shows, supercapacitors connected in series were charged by the electrochemical workstation to reach the operating voltage of the light-emitting diode (LED). Then, the power was cut off, and the LED was lighted by the supercapacitors. To get a better view of the LED shining, the whole process was carried out in a dark environment. Figure 4e shows the voltage curves of these supercapacitors in series. These curves correspond to voltages of one, two, and four capacitors. From Figure 4e, we can find that the voltage dividing between the four capacitors are basically equal. The total voltage of the supercapacitors was 2.23 V after being charged. When they were cut off from the electrochemical workstation and connected with the LED, the voltage decreased to 1.76 V within a short time, and the LED was lighted simultaneously. Afterward, a slow discharge process was observed. The LED was gradually dimming as time passed, as shown in Figure 4e. As expected, the PPE-Fe₃O₄ SCs exhibited a stable power supply effect.

**CONCLUSIONS**

In summary, this article puts forward a method to fabricate flexible supercapacitors based on PU@PMA@EGaIn fibers. As the circuit skeleton, PU fibers decided the construction of supercapacitors. EGaIn, as the electrode material, was outstanding for its excellent conductivity and fluidity. The
The introduction of \( \text{MnO}_2 \) and \( \text{Fe}_3\text{O}_4 \) was to increase the surface area and store electricity by the effect of pseudo-capacitance, thus improving specific capacitance. Moreover, the gel electrolyte used in our experiment was \( \text{NaCl-CaCl}_2 \) PVA gel electrolyte. Its mechanical properties and ionic conductivity could be regulated through the ratio between NaCl and CaCl\(_2\), and the mixing ratio adopted was 2:3. The obtained PPE SCs were characterized and proven to have stable electrochemical and mechanical properties. At a scan rate of 5 mV s\(^{-1}\), the areal specific capacitances of the PPE-Fe\(_3\text{O}_4\) SC and the PPE-Fe\(_2\text{O}_3\) SC (vacuum) were 26.71 and 61.34 mF cm\(^{-2}\). The latter showed a larger specific capacitance through forming a coarser surface and airholes, but the capacitance was not steady. The former showed the best performance with stable specific capacitance and a strain of 120%. Finally, the PPE-Fe\(_2\text{O}_3\) SCs were adopted to light the LED, demonstrating that those supercapacitors could work successfully. Advantages of the PPE SCs include a simple fabrication process, excellent flexibility, high areal specific capacitance, and a stable working performance. Future research may focus on the further improvement of the specific capacitance of supercapacitors based on liquid metal. Generally speaking, liquid metal is a promising candidate for flexible supercapacitors, which deserves more attention and effort in the near future.

**EXPERIMENTAL SECTION**

**Preparation of EGaIn.** The EGaIn used in this experiment was composed of 75.5% gallium and 24.5% indium by weight. Gallium and indium were heated and stirred at 200 °C for 2 h. After cooling down to room temperature, the alloy remained in the liquid state and EGaIn was formed.

**Preparation of O-EGaIn.** EGaIn and metal oxide microparticles were placed into a beaker. They were stirred until EGaIn and the microparticles were intensively mixing. Finally, EGaIn in its liquid form turned into O-EGaIn in the paste form.

**Preparation of NaCl-CaCl\(_2\) PVA Gel Electrolyte.** First, 5 g of PVA powder was added to 35 mL of deionized water at 95 °C and was stirred until PVA was sufficiently dissolved. Then, 2 g of NaCl and 3 g of CaCl\(_2\) were dissolved in deionized water separately and then added to the PVA solution. Finally, the mixture was stirred for 2 h to obtain NaCl-CaCl\(_2\) PVA gel electrolyte.

**Fabrication of PU@PMA@EGaIn Fiber Electrodes.** To improve the adhesion between EGaIn and PU fibers, PMA was smeared onto 20D PU fibers first. PU fibers were immersed in PMA solution for 20 min and then dried at 80 °C. Then, the PU@PMA fibers were soaked into an O-EGaIn bath pool, and EGaIn would uniformly adhere to the surface of the fibers to obtain PU@PMA@EGaIn fibers.

**Supercapacitor Assembly.** Two PU@PMA@EGaIn fibers were positioned parallel to each other and coated by NaCl-CaCl\(_2\) PVA gel electrolyte. The whole system was then dried to vaporize excess water. At this point, the fabrication of soft PU@PMA@EGaIn supercapacitors was completed.

**Characterization.** SEM and EDS images were obtained using a scanning electron microscope and an energy dispersive spectrometer. To evaluate the electrochemical properties of PPE supercapacitors, we adopted a two-electrode system and an electrochemical workstation (CHI 660E, CH Instrument). Testing items included cyclic voltammetry, chronopotentiometry, and A.C. impedance. The voltage curve during the process of lighting the LED was recorded using a signal oscilloscope.

**Calculation of the Electrochemical Performance.** The specific capacitance was calculated from CV curves according to the following equation:

\[
C_{sp} = \frac{1}{2SV} \int_{V_{-}}^{V_{+}} i(V) dV
\]

where \( V_+ \) is the maximum voltage of the voltage window, \( V_- \) is the minimum voltage, \( (V_+ - V_-) \) is the width of the voltage window, \( \nu \) is scan rate, and \( S \) is the surface area.

**ASSOCIATED CONTENT**

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.1c02834.

Preparation of O-EGaIn, SEM images of different fibers, and CV curves of PPE SC and PPE-MnO\(_2\) SC (PDF)

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**Author Contributions**

M.D. and Y.R. contributed equally to this work. M.D. contributed to conceptualization, methodology, investigation, data curation, and writing of the original draft of the study. Y.R. also contributed to the conceptualization, methodology,
investigation, and writing of the original draft. X.S. contributed in designing the methodology, visualization, and funding acquisition. X.Z. participated in designing the methodology. X.W. acquired resources. L.S. contributed in funding acquisition. J.L. participated in review & editing the paper.

Notes
The authors declare no competing financial interest.

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