An Improved Liquid Metal Mask Printing enabled Fast Fabrication of Wearable Electronics on Fabrics *

Rui Guo, Siyuan Yao, Xuyang Sun and Jing Liu

Abstract— **In this paper, a new improved mask printing method of liquid metal is developed, which realizes the fast fabrication of flexible electronics on fabrics. Here, polymethacrylates (PMA) glue is printed on fabrics to improve the adhesion effect of liquid metal (EGaIn) on fabrics. Combined with mask printing, liquid metal can be directly and rapidly printed on the fabrics with PMA glue to manufacture flexible electronics, such as LED array circuit, strain sensor and temperature monitoring circuit. With combined the advantages of favorable stretchability and rapid manufacture, the improved liquid metal mask printing method provides an approach with valuable prospects for individualized wearable health care devices. Besides, this method has extensive application prospect in mass production of smart electronic fabrics.**

I. Introduction

With the advantages of low cost and widely available, fabrics represent an attractive direction in wearable electronics. At present, various manufacturing methods of fabrics electronics showing high stretchability have been proposed. For example, some researches embed electrically connected fibres in fabrics [1]. Another strategy is to encapsulate the conductive structures in flexible material and then attached to the fabrics surface [2]. These conductive structures are generally made up of inorganic and organic conductive materials [3-4].

Recently, eutectic gallium–indium (EGaIn) have been widely applied in flexible electronics and soft robots. With advantages of excellent fluidity [5], high electric conductivity (EGaIn: 3.4×10^6 S m−1) [6], low toxicity [7], soft and superior stretchability [8], at present, liquid metal is convenient to fabricate flexible electronics, such as flexible pressure sensors [9], stretchable wireless energy transducer [10], and electromagnetic actuator [11]. Liquid metal based printing approaches have been proposed such as microchannel injection [12], mask printing [13-14] and transfer printing [15]. Particularly, the liquid metal used in the previous studies about liquid metal mask printing methods needs special treatments, such as air pressure ruptured and mixed metal particles, which increase preparation cost and add complexity.

So far, there are only several suitable substrates to offer high continuity, robustness and complexity liquid metal circuits use existing mask printing approaches, for example, PVC, PDMS, glass and paper. Beyond these, another promising future of liquid metal conductive ink will be the manufacture of wearable electronics on fabrics. On the other hand, the relatively recently developed fabrics electronics encapsulated with conductive fillers, which have high-performance in both conductivity and flexibility, are not a convenient or economically viable solution for the much larger market requirements of flexible wearable devices. Therefore, all these factors come together to compose the basis of the brand-new technology proposed and further tested in this article. Here, we developed an improved liquid metal mask printing method for fast fabrication of wearable electronics on fabrics. Different from traditional mask printing method, we pioneered an adhesion assisted interface processing method using PMA glue to mask print liquid metal on fabrics directly. Relevant testing results showed that the liquid metal conductors on fabrics could maintain stable electrical properties and had excellent stretchability, which were used as tension sensor in this paper. Furthermore, the LED array circuits and temperature monitoring circuit were also designed to demonstrate its applications in wearable electronics and health monitoring. Hence, we believed that this method paved a new way to rapidly fabricate wearable electronics on fabrics directly based on liquid metal.

II. Materials and Methods

A. The improved liquid metal mask printing

Figure 1A demonstrated the schematic illustration of the liquid metal electronics on fabrics. The liquid metal utilized here was gallium-indium eutectic alloys (EGaIn) with the melting point 15 °C and composed of 75.5 wt% gallium and 24.5 wt% indium (Anhui Minor New Materials Co. LTD, 99.999% purity). The main process of the improved liquid metal mask printing was shown in Figure 1B. The PMA glue was firstly uniformly printed on the surface of Polyurethane (PU) fabrics, which is a kind of elastic woven material. Then, the PMA glue would permeate into the pores of PU fabrics, cover its surface completely, and further curing at room temperature after 10 minutes. Afterwards, liquid metal was printed with a rolling brush to the surface of a stainless steel mask, which was tightly attached to the PU fabrics substrate. After removing the stainless steel mask, the liquid metal was uniformly printed on the surface of PU fabrics covered with PMA glue. Then, the microchip (MSP430G2553) and LEDs were placed on the specific site of liquid metal layer. Cu wires were connected to the pin of liquid metal wires, facilitating the stability of the connection between rigid components and liquid metal wires. Finally, a film of Ecoflex (Ecoflex 00-30, Smooth-On, PA, USA, mixed at 1:1) membrane was covered on the surface of the liquid metal layer, and curing at room temperature for another 15 minutes.

