Flexible substrates enabled highly integrated patterns with submicron precision toward intrinsically stretchable circuits

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Abstract
Fabricating high integration density, high resolution, and intrinsically stretchable patterns by patterned technologies remain challenging. Template printing enabled high-precision patterned fabrication at a facile operation. However, the pattern spacing constraint is the major limitation to high integration density. In this study, we develop an elastomer-assisted strategy to improve the template printing process, which involves patterning on the prestrain elastic substrate. This strategy overcomes the spacing limitation and enables the realization of a centimeter-scale pattern with submicron precision. Particularly, the integration density of fabricated intrinsically stretchable patterns can reach 1932 lines on a substrate of 0.5 cm2; the assembly lines with a feature size of 880 nm and an interval of 955 nm. Furthermore, we demonstrate a facile approach for constructing silver nanoparticle/liquid metal alloy composite conductive patterns. The as-prepared flexible electrodes can withstand up to 150% strain and a 2-mm bend radius. This method provides new insights into template printing technology. Additionally, it opens a route for the simultaneous construction of functional patterned arrays with large scale, high integration density, and intrinsic stretchability, which will be useful for the integrated fabrication of various flexible electronic devices.

KEYWORDS
flexible devices, highly integrated patterns, liquid metal alloys, printed electronics, stretchable electronics
1 | INTRODUCTION

Printed electronics as an efficient and economical technology has the characteristics of excellent universality, diversity, and stability for fabricating flexible electronic devices. Printed flexible electronic devices play critical roles in diverse fields of healthcare, communication, energy, actuating, and security, and are exploited for myriad applications, such as solar cells, organic light-emitting diode, sensors, and displays. Flexible circuits are essential components for stretchable electronic devices. In recent years, stretchable electronic devices have been developed toward miniaturization and integration, and the soft circuits need to simultaneously fulfill electrical conductivity, stretchability, and high integration density. Noteworthy, achieving all these requirements in a facile patterning approach is still fundamentally challenging.

Advanced patterning strategies can promote integrated fabrication of functional patterns, such as photolithography-based technologies, nanoimprint lithography, and nanotransfer printing. But the high cost of lithography equipment and operating facilities might create an economic barrier for the patterning process. Nanoimprint and nanotransfer techniques rely on the precision of the master mold and suitable materials. As an alternative, we developed a bottom-up template printing method with wide material compatibility and realized nanoscale patterning resolution from microscale templates by taking advantage of the liquid bridge confined assembly process. The patterns with large scale and high precision on rigid substrates have been achieved. However, the pattern spacing in the template-induced assembly process is limited, and high-precision, highly integrated assembly on flexible substrates in a large area has not yet been achieved.

In this study, we demonstrate an elastomer-assisted template printing (EATP) strategy to construct highly integrated intrinsically stretchable silver nanoparticles (Ag NPs) arrays on flexible thermoplastic polyurethane (TPU) substrates. This method can afford patterns with a feature size of 880 nm and an interval of 955 nm using microscale templates, and the ratio of line width to space is approaching 1:1. The maximum integration density of the pattern reaches 1932 lines on a substrate of 0.5 cm². Moreover, the assembled Ag NPs arrays can act as a template for deposition liquid metal alloys, and the synergy of hard Ag NPs and soft liquid metal can achieve stretchable, high electrical conductivity, and high-resolution composite arrays. The as-prepared flexible electrodes can withstand a strain up to 150% and a bending radius of 2 mm. This study provides a promising patterning approach to fabricate functional patterns with large scale, high integration density, and intrinsic stretchability, enabling great potential applications in flexible electronics devices.

2 | EXPERIMENTAL SECTION

2.1 | Preparation of Ag NPs

Ag NPs were prepared according to the literature. In a typical synthesis route of Ag NPs, polyvinylpyrrolidone (20.0 g) was dissolved in ethylene glycol (50.0 mL), and AgNO₃ (1.2 g) was added to the solution. The suspension was then stirred at 60 °C until the silver nitrate was fully dissolved. The reaction system was then heated to 120 °C and held at that temperature for 1 h to allow the reaction to complete. After the mixture has been cooled to room temperature, the Ag NPs can be easily separated from the system by using a mixture of acetone and ethanol. Following the vacuum drying at 60 °C for 0.5 h, the Ag NPs can be obtained.

