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Environment-friendly Liquid Metal Coating Material for Patterning Soft and Stretchable Electronics at Low Cost

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***Abstract*— Wearable flexible electronics can enrich and facilitate people's daily life. However, although exquisite design and complex processes enrich the functions of wearable electronics, high costs also hinder the commercialization of wearable flexible electronics and their applications in everyday life. Herein, we report a low-cost and rapid fabrication approach for wearable flexible electronics. In this approach, liquid metal coating material composed of liquid metal particles and polyvinyl alcohol solution can be mask coated on some substrate materials commonly used in daily life to fabricate desired soft and stretchable patterns for wearable electronics. Further, we characterized the electrical performance of liquid metal coating material for strain sensors, and fabricated soft LED arrays and finger joint motion sensors on disposable nitrile gloves commonly used in the laboratory to demonstrate that this approach is fully capable of manufacturing low-cost disposable wearable electronics. This approach sheds new light on wearable electronics in high demand for short-lived everyday use.**

I. Introduction

Thanks to its own compliance and stretchability, the application scenarios of flexible electronics can cover almost every aspect of our daily life, from wearable health monitoring, and energy harvesting research to soft robotics, which enriches our vision of a more comfortable life in the future.[1-5] The vigorous development of flexible electronics application field has promoted the research on their design and manufacturing methods to shine in recent years. Whether it is the structural design represented by serpentine lines or the chemical modification of conductive polymers, it has greatly promoted functions of flexible electronics towards diversification and integration.[6-8] Although these state-of-the-art design ideas and ingenious manufacturing processes have presented us with amazing flexible electronic devices, which greatly broadened our understanding of the future trend of flexible electronics, these expensive flexible electronic devices also make it difficult to go into our practical daily life. In other words, low-cost, efficient, and reliable flexible electronics are very meaningful for applications in daily life.

In recent years, liquid metal (LM) has aroused great attention of researchers because of its excellent conductivity and compliance.[9, 10] What is more attractive is that the commercially available eutectic gallium-indium alloy (EGaIn, a type of liquid metal) costs less than US$0.8 per gram, which provides the possibility of its commercial application in daily life. Liquid metal, behaving like its name, exhibits the flow characteristics of a liquid at room temperature (25℃), which is a very nice match for the requirements of sensors and circuit connections in flexible electronics. Moreover, the negligible toxicity of liquid metal allows them to be fabricated into desired conductive patterns like common metals without additional operating environments.[11, 12] Numerous methods have been creatively proposed to pattern liquid metal into desired circuits to achieve specific functions,[13, 14] such as solid additives to reduce liquid metal fluidity,[15, 16] utilizing spontaneously formed oxide films on the surface of liquid metal,[17, 18] selective wetting,[19, 20] etc. Among them, converting bulk liquid metal into slurry composed of liquid metal particles and then patterning this special "ink" is a very generally suitable method, which overcomes the hindrance of the huge surface tension and poor wettability of bulk liquid metal to its patterning process.[21-24] More meaningfully, such liquid metal slurry can be utilized as inks for some existing mature low-cost patterning processes, such as screen printing and mask coating,[25, 26] for mass fabrication of flexible conductive patterns.

In this study, we report a liquid metal coating material (LMCM) for the fabrication of low-cost wearable flexible electronics. This liquid metal coating material is composed of two parts, wherein the slurry composed of liquid metal micro-particles is used as conductive components, and polyvinyl alcohol (PVA) solution is used as a binder to ensure the flat shape of the final liquid metal patterns. Subsequently, we characterized the surface morphology and electrical performance of circuits prepared by mask coating LMCM, and verified its feasibility for flexible strain sensors. Finally, we utilized LMCM to fabricate finger LED arrays and finger joint motion sensors on the disposable nitrile gloves commonly used in the laboratory to meet the application needs of low-cost wearable electronics in daily life.

II. Materials Preparation

Patterning bulk liquid metal into ideal patterns has been challenging due to the adverse effect of huge surface tension.[15, 27] To address this issue, we prepared micron-sized liquid metal particles by sonicating 8g bulk liquid metal (EGaIn, composed of 75% Ga and 25% In in mass proportions) in 10 g absolute ethanol (Fig.1A) for 5 minutes, and the slurry composed of micron-sized liquid metal particles does not appear to spontaneously aggregate into spherical shape like bulk liquid metal on a macroscopic

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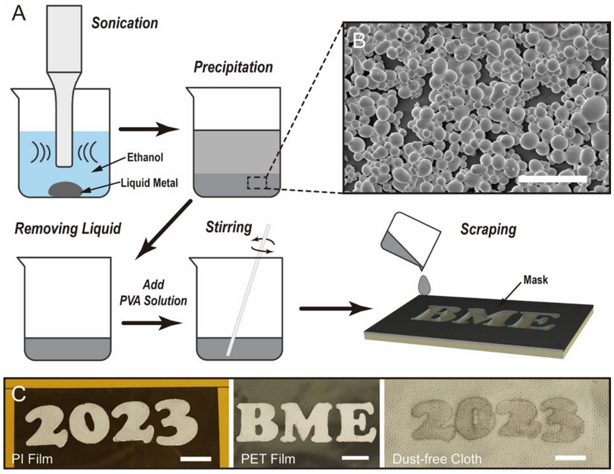