*Research supported by NSFC Key Project under Grant No. 91748206.

R. Guo is with Tsinghua University, Beijing, China.

S. Yao and Xuyang Sun are with the Technical Institute of Physics and Chemistry, Chinese Academy of Sciences, Beijing 100190, China.

J. Liu is with the Department of Biomedical Engineering, School of Medicine, Tsinghua University, Beijing 100084, China; He is also with the Technical Institute of Physics and Chemistry, Chinese Academy of Sciences, Beijing 100190, China (Tel. +86-10-62794896; Fax: +86-10-82543767; E-mail: jliubme@tsinghua.edu.cn).

B. *Performance testing of the liquid metal on fabrics*

In this paper, the photographs (Figure 2A, B, C, D) of SEM (scanning electron microscopes) were taken with SEM S-4300 to characterize microscopic feature of PU fabrics substrate, covered with PMA glue and liquid metal wires. In order to identify the adhesion effect of liquid metal on PU fabrics and PMA glue, the contact angle meter (XG-CAMC) was utilized to measure the contact angle of the liquid metal droplets (diameter of 4mm) on the PU fabrics and PMA glue respectively, as shown in Figure 2E. The resistances of liquid metal wires on PU fabrics were measured with Agilent 34420 (USA). The strain-resistance curve was measured with a universal testing machine (Shimadzu, Model AGS-X). The length of the sample was 36mm, and cross section of 15mm×1mm.

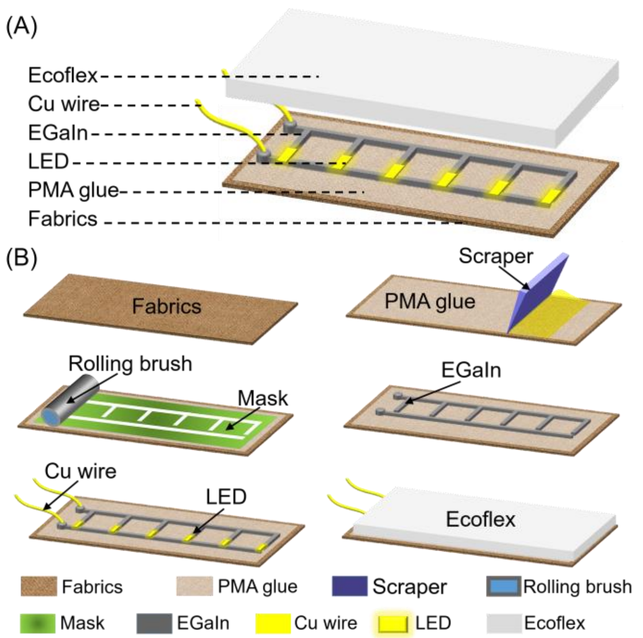

Figure 1. (A) Exploded view of the LED array circuit printed on PU fabrics, consisting of fabrics layer, PMA glue layer, liquid metal layer and Ecoflex layer. (B) The fabrication method of improved liquid metal mask printing.

C. *Design of LED array circuit and temperature monitoring circuit*

In order to exhibit the application prospect of liquid metal enabled flexible electronics on fabrics as wearable health care devices and smart electronic fabrics, we designed LED array circuit and temperature monitoring circuit. Here, the LED array circuit was composed of 7 LEDs and a microchip (MSP430G2553), connected on a miniature circuit board (length: 19 mm, width: 16 mm, height: 2 mm) with other necessary components. A button battery with a DC voltage of 3 V for the microchip and LEDs, are shown in Figure 4A. Besides, using the same LEDs and microchip, we printed a large area LED array circuit on fabrics directly, as shown in Figure 4C. The temperature monitoring circuit consisted of three parts: temperature sensor, microchip and Bluetooth chip. The photograph of temperature monitoring circuit was shown in Figure. 4D. LM35, chosen as the component for the detection of temperature, connected at a miniature circuit board (length: 16 mm, width: 10 mm, height: 7 mm) with some other necessary components. The microchip (MSP430G2553) sent it to the Bluetooth chip. The Bluetooth chip (length: 26 mm, width: 13 mm, height: 2 mm), which transmitted the temperature signal to a computer.