2.2 | Fabrication of highly integrated Ag NPs arrays

The TPU film was cut into a rectangle shape with the size of 8 cm × 2 cm, and the surface of the sample was cleaned with ethanol and dried in an N₂ atmosphere before use. Then, the samples were stretched to reach a certain strain using a customized stretching platform. Ag NPs functionalized ink was carefully dropped onto the TPU substrate and covered with a silicon template, where the orientation of the microstructure was perpendicular to the direction of stress. The microwall can act as the pinpoints to manipulate the directional shrinkage of the Ag NPs, yielding patterned liquid bridges in the gap between the microwall of the template and a flexible substrate. These arranged liquid bridges provide gradually decreasing spaces for the assembly of Ag NPs. With the evaporation of the solvent, the one-dimensional (1D) assembly arrays were successfully formed on flexible substrates. The substrate was then released to obtain highly integrated and high-precision 1D assembly arrays. Ag NPs functionalized ink was composed of 0.4% (wt or vol)–5% Ag NPs and 0.2% sodium dodecyl sulfate in a water solvent. The temperature was kept in a range of 20–45 °C. The assembly process of poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS), poly(sodium-4-styrene sulfonate)/Ag NPs (PSSNa/Ag NPs), and lithium bis(trifluoromethanesulfonylimide)/Ag NPs (LiTFSI/Ag NPs) on unstretched TPU substrates followed the same route as for Ag NPs.
3 | RESULTS AND DISCUSSION

3.1 | Principle and demonstration of EATP

Figures 1A and S1 schematically illustrate the EATP process. We compared a series of elastomeric polymer materials and chose the stretchable TPU film (Figure S2) as the flexible substrate. The substrates were stretched to reach a certain strain, and then a sandwich structure with Ag NP suspensions (Figure S3) and microwall silicon template (Figure S4) was constructed. Template printing exploits the capillary forces of menisci established under the protrusions of a template placed in contact with a liquid film.22,23 During the assembly process (Figure 1A), the Ag NP solution spontaneously infills the microstructures of the silicon template driven by the Laplace pressure. With the solvent evaporating, menisci form under the protrusions of a template placed in contact with a liquid film. Driven by capillary forces, the three-phase contact lines (TCLs) gradually recede, resulting in highly ordered patterned arrays of Ag NPs. The coordination interaction between Ag NP and TPU results in the immobilization of the nanoparticles on the substrate. After releasing the TPU substrate from the prestrain states, the line width and interval are reduced simultaneously, which effectively enhances the assembly integration density and precision of assembly arrays. Highly integrated patterned arrays with a centimeter-scale assembly area are successfully prepared.

The presented fabrication approach is compared to the recently reported solution-processable 1D patterning printing technologies (Figure 1B), such as screen printing,24 gravure printing,25,26 inkjet printing,27–29 template printing,20,30–32 transfer printing,33 and electrohydrodynamic printing.34 Screen printing and gravure printing are low-density patterning techniques with a spatial resolution of hundreds of micrometers. The distance between two adjacent lines is usually in dozens of microns by inkjet printing. Template printing and transfer printing provide a high-precision period of patterns, typically in the range of 5–10 μm. Although electrohydrodynamics can create arrays with ultrahigh precision, achieving a linewidth/space ratio of 1 is still challenging. The EATP approach enables the fabrication of highly integrated, large-area, intrinsically stretchable assembly arrays with an interval resolution of less than 1 μm. The ratio of line width to space is approaching 1:1, which overcomes the spacing restrictions of template printing.

3.2 | Surface characteristics and resilience of flexible TPU substrates

Polydimethylsiloxane (PDMS) is a typical flexible substrate in printing and flexible electronics. However, PDMS has low elasticity and tends to form corrugation on the surface after a stretch-release process.35 Therefore, three elastic polymer materials are investigated, including TPU, ECOFLEX, and natural rubber (NR). The mechanical property curves are shown in Figure 2A. All of them exhibited soft and tough features without yielding. But the ECOFLEX substrate exhibited a corrugated surface after releasing
from prestrain of 100% (Figure S5A,D). NR needs to be mixed with the reinforcing fillers during the fabrication process, which results in many particles on the surface (Figure S5B,E). These particles can act as pinpoints and hinder the TCL receding. In contrast, the flexible TPU substrate exhibits high tensile strength (51.8 MPa) and elongation (2370%), as well as smooth surface morphology (Figures S5C,F and 2B). Furthermore, the TPU shows little change in surface roughness after a stretch-release procedure (Figures 2C and S6).

