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Gallium Liquid Metal: The Devil's Elixir

Shi-Yang Tang,¹ Christopher Tabor,²
Kourosh Kalantar-Zadeh,³ and Michael D. Dickey⁴

¹Department of Electronic, Electrical, and Systems Engineering, University of Birmingham, Birmingham B15 2TT, United Kingdom

²Materials and Manufacturing Directorate, Air Force Research Laboratory, Wright Patterson, Ohio 45433, USA

³School of Chemical Engineering, University of New South Wales, Sydney, NSW 2052, Australia

⁴Department of Chemical and Biomolecular Engineering, North Carolina State University, Raleigh, North Carolina 27695, USA; email: mddickey@ncsu.edu

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Abstract

Gallium is a metal that literally melts in your hand. It has low toxicity, near-zero vapor pressure, and a viscosity similar to water. Despite possessing a surface tension larger than any other liquid (near room temperature), gallium can form nonspherical shapes due to the thin, solid native oxide skin that forms rapidly in oxygen. These properties enable new ways to pattern metals (e.g., injection and printing) to create stretchable and soft devices with an unmatched combination of mechanical and electrical properties. The oxide skin can be transferred to other substrates and manipulated electrochemically to lower the interfacial tension to near zero. The reactivity of gallium can drive a wide range of reactions. The liquid state of gallium makes it easy to break into particles for making colloids and soft composites that have unusual properties due to the deformable nature of the filler. This review summarizes the truly unique and exciting properties of gallium liquid metals.

1. INTRODUCTION

Galinstan: a brand name that is nonetheless generally used to describe low-melting-point alloys containing Ga, In, and Sn, for example, 68.5% Ga, 21.5% In, and 10.0% Sn (by weight). The melting point of such a composition is often reported at -19°C , but we cannot find any original source for this number and suspect it may reflect supercooling, with an actual melting point closer to 10°C

Liquid metal (LM): here, this generic term excludes Hg or higher-melting-point metals. Instead, we use it to refer to liquid Ga and Ga alloys since most of these metals have similar physical properties to a first approximation (low viscosity and metallic conductivity). In most cases, the native oxide on these LMs is Ga oxide

Gallium (Ga) is perhaps the most interesting and unique element on the periodic table. Dr. Oliver Sacks, a popular neurologist and science historian, claimed that Ga—an element discovered in 1875 and named after the Latin word for France (Gallia)—was the first element that came to mind when he tried to identify a favorite element (1). Sam Kean, the *New York Times* best-selling author, chose the title *The Disappearing Spoon* in reference to Ga for his book on the history of the periodic table. Recently, the American Chemical Society highlighted work using Ga as one of the top research stories of the year (2), and another perspective article noted that such materials are experiencing a renaissance (3). Literature references to such materials have grown exponentially (with approximately 21% annual growth) over the past decade (4). So what makes Ga so interesting?

Ga is a posttransition metal that resides directly below Al on the periodic table and thus has several similarities, with one notable difference: melting point. Ga melts at 29.8°C , making it a liquid on a warm day with a water-like bulk viscosity (viscosity 1.99 cps versus 1.00 cps for water) (5). The low melting point is thought to relate to the mixed covalent and metallic bonding found in solid Ga in which dimeric species of Ga affect phase behavior (6). Ga can be combined with several other metals such as In, Sn, Zn, Cd, and Pb to form alloys with even lower melting points; the lowest is Galinstan, a mix of Ga, In, and Sn, with a melting point near 10°C . Despite having a low melting point, Ga boils at $2,400^{\circ}\text{C}$. Remarkably, Ga has effectively zero vapor pressure at room temperature, thereby eliminating concerns about breathing it, which, paired with its low toxicity (7) and antimicrobial properties (8), allows it to be handled safely. The near-zero vapor pressure also allows it to be used in high vacuum without risk of evaporation (9). Like most molten metals, Ga supercools, which means it can be cooled significantly below its melting point before freezing. One study measured 58°C of supercooling (10). Like water, Ga expands when it freezes. As a consequence, the electrical conductivity decreases when Ga freezes, which is atypical of metals.

Given these remarkable properties, why is Ga often overlooked aside from its use in semiconductors (e.g., GaAs, GaN)? This situation can likely be attributed to several factors. Despite being relatively earth abundant, Ga costs $\sim \$0.25/\text{g}$ (2020 price) since it is produced by extracting Ga oxide as an impurity found in alumina. Plus, there is an overgeneralized historical association of liquid metals (LMs) with toxicity (e.g., the case of mercury). However, probably the most pragmatic explanation as to why Ga is not widely employed commercially is a result of its surface reactivity. A paper published in *Science* in 1954 stated: “Despite all precautions, this gallium electrode always behaved erratically” (11, p. 390). We now know that this “erratic” behavior is the result of the high reactivity of Ga with its environment. This reactivity most typically manifests itself in the formation of an $\sim 1\text{--}5\text{-nm}$ -thick native oxide that affects the surface chemistry, wetting, and rheology of Ga. Finally, Ga may get overlooked because it can diffuse into the grain boundaries of certain solid metals, such as Al, leading to significant degradation of mechanical properties.

Researchers have just recently begun to uncover the complexity of the Ga surface reactivity and to harness it for a rapidly expanding array of applications. The ability to manipulate and control LM and its interface can lead to a wealth of electrochemical, electromechanical, thermomechanical, catalytic, and biochemically responsive properties.

