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Directed Assembly of Liquid Metal-Elastomer Conductors for Stretchable and Self-healing Electronics

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Abstract

Due to the growing interest in soft robotics, stretchable electronics, and electronic skins, there is demand for compliant electrodes and interconnects that are soft, stretchable and conductive. Here, we use dielectrophoresis (DEP) to assemble, align, and sinter microdroplets of liquid metal—eutectic Ga-In (EGaIn)—in uncured polydimethylsiloxane (PDMS) to form electrically conducting microwires. There are several noteworthy aspects of this approach: (1) Generally, liquid metal droplets in silicone at loadings approaching 90 wt% remain insulating and form conductive network only when subjected to sintering. Here the use of DEP facilitates assembly of the filler droplets into conductive microwires at loadings as low as 10 wt% EGaIn. (2) Dielectrophoresis is done in silicone for the first time, enabling the microwires to be cured in a stretchable matrix. (3) Because the droplets are liquid, they sinter during dielectrophoresis to form a stretchable metallic microwire that retains its shape after curing the silicone and does not change resistance during mechanical strain. (4) The use of liquid metal eliminates the issue of compliance mismatch observed in soft polymers with solid fillers. (5) The silicone-EGaIn “ink” can be placed in holes created by severely damaged regions of stretchable wires to create stretchable interconnects that heal the damage both mechanically and electrically. We characterize the DEP process of this unique set of materials and demonstrate the interesting attributes enabled by such liquid microwires.

1. Introduction

Relative to conventional rigid electronic devices, soft and stretchable electronics transform how we utilize and fabricate electronics, paving the way for various futuristic applications such as conformable devices, electronic skins and soft robotics^[1-7]. The ability to minimize loss in electronic function under large mechanical strain is essential to the progress of soft and stretchable devices^[8, 9]; for example, “on-skin” electronics require uninterrupted electrical conduction while stretching to remain functional while conforming to the skin^[2, 3, 5]. Approaches to make stretchable conductors have included the patterning of metals such as gold in serpentine electrode architectures as well as by dispersing rigid conductive particles such as metallic or carbon-based fillers in elastomeric matrix ^[9-17]. Unfortunately, such approaches have limited stretchability (32% for serpentine electrodes), electrical resistance typically increases with mechanical strain (R/R_0 can increase by 10 at 30% strain), and elastomeric properties are often compromised with higher wt% conductive fillers (Young’s modulus increase of 50 MPa)^[8, 9, 14, 15, 18-21].

These drawbacks have led researchers to explore the use of liquid metals (LMs) as conductive fillers to fabricate electrically conductive composites^[22-25]. Since LMs like EGaIn (eutectic gallium indium, m.p. 15.7 C) are liquid at room temperature^[26], they have negligible mechanical resistance to strain which helps preserve the elasticity of the elastomer matrix^[27]. Furthermore, mixing LM in a polymer matrix could be achieved with relative ease by shear mixing (i.e. stirring)^[28]. However, these composites tend to require very high loading of LM, typically greater than 85 wt%^[22, 23, 25, 29], which is cost-ineffective and can lead to metal leaking at such high loadings. Significantly, even with such high loading of LM, the composites intrinsically remain electrically insulating and require external stimuli such as pressure in the case of mechanical sintering^[22, 23] or laser irradiation^[30] to form conductive pathways. In principle, liquid metal elastomers with high loading could inadvertently sinter during handling, causing regions of the composite that should be insulating to become conductive.

Thus, there is a need to develop a process that can modulate the placement and concentration of individual EGaIn droplets within an elastomeric matrix. This in turn, could reduce the concentration of fillers required to form conductive pathways and avoid the need for separate sintering steps.

Here, we use dielectrophoresis (DEP) to align, assemble (increase the local density), and sinter particles into conductive wires with micro-scale diameters. DEP is easy to control because of its field-driven nature^[31]. Additionally, it enables control over the location and direction of conductive pathways in the matrix^[32] while keeping other regions of the composite electrically insulating, which produces electrical anisotropy. Traditionally, DEP has been utilized to align solid conductive particles (gold nanoparticles^[31-33] and carbonaceous nanoparticles^[34, 35]) inside dielectric fluids to form conductive microwires. However, DEP has not been used to assemble stretchable and flexible conductive wires within polymeric matrices, nor has it been done in an elastomeric matrix that can be cured *in situ* during alignment. As demonstrated herein, the use of liquid metal particles results in several advantages relative to conventional conductive particles.

Herein, we report the fabrication of flexible and stretchable microwires through DEP-assisted alignment of EGaIn microdroplets inside an elastomeric PDMS (polydimethylsiloxane) matrix. We used alternating current to avoid the effects of electroosmosis and electrolysis (caused by direct current)^[31, 32, 36]. Upon crosslinking of the PDMS matrix post-alignment, the EGaIn microwires retain their conductivity and do not require additional post-processing (such as mechanical sintering). The EGaIn microwires outmatch solid fillers, such as Ag flakes, which fail to maintain conductive paths when aligned and cured in PDMS. This DEP approach results in low resistance values ($3 - 8 \Omega$) of the EGaIn microwires, achieved at significantly lower EGaIn filler loading (10 wt%) than prior composite work (>85 wt%)^[22, 23]. Furthermore, the composite remains conductive until the elastomeric matrix ruptures ($\sim 100\%$) (a strain at failure similar to that of pristine PDMS^[37]), as the inherent fluidity of EGaIn eliminates compliance

mismatch and maintains metallic conductivity during deformation^[27]. Although liquid metal wires often change resistance during elongation due to geometric changes, the resistance values of the wires reported here do not change when stretched up to 100% strain and bent at different bending radii for hundreds of strain cycles due to the meandering shape. Additionally, microwires can be reformed *in situ* to electrically reconnect two conductors and thereby heal severe mechanical damage, recovering 74% of the original conductance. DEP-assisted alignment of EGaIn microdroplets inside a polymer matrix also unlocks the potential for creating on-demand circuits in soft matrix.

2. Results and Discussion

2.1 Dielectrophoresis (DEP) and Design Principles

In non-uniform electric fields, particles suspended in a dielectric medium get polarized and experience a force, and this phenomenon is referred to as dielectrophoresis (DEP)^[38-40]. The resultant DEP force (Figure S1, Supporting Information) depends on the position of these particles in the electric field and the relative polarizabilities of particle and medium^[36, 38, 41]. When the particles are more polarizable than the dielectric medium, they move towards regions of higher electric field gradient, termed as positive DEP (pDEP)^[36, 38]. Within metallic particles, electrons move freely and charges can readily accumulate at the dielectric-metal interfaces upon applying an E-field. Hence, metal particles are infinitely more “polarizable” than the silicone pre-polymer matrix, leading to pDEP phenomenon^[36]. In addition to acting as a dielectric medium during the alignment of EGaIn microdroplets, the silicone can be crosslinked after alignment of the particles to secure and encapsulate the wires.