### 3.3 Wettability of the substrate and template determining the assembly process

The wettability of substrate and template can affect the uniformity and regularity of assembled patterns. Like most polymer substrates, TPU substrates have a hydrophobic surface with an initial water contact angle of $73^\circ \pm 3^\circ$. It is difficult for plasma treatment to achieve consistent and stable wettability. A nonionic polymeric surfactant (Triton) is selected to modify the wettability of TPU substrates. By adjusting the amount of Triton in TPU, substrates with different water contact angles are prepared (Figure 3A). Triton changes the wettability of flexible TPU substrates via physical blending (Figure 3A, inset) and has a negligible effect on the mechanical property (Figure S7). Hence, the original resilience of the substrate is preserved. As illustrated in Figure 3B, the dependence of the water contact angle of substrates and solution concentrations on the assembly process of Ag NPs arrays are investigated. When the water contact angle is large, arrays can be assembled with low Ag NPs concentrations. As the concentration...
of Ag NPs increases, a small water contact angle is favorable for the successful assembly because the contact angle of the solution changes in a hydrophobic direction with the increase of concentration (Figure S8). For flexible TPU substrates, optimized assembled arrays of Ag NPs can be obtained with a water contact angle of 40°–50°.

The wettability of silicon templates can be adjusted by changing the plasma treatment energy. On flexible TPU substrates, two typical assembled morphologies are achieved (Figure 3C) using silicon templates treated with plasma under different conditions, including spreading morphology in a “rectangular” shape and dewetting one with a “triangular” stack (Figure S9). Figure 3D illustrates the formation process of spreading and dewetting assembly lines. Plasma can modify the hydrophilicity of the material by grafting hydrophilic groups. Therefore, the templates treated with low or high energy are superhydrophilic (Figure S10). Consequently, the change in stacking shape is probably caused by the different behaviors of the liquid bridge manipulated by the micro-walls. With a low-energy plasma treatment, the pulling action of the microwall on the liquid bridges is weak and thus easy to break, resulting in a dispersed morphology. When the treatment energy increases, the microwall exerts a strong pulling and tugging force on the liquid bridge, making the liquid bridge hard to break, resulting in a dewetting morphology. However, the dewetting assembly lines are stacked in a brittle structure that cannot tolerate minor deformation and is prone to fall away from the TPU substrate after being released from a large prestrain. Numerous microcracks are discovered on dewetting lines following a simple deformation of the TPU substrate (Figure S11). In comparison, the spread assembly lines are layered in a nearly monolayer configuration so that the assembly arrays can accommodate large strain.
3.4 | Large-scale, intrinsically stretchable Ag NPs arrays with high integration density fabricated by EATP

The microwall as pinpoints to manipulate the liquid bridge is affected by the interval/linewidth ratio, and when the ratio is between 3 and 6, it is favorable for assembly. While assembling the Ag NP lines on the TPU flexible substrate, the interval/linewidth ratio is no longer a limiting factor, and orderly assembled lines can be obtained with interval/linewidth ratio ranging from 1.2 to 10 (Figures S12 and S13), and the assembly microarrays can easily reach the large scale (Figure S14). That may be related to the interactions at the interface. The functional imines (–NH–) groups of TPU can coordinate with the Ag atoms (Figure S15). Therefore, the Ag NPs can immobilize on the substrate, and the spreading assembly lines achieve large scale and intrinsic stretchability. During the EATP process, prestrains of 0%, 50%, 100%, 150%, 200%, and 250% are applied. Figure 4A shows SEM images of the Ag NPs assembly arrays on the unstretched TPU substrate and released TPU substrate (prestrain 250%). When the TPU substrate is fully released, the spacing and the line width are simultaneously decreased. The interval is reduced from 10.05 to 4.05 μm, while the resolution of the line width increased from 2.00 to 0.91 μm. More assembly results on the flexible substrate with various prestrains are displayed in Figure S16. As illustrated in Figure 4B, a relationship between the distances (line width and interval) and the prestrain is established.

