

Printing liquid metals in three-dimensional structures

June 27 2019, by Thamarasee Jeewandara



Using the 3-D printing technique to reconfigure a square coil antenna. Credit: Science Advances, doi: 10.1126/sciadv.aaw2844

In a recent study on materials science and nanomedicine, Young-Geun Park and co-workers at the departments of Nanoscience, Nanomedicine and Materials Science and Engineering in the Republic of Korea developed an unconventional <u>3-D printing</u> approach. The scientists engineered a high-resolution, reconfigurable 3-D printing strategy using liquid metals to form stretchable, 3-D constructs. Using the technique, they formed a minimum line width of 1.9 µm using direct printing and



printed patterns for reconfiguration in to diverse 3-D structures while maintaining pristine resolutions.

They performed reconfigurations many times to generate a thin oxide interface and preserve electrical properties of the material under ambient conditions. The free-standing features could be encapsulated in stretchable, conformal configurations. Park et al. demonstrated applications in the form of reconfigurable antenna, tunable by changing geometries and reversibly movable interconnections to use the constructs as mechanical switches. The free-standing 3-D structures were advantageous to minimize the number and space between interconnections for higher integration, as seen with microLED arrays. The results are now published on *Science Advances*.

Advanced technologies that form 3-D conductive structures with <u>high-resolution</u>, high aspect ratios and minimal error of displacement are important to <u>increase device integrity</u>. Device deformability is a key consideration for free-form electronics, including <u>stretchable electronics</u>, wearable electronics, soft actuators and <u>robotics</u>. These electronic devices typically require conformation with movable, arbitrary shapes such as joints or arms, or the soft surfaces of living organisms. Realizing such stretchable devices with conventional materials such as silicon are a challenge due to their brittleness. Materials scientists have therefore developed diverse conductive materials with excellent stretchability in the form of <u>wavy thin metals</u>, <u>metallic networks</u> and <u>elastomeric composites</u>, yet these processes are unable to form scalable 3-D structures. In addition, 3-D printed, and thermally annealed metals are relatively stiff and rigid <u>causing damage to soft</u>, tissue-like substrates.





High-resolution printing of liquid metals. (A) Schematic illustration of a printing system. (B) SEM image of 2D and 3D high-resolution EGaIn patterns. Scale bar, 100 μ m. Inset: Magnified SEM image of the 3D structures. Scale bar, 100 μ m. (C) AFM image and cross-sectional profile of printed EGaIn line. Scale bar, 2 μ m. (D) SEM image of 1.9- μ m-wide EGaIn patterns. Scale bar, 10 μ m. (E) SEM image of 3D patterns of EGaIn on a PET film and epoxy (SU-8). Scale bar, 10 μ m. (F) Photograph of printed high-resolution EGaIn patterns in (B). Scale bar, 1 cm. (G) Photograph of interconnect patterns of EGaIn. Inset: Top-view photograph. Scale bars, 5 mm. (H) Optical micrographs of printed EGaIn lines according to printing velocities. Scale bar, 40 μ m. (I) The plot of line widths versus printing velocities. (J) The plot of line widths versus inner diameters of nozzles. Error bars in (I) and (J) indicate the SD. (Photo credit: Young-Geun Park, Yonsei University). Credit: Science Advances, doi: 10.1126/sciadv.aaw2844



Comparatively, liquid metals such as eutectic gallium-indium alloy (EGaIn) or gallium-indium-tin alloy (Galinstan) are intrinsically stretchable, with low toxicity and minimal volatility for superb <u>electrical</u> conductivity—comparable to solid metals. Direct ink printing with a nozzle can form <u>free-standing 3-D structures</u> at <u>room temperature</u> by stacking droplets of liquid metal upon one another but the resulting resolution is not suited to build electronic devices. In the present work therefore, Park et al. report a high-resolution printing method with liquid metal for its direct reconfiguration into 3-D electrode patterns through a nozzle, under ambient conditions.

In the experimental setup, Park et al. connected a nozzle to an ink reservoir or pressure controller. The materials scientists used EGaIn (75.5 percent gallium and 24.5 percent indium alloy by weight) as the ink and controlled the distance between the nozzle tip and polymer substrate to deliver the ink. Using scanning electron microscopy (SEM), they viewed the EGaIn pattern printed with complex 2-D and 3-D geometries and used the technique to print more diverse patterns such as interconnects of electric circuits with high resolution.





Reconfiguration of EGaIn printed in the work. Credit: Science Advances, doi: 10.1126/sciadv.aaw2844

After directly printing EGaIn through a nozzle, the scientists lifted the nozzle tip for its relocation to the desired position of the substrate to continue printing. The fracture energy of the oxide skin connected the nozzle tip as a "rope" during lift-off. Park et al. measured the maximum velocities for different diameters of filaments to demonstrate different examples and formed 2-D and 3-D features with repeatable reconfiguration. During the process of reconfiguration, the scientists could lift-off a preprinted filament upright from a substrate without fracturing the construct. The observed stable electrodes could withstand electrical load to become increasingly integrated and miniaturized in electric devices. To verify the suitability of EGaIn electrodes as interconnects, Park et al. conducted electrical breakdown tests thereafter.