Figure 1a presents the device for studying DEP assembly of electrically conducting micro-interconnects. Au/Cu electrodes were deposited on a glass slide for studying the conductive network formation (and physical characterization). After these initial experiments, we replaced the rigid glass substrates with elastomeric substrates to demonstrate flexible and stretchable interconnects. To create microwires, EGaIn-PDMS mixtures are placed across the electrodes,

defined by a laser-cut acrylic (3M VHB 4905) spacer to confine the mixture. To confirm the presence of pDEP and visualise the alignment pattern, EGaIn droplets dispersed PDMS were subjected to an electric field (Figure 1b). The EGaIn droplets accumulate at the edges of the electrodes where the E-field gradient is largest and trace out the electric field lines. These observations are in agreement with the definition of pDEP and previous works on metallic particles^[31, 36, 42].

Figure 1. Dielectrophoretic (DEP) assembly of liquid metal (EGaIn) microwires. (a) Schematic of DEP-assisted assembly of EGaIn microwires inside PDMS matrix. Electrodes were deposited on a glass substrate and laser-cut acrylic spacers were used to contain the EGaIn-PDMS mixture and define its volume. Upon application of electric field, EGaIn droplets align along the direction of the field lines and join to form a conductive pathway. After the alignment, the EGaIn droplets stay in place and the conductive pathway is retained. (b) Optical microscope images of EGaIn nanodroplets in PDMS (uncured) tracing out the electric field. EGaIn nanodroplets accumulate at the edges of the electrodes where the E-field gradient is largest. Scalebar - 500 μm .

Similar to other dielectric and metallic particles in DEP experiments, where induced motion of the particles depends on variables like concentration, voltage and frequency, we established

general relations between these parameters and alignment process^[31, 33, 36, 43-45]. Formation of an electrically continuous wire between the electrodes was characterized by a sudden increase in current (measured between the electrodes). In addition to measuring resistance, the process was continuously observed under a microscope. We only considered conditions successful that formed conductive wires within minutes (<5 min) to maintain the practicality of this approach. Increasing the EGaIn concentration (still well below the isotropic percolation threshold), led to reduction of time required for formation of conductive pathway. (Figure S2, Supporting Information). At 10 wt% and 15 wt%, the microwires were able to form within minutes. However, at 5 wt%, wires did not always form as the EGaIn droplets are too dispersed. As expected, increasing the electric field strength (by increasing the applied voltage) across the electrodes led to significant decrease in the time required to form conductive pathways (Figure S3, Supporting Information). Furthermore, conductive pathways formed at a wide range of frequencies (Figure S4, Supporting Information), with the time being minimized for high frequencies (1 kHz & 10 kHz)^[38, 46]. It is worthwhile to note that similar to other dielectrophoretic experiments reported in the literature, the EGaIn particles were not able to align at very high frequencies (100 kHz)^[43]. Additional details about optimized DEP parameters can be found in the SI. We found that 10 wt% loading, 250 V_{pp} applied voltage and 1 kHz frequency with electrode gaps of 1 mm created wires reliably and is therefore used henceforth.

2.2 Formation of conductive pathways for solid and liquid particles

Conventionally, DEP has been demonstrated with solid particles, hence, it is insightful to compare the alignment process of liquid EGaIn particles against solid particles and the conductance of their resulting composites. It has been previously demonstrated that shear mixing of EGaIn into PDMS results in composites with microdroplets (10 μm – 30 μm) of EGaIn^[28]. Hence, solid silver (Ag) microparticles of similar size distribution was utilized to prepare the composite (10 wt% loading) and thoroughly dispersed in PDMS by planetary mixing (**Figure 2 a**). An EGaIn-PDMS mixture with 10 wt% loading of EGaIn was also

prepared by planetary mixing (Figure 2 b). Particle size analysis reveals that most of the Ag and EGaIn particles have diameters around 50 μm (Figure 2 a-b & Figure S5, Supporting Information), although we note the agglomeration of Ag particles and many smaller EGaIn microdroplets (0 – 10 μm). The software (ImageJ) identifies agglomerated particles as one larger particle. Alignment process for both Ag and EGaIn particles showed pDEP response (as expected), captured by microscope images at regular time intervals (Figure S6 & Figure S7, Supporting Information). They move towards areas with greater E-field gradient (corners of the electrodes). As chains of microdroplets formed at both electrodes, local E-field of high intensity and gradient developed at the tips of the growing chains, thereby attracting more EGaIn microdroplets to the tips of the chain and facilitating the growth of the microwire (Movie 1 & Movie 2, Supporting Information).

Planetary mixing produces EGaIn microdroplets with highly irregular shapes and smaller size accompanied with significant variation in size (Figure 2 b). Hence, to produce more uniform EGaIn microdroplets of larger size, we used the method of flow focusing (FF) (Figure 2 c). Flow focusing^[47] is a hydrodynamic process in which a continuous phase (dimethylsiloxane, DMS) surrounds the dispersed phase (EGaIn) (with which it is immiscible) and gives rise to discrete droplets of dispersed phase at the orifice through which both fluids are extruded ^[48]. The EGaIn microdroplets were dispersed in DMS as they were formed and the fluids exiting the outlet were subsequently diluted with PDMS to reach 10 wt% EGaIn. As observed from the particle size analysis in Figure 2 c, EGaIn microdroplets produced by FF are considerably bigger in size (~150 μm – 200 μm), shaped like peanuts with ribbons of exfoliated oxide streaming behind. FF produced droplets with consistent form factor and relatively narrow size distribution. Similar to the microdroplets obtained by shear mixing, microdroplets produced by FF gets aligned upon application of an electric field (Figure S8 & Movie 3, Supporting Information). We modified the FF procedure to produce rounder particles, which assembled and

behaved in a similar manner (Figure S9, Figure S10, Figure S11 & Movie 4, Supporting Information).

Figure 2. Assembly of microwires by DEP of liquid metal particles in silicone. (Top row) Optical microscope images of (a) Ag microparticles (10 wt%) dispersed in PDMS through planetary mixing, (b) EGaIn microdroplets (10 wt%) formed and dispersed in PDMS through planetary mixing, (c) EGaIn microdroplets (10 wt%) formed by flow focusing. (Second row) Particle size distribution for each image shown above. (d) Optical microscope images of particles before and after application of E-field. The bottom image (Ag microparticles) is marked to help visualize the wire. (e) I-V plots of microwires. EGaIn remained conductive before and after curing whereas Ag microwires could no longer conduct after curing of the

PDMS matrix. (f) Conductance of microwires after the alignment process and after cross-linking of polymer. Electrode gap was maintained at 1 mm and applied voltage at 250 V_{pp} @ 1 kHz. Scalebar - 500 μ m.