This review seeks to highlight exciting aspects of LM research to illustrate why it is experiencing a renaissance. We use the generic term LM throughout because most LMs containing Ga have similar properties to a first approximation (12). We focus on research that provides

conceptual demonstrations. Aspects of LMs have been reviewed elsewhere (13–28); throughout this article, we point the reader to relevant reviews in context while briefly covering the topics here.

2. OVERVIEW

Figure 1a depicts a tree with roots representing the key properties of LMs (discussed briefly in the introduction). The trunk of the tree represents the common embodiments of LMs (bulk fluidic structures, particles, and composites) that have been used in various applications (the foliage). We organize the remainder of this review around these three embodiments while noting that some applications use more than one embodiment.

3. THREE TYPICAL EMBODIMENTS OF LIQUID METALS

3.1. Bulk (Fluidic) Embodiments

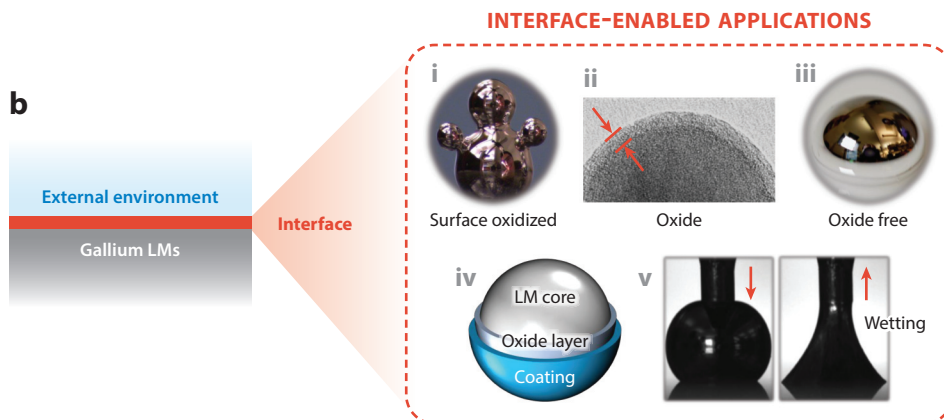
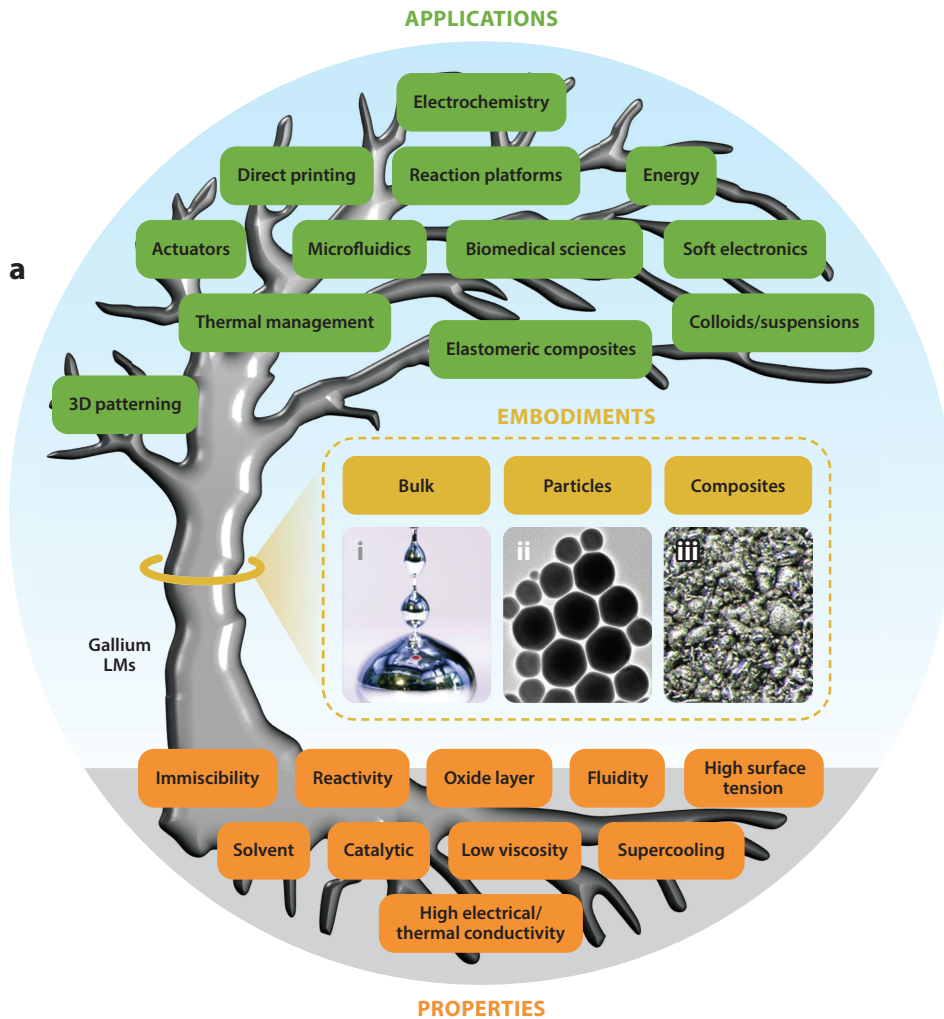
Figure 1a,i shows a photograph of a bulk volume of LM. Here, bulk embodiments (typically) refer to a single, continuous volume or stream of LM. Bulk LM can provide metallic properties (electrical, thermal, optical, chemical) while providing fluidic attributes (softness, flowing, wetting, freezing/melting). A bulk embodiment does not have to be large in scale. In fact, it is important to consider the importance of forces at different length scales to understand the importance of interfacial forces relative to body forces such as gravity. Gravitational forces acting on bulk LM can overcome interfacial forces and cause the metal to flow laterally when a LM volume is taller than ~ 3 mm, the so-called capillary length. Thus, LM is typically confined in a container unless it is used at small, submillimeter length scales and/or small volumes.

