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Rebecca K. Kramer

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Rebecca K. Kramer

School of Mechanical Engineering, Purdue University
585 Purdue Mall, West Lafayette, IN, USA

ABSTRACT

As advanced as modern machines are, the building blocks have changed little since the industrial revolution, leading to rigid, bulky, and complex devices. Future machines will include electromechanical systems that are soft and elastically deformable, lending them to applications such as soft robotics, wearable/implantable devices, sensory skins, and energy storage and transport systems. One key step toward the realization of soft systems is the development of stretchable electronics that remain functional even when subject to high strains. Liquid-metal traces embedded in elastic polymers present a unique opportunity to retain the function of rigid metal conductors while leveraging the deformable properties of liquid-elastomer composites. However, in order to achieve the potential benefits of liquid-metal, scalable processing and manufacturing methods must be identified.

Keywords: Soft electronics, stretchable electronics, soft robotics

1. INTRODUCTION

Soft functional materials have the potential to fundamentally change the way electromechanical systems are made while reducing manufacturing costs. The realization of soft electromechanical systems has thus far been hindered by a mismatch between the existing materials processing techniques of today (tailored for rigid materials) and the soft machines envisioned for tomorrow. Next-generation machines will require new scalable manufacturing techniques for soft, responsive materials.

This paper specifically considers the case of soft electronics, which are necessary components of emerging soft machines. Current soft electronics utilize polymeric electrode materials, resulting in increased flexibility and decreased obscuration compared to conventional metal electrodes. However, these conductive polymers often have sheet resistances far in excess of metals, leading to significant losses and limited stretchability. The use of liquid-metal alloys encased in hyperelastic polymers addresses both of these challenges and compliments many flexible/stretchable applications by exploiting the inherent properties of liquid-metals, such as their metallic conductance, deformability, and potential for self-healing and shape-reconfiguration.

2. BACKGROUND

2.1 Background on Soft Machines

Accepting the feasibility of soft machines,¹⁻³ we can now trade off rigidity and the “nuts and bolts” approach to assembly for sophisticated functional materials, where power storage and transport, sensing, communication, actuation and logic may all be embedded into one conformable material. For autonomous systems, many current solutions utilize environmental stimuli to power soft material actuators, such as with responsive hydrogels,⁴⁻⁶ while other highly deformable dynamic systems are powered by compressed air.⁷⁻¹⁰ Alternatively, there are host of soft mechanical systems actuated by dielectric elastomers,¹¹⁻¹³ electroactive polymers,^{14,15} and shape memory alloys,¹⁶⁻²⁰ all of which require electrical energy to operate.

Along with efficient power transport, soft machines require sensing (e.g. resistance- and capacitance-based) and communication devices (antennae). In traditional mechanical systems, all of these components would be made with highly-conductive metals. In the context of soft electromechanical systems, however, electronics require stretchability and flexibility to accommodate high deformations around joints and elastic bodies.^{21,22} Conformability is also a prerequisite for biocompatibility,²³ integration with wearable systems,²⁴ and sensory skins.²⁵

Further author information: (Send correspondence to R.K.K)

