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Recyclable Liquid Metal-Based Circuit on Paper

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Paper electronics is considered very environmentally friendly, but effective recyclability of metallic circuits on paper still needs more improvement. Therefore, liquid metal (Galinstan) circuits based on the reversible conversion from particles to wires are fabricated on paper using mechanical methods, i.e., mechanical sintering and sonication. Line-width of liquid metal (LM) circuit is kept in the range of 10 µm to ≥0.5 mm by controlling the sintering force. The results demonstrate that LM circuits exhibit high electrical stability during deformation, as resistance changes only ≤4% after the passage of 10 000 folding cycles. Meanwhile, LM particles spread on paper's porous structures, have enhanced the thermal diffusivity of paper and make paper electronics work in a facile temperature when integrated with high density units. More importantly, the reborn circuits exhibit almost identical electrical stability under deformation and thermal characteristic with pristine ones, thus making LM circuits environmentally friendly during their whole life span.

1. Introduction

Paper electronics is thriving in electronic security, flexible display, and biological diagnose for its outstanding features such as low-cost, lightweight, and disposability.[1–5] A series of investigations were dedicated to create functional modules on paper like 3D antenna, sensor, actuator, memory, and transistor.[5–14] Recently, by integrating many functional modules together through paper-based circuits,[7,9,15,16] more complex functional systems were realized, such as a keyboard,[12] sensor system,[17] and touch panel.[18] As people expected,[17] the processability, performance, and stability of electronic materials are basic characteristics of paper electronics which demand that paper-based circuits must have controllable scale, low resistance and high stability under deformation. Although most paper electronics are disposable, the circuits on paper will also raise environmental concern as they cannot be degraded or recycled like their substrate paper.[19] Burning is the popular method to recycle functional materials such as silver nanoparticles on paper,[1,20,21] it will be detrimental to environment and functional materials which are also difficult to be separated from the debris of paper. Therefore, an environmentally friendly method for fabricating and recycling of paper-based circuits is urgently needed.

Basically, all paper electronic devices contain functional modules and circuits. The circuit with controllable scale and high folding stability is the key point on deciding the integration level of devices and electrical stability under deformation. Recently, the fabrication of flexible circuits on paper is mostly based on the connection of tiny conductors, such as metal nanoparticles,[3,7,9,18,22,23] metal nanowires,[15,24] and graphene.[16] Alternatively, conductive liquids (especially liquid metals) were expected to realize circuits with high folding stability for intrinsically combined characteristics of both high electrical conductivity and excellent fluidity.[25] Liquid metal (LM, Galinstan: 68.2 wt% Ga, 21.8 wt% In, 10 wt% Sn[26]) exhibits favorable properties such as low melting point,[27] high electrical conductivity,[28] and low toxicity.[29] LM has been patterned on a variety of smooth substrates such as polydimethylsiloxane and polyvinyl chloride[25,26,30–37] to function as circuits, sensors, and antennas.[25,31,32,37–39] However, impeded by the porous topography of paper, it is hard to reduce the line-width of paper based LM circuits thinner than 200 µm with methods like direct writing,[40] desktop printing,[37,41] and screen printing.[42]

With the LM, here the method for fabricating circuits with controllable line-width and high stability is demonstrated, and these circuits can be recycled based on the transforming mechanism between different morphologies of LM (bulk, wires, and microparticles). Bulk LM can be deformed into LM particles (LMPs) by sonication.[43] Then, LMPs can be deposited on paper which can further be selectively transformed into LM wires through mechanically sintering.[38,44] Two strategies were introduced in the mechanically sintering process to control the width of circuit (pristine circuit) by controlling the contact

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area between tip and LMP film. More importantly, the LM circuit and LMP film on paper e-wastes can further be recycled by sonication which can transform LM back into LMP suspension. The LMP suspension will be used to fabricate another circuit (reborn circuit) and these LMP suspension will be used to fabricate another circuit (reborn circuit) again. Both the pristine circuit and the reborn circuit show high folding stability, which confirm that sonicating has no harm to the functional materials when recycling e-wastes. Furthermore, the untreated part of LMP film demonstrates roles as both insulator and thermal conductor which help to solve the intense joule heat generation resulting from high-density integrated circuits.