Bulk embodiments often take advantage of the properties of LM (e.g., low viscosity, high density, enormous surface tension, metallic conductivity) in conventional fluidic applications because these properties are significantly different from common fluids such as water or oil. As a conductive fluid, it allows for the formation of soft and stretchable electronic components encased in an elastomer. Examples include wires, antennas, and circuits (15, 29). LM can also make gentle electrical contacts on surfaces at room temperature (30). In electrolyte, the electrically conductive LM interface can generate Marangoni forces (gradients in tension) for pumping and mixing by applying potential gradients (31). The high, yet electrically tunable, surface tension and high density allow for exploration of fluid phenomena at dimensionless numbers that differ from common fluids, such as water, without the need to change the length scales or velocities of fluid streams (32, 33). Pumping LM is useful for heat transfer and heat switches due to the metallic thermal conductivity of LM (34). Likewise, LM can change shape to enable physically reconfigurable circuits (35).

3.2. Particle Embodiments

Figure 1a,ii shows an image of LM particles. LMs, because of their low viscosity, can easily be broken up into colloidal droplets via a number of routes. Such particles were recently reviewed (27), including their use in the realm of nanotechnology (17). Once formed, particles do not readily merge back together, even in the absence of surface ligands, as long as the native oxide reforms on the particles.

Particles are an interesting embodiment because they provide high surface area (which is useful for catalyzing or initiating reactions), plasmonic optical properties (at very small length scales),



(Caption appears on following page)

Figure 1 (Figure appears on preceding page)

(a) Properties, typical embodiments, and applications of Ga liquid metals (LMs). The roots represent the key properties of LMs, the trunk represents key embodiments of LMs, and the branches represent applications of LMs that build on these properties and embodiments. The three typical embodiments are (i) bulk, (ii) particles, and (iii) composites. (b) A common thread that unites LM research is the importance of interfaces. The inset shows the importance of the interface for enabling most applications. (i) LM droplets (with diameters less than 1 mm) are extruded from a nozzle and “stacked” on top of one another. The presence of an $\sim 1\text{--}3\text{-nm}$ -thick surface oxide layer allows the liquid to maintain this snowman shape, despite being a liquid with enormous surface tension. (ii) The oxide, which is between the red arrows, is evident on a LM particle imaged by a transmission micrograph. (iii) In the absence of the oxide layer (removed by 1 M HCl), the metal beads up due to the large surface tension of the bare metal. (iv) In air, LM consists of a liquid core surrounded by a native oxide shell, which can be chemically coated or functionalized (not to scale). (v) The process of advancing and receding LM onto a substrate illustrates the role of this oxide in setting interfacial behavior. Without the oxide, LM would not adhere to most substrates. The downward pointing red arrow denotes liquid being pumped out of the nozzle while advancing, and the upward arrow denotes liquid being retracted back into the nozzle while receding.

and interesting phase behavior (including enhanced supercooling at small particle sizes). They also offer a metallic alternative to conventional polymeric or oxide colloids while retaining soft mechanical properties.

3.3. Composite Embodiments

Figure 1a,iii shows a photograph of a composite consisting of LM filler in a polymer matrix. LMs are unique fillers for composites for at least two reasons. First, whereas rigid fillers stiffen composites, LMs can be added to change certain properties of elastomers (dielectric constant, electrical conductivity, thermal conductivity) while maintaining mechanical properties that are soft and stretchable. Second, LM fillers can change shape when the composite is deformed, which can lead to unique strain-dependent properties. Recent reviews highlight LM composites (16, 19).

4. THE ROLE OF THE INTERFACE: THE DEVIL'S ELIXIR

The Nobel laureate Wolfgang Pauli said, “God made the bulk; the surface was invented by the devil” (36, p. 291). If this statement is true, the devil may be smiling because most applications and embodiments of LM are interface enabled. While the native oxide has led to a great deal of frustration in the past from researchers wanting a simple nontoxic drop-in replacement for mercury (11), here we show that as the oxide has become better understood, these interfacial properties are becoming ever more useful.

4.1. Reactivity

Ga is inherently reactive since it readily gives up electrons (oxidizes) to form Ga^{3+} , the most stable oxidation state of Ga. Ga LM can react with ambient molecules to form a self-limiting (passivating) film (**Figure 1b,ii**). In air, this process results in the nearly instantaneous formation of a native oxide at the interface, typically in the form of $1\text{--}5\text{-nm}$ -thick Ga_2O_3 , which is likely an amorphous or poorly crystallized film. There are a number of ways to functionalize the outer surface of LMs with small molecules or macromolecules to form a coating layer and either utilize or modify this reactivity to suit one's needs (18, 27) (**Figure 1b,iv**). In other environments, such reactivity can lead to redox reactions and changes in composition and can also initiate polymerization (37). LM can also be used as a nonfouling catalyst or electrocatalyst due to poor adhesion between the metal interface and carbonaceous products and by-products (38).

4.2. Interfacial Tension and Shaping Liquids

The bare metal has the largest tension of any liquid at room temperature (>700 mN/m) (39) yet can form metastable shapes in ambient conditions because of the formation of the surface oxide. **Figure 1b,i** shows a photograph of stacked droplets that maintain their shape due to the oxide shell. This oxide is amphoteric; that is, it can be removed using strong acids or bases. In the absence of oxide, surface tension drives the stacked droplets to combine into a sphere (**Figure 1b,iii**). Surface segregation of alloy species [e.g., In in the case of oxide-free Ga-In (40)] and surface layering (41) can further complicate the interfacial behavior.