R.K.K.: E-mail: rebeccakramer@purdue.edu, Telephone: 1-765-494-2219

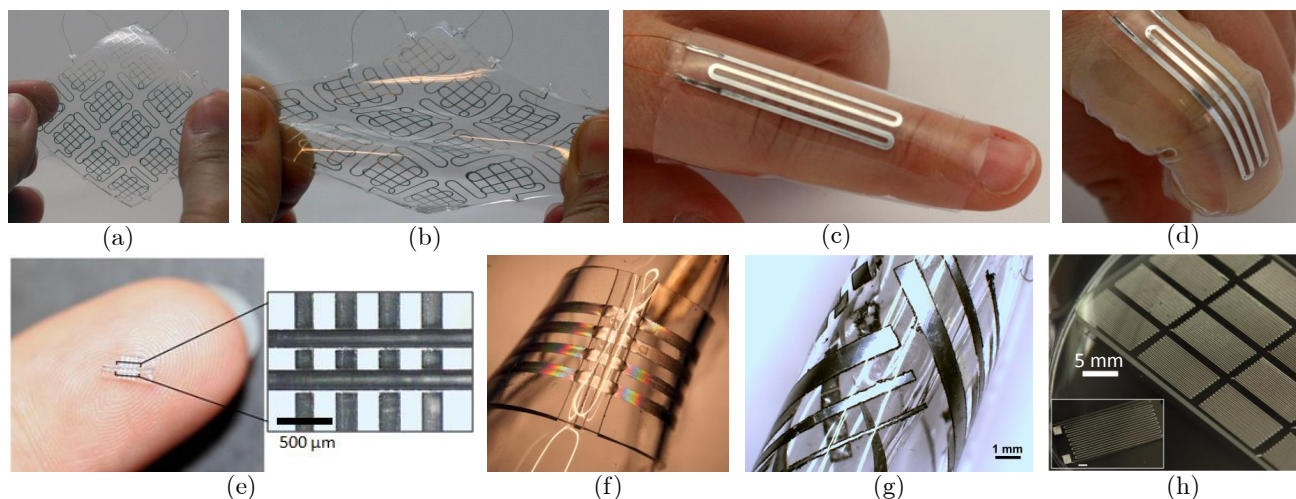


Figure 1. Liquid-metal-embedded elastomer devices. (a-b) Hyperelastic pressure sensitive keypad.³⁴ (c-d) Elastomer-based strain sensor worn around a finger for joint angle proprioception.³⁵ (e) Soft tactile sensor array for micro-manipulation.³⁵ (f) Hyperelastic curvature sensor.³⁸ (g) Rolled elastomer sheet with liquid-metal features patterned by selective wetting.⁴⁰ (h) Strain gauge array patterned by direct writing.⁴²

2.2 Background on Stretchable Electronics

Highly-deformable soft machines require equally deformable and soft electronics for sensing and power transport. There are generally two widely accepted approaches to stretchable (which is different than flexible) conductors: (1) patterning ultrathin films of metal in waves or nets on an elastomer substrate, such as in the work of Rogers and colleagues,^{26,27} and (2) dispersing conductive particles within an elastomeric matrix, as shown by Someya, *et al*²⁸⁻³⁰ and Bao, *et al*.³¹

In the first case, the use of ultrathin metal films patterned into waves and nets employs highly conductive materials, such as silicon or gold, yet the elasticity of the integrated device is dependent on the geometry of the brittle films rather than the inherent elasticity of the elastomer substrate. Therefore, these systems are typically best suited for applications where the maximum strains do not exceed 10%-20%, such as in curved-surface applications.^{32,33} In the second case, dispersions of conductive particles within an elastomeric matrix present many advantages in terms of compatibility with polymer systems, yet several disadvantages with respect to efficiency. Mainly, these conductive polymers have sheet resistances far in excess of metals, leading to losses as current is transferred between loads.

2.3 Background on Liquid-Metal-Embedded Elastomers

Liquid-metal traces embedded in elastic polymers present a unique opportunity to retain the function of rigid metal conductors while leveraging the deformable properties of liquid-elastomer composites. Such systems may function as highly stretchable electrical wiring or sensors that change electrical conductivity in response to elastic deformation. Our group has demonstrated liquid-metal-embedded elastomer devices in applications such as pressure sensitive artificial skin,³⁴⁻³⁷ proprioceptive motion feedback,³⁸ and electronics.³⁹⁻⁴² Other recent demonstrations include reversibly-deformable and mechanically-tunable fluidic antennae,⁴³ hyperelastic multi-modal sensors,^{44,45} 2D/3D highly stretchable conductors,⁴⁶⁻⁵⁰ a stretchable radio frequency (RF) radiation sensor,⁵¹ and self-healing wires.⁵² Because the sensors and conductors are soft and stretchable, they may conform to a host without interfering with the natural mechanics of motion. In addition, maximum strains in these devices are defined by the elastomer material properties, rather than the properties of the conductive element.

The stretchable electronics described above utilize eutectic liquid-metal alloys due to their high conductivity, inherent deformability, and self-healing properties. Liquid-metals at room temperature are most commonly a blend of gallium and indium, and sometimes tin. These low melting temperature alloys have reduced toxicity and a lower vapor pressure than mercury. In fact, according to the CRC Handbook of Chemistry and Physics, the

components of eutectic gallium-indium have “low orders of toxicity”;⁵³ indium has been used in dental fillings and gallium is a trace nutrient. Indium is also a common solder base in the semiconductor industry.