In addition, we used templates with an interval/linewidth ratio of 1.5 and successfully fabricated a highly integrated pattern with a density of 1932 lines/cm². After fully releasing the TPU substrate, the assembly area is reduced to 0.5 cm², resulting in a considerable increase in integration density, the resolutions of line width and interval achieve submicron simultaneously (Figure 4C).
The mechanism of the assembly process is depicted in Figure 4D. The capillary force dominates the receding of TCL before the liquid bridge breaks. After the solution is evaporated, the nanoparticles are immobilized on the substrate. Thus, the spreading assembly lines exhibited a remarkable tolerance to large strain and retained their original structural characteristics.

The flexible TPU substrates also show a general versatility for assembling various functional materials. We successfully fabricated conductive polymers (PEDOT:PSS), polymers/NP (PSSNa/Ag NPs), and NP/NP (LiTFSI/Ag NPs) arrays (Figure S17). PEDOT: PSS arrays can remain structurally undamaged after TPU substrates were released from a prestrain of 100% (Figure S18).

3.5 Design of composite conductive arrays and the electromechanical response of flexible electrodes

The assembled Ag NPs have a low electrical conductivity. Fabricating patterns with metal-like conductivity, high stretchability, and facile patternability remains a critical challenge. Typically, there are trade-offs between these properties, and relatively few reports on stretchable circuits have achieved these goals simultaneously. Liquid metals with the conductivity of metals and the conformability of fluids make them promising candidates for stretchable conductors. Nevertheless, liquid metal has low wettability and is difficult to spread on the surface of materials, while it shows better wettability on Ag NPs and easy to prepare patterns. We used liquid metals (eutectic gallium–indium alloy, EGaIn) and assembled Ag NP arrays to fabricate the composite conductive arrays. The fabrication process is depicted in Figure S19. The EGaIn can selectively deposit on the assembled region (Figures 5A and S20). When EGaIn is deposited, it fills the gaps between the Ag NPs and merges with the Ag NPs to form thin, continuous, semisolid conductive microtraces (Figures 5B and S21). EGaIn deposition leads to a five order-of-magnitude increase in conductivity from 0.23 (sintering, 120 °C, 2 h) to 1.05 × 10⁴ S/m for Ag NP/EGaIn arrays (inset in Figure 5C). Moreover, the various sizes of conductive arrays can be fabricated according to the Ag NP master template (Figure S22). Micron-scale EGaIn patterning
with feature sizes as narrow as 1 and 2 μm line spacings is achieved, resulting in the highest resolution EGaIn patterning additive method to date (Figure S23).

To demonstrate the electrical properties of the composite arrays, we investigated the electromechanical behavior of flexible electrodes with Ag NPs/EGaIn patterns (line width 10 μm and pitch 12 μm) under large deformations. Transmission response over the entire visible wavelength is shown in Figure 5C and indicates no significant decrease in transmittance before \((T = 82.9\%), \text{at } 550 \text{ nm}\) and after \((T = 80.3\%), \text{at } 550 \text{ nm}\) EGaIn deposition. In comparison, TPU substrates (without assembly arrays, blank) present nearly 97.1% transmittance under the same conditions. To demonstrate the potential of Ag NPs/EGaIn arrays for utility as soft circuits in flexible electronics, we measured the mechanical stability of relative resistance change \((\Delta R/R_0)\) during the stretching and bending tests for Ag NPs/EGaIn electrodes. The electrode remains highly conductive of a global strain of up to 150%. As the global strain increases from 30% to 150%, the \(\Delta R/R_0\) increases from 0.15 to 1.58 in parallel and 0.14 to 0.49 in the vertical direction (Figure 5D). Minimum bend radius up to 2 mm, \(\Delta R/R_0\) of 3.06 in the parallel direction, and \(\Delta R/R_0\) of 1.81 in the vertical direction are obtained (Figure 5E). The patterned electrodes are demonstrated with light-emitting diodes (Figure S24). In addition, the flexible electrodes also possess cycling durability with about 0.4 times relative resistance change after reversibly stretching tests to 50% strain and 0.8 times relative resistance change after bending tests to a 3-mm bend radius for 500 cycles (Figure 5F). Figure S25 shows Ag NPs/EGaIn conductive line morphologies before and after cycling. Liquid-phase alloys can operate as dynamic and durable electrical anchors between Ag NPs. The pinning effect of Ag NPs prevents the EGaIn from splitting into microdroplets under large strain. Therefore, the composite circuits simultaneously have excellent conductivity, stretchability, and mechanical robustness. These results indicate the potential of using Ag NP/EGaIn conductive composite conductive arrays to fabricate flexible electrodes.