All the particles, Ag ($R_{\text{mean}} = 37.6 \Omega$), EGaIn (planetary mixed) ($R_{\text{mean}} = 31.74 \Omega$) and EGaIn (FF) ($R_{\text{mean}} = 4.614 \Omega$), aligned upon application of electric field and could produce electrically conductive microwires, initially, as shown in Figure 2 d. Images were taken after dispensing mixture on top of electrodes and after the alignment process to clearly observe the conductive pathway (Figure 2 d). However, the Ag microwires were no longer conductive after curing of the mixture (Figure 2 e-f). Only EGaIn samples remained conductive through the curing process. The loss of electrical conductance happened regardless of whether the Ag microwire samples were left to cure in the oven or at room temperature. On the other hand, both the EGaIn microwires (planetary mixed ($R_{\text{mean}} = 20.2 \Omega$) and FF ($R_{\text{mean}} = 4.927 \Omega$)) remained conductive after curing (Figure 2 e-f). A possible explanation for this could be the fusion of EGaIn microdroplets occurring at the junctions, which does not occur at the solid-solid interface of Ag microparticles. In an uncured silicone, when a DEP-aligned chain of EGaIn microdroplets was vigorously disturbed, the chain did not simply disintegrate into a collection of discrete droplets; rather, it broke into chain segments (Figure S12, Supporting Information). This result suggests that the EGaIn microdroplets sintered at the droplet-droplet junctions. The current breakdown density for the EGaIn microwires (planetary mixed, 1 mm) was calculated to be $8.1 \times 10^3 \text{ A/cm}^2$ (Figure S13, Supporting Information).

We measured the electrical conductance of the microwires (after curing) as shown in Figure 2f. Before curing, solid Ag particles produced electrically conductive networks (Figure S14, Supporting Information) with similar conductance values to the planetary mixed EGaIn droplets; mean conductance for EGaIn (planetary mixed) and Ag microwires were 0.0393 S and 0.0267 S, respectively. Nevertheless, there is a significant variation in electrical conductance among samples of similar compositions, which can be attributed to the variation in particle distribution between the electrodes (Figure S15 & Figure S16, Supporting

Information). Although Ag ($1.59 \times 10^{-6} \Omega\text{-cm}$) has significantly lower resistivity value compared to EGaIn ($29.4 \times 10^{-6} \Omega\text{-cm}$), similar conductance for the microwires can be attributed to presence of higher number of particle-particle junctions for Ag and possible sintering between EGaIn droplets. Conductive microwires composed of aligned EGaIn (FF) microdroplets had considerably higher conductance values (mean conductance of 0.227 S) compared to EGaIn (planetary mixed) microwires (mean conductance of 0.0393 S) (Figure 2f), which can be attributed to the larger cross-sectional area of the EGaIn (FF) microwire and fewer junction points due to the size of the particles (Figure S17, Supporting Information). During the course of our experiments, we found that EGaIn particles with very small ($\sim 100\text{-}800$ nm) and very big (~ 500 μm) diameters failed to form a conductive pathway even when the electric field was applied for a very long duration (up to 2 hours) (Figure S18, Supporting Information). Smaller particles aligned with the field, but did not merge into conductive paths, which is consistent with the larger forces necessary to sinter small particles ^[49, 50]. Larger particles simply did not move in response to the electric fields, presumably due to friction and viscous drag.

2.3 Stretchable, flexible and healable conductive microwires

Following the fabrication of DEP-aligned EGaIn-PDMS conductive microwires, the process was scaled up to increase the length of these conductive microwires from 1 to 5 mm. The experimental conditions used to produce longer wires (3 and 5 mm) are reported in the methods section. The set of parameters utilized for varying electrode gaps have been listed in Table S1, Supporting Information. Owing to their longer length, the 5 mm-long conductive microwires have measured resistance values of $\sim 90 \Omega$ (Figure S19, Supporting Information). As expected, the samples retained the same conductivity before and after crosslinking the PDMS. We measured the electrical performance of these longer microwires under tensile and flexural strain. The resistance of these DEP-aligned EGaIn-PDMS microwires did not vary significantly with linearly increasing strain (**Figure 3 a**) and hovered around 90Ω to 100% strain (we did

not go further due to PDMS failing above this strain^[51]). The inset images show the un-stretched sample and the sample stretched to 100% strain. A possible explanation for the lack of noticeable increase in resistance of the DEP-aligned microwires under tensile strain could be the wavy geometry of the wires. Another possible explanation is the initial resistance is dominated by the narrow portions of the wires, where the particles sinter; thus, elongating the other portions of the wire do not change the resistance significantly as long as the sintered regions do not constrict. These conductive microwires are also highly flexible. We bent the 5 mm-long sample over varying radii of curvature, from 12 mm to 3 mm, with no change in the measured resistance (Figure 3 b). In addition, the wires showed no change in resistance while cyclic bending over a 10 mm radius for 500 cycles (Figure 3 c, inset). The high stretchability and flexibility of the DEP-aligned EGaIn-PDMS microwires could be attributed to the elastomeric behaviour of PDMS and the inherent fluidity of EGaIn. Based on other reports of EGaIn-polymer composites, we expect the microwires to vary in conductivity when subjected to compressive load without loss of electrical continuity^[22, 52, 53]. Prior studies show that liquid metal wires can change resistance when compressed due to geometric changes to the cross-section of the wire^[52]. On the other hand, conductive elastomer sponges loaded with liquid metal show a very small fluctuation in the normalized resistance when subjected to compression (up to 50% strain)^[53]. In contrast, EGaIn-polymer composites are naturally insulating since the metal remains encapsulated in a closed-cell foam-like matrix of the polymer^[22]. Localized compressive stresses applied to the structure rupture the foam-cell walls to form a conductive pathway, but only at high concentrations of liquid metal. Thus, the stray particles (i.e., those that do not get aligned into microwires) remain isolated due to the low concentrations of metal used here.

Figure 3. Stretchable and self-healing microwires. (a) Resistance of a 5 mm-long DEP-aligned EGaIn microwire versus strain. (Inset) Image of the sample in unstretched state and under 100% tensile strain. (b) Resistance of a 5 mm-long DEP-aligned EGaIn microwire against varying bending radii. (c) Resistance of a 5 mm-long DEP-aligned EGaIn microwire under cyclic bending at 10 mm radius over 500 cycles. (Inset) Image of the sample bent on a cylinder of 10 mm radius. (d) Healing demonstration. (Top) Conductance of a 3 mm-long DEP-aligned EGaIn microwire in pristine condition, when cut, and after being healed. (Bottom row) Optical microscope images for the same 3 mm-long microwire in pristine condition, when cut, and after being healed.

Conductive microwires can also be repaired in case of severe mechanical damage. To demonstrate the possibility of healing (Figure 3 d & Movie 5, Supporting Information), 3 mm-long EGaIn-PDMS conductive microwires were fabricated by DEP-assisted alignment of EGaIn microdroplets inside PDMS followed by crosslinking. After establishing the electrical conductance of the microwire, a chunk of the microwire was removed by cutting, creating an open circuit. After filling the gap with uncured EGaIn-PDMS mixture (10 wt%), voltage was applied across the electrodes to induce the alignment of EGaIn microdroplets. The EGaIn microdroplets aligned and connected with the exposed, broken ends of the original wire,

restoring the conductivity (Figure 3d & Figure S20, Supporting Information). The resistance values of the healed microwires were always higher than that of the pristine microwires, with an average increase of 35.6% in resistance for healed samples compared to their pristine counterparts (Figure S21, Supporting Information). This slight increase in resistance was expected because the cured PDMS surrounding the original wire segments restricts their motion and limits the possible area and angle from which the newly formed segments could contact the original wire segments. After the healing process, the samples did not fail up to 100% strain although the resistance increased with strain (Figure S22, Supporting Information).