4.3. Rheology

The oxide also impacts the rheology of LM. Due to the low viscosity of the metal, it can flow easily as long as the applied pressure is sufficient to rupture the oxide layer. In ambient conditions, the pressure required to break the native oxide scales with the interfacial curvature multiplied by the surface yield stress, 0.2–0.6 N/m; for comparison, the interfacial tension of water is 0.072 N/m (12, 40). These attributes imbue the oxide-coated LM with yield stress behavior (42). Above this stress, the metal flows and can be injected, sprayed, smeared, and extruded. Below this critical stress, the oxide allows the metal to maintain its shape, which is important for patterning liquids.

4.4. Wetting

The oxide affects and complicates the wetting behavior. Bare LMs have large contact angles on surfaces (that is, they do not wet them), with the exception of some metals, in which case the wetting is reactive due to metal-metal interactions. In contrast, oxide-coated LM adheres to nearly all flat surfaces. Advancing the metal against surfaces helps improve adhesion due to the formation of new oxide, which presumably makes more conformal contact with the substrate (43). The advancing angle is typically large, regardless of the substrate chemistry, due to the need to rupture the oxide in tension. Yet the oxide causes the metal to pin to the surface during receding, resulting in huge contact angle hysteresis (**Figure 1b,v**). The oxide's presence lowers the interfacial tension of the metal at the metal-oxide interface, yet as a thin and brittle solid, it can break, reform, and wrinkle. A few strategies have been developed to overcome this adhesion of the oxide, including using rough surfaces (44, 45) and thin liquid lubrication layers (46) and functionalizing the surface of the oxide (47).

5. IMPLICATIONS, PROCESSES, AND APPLICATIONS OF LIQUID METALS

5.1. Advanced Manufacturing and Applications of Bulk (Fluidic) Liquid Metals

The fluid nature of Ga allows it to be used and processed in a variety of ways that are not possible with conventional, solid metals. **Figure 2** shows several examples of ways to pattern Ga LMs that uniquely take advantage of the liquid nature, as reviewed in depth elsewhere (14, 22).

5.1.1. Patterning. LM can be injected into capillaries—including those formed via 3D printing—by applying a pressure differential to yield the oxide. The pressure can be applied using positive pressure (40) (**Figure 2a**) or even more simply by vacuum filling (48) (**Figure 2b**).

LMs can also be patterned additively, as reviewed recently (28). Direct writing with LM can be achieved by bring the nozzle in close enough proximity to a substrate. The oxide adheres to many substrates, and subsequent movement of the nozzle parallel to the surface creates LM patterns