III. RESULTS AND DISCUSSION

A. *Adhesion tests of liquid metal on PU fabrics and PMA glue*

Different from previous mask printing methods of liquid metal, this novel improved mask printing method of liquid metal demonstrated in this paper aimed to enhance the adhesive force of liquid metal on fabrics. The previous research about the adhesion of liquid metal on multi materials and chemical surfaces, such as paper, PDMS, glass and metal [15] showed that the surface of liquid metal was covered with an oxide skin layer in air and the adhesion effect between the two surfaces generally depends on the chemical interaction as well as the morphology of their microstructures.

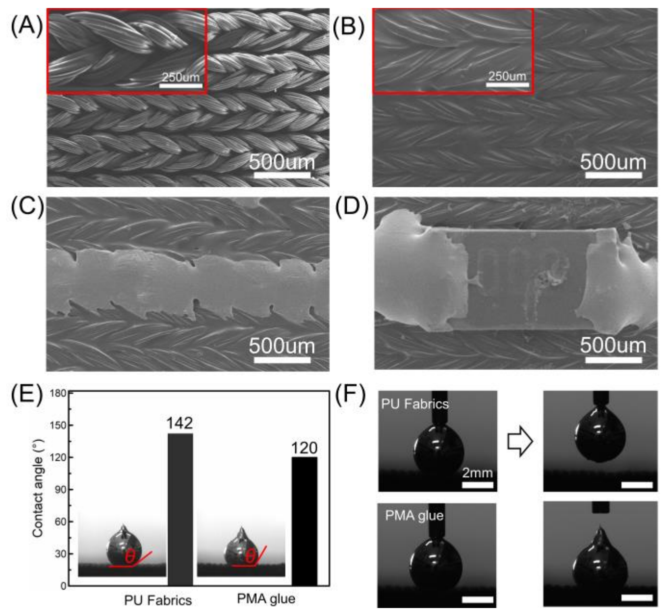

Figure 2. Adhesion testing results of liquid metal on PU fabrics and PMA glue (A) The SEM photograph of the PU fabrics. (B) The SEM photograph of the PU fabrics covered with PMA glue. (C) The SEM photograph of the liquid metal printed on PU fabrics covered with PMA glue. (D) The SEM photograph of a small chip resistor attached to the liquid metal conductor. (E) The contact angle of the liquid metal on PU fabrics and PMA glue. (F) The lifting experiment of liquid metal droplets on PU fabrics and PMA glue.

In order to reveal the significant adhesion difference between liquid metal on PU fabrics and PMA glue, the morphological variation of microstructures among PU fabrics, PU fabrics covered with PMA glue and printed liquid metal were characterized by the SEM photographs in Figure 2A, B, C. As can be seen from Figure 2A, the PU fabrics was knitted from fibers (diameter of 30 um), which resulted in the very rough and uneven surface shape of the PU fabrics. It was obvious that the rough surface of the PU fabrics reduced the contact area between liquid metal and PU fabrics, and the gallium oxide layer hindered the liquid metal from permeating into the gap between PU fibers. In contrast to the rough surface morphology of the PU fabrics, the printed PMA glue could fill in the gap between PU fibers and covered the PU fabrics surface, as shown in Figure 2B. Therefore, the smooth surface of PMA glue increased the contact area with liquid metal. Hence, the liquid metal lines could be adhered on the PU fabrics with PMA glue easily, as shown in Figure 2C. More importantly, the SEM imaging in Figure 2D showed that the PMA glue could adhere the components to the PU fabrics surface closely, and the liquid metal could wet the pins of the component. In order to further investigate the adhesion difference between liquid metal on PMA glue and PU fabrics, the contact angles of liquid metal on PU fabrics and PMA glue were measured, as shown in Figure 2E. The lower contact angle means better wettability and spread on substrate more easily. The results demonstrated that the contact angle of liquid metal on PMA glue (120°) was substantially below that on PU fabrics (142°), which suggested that the liquid metal had better wettability on PMA glue than PU fabrics.