2.4 Healing of stretchable interconnects and assembly of circuits on-demand

Stretchable conductors based on LM-filled microchannels are very popular owing to their ability to maintain their resistivities over very large strains^[54-58]. However damage to these stretchable wires tend to be permanent in nature, with some noted exceptions^[23, 59]. Here, we demonstrate the utilization of DEP-assisted alignment of EGaIn microparticles in EGaIn-PDMS mixture to heal such a damaged stretchable wire (**Figure 4 a**). EGaIn-filled PDMS microchannel served as a stretchable wire and was damaged in the middle intentionally. A square region was cut and completely removed rendering the wire non-conductive. The damaged region was then filled with EGaIn-PDMS mixture (10 wt%) and subsequently aligned by application of electric field (400 V_{pp} @ 1 kHz). The EGaIn droplets aligned themselves along the electric field lines to heal the conductive pathway. The PDMS in the gap crosslinked and seamlessly connected with the surrounding PDMS matrix to produce a mechanically and electrically healed structure.

As the EGaIn microdroplets only align in presence of an electric field, specific pairs of electrodes can be selectively connected for on-demand connectivity (Figure 4 b & Movie 6, Supporting Information). Voltage was applied across each pair of electrodes (1 mm gap) selectively resulting in the sequential formation of conductive pathways (Figure 4 b), indicated

by the lighting of individual LEDs. A 2 mm vertical gap (twice the horizontal gap) was maintained between the pair of electrodes to prevent cross-alignment. These conductive microwires formed by selective alignment of EGaIn microdroplets had very similar ohmic resistance values ($14\ \Omega$, $21\ \Omega$ and $20\ \Omega$), as shown in the I-V characterization in Figure 4 b. Similar to the conductance plot of standalone 1 mm microwires, the variation in the resistance can be attributed to the random distribution of the EGaIn droplets between the electrode gaps.

Figure 4. (a) Healing of a damaged stretchable wire by DEP-assisted alignment of EGaIn microdroplets in a EGaIn-PDMS mixture. This wire was fabricated by filling a PDMS microchannel with EGaIn. A square chunk (2 mm x 3 mm) was cut out of the centre of the wire

to disrupt the conductive pathway. It was then filled with the originally nonconducting EGaIn-PDMS mixture, followed by DEP-driven alignment to repair the wire and restore the conductive pathway. (b) Sequential formation of conductive pathways between three pairs of electrodes by selective application of electric field to demonstrate the potential for fabricating circuits on-demand. Alignment of EGaIn droplets between a chosen pair of electrodes was done by selectively activating the electric field between them. The corresponding LED lit up when a conductive pathway had been formed between the electrodes. The 3 microwires have similar resistance values.

3. Conclusion

In conclusion, alignment of EGaIn microdroplets inside an EGaIn-PDMS mixture by DEP was demonstrated to form conductive microwires. These EGaIn microdroplets aligned along the electric field lines and sintered to form low-resistance conductive pathways. The microwires stayed conductive even after the removal of electric field. The cross-linking of the PDMS matrix further secured the alignment of the EGaIn microdroplets. Whereas aligned Ag particles failed to maintain conductive paths through the curing process, EGaIn microdroplets stayed connected and the microwires remained conductive. These conductive paths form at significantly lower EGaIn loading (~10%) than liquid metal elastomer composites (~85%) that form conductive paths only after mechanical sintering.

These conductive microwires exhibited good electrical performance under tension and flexure; there was no change in measured resistance when stretched up to 100% linear strain and bent at different radii. The resilience of these microwires was also evident from the lack of resistance change under cyclic bending. Additionally, these microwires could be healed using the DEP-assisted alignment process, restoring their structural integrity and functionality. Furthermore, mechanically damaged and disconnected stretchable conductors fabricated by other routes, could be healed using DEP-assisted alignment as well. Finally, this working principle has been utilized to form circuits on-demand by selective application of voltage to chosen pairs of electrodes to assemble conductive pathways exclusively in places where they are desired.

DEP-assisted microwire alignment in silicone elastomer could be used to form new electrical connections without the need for patterning and repair the mechanical and electrical function

of soft elastomeric devices. The principles shown here can enable rewiring circuits in novel ways and provide a non-lithographic route for *in-situ* assembly of liquid metal drops, which could have applications in optics (wire polarizers or deformable colloidal assemblies), microfluidics (mixers, pumps, and electrodes), and anisotropic thermal conductors.

4. Experimental Section

Preparation of Ag-PDMS and EGaIn-PDMS mixtures

Silver (Ag) microparticles (Smart Memories) and polydimethylsiloxane (PDMS) (Sylgard 184, Dow Corning) were utilized as received. PDMS consists of two parts: elastomer base and the curing agent, which is required to be mixed in the ratio 10:1 as per the manufacturer's instructions. For Ag-PDMS mixtures, 10 wt% of Ag microparticles was added to the uncured PDMS mix and thoroughly mixed (1 min - 0.5 mins mixing & 0.5 mins degassing) using planetary shear mixing (Thinky mixer, AR 250). Commercially available eutectic gallium indium (EGaIn) (Indium Corporation) was processed into smaller particles by different routes. For EGaIn nanoparticles, 10 wt% equivalent of EGaIn was added to 15 ml of solvent (Toluene, Sigma Aldrich). The solution was then agitated (Sonication Time – 15 mins, 2s (On pulse) : 3s (Off pulse), Amplitude – 35%) using a probe sonicator (Sonics, VCX-500 Vibra-Cell Ultrasonic Liquid Processors) to break EGaIn into nanoparticles. The sonicated EGaIn-toluene solution containing EGaIn nanoparticles was then blended into the elastomer base using planetary shear mixing (1 min - 0.5 mins mixing & 0.5 mins degassing). The mixture was then stirred on a hot-plate (@ 120 °C) for the solvent to evaporate completely. Finally, the mixture was cooled down to room temperature and cross-linking agent was added to the mixture.

For smaller EGaIn microparticles, 10wt% of EGaIn was mixed together with the elastomer base and curing agent using planetary shear mixing (5 mins – 2.5 mins mixing & 2.5 mins degassing).

For bigger EGaIn microparticles, flow focusing was performed which also provides consistent size distribution. The fabrication process for flow focusing microchannels follows the route of

soft lithography, explained in detail elsewhere. Cured PDMS microchannels are extracted from the silicon wafer mold and sealed with a pristine PDMS block by bonding them together after oxygen plasma treatment (Diener Electronics, Femto-A, 1 min at 100% power). It is then annealed in the oven @ 70 °C for an hour. High viscosity of the elastomer base makes it challenging to pump them into the microchannels and is higher than the required viscosity for the continuous phase reported previously. Hence, dimethylsiloxane (DMS-S21, Gelest) was utilized as the continuous phase owing to its lower viscosity and ability to mix well with PDMS without interfering with the crosslinking process. Flow rate of the individual phases are controlled separately by two syringe pumps (New Era Pump Systems NE-300) as shown in the image of the complete setup (Figure S23, Supporting Information). Flow rates for the continuous and the dispersed (EGaIn) phase were maintained at 900 $\mu\text{L/hr}$ and 300 $\mu\text{L/hr}$, respectively. After a steady flow of EGaIn microdroplets had been established at the outlet, the EGaIn microdroplets were collected in a vial containing pre-measured quantity of uncured PDMS until the appropriate loading (equivalent to 10 wt%) of EGaIn was collected in the vial (Movie 7, Supporting Information).