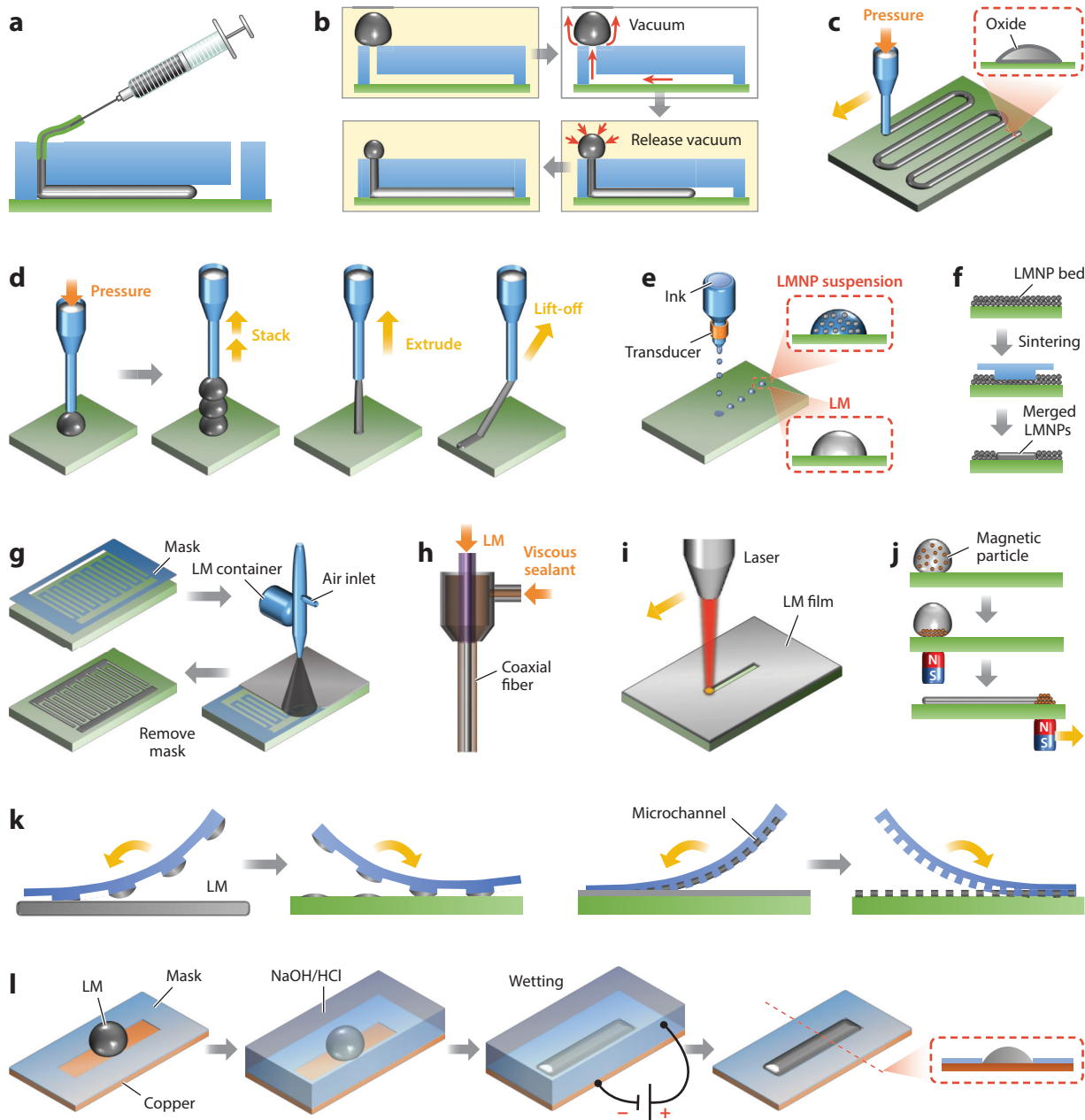


Figure 2

Liquid metals (LMs) can be patterned in unique ways relative to solid metals. (a,b) Injecting (a) and vacuum filling (b) of LMs into a microfluidic channel. (c,d) Direct writing (c) and additive patterning (d) of LMs onto a substrate. (e) Inkjet printing of LM or LM nanoparticle (LMNP) suspension onto a substrate. (f) The printed LMNP bed can be rendered conductive by sintering the particles (rupturing them so they merge). (g) Spray printing of LM onto a stencil. (h) Extruding concentric streams of polymer and LM to form fibers. (i) Laser ablation of LM films. (j) Magnetically dragging a magnetic LM droplet across a surface to leave behind traces of LM. (k) Printing LM using topographical molds featuring selective wetting patterns. (l) Inducing wetting of LM on existing Cu traces by reducing the oxide layer.

(Figure 2c). Droplets naturally form by extruding metal from a nozzle but can be rendered into wires by applying tensile or shear force to the oxide shell (49) (Figure 2d). This process requires adhesion of the oxide-coated metal to the underlying substrate. Stresses arise by moving the nozzle relative to the substrate in tension or shear (28). Interestingly, recent work has shown that small, printed structures can be lifted off the substrate and moved to another location for reconfigurable printing (50). The conductivity of LM also allows for application of voltage between the LM and the substrate to control and enhance wetting during printing (51).

Although the tension of LMs is too large for conventional inkjet printing, LM nanoparticles (LMNPs) can be printed in a carrier solvent such as ethanol (52) (Figure 2e). Such structures are not inherently conductive. Yet LMNPs can merge by mechanical sintering, which occurs by simply applying a compressive force (52) (Figure 2f). Lasers can also cause sintering (53). The pressure required to sinter the particles can be tuned by modifying the oxide shell (54). Similar mechanical sintering can be completed by applying tension on covalently linked particles (55). These techniques can result in electrical behavior that demonstrates invariable resistance to strain, as the connected path that forms is hypothesized to form a tortuous, contiguous 3D morphology. LMs can also be spray painted over a stencil (56) (Figure 2g). Although liquids typically exit nozzles as droplets, wires can be formed by coextruding a coaxial stream of polymer that encapsulates the metal (57) (Figure 2h). Subtractive patterning is also possible; with sufficient energy, lasers can ablate portions of LM films (Figure 2i) to create features small enough to make transparent conductive surfaces (58, 59).

LM can also be printed using a configuration similar to a ballpoint pen (60). Similarly, by dispersing magnetic particles in LM droplets, it is possible to magnetically drag such droplets across a surface to leave behind traces of LM (61) (Figure 2j). This approach does not have the same resolution as the others discussed but is distinguished by being a completely noncontact approach for patterning.

Whereas nozzle-based printing techniques are serial, other additive techniques can print over larger areas in parallel. Stencil printing of LMs that relies on the adhesion of LMs to exposed areas of a substrate has been demonstrated (62, 63). Stamping techniques can pattern LMs using either a topographical mold [recesses or protrusions (Figure 2k)] or a stamp featuring selective wetting patterns (64, 65). Imprinting molds have created structures as small as 1 μm . LMs can also selectively wet or deposit on existing metal traces such as Cu or solid metal particles to create biphasic conductors (66–68) (Figure 2l).

5.1.2. Applications. LMs are well suited for a variety of microfluidic applications, as illustrated in Figure 3 and highlighted in a recent review (20). LM electrodes in microchannels are particularly useful. For example, injecting LM into microchannels at room temperature can make well-defined, soft electrical contacts to self-assembled monolayers or underlying electronic materials (69, 70). Caps of LM affixed to Cu electrodes can induce alternating Marangoni flows for mixing adjacent fluids at submillimeter length scales (71) (Figure 3a) or promote heat transport (31, 72) (Figure 3b). Injected LM electrodes, shown top down in Figure 3c, can span from the top to the bottom of a microchannel, which is a challenging geometry to form via conventional microfabrication. These electrodes can apply electric fields that can amplify small undulations at the interface of colaminar streams to induce mixing (73). Electric fields can also induce dielectrophoretic separation of microdroplets or particles from a stream (74) (Figure 3d). The electrodes can also be used for biological applications, such as stimulating neurons grown in microchannels (75).