Given the previous researches about the adhesion effect of EGaIn on various substrate materials, it was noteworthy that the molecular interaction had a significant impact on the adhesion of liquid metal. The previous research showed that there was hydrogen bond interaction between liquid metal and PMA glue, [15] which resulted in the good wettability of liquid metal on PMA glue (Figure 2E) and helped the liquid metal adhere to the substrates. Besides, a lifting experiment was added to show the significant adhesion difference of liquid metal on fabrics and PMA glue. As could be seen in Figure 2F, the liquid metal droplet on fabric could be lift up, while not that on PMA glue.

B. Electrical performance tests of liquid metal wearable electronics on fabrics and the biomedical applications

In this paper, the PU fabrics, PMA glue and Ecoflex membrane are all stretchable materials. Therefore, we designed a tension sensor using the improved liquid metal mask printing method, as could be seen from the pictures in Figure 3A. The tension sensor was made up of a serpentine liquid metal conductor (line width of 1mm, line length of 66cm), which could be stretched following the PU fabrics. The curve in Figure 3A showed that the liquid metal conductor could keep the circuit connected during the tensile process. Besides, it must also be mentioned that the resistance of liquid metal conductor significantly increased when stretching the sensor from 0% to 100% (4.6 Ω at original length and 14.4 Ω at 100% strain). A mechanical test (sample length of 36mm, cross section of 15mm×1mm) was added and the stress-strain curve was showed in Figure 3B. To further confirm its electrical stability, we stretched the tension sensor over 2000 times at original length, 20% strain, 50% strain and 100% strain, respectively. These curves in Figure 3C showed that there was no obvious change on the resistance of the tension sensor after 2000 repetitive stretching cycles at original length, 20% strain and 50% strain. It should be pointed out that the tension sensor had an obvious resistance variation at 100% strain. The minimum resistance of the tension sensor was 14.1 Ω after 800 repetitive stretching cycles, and the maximum resistance was 14.8 Ω after 1400 repetitive stretching cycles. The maximum resistance variation ratio about 4.8% was negligible compared to the resistance of other functional components, such as LED lights (above 3 kΩ). In addition, the V–I (voltage–current) curves of the printed liquid metal conductors with LED light were tested to further demonstrate its good electrical stability at different tensile states, as shown in Figure 3D. It was obvious that the resistance variation of the liquid metal conductors had unapparent effects to the LED light, and would not affect the functionality of the electronic components.

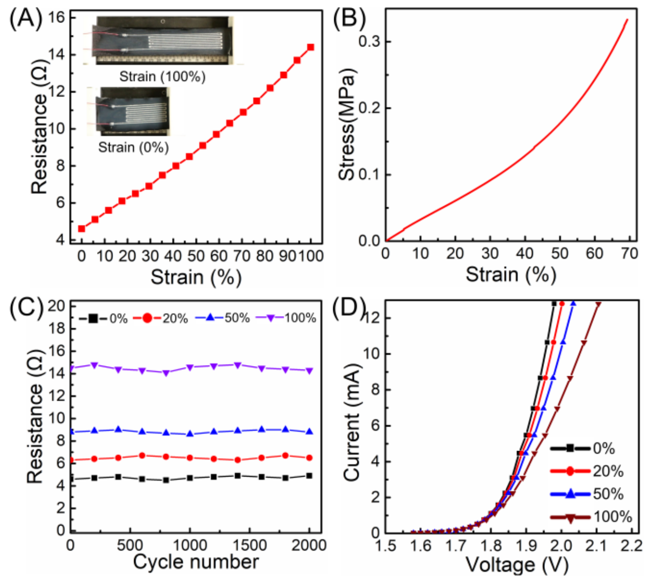

Figure 3. Electrical performance tests of liquid metal wearable electronics on fabrics. (A) The resistance variation of the liquid metal tension sensor under varied strain states. (B) The stress-strain curve of liquid metal tension sensor. (C) The resistance variation of the liquid metal tension sensor stretched over 2000 times at original length, 20% strain, 50% strain and 100% strain, respectively. (D) The V–I (voltage–current) curves of the printed liquid metal conductors with a LED light at original length, 20%, 50% and 100% strain, respectively.