Alignment of particles using dielectrophoresis (DEP)

The device architecture adopted for dielectrophoresis-assisted alignment of EGaIn microparticles has been explained in section 2.1 and Figure 1 a. For microscope images, thermally deposited gold (Au) electrodes were utilized whereas for conductivity measurements and other demonstrations, copper tapes were utilized. Au contacts (100 nm thickness, 0.8 mm width & 20 mm length) were evaporated on the substrate through custom designed shadow masks. The gold evaporation was done by E-Beam evaporation system (HHV) at initial rate of 0.1 $\text{\AA/s}$ for initial 15 nm, 0.15 $\text{\AA/s}$ for next 15 nm, 0.3 $\text{\AA/s}$ for the next 20 nm and 0.5 $\text{\AA/s}$ for the final 50 nm. Copper tape was attached to the edges of the Au electrodes using silver conductive adhesive to provide electrical contacts. Cleaned microscopic glass slides were used as substrates everywhere, unless mentioned otherwise. Circular spacers of 10 mm diameter

were laser-cut (by) from 3M VHB 4905 tape then pasted to define the boundary around the test electrodes. Spacers were pasted such that the gap between the test electrodes lied at the center of the spacer.

The setup for the alignment process is shown in Figure S24 (Supporting Information). Input to the high voltage amplifier (Trek Model 609E-6) was supplied by the function generator (Agilent 33220A) to produce the desired magnitude of AC potential necessary. With increased electrode gaps (1, 3, and 5 mm), we utilized increased voltage to align a conductive microwire. It was observed that the alignment process requires hundreds of volts per mm, although exact value depends on additional considerations (Figure S25 and Table S1, Supporting Information). A 100 kOhm resistor was connected in series to limit the short circuit current when the conductive microwire forms and prevent the sample from burning. For current-time measurements, a electrometer (Keithley 6517a) was connected in series with the circuit (in continuous mode). The formation of the conductive pathway was characterized by the sudden increase in current reading. Finally, the sample was placed in the oven (@ 50 °C) for the PDMS to cross-link.

Optical and physical characterization

DEP-assisted alignment of metallic microparticles was captured using an optical microscope (Olympus BX51) at 10X magnification fitted with a digital camera (Canon EOS 70D).

Resistance of the conductive microwires were measured using Keithley 2400 Sourcemeter (Four-point probe) by performing an I-V sweep (source I from -10 mA to 10 mA & measure V) across the sample.

Measuring electrical performance under mechanical strain

For stretchability, bending and cyclic bending tests, 5 mm long EGaIn-PDMS conductive microwires were utilized. Particle loading was increased to 30 wt% (EGaIn) and applied voltage was increased to 700V_{pp} (while keeping frequency unchanged @ 1 kHz), while the substrate was changed to a highly soft and stretchable elastomer (3M VHB 4910 acrylic tape). As mentioned, bigger EGaIn microparticles result in lower resistance values for the conductive

microwires. Hence, for these longer microwires, EGaIn particles were simply shear mixed into PDMS by hand mixing. Copper tape was utilized as the rigid, test electrodes and acrylic tape was used for securing a boundary for the mixture. After the alignment of the EGaIn microparticles to form a conducting pathway, the sample was placed in the oven (@ 50 °C) for the PDMS to cross-link.

Healing demonstration

3 mm-long microwire samples were utilized for the healing demonstration. 10 wt% of EGaIn was shear mixed into the PDMS by simple hand mixing mixed to break the EGaIn into smaller droplets. To make the electrodes, a pair of copper tape strips were pasted onto a clean glass slide, spaced 3 mm apart, and a spacer was pasted on top of the electrodes such that the gap between the electrodes was in the middle of the spacer. The hand-mixed 10wt% EGaIn-PDMS mixture was poured into the spacer and 400V_{pp}, 1 kHz AC potential was applied across the copper electrodes. After a conducting microwire was assembled between the electrodes, the sample was placed in the oven at 60-70 °C for the PDMS to cure.

Incisions were made to cured and conducting samples. Care was taken to ensure that the incisions were between the test electrodes and that no part of the test electrodes were exposed outside of the original layer of cured sample. This is to eliminate the possibility that any recovery of the sample's conductance was due to formation of new wire directly in contact with the test electrodes. After cutting the sample, resistance between the test electrodes was checked to confirm that the sample was no longer conducting. Fresh, uncured mixture of 10wt% EGaIn in PDMS was poured into the area of the sample's incision. Subsequently, voltage was applied across the test electrodes to induce alignment of EGaIn microdroplets in the uncured mixture at the incision.

Circuit-on-demand demonstration

For this demonstration, we used 10 wt% EGaIn hand mixed in uncured PDMS. Three pairs of copper electrodes were pasted 2 mm vertically apart. The device design for electrode placement

followed the schematic provided in Figure S26 (Supporting Information). Pairs of copper electrodes were patterned 2 mm vertically apart. This spacing was implemented to prevent any cross-alignment between the electrodes. For each pair of electrode, the electrodes were placed 1 mm horizontally apart. A spacer were placed on top of the three pairs of electrodes such that the three pairs of electrodes were in the middle of the spacer. The circuit was set up as shown in Figure 4. Each pair of electrodes was connected to one white LED light bulb.

We first applied an alternating voltage of 250 V_{pp} at 1 kHz across a pair of electrodes. After the LED bulb lighted up, we disconnected the circuit. Another pair of electrodes was connected to the circuit. We repeated the aforementioned procedure until three microwires bridging three electrode gaps were successfully formed.

Supporting Information

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

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References

- [1] S. Gupta, W. T. Navaraj, L. Lorenzelli, R. Dahiya, npj Flexible Electronics 2018, 2, 1.
- [2] H. Lee, T. K. Choi, Y. B. Lee, H. R. Cho, R. Ghaffari, L. Wang, H. J. Choi, T. D. Chung, N. Lu, T. Hyeon, Nature nanotechnology 2016, 11, 566.
- [3] W. Gao, S. Emaminejad, H. Y. Y. Nyein, S. Challa, K. Chen, A. Peck, H. M. Fahad, H. Ota, H. Shiraki, D. Kiriya, Nature 2016, 529, 509.
- [4] F. Carpi, D. De Rossi, R. Kornbluh, R. E. Pelrine, P. Sommer-Larsen, *Dielectric elastomers as electromechanical transducers: Fundamentals, materials, devices, models and applications of an emerging electroactive polymer technology*, Elsevier, 2011.
- [5] J. Y. Sun, C. Keplinger, G. M. Whitesides, Z. Suo, Advanced Materials 2014, 26, 7608.