Figure 1. Preparation of liquid metal coating material (LMCM). (A) Schematic diagram of the preparation procedure of LMCM and patterning process. (B) Scanning electron microscope (SEM) image of liquid metal particles after sonication. The scale bar is 50 μm. (C) Patterns prepared by depositing LMCM on different substrate materials. The scale bars are 10mm.

scale. The scanning electron microscope (SEM) image of micron-sized liquid metal particles prepared by sonication is shown in Fig. 1B. The prepared dispersion of liquid metal particles in ethanol was placed to stand for stratification, the light liquid in the upper layer was discarded, and the bottom slurry was kept. Then we added 1 mL polyvinyl alcohol solution (10% w/v) into slurry composed of liquid metal particles, and stirred the mixture until a homogeneous liquid metal coating material was obtained. Next, we attached masks to different substrate materials, and then prepared desired soft conductive patterns by scraping LMCM on them. The liquid metal patterns obtained by depositing LMCM on different substrate materials (PI film, PET film, and dust-free cloth) are shown in Fig. 1C.

In the LMCM system, liquid metal particles are conductive components, and PVA acts as a binder to enhance the adhesion between liquid metal particles. To better highlight the effect of PVA solution on LMCM system, we coated the liquid metal slurry (without PVA component) and LMCM on the SEBS substrate material, and after all the solvents were completely volatilized, the traces prepared by the two materials are shown in Fig. 2A, and SEM images of their surface morphology are shown in Fig. 2B&C. In contrast to the trace prepared with liquid metal slurry (without PVA component), which shows obvious cracks on its surface, the trace prepared with LMCM exhibits a flat and uniform surface morphology. Even the amount of PVA added into liquid metal slurry is very tiny, the adhesion effect is very evident. The experimental results show that a small amount of adhesive is very important to form a uniform and flat liquid metal pattern through micronizing the liquid metal method.

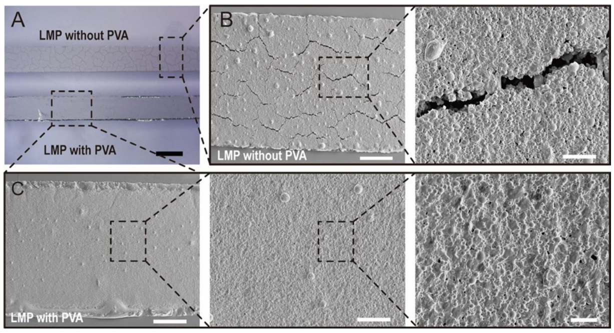

Figure 2. Adhesive effect of PVA on liquid metal particles. (A) Optical microscope images of liquid metal particles (LMP) with and without PVA solution after drying. The scale bar is 2 mm. (B)SEM images of trace by depositing liquid metal particles without PVA solution. The scale bars are 500 μm and 50 μm respectively. (C) SEM images of the trace by depositing liquid metal particles with PVA solution. The scale bars are 500 μm, 100 μm and 20 μm respectively.

Notably, although the method of converting bulk liquid metal into micro-particles can facilitate the patterning process of liquid metal, the liquid metal pattern obtained after solvent evaporation is generally insulating, because electrical connections among liquid metal particles are very weak, where liquid metal particles are isolated from each other by some adhesive or their own oxide films,[21, 23, 28] therefore, some necessary mechanical activation operations, also called sintering operations, are necessary to restore the conductivity of the pattern, such operations as stretching, pressing, scraping, etc.[24, 27, 29]

III. ELECTRICAL PERFORMANCE

We deposited LMCM on the SEBS substrate to create a liquid metal pattern as shown in the inset of Fig. 3A. After liquid metal coating material was completely dried, the conductivity of the circuit was activated by stretching, and SEBS was also used as the encapsulation layer. First, we tested the resistance change of the LMCM-based conductive path under different tensile strains, and the results showed that its resistance responsiveness to strain is acceptable when used as strain sensors. Subsequently, we kept the LMCM-based conductive path under five different tensile strains for 20 s (Fig. 3B), that is, to test the data retention ability of the LMCM-based strain sensor. Next, we tested the resistance changes of this soft strain sensor in a reverse compression