In this paper, the liquid metal printed on the PU fabrics with PMA glue was utilized to fabricate LED array, as shown in Figure 4A. Besides, the photo in the Figure 4B showing that the wearable electronics on fabrics were waterproof. Here, liquid metal conductors were width of 1mm, and all the LED lights were kept in normal working condition even at 40% strain state and bent state, as shown in Figure 4B. More importantly, this improved liquid metal mask printing method offered exciting opportunities to produce large area wearable electronics. Another liquid metal LED array circuit (Liquid metal conductors were width of 1.5mm) printed on a T-shirt was showed in Figure 4C. As could be seen from these pictures, the large area LED array circuit was also kept in proper working condition, when it was worn on body or pulled. Finally, to demonstrate the biomedical applications of liquid metal wearable electronics, a temperature monitoring circuit (Liquid metal conductors were width of 1mm) printed on PU fabrics was tested, as shown in Figure 4D. The curve in Figure 4E showed that the temperature increased when the hand was put on the sensor, and the temperature decreased when the hand was removed from the sensor. These results suggested that this improved liquid metal mask printing method could be applied in individual healthcare monitoring, which could provide low cost, great convenience and point of care as wearable electronics.

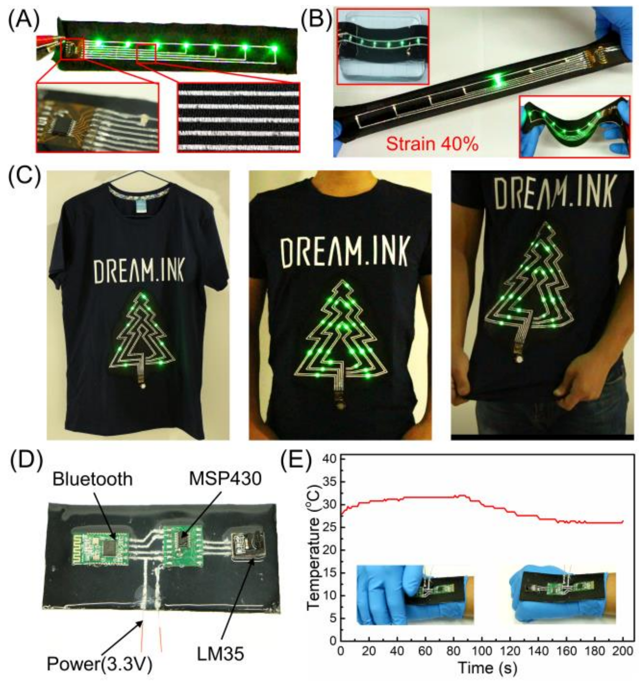

Figure 4. Applications of the liquid metal wearable electronics on fabrics. (A) The pictures of liquid metal LED array circuit. (B) The pictures of the liquid metal LED array circuit at 40% strain state and bent state. (C) The pictures of the large area LED array circuit printed on a T-shirt, when it was worn on body and pulled. (D) The picture of the temperature monitoring circuit printed on PU fabrics. (E) The curve of the temperature variation of the temperature monitoring circuit.

IV. Conclusion

In summary, we developed an improved liquid metal mask printing method for wearable electronics on fabrics. Through printing PMA glue on fabrics, the adhesion of liquid metal on fabrics was increased, which made it possible to print the liquid metal using a rolling brush directly. The results of electrical performance tests showed that the liquid metal conductors could maintain excellent electrical stability. Furthermore, this method could be used to fabricate large area wearable electronics on fabrics and individual healthcare monitoring. In fact, we also noted that the width of the liquid metal conductors was not thin enough for microelectronic devices due to the rough surface of the fabrics. With the improvement in adhesion, we expected that this method could have potential applications in fabricating flexible electronics on various soft substrates.

References

[1] M. Rein, V. D. Favrod, C. Hou, T. Khudiyev, A. Stolyarov, J. Cox, C.-C. Chung, C. Chhav, M. Ellis. J. Joannopoulos, and Y. Fink "Diode fibres for fabric-based optical communications," *Nature,* vol. 560, pp. 214–218, Aug. 2018.