- [6] A. Ankit, J. Y. Chan, L. L. Nguyen, F. Krisnadi, N. Mathews, *Large-area, flexible, integrable and transparent DEAs for haptics*, Vol. 10966, SPIE, 2019.
- [7] Ankit, N. Tiwari, M. Rajput, N. A. Chien, N. Mathews, *Small* 2018, 14, 1702312.
- [8] J.-H. Ahn, J. H. Je, *Journal of Physics D: Applied Physics* 2012, 45, 103001.
- [9] S. Rosset, H. R. Shea, *Applied Physics A* 2013, 110, 281.
- [10] C. Martin, J. Sandler, A. Windle, M.-K. Schwarz, W. Bauhofer, K. Schulte, M. Shaffer, *Polymer* 2005, 46, 877.
- [11] K. Tsuchiya, A. Sakai, T. Nagaoka, K. Uchida, T. Furukawa, H. Yajima, *Composites Science and Technology* 2011, 71, 1098.
- [12] M. Iqbal, G. Mamoor, T. Bashir, M. Irfan, M. Manzoor, *Journal of Chemical Engineering* 2010, 61.
- [13] M. Park, J. Im, M. Shin, Y. Min, J. Park, H. Cho, S. Park, M.-B. Shim, S. Jeon, D.-Y. Chung, J. Bae, J. Park, U. Jeong, K. Kim, *Nature Nanotechnology* 2012, 7, 803.
- [14] P. Lee, J. Lee, H. Lee, J. Yeo, S. Hong, K. H. Nam, D. Lee, S. S. Lee, S. H. Ko, *Advanced materials* 2012, 24, 3326.
- [15] S.-M. Park, N.-S. Jang, S.-H. Ha, K. H. Kim, D.-W. Jeong, J. Kim, J. Lee, S. H. Kim, J.-M. Kim, *Journal of Materials Chemistry C* 2015, 3, 8241.
- [16] Q. Yang, M. H. Beers, V. Mehta, T. Gao, D. Parkinson, *ACS applied materials & interfaces* 2017, 9, 958.
- [17] D. Amoabeng, S. S. Velankar, *Colloids and Surfaces A: Physicochemical and Engineering Aspects* 2018, 553, 624.
- [18] W. Thongruang, R. J. Spontak, C. M. Balik, *Polymer* 2002, 43, 3717.
- [19] L. A. Toth, A. A. Goldenberg, "Control system design for a dielectric elastomer actuator: the sensory subsystem", presented at *Smart Structures and Materials 2002: Electroactive Polymer Actuators and Devices (EAPAD)*, 2002.
- [20] J. A. Fan, W.-H. Yeo, Y. Su, Y. Hattori, W. Lee, S.-Y. Jung, Y. Zhang, Z. Liu, H. Cheng, L. Falgout, M. Bajema, T. Coleman, D. Gregoire, R. J. Larsen, Y. Huang, J. A. Rogers, *Nature Communications* 2014, 5, 3266.
- [21] Y. Mengüç, Y.-L. Park, H. Pei, D. Vogt, P. M. Aubin, E. Winchell, L. Fluke, L. Stirling, R. J. Wood, C. J. Walsh, *The International Journal of Robotics Research* 2014, 33, 1748.
- [22] A. Fassler, C. Majidi, *Advanced Materials* 2015, 27, 1928.
- [23] E. J. Markvicka, M. D. Bartlett, X. Huang, C. Majidi, *Nature materials* 2018, 17, 618.
- [24] M. Khondoker, D. Sameoto, *Smart Materials and Structures* 2016, 25, 093001.
- [25] H. Wang, Y. Yao, Z. He, W. Rao, L. Hu, S. Chen, J. Lin, J. Gao, P. Zhang, X. Sun, X. Wang, Y. Cui, Q. Wang, S. Dong, G. Chen, J. Liu, *Advanced Materials* 2019, 31, 1901337.
- [26] M. D. Dickey, *Advanced Materials* 2017, 29, 1606425.
- [27] S. Zhu, J. H. So, R. Mays, S. Desai, W. R. Barnes, B. Pourdeyhimi, M. D. Dickey, *Advanced Functional Materials* 2013, 23, 2308.
- [28] M. D. Bartlett, A. Fassler, N. Kazem, E. J. Markvicka, P. Mandal, C. Majidi, *Advanced Materials* 2016, 28, 3726.
- [29] J. Wang, G. Cai, S. Li, D. Gao, J. Xiong, P. S. Lee, *Advanced Materials* 2018, 30, 1706157.
- [30] S. Liu, M. C. Yuen, E. L. White, J. W. Boley, B. Deng, G. J. Cheng, R. Kramer-Bottiglio, *ACS applied materials & interfaces* 2018, 10, 28232.
- [31] K. D. Hermanson, S. O. Lumsdon, J. P. Williams, E. W. Kaler, O. D. Velev, *Science* 2001, 294, 1082.
- [32] S. O. Lumsdon, D. M. Scott, *Langmuir* 2005, 21, 4874.
- [33] S.-H. Cha, S.-H. Kang, Y. J. Lee, J.-H. Kim, E.-Y. Ahn, Y. Park, S. Cho, *Scientific reports* 2019, 9, 1.