In addition to LMs forming electrodes, the properties of LMs can be harnessed for other microfluidic applications. Patterned LMs within an elastomer can be removed as a fugitive ink to create small microchannels or cavities with optically smooth interfaces (76) (Figure 3e). The

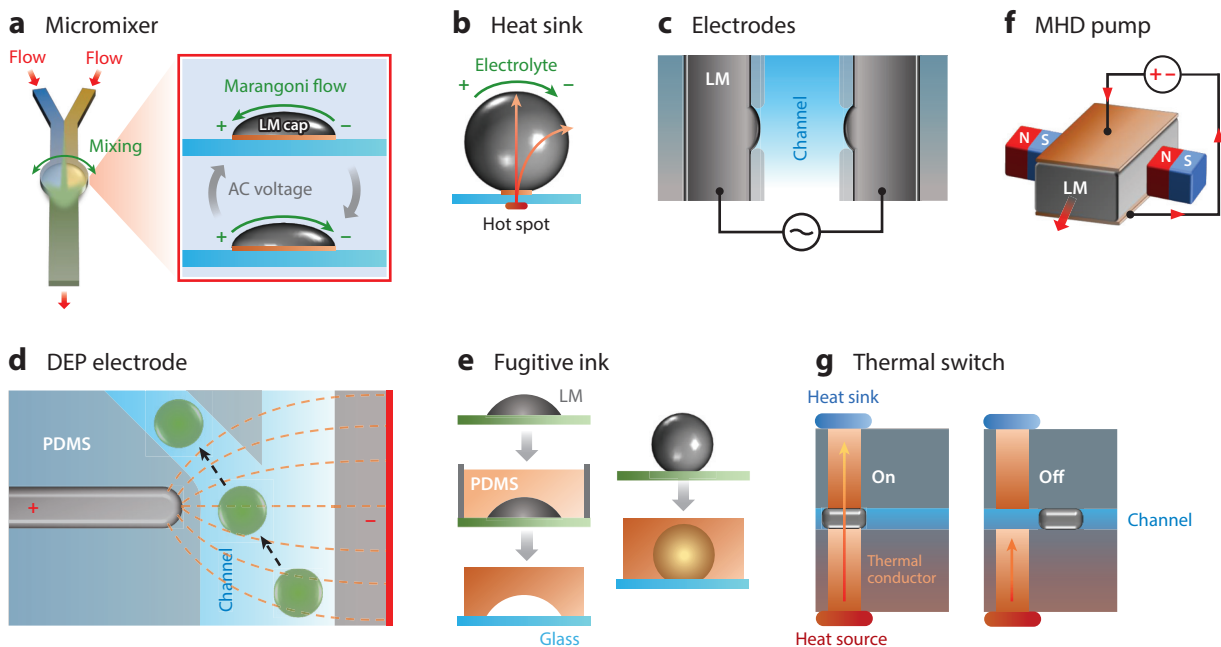


Figure 3

Applications enabled by LM in microfluidics. (a) A LM-enabled micromixer using alternating Marangoni flows for mixing laminar flows. (b) Marangoni flows resulting in improved heat transfer. (c) Top-down view of soft 3D microelectrodes formed by injecting LM into microfluidic channels. (d) DEP LM electrode for sorting microdroplets or particles. (e) Microfabrication of smooth molded surfaces using LM as fugitive inks. (f) Pumping LM using MHD. (g) A thermal switch realized by moving a slug of LM within a microchannel. Abbreviations: DEP, dielectrophoretic; LM, liquid metal; MHD, magnetohydrodynamic; N, north; PDMS, polydimethylsiloxane; S, south.

high electrical conductivity of LMs can handle the large currents needed for magnetohydrodynamic pumping (77) (**Figure 3f**). LM plugs can also be moved by pumping in a carrier fluid or by continuous electrowetting (25). These moving slugs of metal can make a thermal switch (34) (**Figure 3g**).

The ability of liquids to move, conduct electricity, and conform to surfaces can be harnessed to convert mechanical energy to electricity and vice versa. For example, electrowetting uses electric fields to move droplets. Reverse electrowetting instead uses mechanical energy to move a droplet of metal into gaps between insulated electrodes. The presence of the metal changes the capacitance between the rigid electrodes and thereby induces charge from a battery to move into the capacitor (78, 79). As the metal moves away from the electrodes, the capacitor discharges back to the battery, generating electricity (**Figure 4a**). Dipping dielectric objects into LM can induce tribocharging, which can be harnessed to generate electricity (80) (**Figure 4b**). LM electrodes have also been used in stretchable tribocharging devices (81). Finally, applying a potential drop across LM in electrolyte can create an interfacial tension differential via a process known as continuous electrowetting (25). This gradient in surface tension can cause the metal to move. **Figure 4c** shows that this movement can be used to drive robotic systems directly or along a circular path to change the center of gravity inside a wheel, thereby causing the wheel to turn (82). This design is a type of simple motor that transduces electricity into mechanical movement. Finally, LMs can be used for batteries, which is discussed in Section 5.5.