2. Results and Discussion

Due to environmental concern, mechanical methods (sonicating and mechanically sintering) were employed to fabricate LM circuits and recycle LM on paper e-wastes. The productions from the recycling step are LMPs that can be used to make other circuits, thus making the whole life span of paper electronics a closed loop. **Figure 1** shows schematically the proposed closed-loop processing method for recyclable paper based flexible LM circuits. First, the LMP suspension in Figure 1a was fabricated by sonicating bulk LM in alcohol for 3 min in an ultrasonic cell disruptor, where particles more than 90% of the total mass have a diameter between 1.64 and 5.5 µm as shown in Table S1 of the Supporting Information. Second, the suspension was deposited on paper to form the LMPs film. Oxide layer was formed on the surface of LMPs when being sonicated. And this oxide layer makes the LMPs film electrically insulating (Figure 1b).[38,45] Third, desired LM circuits were readily obtained by a simple mechanical sintering process,[41] which selectively transforming LMP film into LM lines by breaking the surface oxide layer of LMPs and connecting them together, as shown in Figure 1c. With this method, the size of LM circuits (pristine circuits) was able to be adjusted easily by controlling the applied force on the tip, which determined the contact area between the LMP film and the tip. Fourth, rigid LEDs or other components were integrated on the obtained LM circuits to create hybrid multifunctional systems (Figure 1d). Meanwhile, the thermal conductivity of paper was enhanced by LMP film that helps to cool down the working device, which is vital especially for highly integrated circuits. Finally, valuable material LM on paper e-wastes were designed to be recycled by transfer bulk LM (circuits) back into LM particles in alcohol solvent with sonication, as shown in Figure 1e. In this step, LMPs were dispersed in alcohol again and could be used directly to fabricate other circuits (reborn circuits) and thus the paper based LM circuits became environmentally friendly in its whole life span.

The size of LM circuits was successfully controlled for integrating components with different scales. **Figure 2**a shows the variation of LMP film thickness with the LM mass per unit area, where LMPs pile up layer by layer with the assistance of gravity. One can see that the thickness can be effectively controlled from 7 µm to larger than 115 µm by piling up the LMPs with diameters smaller than 5.5 µm. The inset is a cross-sectional SEM image of LMP film on paper. As the line-width of the circuit determines the integration level of electronics, two strategies are feasible to effectively control it (Figure 2b). The first strategy relies on controlling the applied force on the tips which influenced the contacting area between tips and LMP film. For example, circuits with line-width of 10 and 30 µm were manufactured by the tip of step profiler with applied forces of 10 and 30 µN, respectively ((i) and (ii) in Figure 2b). However, such force controlling strategy is only effective for adjusting line-width in the same order of magnitude. To create circuits with line-widths in different orders of magnitude, changing tips with different scales is the more feasible choice. For example, macroscale circuits (200 µm) can be created with the tip of a ballpoint pen (radius of head: 500 µm; (iii) in Figure 2b).

With a rough surface of substrate paper, mechanically sintered film (circuit) can be disconnected by organic fibers, especially for a thin LMP film. Figure 2c shows the effect of

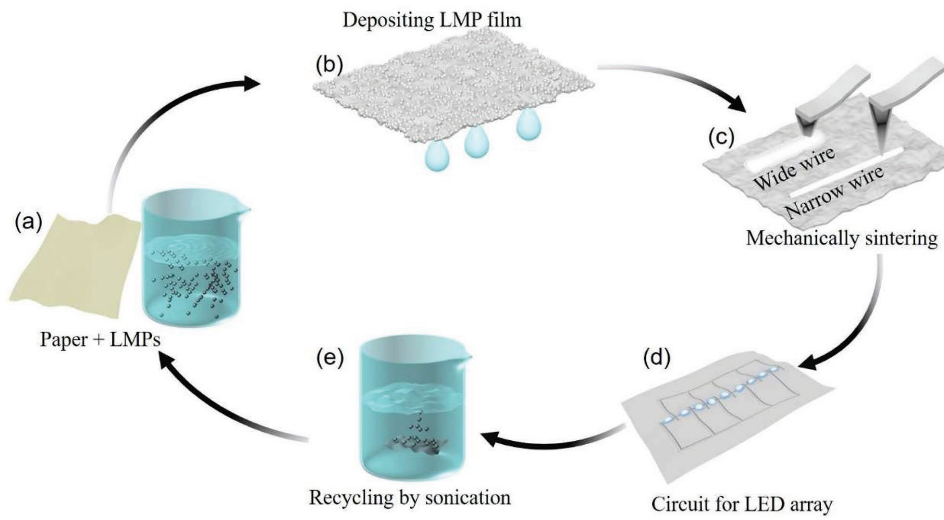

Figure 1. Closed-loop process for fabricating and recycling paper based LM circuit. a) Paper and LMP suspension made by sonicating bulk LM in ethanol. b) Depositing LMP film on paper. c) Fabrication of LM circuit by mechanically sintering the LMP film. d) Integrating LEDs on paper with interdigital LM circuit. e) Recycling LMP film and circuit by sonicating in ethanol.

