ACS APPLIED MATERIALS & INTERFACES
Research Article
www.acsami.org

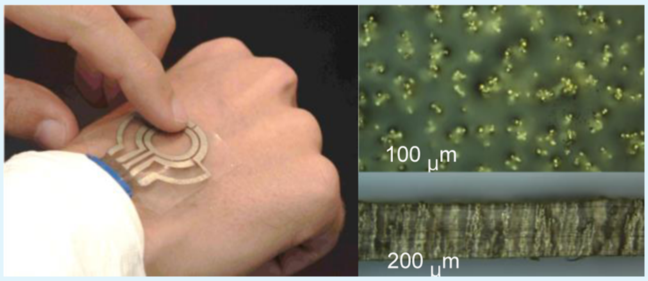
Soft Anisotropic Conductors as Electric Vias for Ga-Based Liquid Metal Circuits

Tong Lu, James Wissman, Ruthika, and Carmel Majidi*

Soft Machines Lab, Department of Mechanical Engineering, Carnegie Mellon University, Pittsburgh, Pennsylvania 15213 United States

Supporting Information

ABSTRACT: We introduce a method for sealing liquid metal (LM) circuits with soft anisotropic conductors that prevent leaking, while simultaneously allowing for electrical contact with skin and surface mounted electronics. These films are composed of polydimethylsiloxane (PDMS) embedded with vertically aligned columns of ferromagnetic Ag–Ni microparticles. The microparticles are magnetically aligned and support electrical conductivity only through the thickness (z-axis) of the elastomer film. Measurements on 10–40% (by wt) composites show moderate volumetric resistivity (as low as ρ = 0.03 Ω/m) through the thickness and no conductivity between adjacent traces. Functionality is demonstrated with several illustrative applications related to tactile sensing and electronics hardware integration.



KEYWORDS: *eutectic gallium–indium, liquid metal, conductive elastomers, anisotropic conductivity, stretchable electronics*

1. INTRODUCTION

Liquid-phase electronics are composed of microfluidic channels of liquid metal (LM) or ionic solution embedded in a soft elastomer.[1−3] Ga-based LM alloys are especially attractive due to their high conductivity, low toxicity, and "moldable" properties.[4−7] Together, these features enable room temperature liquids like eutectic gallium–indium (EGaIn) (75%Ga-25%In, by wt; 15 °C MP) and Galinstan (68%Ga-22%In-10% Sn; −19 °C MP) to be patterned using a broad range of fabrication techniques[8,9] and function as stretchable circuit wiring and sensors for soft wearable technologies.[10−14] In contrast to other approaches to stretchability, liquid-phase electronics need not rely on deterministic architectures based on thin films, wavy circuits, or percolating networks of conductive fibers to avoid failure during extreme deformations.[15−18] Moreover, in the case of EGaIn, pH and/or electric bias can be used to reversibly manipulate the shape, position, and surface composition of a liquid droplet suspended in an ionic medium. This occurs through redox reactions at the LM-medium interface and has been used to tune conductivity for a soft-matter memristor[19] and diode[20] as well as to perform low-voltage electrowetting and microfluidic pumping.[21−25]

Although promising for applications in wearable computing and soft microfluidics, further progress in Ga-based LM electronics depends on robust methods for sealing and electrical interfacing. Sealing is essential to prevent the liquid from leaking, staining skin and clothing, or reacting with other metals through corrosion (e.g., Al) or alloying (e.g., Cu). At present, the terminals of LM circuits are typically accessed by manually inserting rigid wires. While reliable for prototyping and proof-of-concept demonstrations, these connections are not mechanically robust and introduce fluidic-rubbery-rigid boundaries that are susceptible to leaking, changes in electrical contact resistance, or even wire pull-out. Increasing the rigidity of the elastomer near the terminal can reduce the interfacial stress concentration between the polymer and solid wiring to prevent tearing and leaking.[12] However, this method is not applicable to tactile sensing or stretchable circuits that rely on electrical contact with skin or preservation of elastic compliance. Another approach is to seal the terminals with conductive elastomers, such as polydimethylsiloxane (PDMS), filled with carbon (CPDMS) and Ag powder (AgPDMS).[26] When embedded above the terminal points, they function as electrical vias for direct electrical contact with skin or external electronics without altering the integrity or elasticity of the surrounding microfluidic circuit. However, conductive elastomers typically exhibit isotropic conductivity and therefore need to be patterned and aligned to prevent shorting between adjacent circuit terminals. This can be performed with a CO_2 laser engraver, which allows for simultaneous patterning of liquid metal and both conductive and insulating elastomer.[26] However, other fabrication methods, such as stencil lithography, microcontact printing,[27] and additive fabrication,[28] require separate patterning and alignment of the liquid metal circuit and the conductive elastomer vias. While possible, the

Received: August 13, 2015
Accepted: November 16, 2015
Published: November 16, 2015