- [34] J. Kimbrough, S. Chance, B. Whitaker, Z. Duncan, K. Davis, A. Henderson, Z. Xiao, Q. Yuan, F. Camino, "Deposition and Alignment of Carbon Nanotubes with Dielectrophoresis for Fabrication of Carbon Nanotube Field-Effect Transistors", presented at *2018 IEEE International Conference on Manipulation, Manufacturing and Measurement on the Nanoscale (3M-NANO)*, 2018.
- [35] A. Oliva-Avilés, F. Avilés, V. Sosa, A. Oliva, F. Gamboa, *Nanotechnology* 2012, 23, 465710.
- [36] K. H. Bhatt, O. D. Veleev, *Langmuir* 2004, 20, 467.
- [37] I. D. Johnston, D. K. McCluskey, C. K. L. Tan, M. C. Tracey, *Journal of Micromechanics and Microengineering* 2014, 24, 035017.
- [38] R. R. Pethig, *Dielectrophoresis: Theory, methodology and biological applications*, John Wiley & Sons, 2017.
- [39] O. D. Veleev, S. Gangwal, D. N. Petsev, *Annual Reports Section "C" (Physical Chemistry)* 2009, 105, 213.
- [40] S.-H. Cha, S.-H. Kang, Y. J. Lee, J.-H. Kim, E.-Y. Ahn, Y. Park, S. Cho, *Scientific reports* 2019, 9, 3629.
- [41] H. A. Pohl, K. Pollock, J. S. Crane, *Journal of Biological Physics* 1978, 6, 133.
- [42] S.-Y. Tang, J. Zhu, V. Sivan, B. Gol, R. Soffe, W. Zhang, A. Mitchell, K. Khoshmanesh, *Advanced Functional Materials* 2015, 25, 4445.
- [43] M. Abe, A. Yamamoto, M. Orita, T. Ohkubo, H. Sakai, N. Momozawa, *Langmuir* 2004, 20, 7021.
- [44] Y. Guo, S. Batra, Y. Chen, E. Wang, M. Cakmak, *ACS Applied Materials & Interfaces* 2016, 8, 18471.
- [45] S. Batra, E. Unsal, M. Cakmak, *Advanced Functional Materials* 2014, 24, 7698.
- [46] A. W. Maijenburg, M. G. Maas, E. J. B. Rodijk, W. Ahmed, E. S. Kooij, E. T. Carlen, D. H. A. Blank, J. E. ten Elshof, *Journal of Colloid and Interface Science* 2011, 355, 486.
- [47] J. Thelen, M. D. Dickey, T. Ward, *Lab on a Chip* 2012, 12, 3961.
- [48] T. Hutter, W.-A. C. Bauer, S. R. Elliott, W. T. S. Huck, *Advanced Functional Materials* 2012, 22, 2624.
- [49] J. W. Boley, E. L. White, R. K. Kramer, *Advanced Materials* 2015, 27, 2355.
- [50] Y. Lin, J. Genzer, M. D. Dickey, *Advanced Science*, n/a, 2000192.
- [51] S. Park, K. Mondal, R. M. Treadway, V. Kumar, S. Ma, J. D. Holbery, M. D. Dickey, *ACS Applied Materials & Interfaces* 2018, 10, 11261.
- [52] Y. Jin, Y. Lin, A. Kiani, I. D. Joshipura, M. Ge, M. D. Dickey, *Nature Communications* 2019, 10, 4187.
- [53] S. Liang, Y. Li, Y. Chen, J. Yang, T. Zhu, D. Zhu, C. He, Y. Liu, S. Handschuh-Wang, X. Zhou, *Journal of Materials Chemistry C* 2017, 5, 1586.
- [54] M. D. Dickey, R. C. Chiechi, R. J. Larsen, E. A. Weiss, D. A. Weitz, G. M. Whitesides, *Advanced Functional Materials* 2008, 18, 1097.
- [55] S. W. Jin, J. Park, S. Y. Hong, H. Park, Y. R. Jeong, J. Park, S.-S. Lee, J. S. Ha, *Scientific Reports* 2015, 5, 11695.
- [56] K. Khoshmanesh, S.-Y. Tang, J. Y. Zhu, S. Schaefer, A. Mitchell, K. Kalantar-zadeh, M. D. Dickey, *Lab on a Chip* 2017, 17, 974.
- [57] J. Yang, T. Zhou, L. Zhang, D. Zhu, S. Handschuh-Wang, Z. Liu, T. Kong, Y. Liu, J. Zhang, X. Zhou, *Journal of Materials Chemistry C* 2017, 5, 6790.
- [58] S. Zhu, J.-H. So, R. Mays, S. Desai, W. R. Barnes, B. Pourdeyhimi, M. D. Dickey, *Advanced Functional Materials* 2013, 23, 2308.
- [59] E. Palteau, S. Reece, S. C. Desai, M. E. Smith, M. D. Dickey, *Advanced Materials* 2013, 25, 1589.
- [60] Z. J. Farrell, C. Tabor, *Langmuir* 2018, 34, 234.

Table of Content

Dielectrophoresis assisted alignment of EGaln microdroplets in silicone matrix enables fabrication of stretchable and flexible electrically conducting microwires at significantly low weight loadings (10 wt%). Liquid EGaln eliminates compliance mismatch with the elastomeric matrix. This directed assembly within a silicone-EGaln “ink” can heal severely damaged regions of stretchable interconnects to restore mechanical integrity and electrical conductivity.

Febby Krisnadi[#], Linh Lan Nguyen[#], Ankit[#], Jinwoo Ma, Mohit R. Kulkarni, Nripan Mathews, Michael D. Dickey**

Directed Assembly of Liquid Metal-Elastomer Conductors for Stretchable and Self-healing Electronics

Supporting Information

Directed Assembly of Liquid Metal-Elastomer Conductors for Stretchable and Self-healing Electronics

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$$F = 4\pi R^3 \epsilon_0 \epsilon_m [CM] (E \cdot \nabla) E$$

Figure S1. Illustration of polarization giving rise to DEP force and governing equation for DEP force.

Figure S2. Effect of EGaIn concentration on time taken for microwire to form. The more concentrated the EGaIn microparticles, the closer the particles. Hence, the interparticle attraction is enhanced, further shortening the time taken for the wire formation. The time to form the wires is apparent by the sudden increase in current when the wires span the two electrodes.

Figure S3. Wire formation is apparent when the current between the electrodes suddenly increases. The following plot shows the effect of voltage applied on time taken for microwire to form. As the dielectrophoretic force is dependent on electric field strength, the higher the voltage, the greater the dielectrophoretic force. Hence, the shorter the time taken for the wire formation.

Figure S4. Wire formation is apparent when the current between the electrodes suddenly increases, indicative of a wire spanning the two electrodes. This plots shows the effect of frequency of voltage source on time taken for microwire to form. The variation in the magnitude of the current does not necessarily correspond to variation in the resistance values of the microwires formed as the frequency increases. Such variation is attributed to the fact that the measurement time interval of the electrometer is much larger than the period of the alternating current in the high frequency range (above 100 Hz). Hence, the electrometer could not capture the full cycle and the peak of the alternating current. Optimal frequency ranges from 1 Hz to 10 kHz.

Parameter Optimization

Loading: 10% was the lowest weight loading of EGaIn microdroplets at which the process of forming conductive microwires could occur within reasonable time frames. As shown in Figure S2, at 5 wt% loading, a conductive pathway did not form. Thus, we used at least 10 wt% in our experiments.

Voltage: No conductive pathway formation was observed for 150 V_{pp} (Figure S3). At 250 V_{pp}, the particles were found to be aligned within a few minutes. Note that the applied field did not play any significant role in determining the conductance of the microwire.

E-field: The order of magnitude of the E-field, 10⁵ V/m – noticeably larger than other DEP work utilizing aqueous medium (10⁴ V/m) (Hermanson et al., 2001; Bhatt et al., 2004)^[31, 36] – was suitable for our system. This can be attributed to utilization of viscous PDMS as the dielectric medium. According to Stoke's law, a perfect sphere traveling through a viscous fluid experiences drag force proportional to the liquid's viscosity and the sphere's radius. Thus, the larger size of the EGaIn droplet (relative to nanoparticles used in literature) also contributes to larger viscous drag.

Frequency: Between 1 Hz and 10 kHz, the microwires assemble relatively quickly. In this range, we observed that the time taken for a conductive pathway to form decreased with frequency (Figure S4). However, when we increased the frequency by an order of magnitude to 100 kHz, the EGaIn droplets jittered in place instead of moving in a clear direction towards alignment. Thus, 1 kHz was chosen for assembly. Frequency did not play a role in the conductance of the microwire.

Summary: Taken in sum, considering the conductivity of the wires does not depend on any of the parameters, we simply chose conditions that gave microwires quickly. The best combination of parameters – 10wt% loading, 250 V_{pp}, 1 kHz, 1 mm gap – emerged from these studies.

Figure S5. Microscope images of Ag (top) and EGaln (bottom) microparticles at 10X magnification. Scalebar – 250 μm .

Figure S6. DEP-assisted alignment of 10wt% Ag microparticles in uncured Sylgard 184 prepared by planetary mixing. The sample was subjected to 250 Vpp, 1 kHz. Scalebar – 500 μm .

Figure S7. DEP-assisted alignment of 10wt% EGaIn microdroplets in uncured Sylgard 184 prepared by planetary mixing. The sample was subjected to 250 Vpp, 1 kHz. Scalebar – 500 μm .