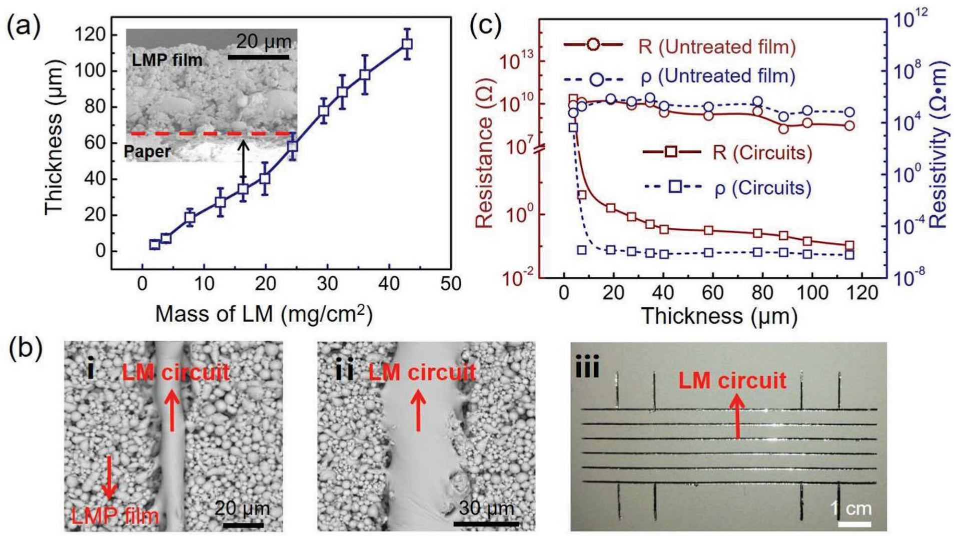

Figure 2. a) Average thickness of LMP film in respect to mass of LM per unit area. The inset is the cross-section view of LMP film with the thickness of 40 μm. b) LM circuits with different line-widths. i,ii) SEM images of LM circuits with the line-width of 10 and 30 μm manufactured by the same diamond tip (radius of head: 2 μm) with applying different forces (10 μN, 30 μN), respectively. iii) Large scale LM circuits fabricated by a ball pen tip (radius of head: 500 μm). c) Resistance and resistivity of untreated film and sintered film (LM circuit) as function of film thickness. The resistance of LM circuit is between two ends of an LM line with the fixed length 1 cm, width 0.5 mm. The resistance of untreated film is between two parallel LM lines with the fixed length 1 cm and distance 0.5 cm. The resistivity is estimated by the equation $\rho = \frac{Rs}{l}$, where l is the length of circuit (or LMP film) and s is the estimated sectional area.

LMP film thickness on the resistance and resistivity of LM circuits. As a comparison, the variation of LMP film resistance and resistivity with film thickness is given as well. Indeed, for films thinner than 7 μm, both the resistance and resistivity of circuit is in same magnitude of LMP film. This is resulted from that sintered conducting segments are hindered by the rather rough surface of paper. But for circuits on films thicker than 7 μm, the resistivity is stable and in same magnitude with bulk LM. The change of circuits' resistance is influenced by the geometry. Overall, for an LMP film thicker than 7 μm, the untreated film and annealed regions maintain their function as insulator and conductor, respectively.