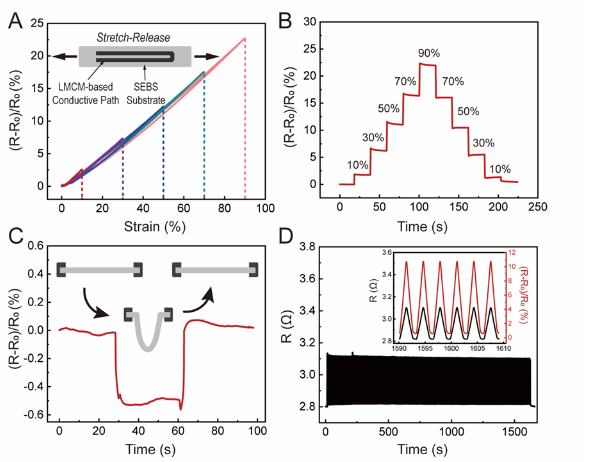

Figure 3. Electrical performance of LMCM circuits. (A) Normalized resistance changes of LMCM-based strain sensor under different tensile deformations. (B) Data retention performance of LMCM-based strain sensor under different tensile deformations. (C) Normalized resistance changes of LMCM-based circuit during a compression-recovery cycle. (D)Resistance changes of the LMCM-based strain sensor during 500 stretching-releasing cycles, where the maximum stretching strain is 50%.

cycle (holding for 30 s), as shown in Fig. 3C, to explore the reverse strain sensing ability of this LMCM-based strain sensor. Finally, to test the stability of the LMCM-based strain sensor during long-term use, we carried out 500 cyclic stretching-releasing experiments with a fixed strain of 50%, and the strain response ability of the strain sensor did not show obvious attenuation during the whole process (Fig. 3D), showing its excellent stability.

IV. Wearable Applications

Wearable electronics, especially for health monitoring and motion tracking, is a very attractive research field for the application of flexible electronics.[30-32] However, the high cost and complicated fabrication process of some wearable electronics obviously limit their further commercial application and daily use. For example, in daily life, some applications may only require wearable flexible devices to complete the sensing function for a certain period of time, and then they will be discarded. In such occasions where there is a demand for disposable wearable devices, although expensive wearable devices that have been precisely designed and manufactured have a huge advantage in accuracy, the high cost is undoubtedly not cost-effective. Therefore, low-cost, fast fabrication of wearable electronics is desired in many simple daily applications.

To verify the application potential of LMCM in low-cost and rapid fabrication of wearable electronics, we attached a circuit mask to a disposable nitrile glove commonly used in the laboratory, and scrape-coated LMCM on it. After the paths are completely dried, a simple stretching operation can restore the conductivity of paths. Then 12 LEDs are mounted on the paths to realize the fabrication of flexible finger LED arrays. Admittedly, the cost of this device is very low, and the manufacturing process is very simple, but it is completely competent for the display of finger gestures in the dark (Fig. 4A).

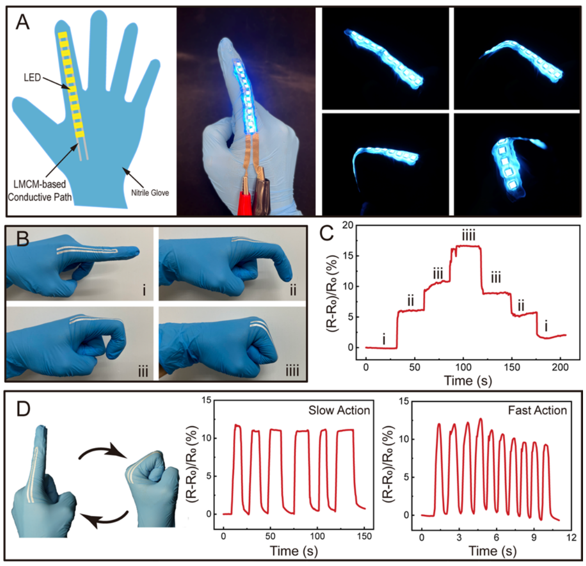

Figure 4. LMCM-based wearable applications. (A) LED arrays on nitrile glove based on LMCM conductive paths. (B) Four different strain states of the LMCM-based strain sensor coating on the index finger of a nitrile glove. (C) Normalized resistance changes of the strain sensor coating on the nitrile glove when the index finger is in four different postures. (D) Monitoring performance of finger joint sensor at different motion speeds.

Another representative wearable application is the finger joint sensor, as shown in Fig. 4B. Similar to the fabrication of finger LED arrays, we also used LMCM to fabricate the sensing circuit tightly attached to the nitrile glove with the help of a mask. Although the manufacturing process is simple, the LMCM-based finger joint sensor can well represent the finger posture state in the form of resistance changes when the index finger is in different postures (Fig. 4C), which is perfectly acceptable for simple, one-time use in everyday life. Furthermore, we tested the ability of the finger joint sensor to capture the motion of the index finger, as shown in Fig. 4D, when the index finger repeated the straightening-bending motion at two different speeds, fast and slow, this low-cost strain sensor coating on disposable nitrile glove can track finger motions well. This material system and manufacturing method provide a good idea for low-cost wearable electronics used in daily life.

V. Conclusion

In summary, we reported a simple and environment-friendly liquid metal coating material (LMCM) for fabricating flexible circuits on various substrate materials. The preparation process of LMCM does not involve organic solvents that are toxic to the human body and harmful to the environment. LMCM is simple to use and is very suitable for fabricating flexible circuits or strain sensors on some commonly used substrate materials, which provides a new idea for the fabrication of disposable wearable flexible electronic devices.

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