3. MANUFACTURING CHALLENGES

The emerging field of soft robotics will benefit greatly from new scalable manufacturing techniques for soft, responsive materials. Currently, most soft robotic examples are fabricated one-at-a-time, using techniques borrowed from molding and lithography. This limits both the maximum and minimum size of components that can be fabricated, and hinders batch production, which is critical to gaining wider acceptance for soft robotic systems. Although, both laser ablation and direct writing (extrusion) of soft materials have gained recent attention.⁵⁴

Current liquid-metal-embedded elastomer fabrication approaches rely on injecting liquid-metals into microfluidic channels in low-modulus elastomer substrates. This approach is limited by the “sticky” oxide layer that forms on exposed gallium-indium alloy surfaces,^{53,55,56} which yields high surface tension and increases the pressures required to inject the liquid-metal into the channel. As the channel dimensions decrease, the pressure required to fill the channel increases, resulting in a minimum channel size before delamination of the elastomer layers occurs. In addition, this method requires a discrete inlet and outlet location for each conductive trace, eliminating potential devices that require isolated features or arrays of devices over large areas.

While several methods of processing liquid-metal are actively being explored,^{42,50,56-60} none has produced a solution that enables both feature scalability and parallel processing (i.e. batch scalability). Now that individual sensors and conductors have been demonstrated, it is desirable to move towards more complex sensor networks and electronic skins that require new batch-manufacturing processes. However, there is currently a mismatch between existing materials processing techniques and the soft materials that soft machines will be constructed from. Low-viscosity liquids are easily printed, coated, and rolled. Hard solids are easily cast, formed, cut, and joined via traditional machining and metalworking methods. Viscous or high surface tension liquids, such as liquid-metal, and soft solids, such as elastomers, are less explored from a manufacturing standpoint.