The recycling process based on the transforming mechanism from LM wires to particles is the counterpart of the mechanically sintering step. **Figure 3** shows the recovery rate of LMP film as the function of sonicating time under different sonicating power. For sonicating power 180 W, the recovery rate increases quickly with time and can almost be saturated at 91% in 1 min. The recovery rate can be increased by increasing the time and power of sonication. SEM images in Figure S1b of the Supporting Information are cross-section views before and after recycled by sonication which shows that both the untreated LMP film and LM circuits were removed from paper. The EDX data in Figure S2a of the Supporting Information show that the LM is stable during the recycling process. The SEM images in Figure S2 of the Supporting Information were taken on same position of paper before and after recycling. Further, the EDX data on the paper after recycling show some LM residue. In short, sonication can deform LMP film and circuits on paper e-wastes into LMP suspension in a solvent, which can be used directly to make a reborn LMP film. For comparison, all reborn samples in this paper were made by recycling LM from pristine samples.

In this work, folding stability analysis was conducted on pristine and reborn circuits for high folding stability is the key requirement for flexible circuits as shown in **Figure 4**a. The resistances were obtained by measuring LM lines with a fixed 1 cm length, 200 μm width, and 40 μm thickness. Both the pristine and reborn circuits maintained low resistances (≤1.5 Ω) which changed parallelly as the bending angle was changed from 0° to ±180°. As demonstrated in Figure 4b, while being successively folded and unfolded, both the pristine and reborn circuits maintained low resistance smaller than 1 Ω and changed ≤4% after 10^4 folding tests. At the same time, all

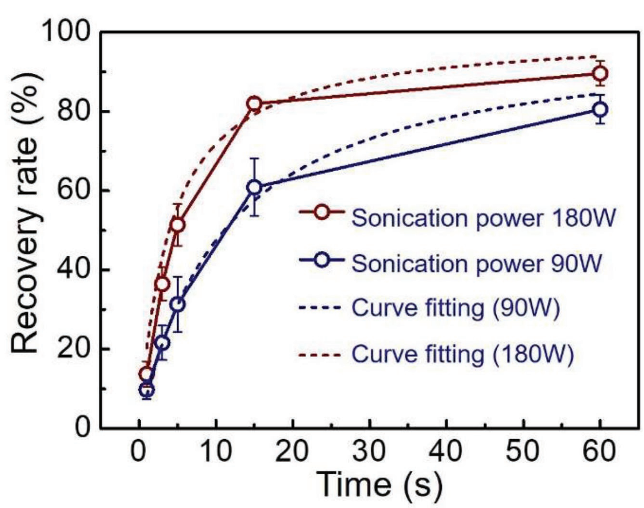

Figure 3. The recovery rate as the function of sonication time at different sonication power.