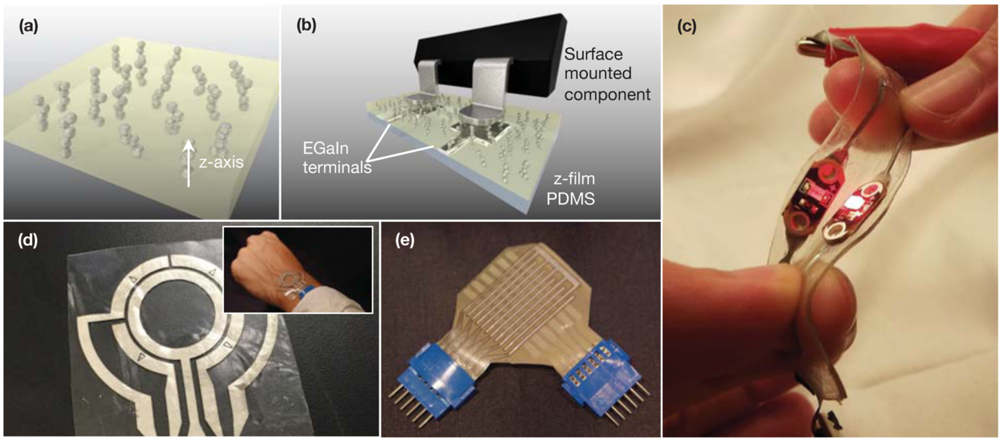


Figure 1. (a) PDMS embedded with an array of vertically aligned columns of Ag−Ni microparticles that support conductivity only through the thickness of the film, that is, along its *z*-axis. (b) Ag−Ni columns function as electrical vias between embedded EGaIn circuit terminals and surface mounted electronics. (c) EGaIn traces attached to LEDs via a commercial *z*-axis conductive film (ECATT 9703; 3M). PDMS/Ag−Ni *z*-axis film ("z-film") functions as a transparent seal and electrical interface for tactile sensing in an (d) arrowpad and (e) 6 × 6 node touchpad.

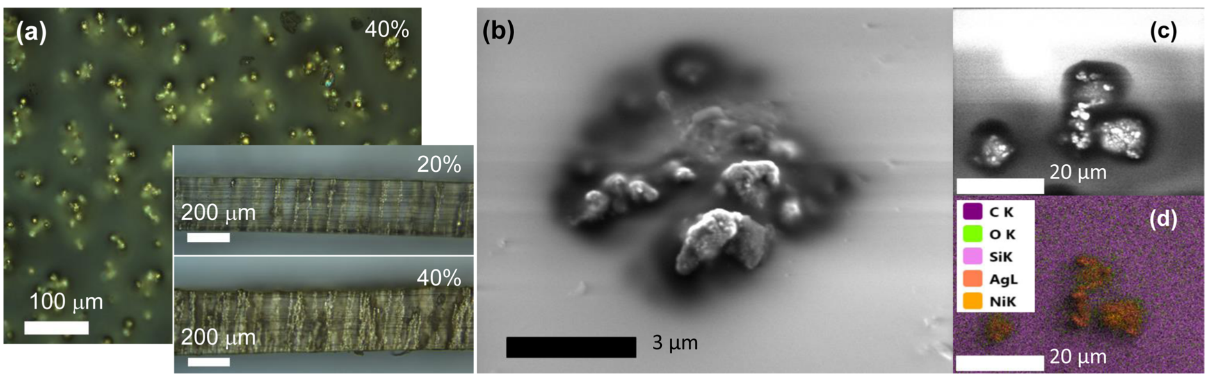


Figure 2. (a)Top down and side views of PDMS film embedded with magnetically aligned columns of 15 μm diameter Ag−Ni microparticles (20 and 40%, by weight). (b) An environmental SEM image of a particle at the surface of the *z*-film. (c, d) An environmental SEM image of the particles at the surface of *z*-film and the corresponding EDS image indicating the distribution of the elements.

additional alignment step requires specialized hardware and remains to be demonstrated.

Here, we introduce a robust and versatile alternative for LM interfacing using an elastomer film that is only conductive through its thickness (*z*-axis) (Figure 1a). An anisotropically conductive "*z*-film" simultaneously prevents leakage while creating electrical vias for accessing LM without the need for alignment. It is composed of polydimethylsiloxane (PDMS) embedded with vertically aligned columns of conductive Ag-coated Ni microparticles. As shown in Figure 1a, the ferromagnetic particles are magnetically aligned to form percolating networks that support electrical conductivity only through the thickness. *Z*-axis anisotropic conductors were first reported in 1988 by researchers at AT&T[29] and have since been used for touch-sensitive screens,[30] tactile shear sensing,[31] solder replacement for surface mount technology (SMT),[32] and electronics packaging.[33,34] In this study, we extend the use of *z*-axis conductors to LM circuits and demonstrate compatibility for illustrative examples presented in the Figure 1 overview. These include surface mounted electronics (Figure 1b and 1c), tactile sensing (Figure 1d and 1e), and joint proprioception, which are described in greater detail in section 2. In all cases, stretchable functionality is preserved by the elasticity and compliance of the soft z-film composite.

2. RESULTS AND DISCUSSION

The *z*-axis conductive films are composed of PDMS (Sylgard 184; 10:1 base-catalyst ratio, by wt.; Dow Corning) embedded with vertically aligned columns of D = 15 μm diameter Ag-coated Ni microparticles (SN15P30:69.5%Ni-30.5%Ag, by wt; Potters Inc.). Optical images (InfiniteFocus, Alicona, Inc.) of the samples are presented in Figure 2a for samples with 40% (top and side views) and 20% (side view only) by weight of Ag−Ni filler. The sample with 40% (f = 0.4) concentration has a thickness of t = 256 μm and an estimated column−column spacing in the range of x = 50−75 μm. Noting that Ag and Ni have χ = 10 times the density of PDMS, the microparticle volume fraction is $v = 1/[1 + \chi(f^{-1} - 1)] = 0.0625$. Assuming hexagonal spacing, each column will contain between $N \approx 20$ and 44 microparticles, where $N = 3\sqrt{3}\,tvx^2/\pi D^3$. This corresponds to staggered columns that are approximately 2−4 particles wide, which is consistent with what is observed in the side view. Since the columns are separated by polymer, no in-plane conductivity is measured. Instead, the capacitance of two coplanar capacitors sealed with z-film were measured to be 1.85 and 1.29 pF with Q factors of 24.7 and 11.4, respectively, indicating that the electrodes were well insulated. Details of the corresponding test are described in section S.1. Lastly, an SEM image of the top of a column (Figure 2b) shows protrusion through the surface and an EDS (energy-dispersive X-ray