Figure S8. DEP-assisted alignment of 10wt% EGaIn microdroplets in uncured Sylgard 184. EGaIn microdroplets were formed by flow focusing in DMS-S21, with flow rates 900 $\mu\text{L/hr}$ and 300 $\mu\text{L/hr}$ for DMS and EGaIn respectively. The sample was subjected to 250 Vpp, 1 kHz. Scalebar – 500 μm .

Figure S9. 10wt% EGaIn microdroplets in uncured Sylgard 184. Microdroplets were formed by flow focusing in DMS-S21 with 5wt% DDT, at flow rates 900 $\mu\text{L/hr}$ and 300 $\mu\text{L/hr}$ for DMS (5wt% DDT) and EGaIn respectively. To achieve more spherical form factor for EGaIn particles through FF, the interfacial dynamics was further modified through addition of a surfactant, dodecanethiol (DDT)^[60]. Similar to the FF experiment without surfactant, the EGaIn microdroplets with DDT were accompanied with ribbon-like particulates, but appeared to be detached from the droplets. Scalebar – 500 μm .

Figure S10. DEP-assisted alignment of 10wt% EGaIn microdroplets in uncured Sylgard 184. EGaIn microdroplets were formed by flow focusing in DMS-S21 with 5wt% DDT, with flow rates 900 $\mu\text{L/hr}$ and 300 $\mu\text{L/hr}$ for DMS (5wt% DDT) and eGaIn respectively. Sample pictured was subjected to 250 Vpp, 1 kHz. Scalebar – 500 μm .

Movement of particles induced by dielectrophoresis

Simulation studies on movement of particles induced by dielectrophoresis predicts that for two particles suspended in a dielectric medium case, each particle's relaxation time (time to rotate) is significantly shorter than the time required for global reorientation (time to translate). Alignment of non-spherical EGaIn microdroplets made by shear mixing (Figure S7 and Movie 1, Supporting Information) was faster compared to aligning of spherical microdroplets made by flow focusing (Figure S10 and Movie 4, Supporting Information). This observation could be because non-spherical EGaIn droplets were aligned due to electrorotation while spherical EGaIn depended mainly on translation.

Furthermore, for two particles positioned such that the line connecting their centres was parallel to the electric field lines, the electric field between the particles was weak, resulting in attractive DEP force. When the line connecting the centres of the particles was perpendicular to the electric field, simulation results showed a region of strong electric field formed between the particles. The resultant mutual DEP force was repulsive; the two particles were repelled away from each other. In line with these predictions, we observed that the EGaIn chains formed were always parallel to the electric field lines. The repulsive force experienced by vertically aligned particles may explain the increase in vertical distance between the EGaIn chains (as we move away from the electrodes), resulting in curved EGaIn chains formed far away from the centre of the electrodes.

Figure S11. Effect of use of surfactant in flow focusing process (5wt% DDT in DMS-S21) on resistance of microwire. Even with DDT in the FF continuous phase, the eGaIn microdroplets were still able to form conducting wires after lining up and touching. Resistance of FF-eGaIn microwires was slightly higher in the presence of DDT in the FF continuous phase.

Figure S12. Microscope images of a DEP-aligned chain of EGaIn microdroplets in uncured PDMS before and after being mechanically disturbed. The chain broke after the mechanical disturbance, however, the droplets in the broken chain segments remained stuck together. Scalebar – 500 μm .

Figure S13. I-V curve for 1 mm long EGaIn microwire measured till breakdown. Mean cross-sectional area ($\sim 1107 \mu\text{m}^2$) calculated from Image J analysis for the aligned microwire.

Calculation of current breakdown density (Fig. S13): Since the current at breakdown (measured) is 90 mA and the mean cross-sectional area of the EGaIn microwire: $\sim 1107 \mu\text{m}^2$ (calculated by Image J analysis), the current breakdown density is $8.1 \times 10^3 \text{ A/cm}^2$.

Figure S14. Conductance of microwires before curing. Samples featured in this graph are of 10wt% concentration of conductive droplets/ particles and 1 mm electrode gap. Dot shows mean and bars show range.

Figure S15. 10wt% Ag microparticles in uncured Sylgard 184 prepared by planetary mixing. Scalebar – 500 μm .

Figure S16. 10wt% EGaIn microdroplets in uncured Sylgard 184 prepared by planetary mixing. Scalebar – 500 μm .

Figure S17. 10wt% EGaIn microdroplets in uncured Sylgard 184. Microdroplets were formed by flow focusing (FF) in DMS-S21, with flow rates 900 $\mu\text{L/hr}$ and 300 $\mu\text{L/hr}$ for DMS and EGaIn respectively. Even though FF produces EGaIn particles with narrow size distribution and uniform shape, in the absence of a surfactant, they are elongated and not spherical. Scalebar – 500 μm .

Figure S18. Effect of size of EGaIn droplets on DEP-assisted alignment of conductive pathways. Dielectrophoresis-assisted alignment of EGaIn nanodroplets (~100-800 nm, produced by probe sonication) in PDMS, failed to produce conductive pathways. Interestingly, efforts to align very huge EGaIn particles (~500 μm) to form a conductive pathway failed as well.

Figure S19. Box plot of resistance of 5 mm microwires made from 30 wt% EGaIn mixture before and after curing.

Figure S20. Microscope images of 3 mm, 10wt% (prepared by hand mixing) EGaIn microwire in pristine condition, when cut, and after being healed. Scalebar – 500 μm .

Figure S21. Box chart of resistance of 3 mm long microwires formed by DEP-assisted alignment of EGaIn microparticles, in pristine condition and after being healed. Sample was prepared by hand mixing of EGaIn (10 wt% loading) into PDMS. Resistance of samples after being healed were always higher than that of the pristine samples. However, they are on the same order of magnitude; on average, between healed and pristine samples there is a 35.6% increase in resistance.

Figure S22. Plot of R/R_0 (resistance/original resistance) for a healed 5 mm long EGaIn microwire versus strain. The images for the sample are provided on the right-hand side.

Figure S23. Image of flow focusing setup for producing more uniformly shaped and sized EGaIn microdroplets.

Figure S24. Schematic of the DEP experimental setup. AC signal from function generator was amplified using a high voltage amplifier. 100 k Ω resistor was connected in series to prevent excessive current from flowing through the sample.

Figure S25. Relationship between electrode gap and applied electric field. If alignment requires a constant electric field, we would expect a linear relation between the gap and the necessary voltage, but in practice there are some additional considerations.

Table S1. Parameters for DEP-assisted alignment for different electrode gaps. Different mixing processes yield different size for EGaIn microdroplets. While planetary mixing gives smaller EGaIn microdroplets (~ 50 μm), flow focusing and hand mixing gives bigger EGaIn microdroplets (~200 μm).

Electrode Gap (mm)	EGaIn loading (wt%)	Mixing process	Applied Voltage (V_{pp})	Resistance (Ω)
1	10	Planetary mixing	250	~20
1	10	Flow focusing	250	~5
3	10	Hand mixing	400	~7
5	30	Planetary mixing	700	~90
5	30	Hand mixing	700	~17

Figure S26. Device design, for electrode placement, followed for the circuits-on-demand demonstration.