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CONFLICTS OF INTEREST
The authors declare no conflicts of interest.

DATA AVAILABILITY STATEMENT
The data that support the findings of this study are available in the supplementary material of this article.

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REFERENCES
1. Huang Q, Zhu Y. Printing conductive nanomaterials for flexible and stretchable electronics: a review of materials, processes, and applications. Adv Mater Technol. 2019;4(5):1800546.
2. Kim J, Kumar R, Bandodkar AJ, Wang J. Advanced materials and printed wearable electrochemical devices: a review. Adv Electron Mater. 2017;3(1):1600260.
3. Su M, Song Y. Printable smart materials and devices: strategies and applications. Chem Rev. 2022;122(5):5144-5164.
4. Li D, Lai W-Y, Zhang Y-Z, Huang W. Printable transparent conductive films for flexible electronics. Adv Mater. 2018;30(10):1704738.
5. Zhou L, Yu M, Chen X, et al. Screen-printed poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) grids as ITO-free anodes for flexible organic light-emitting diodes. Adv Funct Mater. 2018;28(11):1705953.
6. Wang Z, Jiang X, Huang K, et al. A bioinspired adhesive-integrated-agent strategy for constructing robust gas-sensing arrays. Adv Mater. 2021;33(51):e2106067.

4 | CONCLUSION
We demonstrated an effective approach to fabricating stretchable patterns with integration density and high precision. Large-scale Ag NPs patterns are prepared on flexible TPU substrates with a line width of 880 nm and an interval of 955 nm, reaching an integration density of 1932 lines on a substrate of 0.5 cm². We further demonstrated printing stretchable conductive microtraces using liquid metal alloys. The composite circuits have great potential for flexible electronics. The as-prepared liquid metal-based patterned flexible electrodes display a marginal relative resistance change by only 1.58-fold at 150% strain and 1.81-fold at a 2-mm bend radius. This study effectively assembles functional materials into intrinsically stretchable and highly integrated patterns on a flexible substrate, especially for the integration fabrication of various flexible electronic devices.
15. Park HH, Kim HN, Kim H, et al. Thermally assisted hybrid printing and hybrid hot embossing. *Adv Mater*. 2021;33(21):2007772.

16. Huang Z, Su M, Yang Q, et al. A general patterning approach for nanoscale molecular aggregates. *Adv Funct Mater*. 2021;31(36):2105054.

17. Min F, Zhou P, Huang Z, et al. A bubble-assisted approach for patterning nanoscale molecular aggregates. *Angew Chem Int Ed*. 2021;60(11):841-850.

18. Yang Y, Min F, Qiao Y, et al. Embossed transparent electrodes assembled by bubble templates for efficient flexible perovskite solar cells. *Nano Energy*. 2021;89:106384. https://www.sciencedirect.com/science/article/pii/S221128552100639X

19. Zhang Z, Wang H, Su M, et al. Printed nanochain-based colorimetric assay for quantitative virus detection. *Angew Chem Int Ed*. 2021;60(45):24234-24240.

20. Su B, Zhang C, Chen S, et al. A general strategy for assembling nanoparticles in one dimension. *Adv Mater*. 2014;26(16):2501-2507.

21. Guo D, Li C, Wang Y, Li Y, Song Y. Precise assembly of particles for zigzag or linear patterns. *Angew Chem Int Ed*. 2017;56(48):15348-15352.

22. Cavallini M, Gentili D, Greco P, Valle F, Biscarini F. Micro- and nanopatterning by liquid-phase-assisted contact printing. *Nat Protoc*. 2012;7(9):1668-1676.

23. Miele E, Raj S, Baraissou Z, Král P, Mirsaidov U. Dynamics of templated assembly of nanoparticle filaments within nanochannels. *Adv Mater*. 2017;29(37):1702682.

24. Hyun WJ, Lim S, Ahn BY, Lewis JA, Frisbie CD, Francis LF. Screen printing of highly loaded silver inks on plastic substrates using silicon stencils. *ACS Appl Mater Interfaces*. 2015;7(23):12619-12624.