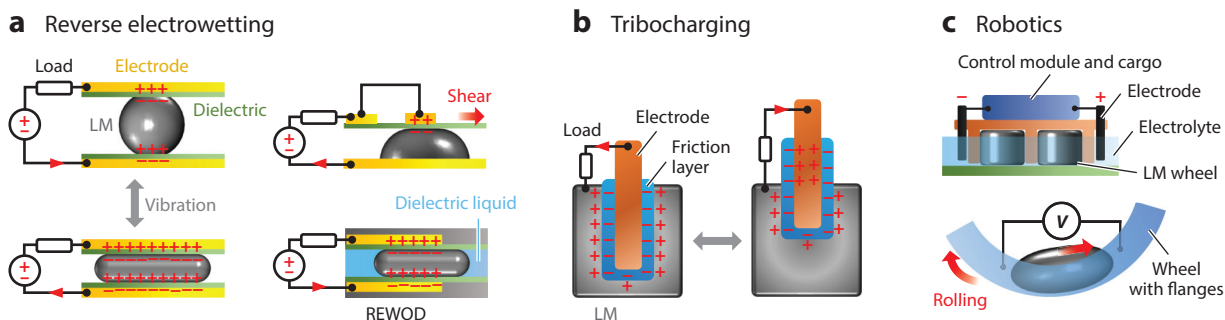


Figure 4

Applications of LM in energy and actuation devices. (a) Energy harvesting using reverse electrowetting. (b) Tribocharging energy harvesting device using LM. (c) Robotic systems driven by LM using the continuous electrowetting effect, in which LM droplets can act as wheels or change the center of gravity of a wheel. Abbreviations: LM, liquid metal; REWOD, reverse electrowetting on dielectric.

One of the most compelling applications of LMs is the ability to create conductors that provide the best combination of stretchability and conductivity (15). The Ashby plot in **Figure 5a** compares stretchability versus conductivity for various conductive elastic materials. While many available material sets have been demonstrated in the literature, the options that provide the best performance for both stretchability and conductivity utilize LMs. **Figure 5b** shows a schematic of a LM conductor encased in elastomer. Although these wires illustrate the principle in a simple geometry, the methods shown in **Figure 2** enable more complex circuitry to be patterned. The wires depicted in **Figure 5c** maintain metallic conductivity to strains limited seemingly only by the encasing polymer [e.g., studies show metallic conductivity up to 700% strain (83) and 1,500% strain (84)]. **Figure 5b** shows that during deformation, such soft circuits change their geometry. Although the resistivity—an intrinsic material property—does not change, the resistance does change due to its dependence on geometry. Geometric changes have been utilized to make soft touch sensors (85), tunable antennas that change their resonant frequency in response to stretching (86, 87), and capacitive sensors of strain and torsion (88). A self-healing elastomer can be used to create stretchable metallic conductors that can be cut completely apart and then reassembled (89) (**Figure 5d**).

Ga melts near room temperature and can therefore be used in applications that benefit from the huge changes in mechanical properties during phase change. So-called variable stiffness materials are of interest for a variety of applications, including soft robotics, due to their similarities to muscle, which can transform from a soft, extensible state to a rigid, load-bearing state. Perhaps the simplest example is an elastomeric substrate filled with LM. In the frozen state, the metal stiffens the composite, but when the metal melts, the composite takes on the mechanical properties of the elastomer (90). The ability to change stiffness can be utilized for shape memory materials (**Figure 5e**). The elastomer fiber can be stored in an extended state by freezing the metal. When the metal melts, the extended elastomer drives the fiber to return to its original state. This approach can result in a huge change in effective modulus ($>700\times$) and very rapid shape change. This transition between mechanical states can be tuned by modifying the composition of the LM alloy and therefore the phase transition of the LM from solid to liquid. The use of LM for variable stiffness materials provides additional advantages because LM can conduct electricity, can serve as antennas or electrodes, and can be triggered to melt by Joule or inductive heating.