DOI: 10.1021/acsami.5b07464
ACS Appl. Mater. Interfaces 2015, 7, 26923−26929

spectroscopy) scan (Figure 2c, d) indicates the abundance of Ag and Ni. Together, these results provide evidence for the presence of Ag–Ni microparticles for forming direct electrical contacts at the surface.

2.1. Conductivity. Figure 3a presents conductivity measurements for z-film composites with a $A = 9$ mm^2 area (3 mm × 3 mm) and $t = 90$ μm thickness. As expected, the z-axis resistance R and volumetric resistivity ($\rho = AR/L$) decrease with increasing mass fraction f. Likewise, for a sample with fixed mass fraction, resistance decreases with increasing area and decreasing thickness. This is demonstrated in Figure 3b for f = 0.4 and samples with dimensions of 1 × 1, 2 × 2, and 3 mm × 3 mm. In the case of a 3 mm × 3 mm sample with a thickness of 69 μm, the electrical resistance is 0.2Ω, comparable to typical values for cm-scale wiring and contacts. Even for samples with a lower concentration of 20 wt %, the electrical resistance is low enough (0.6Ω for $A = 9$ mm^{2}and $t = 90$ μm) to support basic circuit and sensing functionality. The conductivity of z-film with strain and cyclic loading is also measured. Since the Ag–Ni columns are semirigid and do not change size with the deformation of surrounding elastomer, we do not anticipate significant electro-elasto coupling. This is confirmed by the electromechanical measurements presented in section S.2.

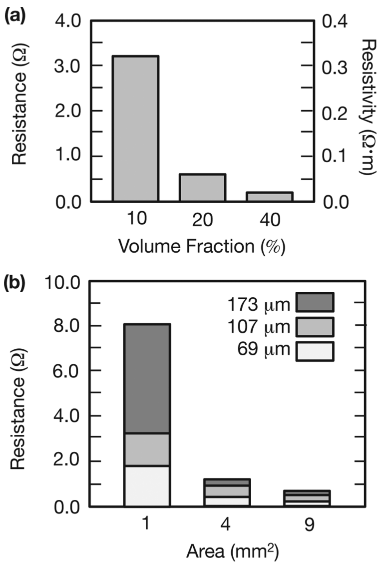


Figure 3. (a) Z-axis resistance of composites for f = 0.1, 0.2, and 0.4; A = 9 mm^2 (3 mm × 3 mm), t = 90 μm thickness. (b) Results for z-film with 40% filler concentration; varying A and t.

Connecting LM circuits to external electronics through ribbon cables is challenging since they typically require mechanical adapters that clip on to the circuit terminals. For liquid-phase soft electronics, the clip will exert stress concentrations that may puncture the seal or otherwise damage the circuit. To avoid this, conductive paper is used to connect the terminal ends of the liquid circuit to the cable adapter, as described in section S.3. The conductive paper is 60 μm thick and composed of Ni/Cu-coated nonwoven polyester fiber (CN-3490; 3M). As with the other materials, it can be patterned with a CO_2 laser and readily bonds to both uncured PDMS and pressure sensitive polyacrylate adhesives (e.g., F-9473PC, VHB 4905, and ECATT 9703).

2.2. Mechanical Durability. The mechanical properties of z-film samples with particle mass fractions of f = 0.2 and 0.4 (three samples for each) are tested using a materials testing system (5969 Instron) and a typical strain–stress curve of sample with f = 0.4 is shown in Figure 4. Samples with both

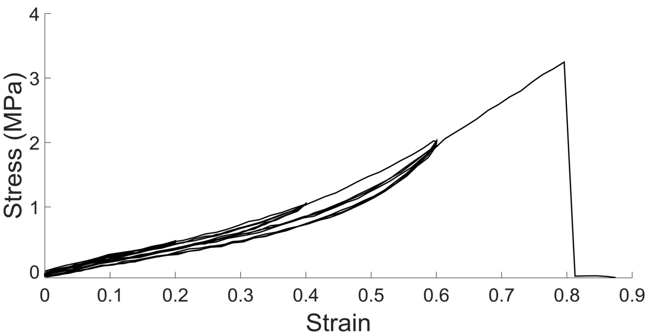


Figure 4. Strain–stress curve of sample with f = 0.4 particle concentration.

contents were cured at 100 °C. Samples with 20 wt % content were stretched 3× to 30%, 60%, and 90% strains in sequence. Samples with 40 wt % content were loaded 3× to strains of 20%, 40%, and 60%. After the nine loading cycles, each sample was stretched to mechanical failure. The average 10%-strain elastic moduli are 1.974 MPa (SD = 0.109 MPa) and 2.596 MPa (SD = 0.172 MPa) for f = 0.2 and 0.4, respectively, which are higher than that of unfilled PDMS cured at 100 °C for 2 h (1.758 MPa). For all samples, a moderate Mullins effect with low subsequent hysteresis was observed and the maximum failure strain was 94% (SD = 5.1%) and 80% (SD = 1.7%) for f = 0.2 and 0.4, respectively.