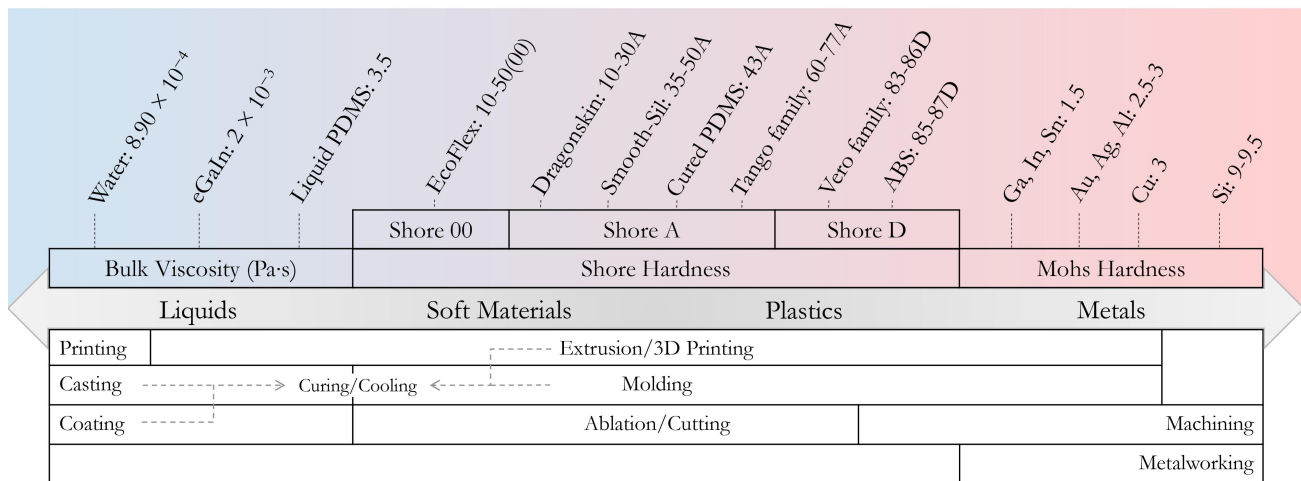


Figure 2. Hardness values for various materials. Liquids are characterized by their bulk viscosity, while solids are characterized by their Shore Hardness (for rubbers and plastics) and Mohs Hardness (for metals). Materials are not spaced to scale. Manufacturing processes are given based on material hardness. The manufacturing categories listed are meant to be representative and not exhaustive. Printing may include ink-jet and offset printing. Casting may include selective wetting and drop-casting. Coating may include spin-coating, gravure, and slot-die coating. Metalworking may include all casting, forming, cutting, and joining processes, although ablation (a sub-category of machining, which itself is a sub-category of cutting), extrusion (a sub-category of forming), and casting are represented independently to clarify their applicability to non-metals. Curing and cooling are shown as transitional processes that represent phase change between liquids and solids. Note that microfabrication is not directly represented, but includes processes such as coating and molding (patterning of photoresists), as well as metalworking (deposition & growth, etching, and micro-cutting).

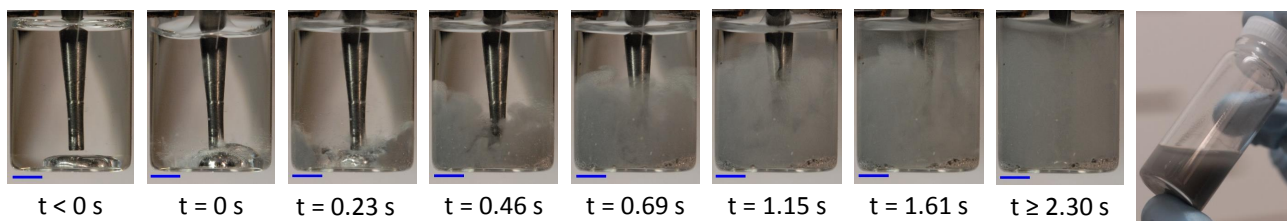


Figure 3. Ultrasonication of gallium-indium alloy in a carrier solvent results in a gallium-indium dispersion ink. Scale Bars are 5 mm in length. Adapted from Boley, *et al.*⁶⁸

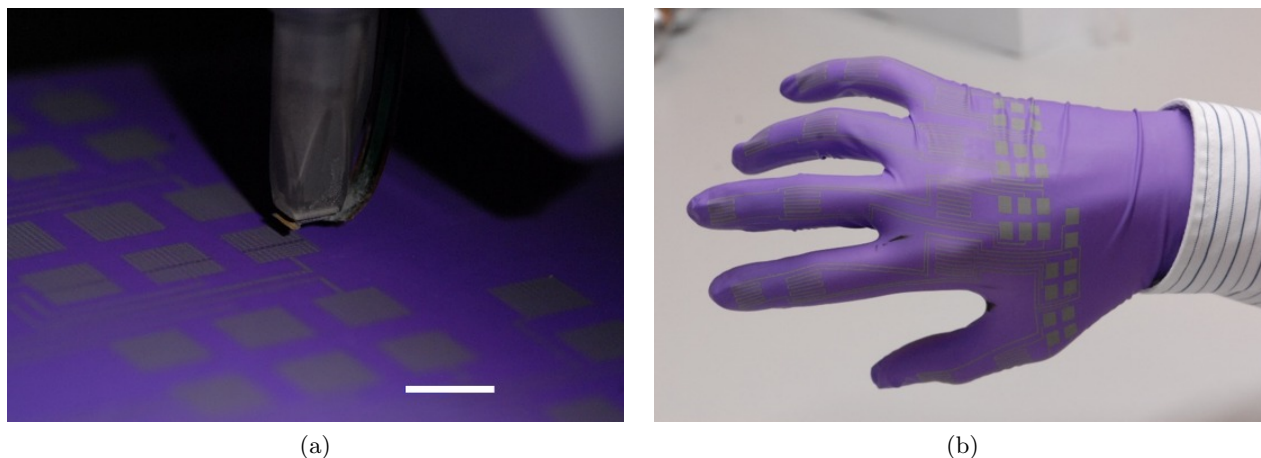


Figure 4. (a) Ink-jet systems printing liquid-metal particle dispersion. Scale bar is 10 mm. (b) Human hand wearing ink-jet functionalized nitrile glove with arrays of strain gauges, intricate wiring, and contact pads comprised of liquid-metal. Adapted from Boley, *et al.*⁶⁸

To address this point, our group is exploring several complementary processing approaches for viscous and high surface tension liquids. These approaches bridge the gap between well-established scalable liquid processing, such as printing, and the processing of emerging soft functional materials that exhibit high surface tension, viscosity, and density properties that typically preclude printability.