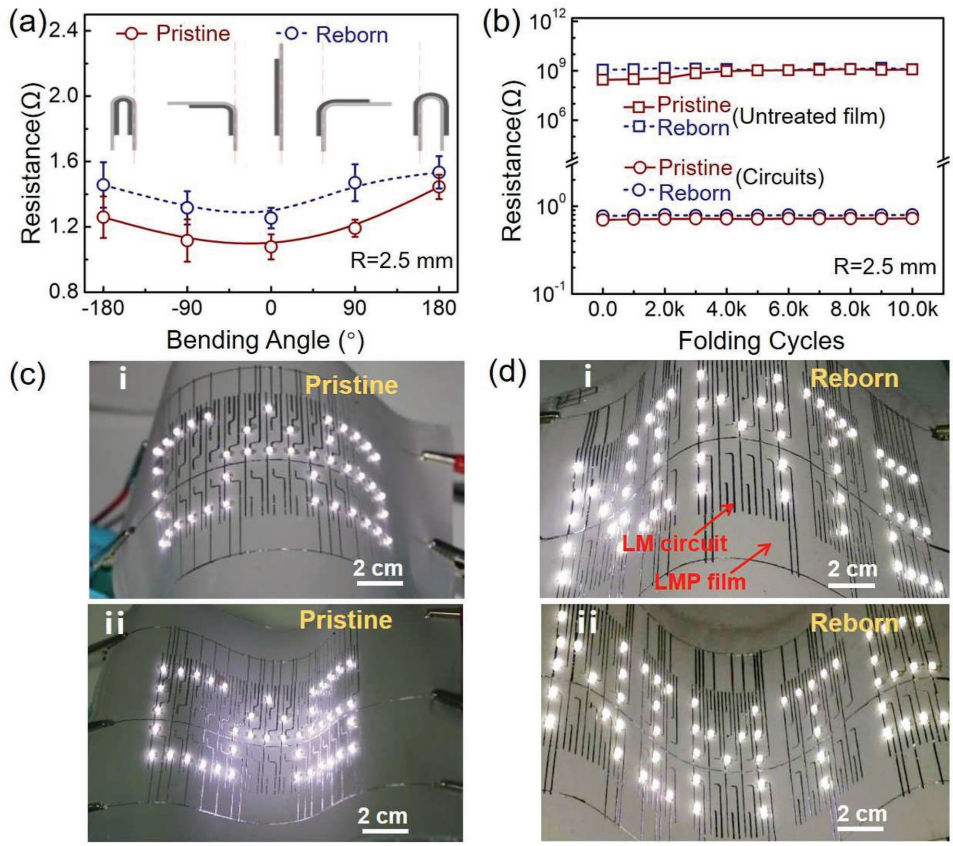

Figure 4. a) Resistance change of pristine and reborn LM circuits with the fixed length 1 cm, width 0.5 cm, and thickness 40 µm for various bending angles. b) Resistance of LM circuits (circle symbols) and untreated LMP film (square symbols) on pristine (wine) and reborn (navy blue) samples as function of folding cycles. c,d) Optical images of a flexible paper display containing LEDs shaped as "CAS" and "NIMTE" respectively. The circuit in (d) (reborn circuit) was made by recycling the circuit (pristine circuit) in (c) with sonication.

untreated film on pristine and reborn sample maintained high resistance (≥10^8 Ω) for being insulator.

The SEM images in Figure S4a,b of the Supporting Information are circuits before and after folding process, which can provide a qualitative explanation to the high stability of the LM circuits. According to former research on LM droplets,[46] the oxide skin of LM will be broken during deformation, which leads to the exposure of bare LM to air and the formation of some oxide skin wrinkles. Then, the bare LM newly exposed to air will be oxidized during the folding test. After experienced abundant circulation of oxide skin broken and reformation, enough oxide skin wrinkles will adhere to the surface of LM circuits, which can maintain its shape during subsequent folding test and prevent the LM circuits from further oxidation.

To demonstrate the scale controllability, recyclability of our LM circuits on large area flexible paper displays, we fabricated two white LED displays (pristine display and reborn display) with LMP circuits and surface-mount LEDs. Letters of "CAS" were aligned on the pristine display (Figure 4c), then the pristine display was recycled by sonication to produce the reborn LED displays with letters of "NIMTE" on it (figure 4d). Both the pristine and reborn displays were found to work stably under different deformations, and the performance of reborn display showed no degradation in contrast to pristine display.

The Aforementioned low scale circuits on paper endow us chances to make the high density integrated circuits, but the accompanied high heat production must be considered as paper is very flammable. The untreated LMP film between sintered film (circuits) can help to alleviate this issue by acting simultaneously as a thermal conductor. The thermal diffusivity of pristine and reborn LMP films are shown in **Figure 5**a as a function of thickness. As expected, the thermal diffusivity of LMP film increases notably with film thickness, and even triples for the film with a thickness of 100 µm when compared to the bare paper. Furthermore, to test the cooling effect for the real components, an LED display was fabricated with the letter "C" on pristine paper (right part in Figure 5b) and another "C" on LMP film-coated paper (left part in Figure 5b). A series of infrared images have been captured to show the heating process of both characters. After being lit for 20 min, the highest temperature on pristine paper reached 40 °C. By contrast, the lights on LMP film-coated paper maintained facile temperature lower that 36.5 °C, which will make paper-based devices user friendly. Further, the system was analyzed by finite element method with COMSOL, and the simulation results agree well with the experiment (Figure S5, Supporting Information).