To assess the durability of the electrical interface, circuits with surface mounted LED chips were fabricated and subject to tensile loading. Two samples were tested, both with a 130-μm-thick PDMS substrate and a 230-μm-thick z-film (f = 0.4) layer embedded with EGaIn traces. LED chips were then mounted above the z-film. One of the samples was coated with an extra encapsulating layer (60 μm) of PDMS to seal the LED contacts. The samples were then fixed on a vertical tensile tester and powered with 2.4 V as they were stretched (Figure 5). In the absence of the extra layer of PDMS, the LED chip remained fully functional until delamination occurred at approximately 30% strain. As delamination developed, the contact between LED and EGaIn was weakened and the circuit completely failed at about 67% strain. For the sample with the PDMS seal, although delamination was observed at a strain of approximately 60%, the circuit remained functional even after elastomer tore below the LED at a maximum strain of approximately 110%.

2.3. Tactile Sensor: LM Circuit with Human Skin. Referring to Figures 1d and 6a, a soft-matter tactile sensors is produced by sealing a laser-patterned film of EGaIn between layers of insulating PDMS and PDMS-based z-film (f = 0.2). An image showing the quality of the line cut with a CO_2 laser (VLS 3.50; Universal Laser Systems) is shown in section S.4. The combination of low elastic modulus and <400 μm thickness allows the tactile sensor to conform to the skin without introducing mechanical constraints or causing discomfort. The sensor contains five leads that are connected to an external microcontroller (Arduino Uno) for recording fingertip-activated changes in voltage drop between the sensor electrodes. The leads interface through a ribbon cable and adapter. The sensor is composed of a central electrode that is

DOI: 10.1021/acsami.5b07464
ACS Appl. Mater. Interfaces 2015, 7, 26923–26929

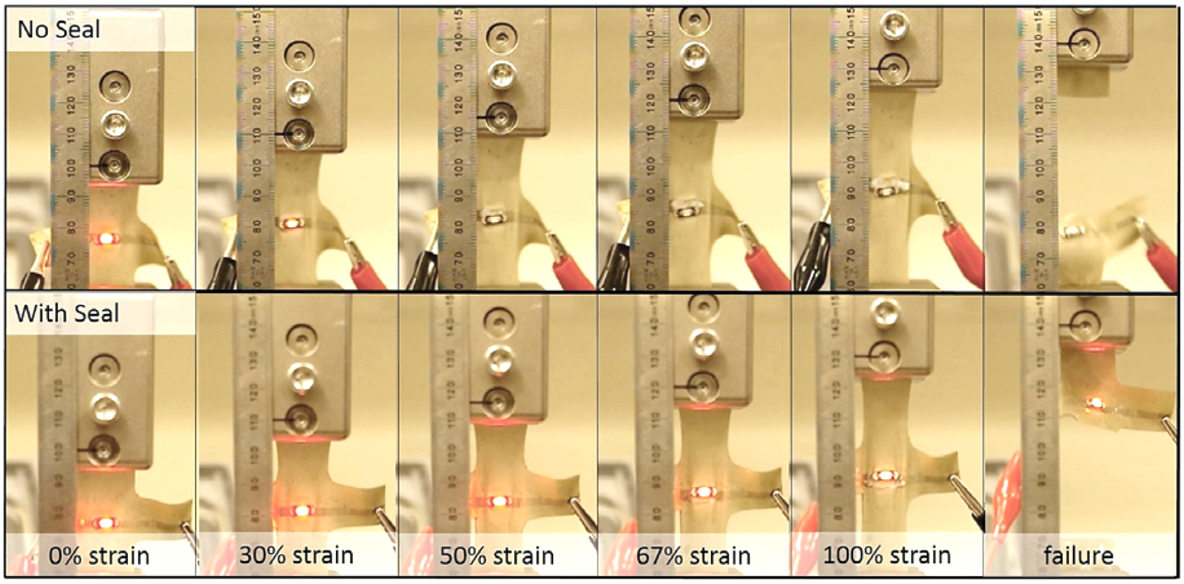


Figure 5. Tensile test of samples with a surface mounted LED.

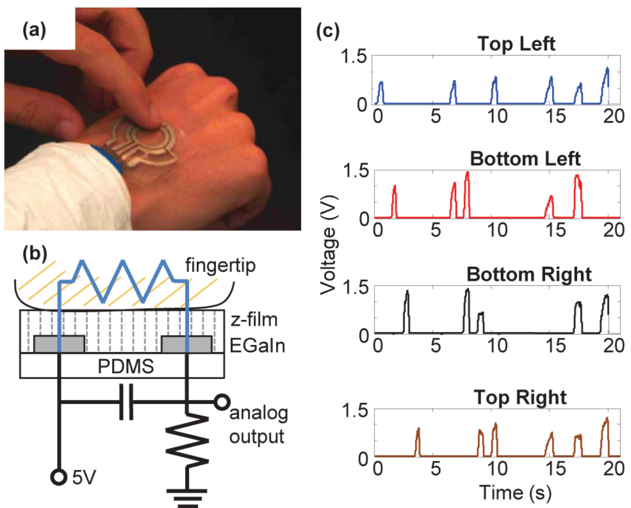


Figure 6. (a) Wearable touchpad with *z*-film seal to allow for direct electrical contact with skin. (b) Schematic of voltage divider architecture for tactile sensing. (c) Voltage output on each electrode for user test with 5 V input and 10MΩ resistor.