3.1 Ink-Jet Printing

Printing is a prominent technology enabling ultra-low-cost, large-area electronics. There has been quite a bit of work utilizing ink-jet printed metal nanoparticle dispersions, where a sintering process must be employed to melt together the metal particles to form conductive traces.^{61–66} However, heated sintering may not be compatible with many integrated devices, such as actuators, sensors, or batteries. Furthermore, the resulting printed conductors are brittle at room temperature and incompatible with applications that require stretchability. We have found that in the context of ink-jet printing, the combination of viscosity, surface tension, and density alone suggest eutectic gallium-indium to be a non-printable fluid without modifying either the material or the atmosphere. Additionally, the fast-forming skin-like oxide can easily clog a nozzle orifice, presenting additional challenges during any small-scale printing or extrusion processes.

We have circumvented this issue by dispersing gallium-indium in a non-metallic carrier solvent. As demonstrated by Hohman *et al.*,⁶⁷ stable ethanolic nanoscale liquid-metal colloids can be produced using sonication, which induces mechanical separation to form liquid-metal nano-droplets due to the presence of high oscillating shear forces (Figure 3). Via this approach, the properties of the base solvent are exploited for processing and the self-healing (coalescing) properties of gallium-indium droplets for patterning.

The dispersion is compatible with ink-jet printing, especially at low concentrations (Figure 4). Once deposited, the encasing gallium-oxide skin prohibits spontaneous coalescence and surface wetting. These non-coalesced films are not conductive due to high interface resistance between the metal droplets (sources of losses include reduced conductivity of gallium-oxide, small contact areas, and electron scattering across oxide interfaces). Given the

small size of the liquid-metal particles, there are many such interfaces in a macro-scale area, leading to significant resistance. However, the particles may be joined by removal or fracture of the oxide skin encasing each particle using both physical and chemical techniques. This is a unique property of the system, as it allows devices to act as switches, binary pressure sensors, or for sub-devices to be activated within larger printed areas.

3.2 Controlled Wetting

Perhaps among the most exciting opportunities with liquid-metal is the ability to reconfigure its shape for reconfigurable electronics and structural components. In Cumby, *et al.*,⁴⁶ reconfigurable circuits were demonstrated using mercury. However, transferability of these results to non-toxic gallium-alloys is challenging due to the adhesive nature of gallium-oxide. Indeed, the difference between receding and advancing contact angles of gallium-based liquid-metal on a silicone-based surface suggests that the adhesion of the oxide layer to the surface is stronger than the wetting of the surface by the gallium-alloy. Our group and others have shown that this oxide adhesion plays an integral role in the formation of stable liquid-metal structures on a substrate.^{42,49} By controlling the presence and formation of the oxide, adhesion of liquid-metal to a polymeric substrate may be controlled, yielding the possibility of reconfigurable devices that may adapt their electronic, mechanical, and structural properties to the demands of their environment.⁶⁹⁻⁷²