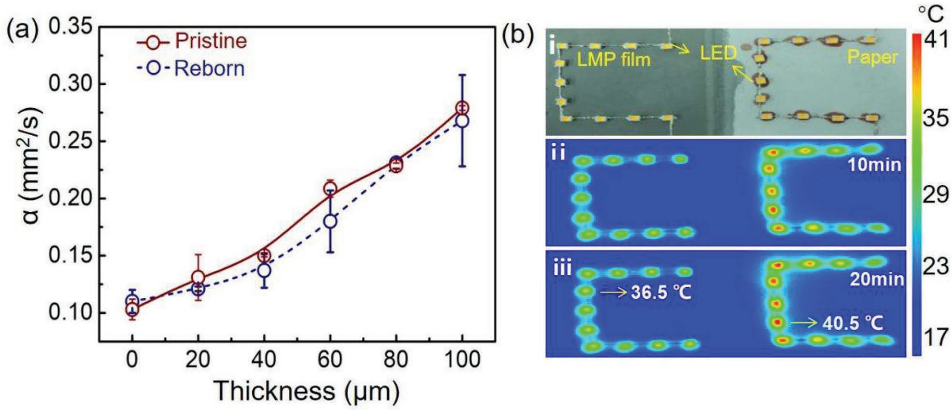

Figure 5. a) Thermal diffusivity of pristine and reborn LMP film with different thicknesses. b) The optical and infrared images of a paper display integrated with LED array. i) The left part is coated with LMP film while the right part is pure paper with LM lines made by a brush. ii, iii) Heat distribution images at different times. In (iii) the highest temperature on left part (LMP film coated) is 36.5 °C while the highest temperature on right part (pure paper with brushed LM lines) is 40.5 °C.

3. Conclusion

In summary, we demonstrate the recyclable LM circuits on paper, and the fabricating process includes LMP film depositing, circuits scale controlling, waste circuits recycling, and the further LMP film redepositing for reborn circuits. By adopting sintering force controlling strategy, line-width of LM circuits can be controlled from 10 μm to larger than 0.2 mm. More importantly, circuits created with this method have high stability. The resistance changes only ≤4% after experienced 10 000 folding tests. SEM images indicate that the high stability comes from LM oxide skin wrinkles that can confine the LM in its conducting channel. Also, the LMP film and those LMPs spread into paper's porous structure can enhance the thermal conductivity of paper and thus make paper electronics work in a facile temperature when integrated with high density units. More importantly, sonicating was applied as an environmentally friendly mechanical recycling method to recycle LM on paper. LMP suspension got from the recycling step was further used to make reborn circuits which showed no degradation in thermal conductivity and folding stability. Finally, the comparison between pristine and reborn circuits showed the damage free ability of sonication for recycling LM on paper e-wastes, and this also confirmed the environmental protection of LM circuit during its whole life span. Nonetheless, LMP film on circuits can be sintered by occasional force in daily life. From the view of application, sealing technologies is necessary for protecting both the LMP film and LM circuits.

4. Experimental Section

Preparation of Liquid Metal Galinstan: Gallium (99.99%; Beijing Founde Star Sci. & Technol. Co., Ltd), indium (99.995%; Beijing Founde Star Sci. & Technol. Co., Ltd), and tin (99.99%; Beijing Founde Star Sci. & Technol. Co., Ltd) were mixed together in the ratio of 68.2:21.8:10 by mass. Then the mixture was heated and stirred for 30 min protected by nitrogen gas at 60 °C to obtain liquid-metal Galinstanm ($Ga_{68.2}In_{21.8}Sn_{10}$).

Preparation of LMPs and LMP Film: A fixed mass (500 mg) of Galinstan and a fixed volume of ethanol (10 mL) were poured into a beaker. The mixture was sonicated for 3 min using an ultrasonic cell disruptor (Xinyi JY92-IIN) at 50% amplitude ratio to get the LMP suspension. Following this, the LMP was spread out on the surface of paper with the assistance of a glass rod. After drying in a fume hood for 1 h, a layer of LMP formed on the surface of the paper. To control the thickness of LMP film, the surface of paper was adjusted to change the mass of LM per area.

Scale Controlling of LM Circuit: The LM circuits showed in Figure 2b were made by two strategies. Strategy 1: to adjust the scale of LM circuits finely, the forced applied on one tip was controlled. The LM circuits showed in Figure 2b(i,ii) are made by applying forces of 10 and 30 μN, respectively, by a tip of step profiler (Bruker DektakXT). Strategy 2: to adjust the scale of LM circuits roughly, tips with different scales were changed. LM circuit showed in Figure 2b(iii) was made by a ball-pen tip on a three-axes movement controller (AMCNC01).

Electrical Testing: The resistance of LMP circuits and film were measured by a current source (Keithley 6221) and nanovoltmeter (Agilent 34420A). To test the stability under different bending angles, samples on plastic models with different angles were curled. Furthermore, a stability test machine was used to folding the sample for 10 000 cycles.

Thermal Testing of LMP Film: The thermal diffusivity of LMP film in Figure 4a was measured by the laser flash diffusivity apparatus (Netzsch LFA457). The infrared photos in Figure 4b(ii,iii) were taken by a thermal infrared imager system (FLIR A325sc).