connected to 5 V input and four outer electrodes that are electrically grounded through a R = 10 MΩ resistors (Figure 6b). The 470 pF capacitor in the diagram is used to filter out high frequency noise. When the fingertip makes simultaneous contact with the central and outer electrode, the outer electrode charges and the voltage rapidly increases above 0 V. Fingertip contact can also be detected by monitoring the electrical conductivity between the central and outer electrodes (Figure 6b). In the absence of fingertip contact, there is no measurable conductivity between the electrodes. When a finger is lightly pressed against a sensing node, a ∼1 MΩ resistance is measured. This resistance is dominated by the resistivity of the skin and its contact resistance with finger contact the z-film. Measurements performed with the tactile sensor are presented in Figure 6c. As shown by the plots, the voltage output on each electrode rises when the electrode is connected to the input trace via the fingertip. With different placements of the finger, the electrodes can be activated either individually or simultaneously.

An example of a multilayer tactile sensor is presented in Figure 1e and a similar sensor is demonstrated in the Supporting Information (section S.5). In this implementation, rows and columns of EGaIn electrodes are embedded inside a *z*-film matrix (f = 0.4). There is a thin strip of insulating PDMS at the bottom of each electrode in the top layer that prevents direct electrical shorting. Each column and row pair become electrically connected to a voltage input by touching the surface with fingertip and forming electrical vias beween the electrodes and the voltage input. As with the other sensor, the multilayer

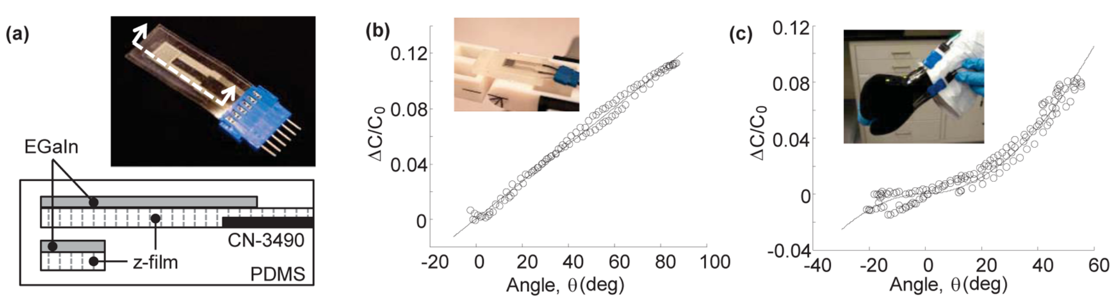


Figure 7. (a) Capacitive strain sensor for measuring joint motion. EGaIn electrodes embedded in PDMS are connected to a ribbon cable via *z*-film and CN-3490 conductive paper. Plots show sensor output for bending on a (b) mechanical joint and (c) wrist.

DOI: 10.1021/acsami.5b07464
ACS Appl. Mater. Interfaces 2015, 7, 26923−26929

sensor shows satisfactory voltage change in each channel as a response to the interaction with human skin.

Tactile sensing is also implemented with commercial acrylate elastomers, with details presented in section S.6. As shown in Figure S6a, the EGaIn circuit is embedded within layers of adhesive acrylate elastomer (F-9473PC and 4905 VHB; 3M) and *z*-axis electrically conductive adhesive transfer tape (ECATT 9703; 3M). The arrowpad is used to operate a Tetris gaming script in Python (Figures S1b, c). Referring to Figure S6d, the circuit is produced through a combination of CO_2 laser patterning and stencil lithography. In contrast to PDMS-based EGaIn circuits, fabrication with commercial acrylates eliminates the need for time-consuming and labor intensive steps with elastomer preparation and curing.

2.4. Wrist Motion Sensor: Interfacinig LM Circuit with External Wiring. Another example of robust electrical interfacing with a soft LM circuit element is the joint motion sensor presented in Figure 7a. The sensor is composed of two overlapping films of EGaIn that function as capacitive electrodes insulated by a thin layer of PDMS. When placed on a wrist, the joint motion causes the electrodes and dielectric to deform and change capacitance. As in the tactile sensor, *z*-film and conductive paper are used to connect the LM circuit element to off-board electronic hardware through a ribbon cable. An LCR meter is used to measure the relative change in capacitance $\Delta C/C_0$ in response to changes in bending angle θ.

When undeformed, the circuit has a capacitance of $C_0 \approx 7$ pF. When placed on a mechanical joint (Figure 7b) the capacitance changes linearly with θ, with an R^2 value of 0.99 and bending-mode gauge factor GF = $\{\Delta C/C_0\}/\theta(\text{rad})$ of 0.077. When placed above the wrist (Figure 7c), the response is nonlinear: $\{\Delta C/C_0\} = \alpha\theta(\text{rad})^2\{\text{sgn}(\theta)\}$, where $\alpha = 1.67 \times 10^{-3}$ and $R^2 = 0.95$. Although the internal contact resistance does not factor into the capacitive measurements, the reliability of these results (smooth, monotonic, low hysteresis) demonstrates the ability of the interface to support bend sensing applications without electrical failure.