[2] J. J. Park, W. J. Hyun, S. C. Mun, Y. T. Park, and O. O. Park, "Highly stretchable and wearable graphene strain sensors with controllable sensitivity for human motion monitoring," *ACS Appl. Mater. Interfaces*, vol. 7, pp. 6317–6324, Mar. 2015.

[3] Y. Wang, C. Zhu, R. Pfattner, H. Yan, L. Jin, S. Chen, F. Molina-Lopez, F. Lissel, J. Liu, N. I. Rabiah, Z. Chen, J. W. Chung, C. Linder, M. F. Toney, B. Murmann, and Z. Bao,, "A highly stretchable, transparent, and conductive polymer," *Sci. Adv,* vol. 3, e1601473, Mar. 2017.

[4] M. L. Hammock, A. Chortos, B. C.-K. Tee, J. B.-H. Tok, and Z. Bao, "25th anniversary article: The evolution of electronic skin (E‐Skin): A brief history, design considerations, and recent progress," *Adv. Mater*, vol. 25, pp. 5997–6038, Oct. 2013.

[5] S.-Y. Tang, V. Sivan, P. Petersen, W. Zhang, P. D. Morrison, K. K.-zadeh, A. Mitchell, and K. Khoshmanesh, "Liquid metal actuator for inducing chaotic advection," *Adv. Funct. Mater,* vol. 24, pp. 5851–5858, July. 2014.

[6] Y. Gao, H. Li, and J. Liu, "Inductor and capacitor to composite functional circuits: A super-simple way for alternative electronics," *PLoS ONE*, vol. 8, e69761, Aug. 2013.

[7] Y. Lu, Q. Hu, Y. Lin, D. B. Pacardo, C. Wang, W. Sun, F. S. Ligler, M. D. Dickey, and G. Zhen, "Transformable liquid-metal nanomedicine," *Nat. Commun*, vol. 6, 10066, Dec. 2015.

[8] S. Zhu, J.-H. So, R. Mays, S. Desai, W. R. Barnes, B. Pourdeyhimi, and M. D. Dickey, "Ultrastretchable fibers with metallic conductivity using a liquid metal alloy core," *Adv. Funct. Mater*, vol. 23, pp. 2308–2314, Dec. 2012.

[9] Y.-L. Park C. Majidi, R. Kramar, P. Bernard, and R. J. Wood, "Hyperelastic pressure sensing with a liquid-embedded elastomer," *J. Micromech. Microeng*, vol. 20, 125029, Nov. 2010.

[10] Y. R. Jeong, J. Kim, Z. Xie, Y. Xue, J. A. Rogers, and J. S. Ha, "A skin-attachable, stretchable integrated system based on liquid GaInSn for wireless human motion monitoring with multi-site sensing capabilities," *NPG Asia Mater*, vol. 9, e443, Aug. 2017.

[11] R. Guo, L. Sheng, H. Gong, and J. Liu, "Liquid metal spiral coil enabled soft electromagnetic actuator," *Sci China. Tech. Sci*, vol. 61, pp. 516–521, Apr. 2018.

[12] M. D. Dickey, R. C. Chiechi, R. J. Larsen, E. A. Weiss, D. A. Weitz, and G. M. Whitesides, "Eutectic Gallium‐Indium (EGaIn): A liquid metal alloy for the formation of stable structures in microchannels at room temperature," *Adv. Funct. Mater*, vol. 18, pp. 1097–1104, Apr. 2008.

[13] R. Guo, X. Wang, H. Chang, W. Yu, S. Liang, R. Wei, and J. Liu, "Ni‐GaIn amalgams enabled rapid and customizable fabrication of wearable and wireless healthcare electronics," *Adv. Eng. Mater*, vol. 20, Mar, 2018.

[14] C. Guo, Y. Yu, and J. Liu, "Rapidly patterning conductive components on skin substrates as physiological testing devices via liquid metal spraying and pre-designed mask," *J. Mater. Chem. B*, vol. 2, pp. 5739–5745, July, 2014.

[15] R. Guo, J. Tang, S. Dong, J. Lin, H. Wang, J. Liu, and R. Wei, "One‐step liquid metal transfer printing: Toward fabrication of flexible electronics on wide range of substrates," *Adv. Mater Technol*, vol. 3, No. 12, Dec, 2018.