Recycling of LM on Paper e-Wastes: Sonication was used to recycle LM circuits and LMP film on paper e-wastes, and reborn samples were made by LMP suspension got from recycling e-wastes. The first step removed the rigid components away. Then the paper was submerged into ethanol in a beaker which was sonicated in a sonication cleaner (KUDOS SK3310HP) for 15 min. After this step the wasted LMP film was transferred into the LMP suspension which can be used to create a reborn LMP film again.

Supporting Information

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

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

flexible circuit, liquid metal, paper circuit, recyclable

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[1] M. Yang, S. W. Jeong, S. J. Chang, K. H. Kim, M. Jang, C. H. Kim, N. H. Bae, G. S. Sim, T. Kang, S. J. Lee, B. G. Choi, K. G. Lee, *ACS Appl. Mater. Interfaces* **2016**, *8*, 34978.
[2] Y. H. Jung, T. H. Chang, H. Zhang, C. Yao, Q. Zheng, V. W. Yang, H. Mi, M. Kim, S. J. Cho, D. W. Park, H. Jiang, J. Lee, Y. Qiu, W. Zhou, Z. Cai, S. Gong, Z. Ma, *Nat. Commun.* **2015**, *6*, 7170.
[3] Y. Ko, M. Kwon, W. K. Bae, B. Lee, S. W. Lee, J. Cho, *Nat. Commun.* **2017**, *8*, 536.
[4] K. Pardee, A. A. Green, M. K. Takahashi, D. Braff, G. Lambert, J. W. Lee, T. Ferrante, D. Ma, N. Donghia, M. Fan, N. M. Daringer, I. Bosch, D. M. Dudley, D. H. O'Connor, L. Gehrke, J. J. Collins, *Cell* **2016**, *165*, 1255.
[5] S. Lee, H. Seong, S. G. Im, H. Moon, S. Yoo, *Nat. Commun.* **2017**, *8*, 725.
[6] W. J. Hyun, E. B. Secor, G. A. Rojas, M. C. Hersam, L. F. Francis, C. D. Frisbie, *Adv. Mater.* **2015**, *27*, 7058.
[7] J. Lessing, A. C. Glavan, S. B. Walker, C. Keplinger, J. A. Lewis, G. M. Whitesides, *Adv. Mater.* **2014**, *26*, 4677.
[8] Z.-K. K. Der-Hsien Lien, T.-H. Huang, Y.-C. Liao, S.-C. Lee, J.-H. He, *ACS Nano* **2014**, *8*, 7613.
[9] A. Russo, B. Y. Ahn, J. J. Adams, E. B. Duoss, J. T. Bernhard, J. A. Lewis, *Adv. Mater.* **2011**, *23*, 3426.
[10] M. Amjadi, M. Sitti, *ACS Nano* **2016**, *10*, 10202.
[11] X. Liao, Z. Zhang, Q. Liao, Q. Liang, Y. Ou, M. Xu, M. Li, G. Zhang, Y. Zhang, *Nanoscale* **2016**, *8*, 13025.
[12] K. Lee, J. Lee, G. Kim, Y. Kim, S. Kang, S. Cho, S. Kim, J. K. Kim, W. Lee, D. E. Kim, S. Kang, D. Kim, T. Lee, W. Shim, *Small* **2017**, *13*, 1700368.
[13] S. R. Das, Q. Nian, A. A. Cargill, J. A. Hondred, S. Ding, M. Saei, G. J. Cheng, J. C. Claussen, *Nanoscale* **2016**, *8*, 15870.
[14] M. A. Khan, U. S. Bhansali, H. N. Alshareef, *Adv. Mater.* **2012**, *24*, 2165.
[15] N. N. Jason, W. Shen, W. Cheng, *ACS Appl. Mater. Interfaces* **2015**, *7*, 16760.
[16] W. J. Hyun, O. O. Park, B. D. Chin, *Adv. Mater.* **2013**, *25*, 4729.
[17] D. Tobjork, R. Osterbacka, *Adv. Mater.* **2011**, *23*, 1935.