3. EXPERIMENTAL SECTION

Z-axis anisotropic conductors are produced by first mixing PDMS (Sylgard 184, 10:1 base-to-catalyst ratio; Dow Corning) with 15 μm diameter Ag-coated Ni microparticles (69.5%Ni−30.5%Ag, by wt; SN15P30, Potters Inc.). Next, the uncured polymer/particle mixture is deposited on a substrate by spin coating (Model KW-4A; SPI Supplies) or with a thin-film applicator (ZUA 2000 Universal Applicator; Zehntner GmbH). Prior to deposition of the mixture, the substrate may be covered with layers of conductive paper (CN-3490; 3M) and insulating PDMS that will bond to the z-film after curing. For mixtures with 40% Ag−Ni powder (by wt), spin coating at 600, 1200, and 1800 rpm for 10s results in the following film thicknesses: 173, 107, and 69 μm, respectively. The sample is then placed on top of a flat magnet (2″ × 2″ × 1/4″ NdFeB; K&J Magnetics, Inc.) and cured in an oven at 100 °C for 25 min. The magnetic field aligns the particles as the elastomer cures, as has been previously discussed in the literature.[35,36] During or prior to curing, surface mounted electronics may be deposited on the *z*-film.

The tactile sensor is produced using the laser patterning techniques previously introduced in ref 26 and summarized in Figure S3. First, the cutting substrate is prepared by covering one edge with CN-3490. Next, z-film is deposited and cured using the steps described in the previous paragraph. EGaIn is then deposited on the cured z-film using a roller (e.g., a PDMS cylinder or lint roller) followed by a layer of insulating PDMS applied with spin coating or a thin-film applicator. After the PDMS seal is partially cured on a hot plate or in a curing oven (at 100 °C for 5 min), the sample is patterned with a CO_2 laser (VLS 3.50; Universal Laser Systems), as illustrated in Figure S3a. After the excess material is removed (Figure S3b), a second layer of insulating PDMS is deposited and cured (Figure S3c). Lastly, the patterned CN-3490 traces are clipped into a ribbon cable adapter for external wiring (Figure S3d).

Anisotropic electrical conductivity is measured by inserting the probes of a multimeter (34401A; Agilent Technologies) into EGaIn droplets deposited above masked portions of the z-film (see Figure S2-1). Serial resistance is accounted for by measuring total resistance with and without the z-PDMS film. The values presented in Figure 3 are only estimates since they include both the volumetric resistivity of the bulk as well as the contact resistance between the z-film and EGaIn. For tactile sensing, voltage measurements are obtained with an Arduino Uno microcontroller. Changes in the capacitance of the wrist motion sensor are measured with an LCR meter (889B; BK Precision).

4. CONCLUSION

Z-axis anisotropic conductors (z-film) are a versatile material for electrically interfacing Ga-based liquid metal (LM) circuits with human skin and external electronics. Previous studies of z-film have focused on solid-state electronics, packaging, and other applications that take advantage of their optical transparency and bonding properties. Here, we show that z-film elastomers are also well suited as electric vias for LM-based circuits since they seal in liquid without introducing elastic rigidity or electrical shorts within the plane of the circuit. This work improves upon previous efforts with EGaIn-based microfluidics by eliminating the need for manual wire insertion. Wire insertion is not only labor intensive and prone to error, but also introduces mechanical mismatches, stress concentrations, and nonbonded interfaces that can result in elastomer rupture, wire slip/pull-out, and LM leakage. Alternative interfacing methods based on isotropically conductive elastomers[26] and rigidity graded materials[12] address these issues but are challenging to implement because of their dependency on precise alignment and specialized materials and fabrication methods. In contrast, a single layer of z-film can be applied to the entire surface of the LM circuit without any additional alignment step.

Since the use of z-film in conventional electronics is already well-established, the purpose of this work is to demonstrate their reliability for forming interfaces with LM alloys. We have not investigated the principles of ferromagnetic microparticle alignment as this has already been addressed in the literature.[11,35] In summary, the *z*-axis column formation is controlled by a magneto-rheological response to external magnetic field. Once the surrounding elastomer has cured, the particles are constrained to their relative configuration and the field can be removed. The length and spacing of the columns is controlled by the composition (i.e., wt % of microparticles) and field intensity. For example, as previously shown, we find that the length of the columns of ferromagnetic microparticles scales with the intensity of the magnetic field. If the field is too weak, the column length will be less than the film thickness and *z*-axis conductivity will not be achieved. If the field is too strong, the microparticles will break the surface tension of the uncured elastomer and the column length will exceed the thickness of the film and form dendrite-like structures.

While successfully implemented with the help of *z*-axis condcutive film, the tactile sensing devices demonstrated in this work still need to be further studied to clarify their tolerances and resistance against environmental factors such as humidity,

DOI: 10.1021/acsami.5b07464
ACS Appl. Mater. Interfaces 2015, 7, 26923−26929

sweat, and oil, etc. User-based studies, by which devices are tested by human participants, should be performed in the future to assess input accuracy for prescribed data entry tasks.[37] Lastly, As with other LM circuits, we observe mechanical and electrical failures when a sample is stretched to beyond the strain limit of the elastomer sealing. In the case of surface mounted electronics, poor bonding between the rigid circuit and *z*-axis conductor will lead to delamination and eventual mechanical failure. This is caused by stress concentrations that initiate at the edges of the interface due to elastic mismatch between the soft elastomer and rigid electronic component. As external loading increases, a mode II crack forms and propagates toward the center of the interface. Further studies are required to characterize the bond strength between *z*-axis adhesives and rigid or semirigid circuit elements. Nonetheless, the current study shows that under tensile strains of up to 50%, the bonding is adequate for supporting electronic functionality.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsami.5b07464.