The electrical contact of direct-printed and reconfigured liquid metals. (A) Schematic illustrations of direct printing (left) and reconfiguration (right). (B) Dependence of total resistance on the length of the channel. Error bars represent the SD. (C) Current-voltage characteristics between Ag pads and direct-printed EGaIn. (D) Current-voltage characteristics between Ag pads and reconfigured EGaIn. (E and F) SEM images of EGaIn on an Ag pad after 7 hours of direct printing. (G and H) SEM images of EGaIn after 7 hours of reconfiguration. Scale bars, 200 µm. Credit: Science Advances, doi: 10.1126/sciadv.aaw2844

When they applied DC or AC bias to monitor electrical breakdown, the temperature too increased in the experimental setup affecting the mechanical stability of the EGaIn 3-D features. The constructs maintained their initial free-standing 3-D structure without structural



collapse at 500[°]C for 30 minutes. After repeated heating and cooling at room temperature, the oxide skin of the 3-D feature slightly wrinkled due to thermal expansion between <u>the oxide shell and EGaIn core</u>. Park et al. tested the electrical contact of direct-printed and reconfigured <u>liquid metals</u> and measured the dependence of total resistance on the length of the printed channel to show that the resistance of EGaIn patterns significantly increased with time under ambient conditions.

As a proof-of-principle of the reconfigurable electronics developed in the present work, Park et al. demonstrated the formation of a reconfigurable antenna with ability to modify its resonance frequency and radiation properties by changing its geometry. For this, the scientists formed a dual coil antenna structure on a glass slide by directly printing EGaIn. During reconfiguration, EGaIn formed a 3-D bridged interconnect, whose resonance frequency the scientists first determined, followed by their use to selectively operate three different light-emitting diodes (LEDs) with red, green and blue light emissions. The reconfigurable, free-standing interconnect maintained its resistance to reliably operate all LEDs at 3V during repeated detachment and connection of multiple reconfiguration steps.





LEFT: 3D reconfiguration of liquid metals for electronics. (A) Schematic illustrations of the reconfigurable antenna. (B) Schematic illustrations of two concentric antennas (top) and the SEM image of the disconnected region (bottom). Scale bar, 300 µm. (C) Schematic illustrations of two concentric antennas that are electrically connected (top) and the SEM image of connected lines by reconfiguration (bottom). Scale bar, 300 µm. (D) Measured scattering parameters of the printed antenna in disconnected and connected states. (E) Schematic illustrations of the reconfiguration process for dynamic switching of LEDs. (F) Colorized SEM image of three LED pixels and EGaIn interconnects. The red, green, blue, and yellow colors correspond to red, green, and blue LEDs and EGaIn, respectively. Scale bar, 1 mm. (G) Photograph of three LED pixels and EGaIn interconnects. Scale bar, 1 mm. (H) Schematic illustrations of reconfiguration and photographs of LED working. Scale bars, 5 mm. (Photo credit: Young-Geun Park, Yonsei University). RIGHT: MicroLED array with 3D liquid metal interconnects. (A) Schematic illustration of the microLED array with reconfigured 3D interconnects. (B) Colorized SEM image of the microLED array and EGaIn interconnects. Blue and yellow colors correspond to microLED and EGaIn, respectively. Scale bar, 300 µm. (C) Colorized SEM image of 3D interconnects. The blue and yellow colors correspond to the microLED and EGaIn, respectively. Scale bar, 300 µm. (D) Photographs of light emission of the microLED array. Scale bars, 1 cm. (E) Current-voltage characteristics of microLED with reconfigured interconnects under flat or bent condition. (Photo credit: Young-Geun Park, Yonsei University). Credit: Science Advances, doi: 10.1126/sciadv.aaw2844

The free-standing 3-D interconnects formed using the process of reconfiguration were advantageous to build cross-geometries in a single XY plane, instead of using multiple layers to thereby prevent unwanted electric contact. For this, Park et al. demonstrated both transverse and longitudinal interconnects of EGaIn for a 4 x 4 array of microLEDs on a flexible polymer film to prevent short circuiting. Using the method, Park et al. minimized the number of interconnections integrated in a miniature device, as the 3-D pattern could efficiently minimize the



number and space of interconnections.

In this way, Young-Geun Park and co-workers demonstrated highresolution 3-D printing using liquid metal and showed its application for stretchable 3-D integrations that are difficult to achieve with conventional engineering. Compared to existing 3-D printing techniques, this method can form fine, free-standing 3-D structures of electrodes with reconfigurable patterns. As an example, Park et al. engineered a reconfigurable antenna capable of modifying its <u>resonance frequency</u> via geometric changes. They also presented reversibly movable 3-D interconnections as mechanical switches that could facilitate higher compact integration in miniaturized devices. The scientists expect the high-resolution 3-D reconfiguration method to offer a promising new additive manufacturing strategy for highly integrated and stretchable next-generation electronic devices.

More information: Young-Geun Park et al. High-resolution, reconfigurable printing of liquid metals with three-dimensional structures, *Science Advances* (2019). DOI: 10.1126/sciadv.aaw2844

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Citation: Printing liquid metals in three-dimensional structures (2019, June 27) retrieved 26 April 2024 from <u>https://phys.org/news/2019-06-liquid-metals-three-dimensional.html</u>



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