25. Huang Q, Zhu Y. Gravure printing of water-based silver nanowire ink on plastic substrate for flexible electronics. *Sci Rep*. 2018;8(1):15167.

26. Ko K-J, Lee HB, Kang J-W. Flexible, wearable organic light-emitting fibers based on PEDOT:PSS/Ag-fiber embedded hybrid electrodes for large-area textile lighting. *Adv Mater Technol*. 2020;5(6):2000168.

27. Ahn BY, Duoss EB, Motala MJ, et al. Omnidirectional printing of flexible, stretchable, and spanning silver microelectrodes. *Science*. 2009;323(5921):1590-1593.

28. Lin Y, Gao Y, Fan Z. Printable fabrication of nanocoral-structured electrodes for high-performance flexible and planar supercapacitor with artistic design. *Adv Mater*. 2017;29(43):1701736.

29. Zhang Z, Zhang X, Xin Z, Deng M, Wen Y, Song Y. Controlled inkjetting of a conductive pattern of silver nanoparticles based on the coffee-ring effect. *Adv Mater*. 2013;25(46):6714-6718.

30. Wu Y, Feng J, Jiang X, et al. Positioning and joining of organic single-crystalline wires. *Nat Commun*. 2015;6(1):6737.

31. Feng J, Jiang X, Yan X, et al. “Capillary-bridge lithography” for patterning organic crystals toward mode-tunable micro-/nanolaser array. *Adv Mater*. 2017;29(1):1603652.

32. Zou C, Yanahashi N, Wu Y, et al. Patterning smectic liquid crystals for OFETs at low temperature. *Adv Funct Mater*. 2019;29(7):1804838.

33. Song D, Mahajan A, Secor EB, Hersam MC, Francis LF, Frisbie CD. High-resolution transfer printing of graphene lines for fully printed, flexible electronics. *ACS Nano*. 2017;11(7):7431-7439.

34. Zou W, Yu H, Zhou P, Zhong Y, Wang Y, Liu L. High-resolution additive direct writing of metal micro/nanostructures by electrohydrodynamic jet printing. *Appl Surf Sci*. 2021;543:148800. https://www.sciencedirect.com/science/article/pii/S0169433220335595

35. Hammock ML, Chortos A, Tee BC-K, Tok JB-H, Bao Z. 25th anniversary article: the evolution of electronic skin (E-Skin): a brief history, design considerations, and recent progress. *Adv Mater*. 2013;25(42):5997-6038.

36. Li Y, Zhang Z, Su M, et al. A general strategy for printing colloidal nanomaterials into one-dimensional micro/nanostructures by electrohydrodynamic jet printing. *Nano Energy*. 2018;50:22374-22380.

37. Guo D, Zheng X, Wang X, et al. Formation of multicomponent size-sorted assembly patterns by tunable templated dewetting. *Angew Chem Int Ed*. 2018;57(49):16126-16130.

38. Yin T, Lavoie SR, Qu S, Suo Z. Photoinitiator-grafted polymer chains for integrating hydrogels with various materials. *Cell Rep Phys Sci*. 2021;2(6):100463.

39. Kang H, Jung S, Jeong S, Kim G, Lee K. Polymer–metal hybrid transparent electrodes for flexible electronics. *Nat Commun*. 2015;6(1):6503.

40. Lyu J, Wang X, Liu L, et al. High strength conductive composites with plasmonic nanoparticles aligned on aramid nanofibers. *Adv Funct Mater*. 2016;26(46):8435-8445.

41. Wang J, Cai G, Li S, Gao D, Xiong J, Lee PS. Printable superelastic conductors with extreme stretchability and robust cycling endurance enabled by liquid-metal particles. *Adv Mater*. 2018;30(16):e1706157.

42. Yuan B, Zhao C, Sun X, Liu J. Lightweight liquid metal entity. *Adv Funct Mater*. 2020;30(14):1910709.

43. Liu S, Shah DS, Kramer-Bottiglio R. Highly stretchable multilayer electronic circuits using biphasic gallium-indium. *Nat Mater*. 2021;20(6):851-858.
44. Zhuang Q, Ma Z, Gao Y, et al. Liquid-metal-superlyophilic and conductivity-strain-enhancing scaffold for permeable superelastic conductors. Adv Funct Mater. 2021;31(47):2105587.

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SUPPORTING INFORMATION
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