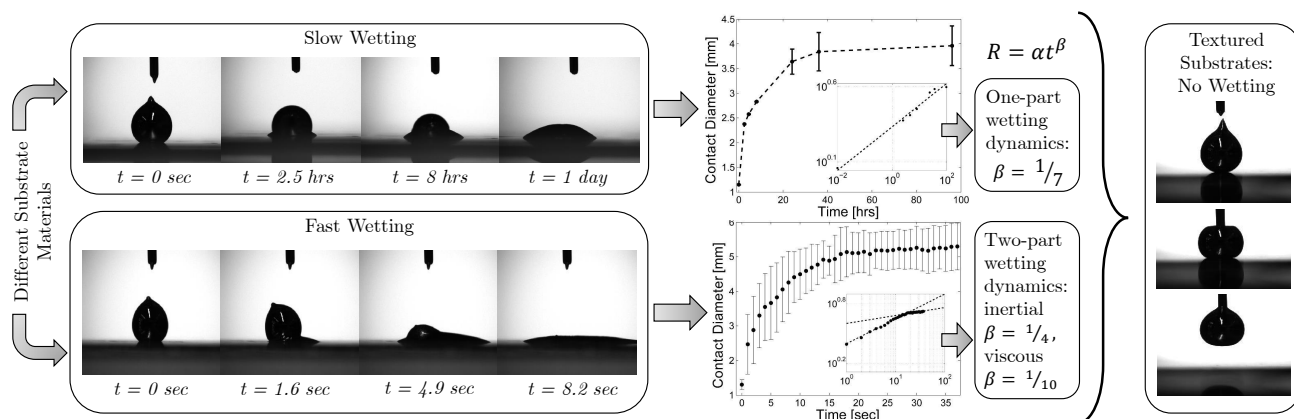


Figure 5. Liquid-metal wetting dynamics may be controlled via tuning of surface composition and texture. Adapted from Kramer, *et al.*⁶⁹

4. OUTLOOK

Scalable manufacturing techniques for soft materials will bridge the gap between the more mature manufacturing methods for low-viscosity liquids and rigid solids, enabling future electromechanical devices to transition from heavy, rigid, and expensive to soft, conformal, and inexpensive. This paper focuses on liquid-metal-embedded elastomers, which are comprised of two materials that fall within the gap and are typically thought to be incompatible with scalable manufacturing processes. Liquid-metal-embedded elastomers present unique opportunities in soft robotics, along with substantial weight reductions, reduced cost, and improved performance beyond what is currently technologically feasible with rigid systems. Specifically, we foresee applications in electrical and signal connectivity, wearable systems, and human-machine interaction.

Connectivity and Wire Harnessing. Most modern electronic devices require conductive interconnects for power and signal transmission. While traditional wire-based interconnects are sufficient for static applications, many modern applications, such as deployable, distributed power generation, require more flexibility. The flexibility provided by a liquid-metal based system has several advantages. First, it allows the system designer more freedom in routing signals through an environment with changing geometry. Second, it lessens the fatigue issues associated with repeated bending of traditional metal conductors. Third, a liquid-metal-embedded elastomer is more robust in high-strain applications and less sensitive to vibrations than its rigid counterparts. Together, these advantages make liquid-metal-based connectors and wire harnesses suitable for a range of emerging applications.

Wearable Systems. Wearable systems pose a significant challenge to traditional conductor technologies. A wearable system must accommodate a range of wearers of different sizes, and must remain conductive as they move without interfering with their motion. Many bending cycles will be induced in conductors traveling across the wearers' joints. Liquid-metal-embedded elastomers provide a solution to this challenge, with hyperelastic deformations and virtually unlimited cycle life. Examples of wearable systems might include ridged components such as radios, batteries, and computers integrated with soft components such as keypads, displays, and antennae. Liquid-metal-embedded elastomers could also be used to enable conformable sensory skins for health-monitoring or dynamic feedback to assist with motions and prolong endurance without restricting the natural mechanics of motion.

Human-Machine Interaction. Systems made out of hyperelastic materials, including liquid-metal-embedded elastomers, have the potential to embed safety a fundamental level – the material level – to create safe human-robot interactions. Currently, sensors and human interface devices are thought of as separate systems, requiring separate physical structures, power supplies, sensors, etc. However, this requirement could be reduced or eliminated through the use of sensory skins. For example, a robot might use a particular sensor-imbued region on the surface both to measure the environment and to interact with an operator through touch gestures. These sensory skins could either be designed into new systems, or applied to the surface of existing systems, resulting in improved capability with little to no modification of the underlying structure.

5. CONCLUSION

A future exists where machines safely work hand-in-hand with humans, spurring new levels of mobility, productivity and quality of life. These mechanical devices will perform services for the physically impaired and the elderly, provide intelligent prosthetic and orthosis for the disabled, maintain and repair our infrastructure, bolster efficiency in manufacturing and farming industries, and be instrumental in disaster relief and clean-up. If we are to realize this future, it will require going further than merely prototyping individual components. Rather, new scalable manufacturing techniques for soft, responsive materials will allow the full potential of soft robotics to be realized.

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