[18] S. Jung, S. J. Chun, C. H. Shon, *ACS Appl. Mater. Interfaces* **2016**, *8*, 20301.
[19] H. Jeong, S. Baek, S. Han, H. Jang, S. H. Kim, H. S. Lee, *Adv. Funct. Mater.* **2018**, *28*, 1704433.
[20] H. Wu, S. W. Chiang, W. Lin, C. Yang, Z. Li, J. Liu, X. Cui, F. Kang, C. P. Wong, *Sci. Rep.* **2014**, *4*, 6275.
[21] Y. Wang, H. Guo, J. J. Chen, E. Sowade, Y. Wang, K. Liang, K. Marcus, R. R. Baumann, Z. S. Feng, *ACS Appl. Mater. Interfaces* **2016**, *8*, 26112.
[22] L. Liu, X. Wan, L. Sun, S. Yang, Z. Dai, Q. Tian, M. Lei, X. Xiao, C. Jiang, W. Wu, *RSC Adv.* **2015**, *5*, 9783.
[23] Y.-L. Tai, Z.-G. Yang, *J. Mater. Chem.* **2011**, *21*, 5938.
[24] R. Z. Li, A. Hu, T. Zhang, K. D. Oakes, *ACS Appl. Mater. Interfaces* **2014**, *6*, 21721.
[25] Q. Wang, Y. Yu, J. Yang, J. Liu, *Adv. Mater.* **2015**, *27*, 7109.
[26] I. D. Joshipura, H. R. Ayers, C. Majidi, M. D. Dickey, *J. Mater. Chem. C* **2015**, *3*, 3834.
[27] T. Liu, P. Sen, C.-J. Kim, *J. Microelectromech. Syst.* **2012**, *21*, 443.
[28] D. Zrnic, D. S. Swatik, *J. Less-Common Met.* **1969**, *18*, 67.
[29] H. H. M. J. E. Chandler, G. Ellender, *J. Dent. Res.* **1994**, *73*, 1554.
[30] A. Tabatabai, A. Fassler, C. Usiak, C. Majidi, *Langmuir* **2013**, *29*, 6194.
[31] Y. Lin, O. Gordon, M. R. Khan, N. Vasquez, J. Genzer, M. D. Dickey, *Lab Chip* **2017**, *17*, 3043.
[32] M. Kubo, X. Li, C. Kim, M. Hashimoto, B. J. Wiley, D. Ham, G. M. Whitesides, *Adv. Mater.* **2010**, *22*, 2749.
[33] R. K. Kramer, C. Majidi, R. J. Wood, *Adv. Funct. Mater.* **2013**, *23*, 5292.
[34] B. A. Gozen, A. Tabatabai, O. B. Ozdoganlar, C. Majidi, *Adv. Mater.* **2014**, *26*, 5211.
[35] J. W. Boley, E. L. White, G. T. C. Chiu, R. K. Kramer, *Adv. Funct. Mater.* **2014**, *24*, 3501.
[36] Y. Zheng, Q. Zhang, J. Liu, *AIP Adv.* **2013**, *3*, 112117.
[37] Y. Zheng, Z. Z. He, J. Yang, J. Liu, *Sci. Rep.* **2014**, *4*, 4588.
[38] J. W. Boley, E. L. White, R. K. Kramer, *Adv. Mater.* **2015**, *27*, 2355.
[39] Q. Zhang, Y. Gao, J. Liu, *Appl. Phys. A* **2013**, *116*, 1091.
[40] H. L. Yunxia Gao, J. Liu, *PLoS One* **2012**, *7*, e45485.
[41] Y. Zheng, Z. He, Y. Gao, J. Liu, *Sci. Rep.* **2013**, *3*, 1786.
[42] L. Wang, J. Liu, *RSC Adv.* **2015**, *5*, 57686.
[43] J. N. Hohman, M. Kim, G. A. Wadsworth, H. R. Bednar, J. Jiang, M. A. LeThai, P. S. Weiss, *Nano Lett.* **2011**, *11*, 5104.
[44] M. G. Mohammed, R. Kramer, *Adv. Mater.* **2017**, *29*, 1604965.
[45] N. Syed, A. Zavabeti, M. Mohiuddin, B. Zhang, Y. Wang, R. S. Datta, P. Atkin, B. J. Carey, C. Tan, J. van Embden, A. S. R. Chesman, J. Z. Ou, T. Daeneke, K. Kalantar-zadeh, *Adv. Funct. Mater.* **2017**, *27*, 1702295.
[46] K. Doudrick, S. Liu, E. M. Mutunga, K. L. Klein, V. Damle, K. K. Varanasi, K. Rykaczewski, *Langmuir* **2014**, *30*, 6867.