Experimental methods for fabrication and testing (PDF)
Video showing sensor use and flexibility (MPG)

■ AUTHOR INFORMATION

Corresponding Author

*Phone: +1 (412) 268 2492. E-mail: cmajidi@andrew.cmu.edu.

Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

The authors thank the Office of Naval Research (Bio-Inspired Autonomous Systems; Program Officer Dr. Tom McKenna; Grant #N000141210614) and Air Force Office of Scientific Research (Mechanics of Multifunctional Materials and Microsystems; Dr. Les Lee; FA9550-13-1-0123) for supporting this research.

■ REFERENCES

(1) Dickey, M. D. Emerging Applications of Liquid Metals Featuring Surface Oxides. *ACS Appl. Mater. Interfaces* **2014**, *6*, 18369−18379.

(2) Cheng, S.; Wu, Z. Microfluidic Electronics. *Lab Chip* **2012**, *12*, 2782−2791.

(3) Hammock, M. L.; Chortos, A.; Tee, B. C.-K.; Tok, J. B.-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*, 5997−6038.

(4) Chiechi, R. C.; Weiss, E. A.; Dickey, M. D.; Whitesides, G. M. Eutectic Gallium-Indium (EGaIn): A Moldable Liquid Metal for Electrical Characterization of Self-Assembled Monolayers. *Angew. Chem., Int. Ed.* **2008**, *47*, 142−144.

(5) Dickey, M. D.; Chiechi, R. C.; Larsen, R. J.; Weiss, E. A.; Weitz, D. A.; Whitesides, G. M. Eutectic Gallium-Indium (EGaIn): A Liquid Metal Alloy for the Formation of Stable Structures in Microchannels at Room Temperature. *Adv. Funct. Mater.* **2008**, *18*, 1097−1104.

(6) Liu, T.; Sen, P.; Kim, C.-J. Characterization of Nontoxic Liquid-Metal Alloy Galinstan for Applications in Microdevices. *J. Microelectromech. Syst.* **2012**, *21*, 443−450.

(7) Kim, D.; Thissen, P.; Viner, G.; Lee, D.-W.; Choi, W.; Chabal, Y. J.; Lee, J.-B. J. Recovery of Nonwetting Characteristics by Surface Modification of Gallium-Based Liquid Metal Droplets Using Hydrochloric Acid Vapor. *ACS Appl. Mater. Interfaces* **2013**, *5*, 179−185.

(8) Joshipura, I. D.; Ayers, H. R.; Majidi, C.; Dickey, M. D. Methods to Pattern Liquid Metals. *J. Mater. Chem. C* **2015**, *3*, 3834−3841.

(9) Doudrick, K.; Liu, S.; Mutunga, E. M.; Klein, K. L.; Damle, V.; Varanasi, K. K.; Rykaczewski, K. Different Shades of Oxide: From Nanoscale Wetting Mechanisms to Contact Printing of Gallium-Based Liquid Metals. *Langmuir* **2014**, *30*, 6867−6877.

(10) Kramer, R. K.; Majidi, C.; Wood, R. J. Wearable Tactile Keypad with Stretchable Artificial Skin. *Proc. IEEE Int. Conf. Rob. Autom.* **2011**, 1103−1107.

(11) Ponce Wong, R. D.; Posner, J. D.; Santos, V. J. Flexible Microfluidic Normal Force Sensor Skin for Tactile Feedback. *Sens. Actuators, A* **2012**, *179*, 62−69.

(12) Mengüç, Y.; Park, Y.-L.; Pei, H.; Vogt, D.; Aubin, P. M.; Winchell, E.; Fluke, L.; Stirling, L.; Wood, R. J.; Walsh, C. J. Wearable Soft Sensing Suit for Human Gait Measurement. *Int. J. Rob. Res.* **2014**, *33*, 1748−1764.

(13) Park, Y.-L.; Chen, B.-R.; Wood, R. J. Design and Fabrication of Soft Artificial Skin Using Embedded Microchannels and Liquid Conductors. *IEEE Sens. J.* **2012**, *12*, 2711−2718.

(14) Park, Y.-L.; Majidi, C.; Kramer, R.; Berad, P.; Wood, R. J. Hyperelastic Pressure Sensing with a Liquid-embedded Elastomer. *J. Micromech. Microeng.* **2010**, *20*, 125029.

(15) Takahashi, T.; Takei, K.; Gillies, A. G.; Fearing, R. S.; Javey, A. Carbon Nanotube Active-Matrix Backplanes for Conformal Electronics and Sensors. *Nano Lett.* **2011**, *11*, 5408−5413.

(16) Kaltenbrunner, M.; Sekitani, T.; Reeder, J.; Yokota, T.; Kuribara, K.; Tokuhara, T.; Drack, M.; Schwödiauer, R.; Graz, I.; Bauer-Gogonea, S.; Bauer, S.; Someya, T. An Ultra-Lightweight Design for Imperceptible Plastic Electronics. *Nature* **2013**, *499*, 458−463.

(17) Lipomi, D. J.; Vosgueritchian, M.; Tee, B. C.-K.; Hellstrom, S. L.; Lee, J. A.; Fox, C. H.; Bao, Z. Skin-Like Pressure and Strain Sensors based on Transparent Elastic Films of Carbon Nanotubes. *Nat. Nanotechnol.* **2011**, *6*, 788−792.

(18) Martinez, V.; Stauffer, F.; Adagunodo, M. O.; Forro, C.; Vörös, J.; Larmagnac, A. Stretchable Silver Nanowire-Elastomer Composite Microelectrodes with Tailored Electrical Properties. *ACS Appl. Mater. Interfaces* **2015**, *7*, 13467−13475.

(19) Koo, H.-J.; So, J.-H.; Dickey, M. D.; Velev, O. D. Towards All-Soft Matter Circuits: Prototypes of Quasi-Liquid Devices with Memristor Characteristics. *Adv. Mater.* **2011**, *23*, 3559−3564.

(20) So, J.-H.; Koo, H.-J.; Dickey, M. D.; Velev, O. D. Ionic Current Rectification in Soft-Matter Diodes with Liquid-Metal Electrodes. *Adv. Funct. Mater.* **2012**, *22*, 625−631.

(21) Tang, S.-Y.; Sivan, V.; Khoshmanesh, K.; O'Mullane, A. P.; Tang, X.; Gol, B.; Eshtiaghi, N.; Lieder, F.; Petersen, P.; Mitchell, A.; Kalantar-zadeh, K. Electrochemically Induced Actuation of Liquid Metal Marbles. *Nanoscale* **2013**, *5*, 5949−5957.

(22) Tang, S.-Y.; Khoshmanesh, K.; Sivan, V.; Petersen, P.; O'Mullane, A. P.; Abbott, D.; Mitchell, A.; Kalantar-zadeh, K. Liquid Metal Enabled Pump. *Proc. Natl. Acad. Sci. U. S. A.* **2014**, *111*, 3304−3309.

(23) Khan, M. R.; Eaker, C. B.; Bowden, E. F.; Dickey, M. D. Giant and Switchable Surface Activity of Liquid Metal via Surface Oxidation. *Proc. Natl. Acad. Sci. U. S. A.* **2014**, *111*, 14047−14051.

(24) Khan, M. R.; Trlica, C.; So, J.-H.; Valeri, M.; Dickey, M. D. Influence of Water on the Interfacial Behavior of Gallium Liquid Metal Alloys. *ACS Appl. Mater. Interfaces* **2014**, *6*, 22467−22473.

(25) Gough, R. C.; Morishita, A. M.; Dang, J. H.; Moorefield, M. R.; Shiroma, W. A.; Ohta, A. T. Rapid Electrocapillary Deformation of Liquid Metal with Reversible Shape Retention. *Micro Nano Syst. Lett.* **2015**, *3*, 1−9.

(26) Lu, T.; Finkenauer, L.; Wissman, J.; Majidi, C. Rapid Prototyping for Soft-Matter Electronics. *Adv. Funct. Mater.* **2014**, *24*, 3351−3356.

(27) Tabatabai, A.; Fassler, A.; Usiak, C.; Majidi, C. Liquid-Phase Gallium-Indium Alloy Electronics with Microcontact Printing. *Langmuir* **2013**, *29*, 6194−6200.

DOI: 10.1021/acsami.5b07464
ACS Appl. Mater. Interfaces 2015, 7, 26923−26929

(28) Ladd, S.-J.-H.; Collin; Muth, J.; Dickey, M. D. Liquid-Phase Gallium-Indium Alloy Electronics with Microcontact Printing. *Adv. Mater.* **2013**, *25*, 5081−5085.
(29) Jin, S.; Sherwood, R. C.; Mottine, J. J.; Tiefel, T. H.; Opila, R. L.; Fulton, J. A. New, Z-Direction Anisotropically Conductive Composites. *J. Appl. Phys.* **1988**, *64*, 6008−6010.
(30) Jin, S.; Tiefel, T. H.; Wolfe, R.; Sherwood, R. C.; Mottine, J. J. Optically Transparent, Electrically Conductive Composite Medium. *Science* **1992**, *255*, 446−448.
(31) Chen, L. H.; Jin, S.; Tiefel, T. H. Tactile Shear Sensing using Anisotropically Conductive Polymer. *Appl. Phys. Lett.* **1993**, *62*, 2440−2442.
(32) Sancaktar, E.; Dilsiz, N. Anisotropic Alignment of Nickel Particles in a Magnetic Field for Electronically Conductive Adhesives Applications. *J. Adhes. Sci. Technol.* **1997**, *11*, 155−166.
(33) Breval, E.; Klimkiewicz, M.; Shi, Y. T.; Arakaki, D.; Dougherty, J. P. Magnetic Alignment of Particles in Composite Films. *J. Mater. Sci.* **2003**, *38*, 1347−1351.
(34) Ramkumar, S. M.; Srihari, K. A Novel Anisotropic Conductive Adhesive for Lead-Free Surface Mount Electronics Packaging. *J. Electron. Packag.* **2007**, *129*, 149−156.
(35) Filipcsei, G.; Csetneki, I.; Szilágyi, A.; Zrínyi, M. *Oligomers−Polymer Composites−Molecular Imprinting*; Springer: Berlin, 2007; Vol. *206*; pp 137−189.
(36) Li, Y.; Lu, D.; Wong, C. P. *Electrical Conductive Adhesives with Nanotechnologies*; Springer US: Boston, MA, 2010; Chapter 5, pp 227−278.
(37) Weigel, M.; Lu, T.; Bailly, G.; Oulasvirta, A.; Majidi, C.; Steimle, J. iSkin: Flexible, Stretchable and Visually Customizable On-Body Touch Sensors for Mobile Computing. *ACM Conf. Hum. Factors Comput. Syst.* **2015**, 2991−3000.

DOI: 10.1021/acsami.5b07464
ACS Appl. Mater. Interfaces 2015, 7, 26923−26929