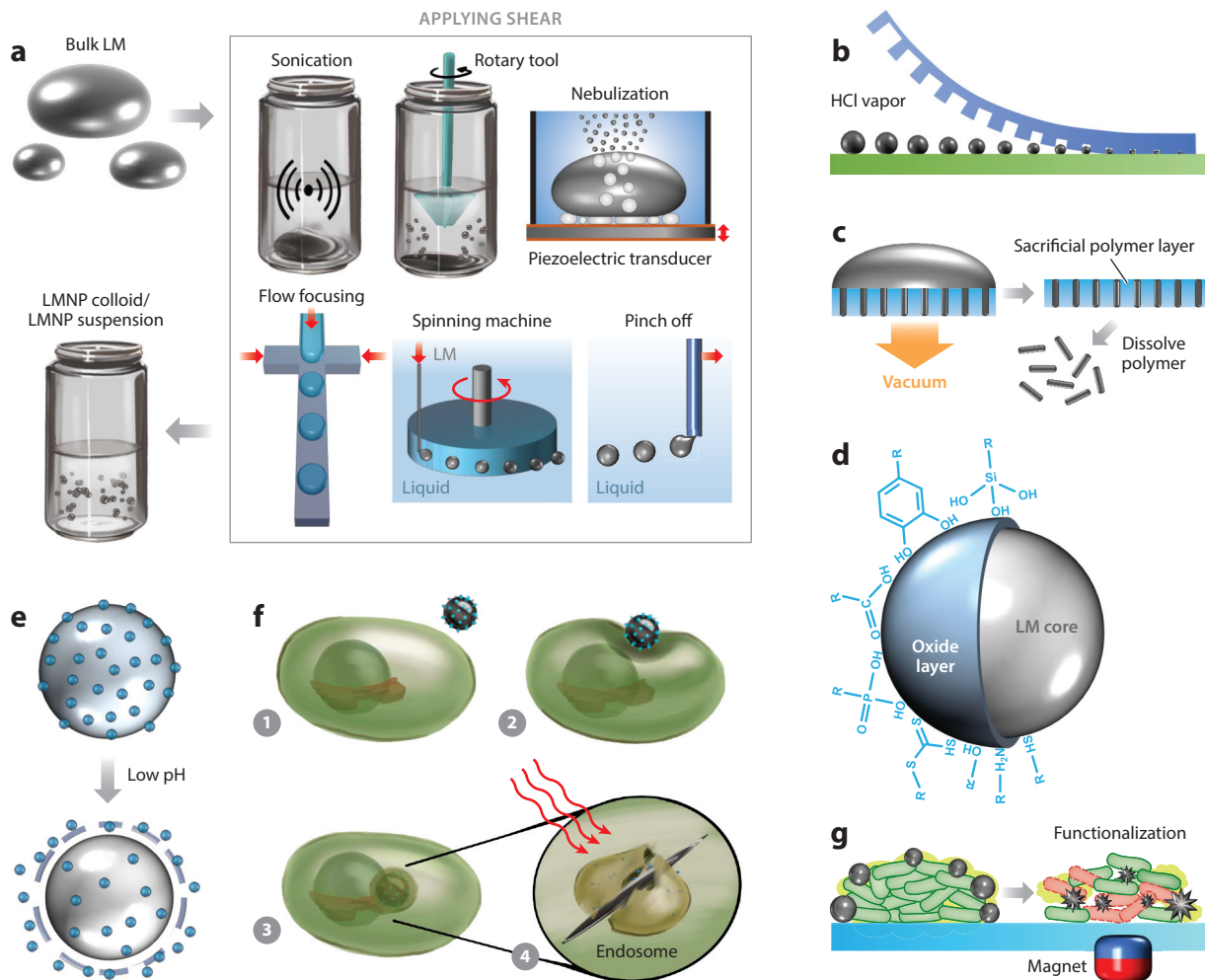


Figure 6

Fabrication and applications of LM particles. (a) Breaking up bulk LM into colloidal droplets by applying shear via routes such as sonication, shearing using high-speed mixing, liquid-based nebulization, flow focusing, and movement of carrier fluids relative to an orifice. (b) Patterning and releasing LM microdroplets from molded structures by using HCl vapor to remove the native oxide. (c) Fabrication of nonspherical LM particles using a sacrificial polymer mold. (d) Use of molecules with different anchoring groups to graft the surface of LMs. (e) Removing the oxide layer at low pH values for releasing surface-attached molecules or drugs. (f) Conversion of soft, spherical LM particles into solid rods of GaOOH for rupturing endosomes upon exposure to light. (g) Destroying bacterial films with LM particles loaded with magnetic materials in response to rotating magnetic fields. Abbreviations: LM, liquid metal; LMNP, liquid metal nanoparticle.

in larger (10–100- μm) particles, the particles can have monodispersed diameters (32). **Figure 6b** shows that molded LM can be released from the mold by using HCl vapor to remove the oxide layer, thereby causing the metal to bead up. A similar strategy can be utilized to mold the metal and then dissolve the mold (**Figure 6c**).

5.2.2. Particle surface functionalization. LM can be surface functionalized in ways that depend on the presence or absence of oxide. Similar to the case of other metals that are free of surface

oxides (such as Au), it is possible to directly form thiol bonds to the bare metal surface (91). In contrast, the oxide presents hydroxyl groups that offer anchoring sites for surface functionalization. Thus, many mature chemistries can be utilized to functionalize oxide-coated LMs and, in particular, high-surface-area LMNPs. **Figure 6d** shows examples such as silanes (92), phosphates (83), catechols (93), and carboxylic acids (27). Hydroxyl-containing polymers such as polyvinyl alcohol can physisorb to the oxide surface (32). Polymers can be grafted to the surface (94), and recent research shows that LMs can even initiate free-radical polymerization (37). Particle functionalization helps promote the colloidal stability of particles (94), helps enable biorecognition events at the interface (95), and helps change the mechanical properties of particles (54).

5.2.3. Responsive and active particles. Particles can respond to changes in pH. The oxide layer on LMNPs is amphoteric and can be removed at high or low pH values. This principle can be used to induce the particles to release the surface coating (**Figure 6e**) and/or recover the particles by having them merge back into a single larger volume of LM. The former has proven useful for drug delivery in which particles that undergo endocytosis experience a change in pH that releases surface-bound drugs (e.g., doxorubicin) inside cancer cells. By also functionalizing the particle surface with targeting molecules for cancer cells, the particles can be selectively endocytosed by those cells (96).

LM particles can also be active. Electrochemical reactions promoted by placing a small piece of Al on a particle of Ga can create tension gradients across the surface of the particle that drive movement of LM through electrolyte (97). Ga can also react with water to form GaOOH, which forms a solid, rodlike crystal (98). Thus, soft, spherical liquid particles can be converted into solid rods. This process can be accelerated by heating the water that surrounds the particles or by adding photoabsorbers (e.g., graphene oxide) that can locally absorb light to heat the particles (99). This principle is shown in **Figure 6f**, which depicts LMNPs first entering a cell by endocytosis. Once the LMNPs are inside the cell, exposure to near-IR light causes them to react to form GaOOH rods that can puncture the walls of endosomes, thereby releasing the carried drugs.

To impart additional responsive properties to LM, a secondary metal particle can readily be mixed into the LM due to metal-metal interactions (100, 101). The fate of the particles—suspension, dissolution, paste, or intermetallic formation—depends on the phase diagram for the species. For example, it can become responsive to magnetic fields by the addition of magnetic particles (102). Recent work has shown that LM particles loaded with a small amount of Fe can spin and form rods in response to a rotating magnetic field (103). These active particles successfully destroy bacterial films, which is a particularly promising physical approach, given the rise of antibiotic resistance of such films (**Figure 6g**).

Other sources of energy can be utilized to activate LM particles. The aforementioned LM rods (**Figure 6c**) can be driven to puncture cells by using acoustic waves (104). LM particles can respond to heating from near-IR laser irradiation (105) and induction (106), which are appealing because both represent types of noncontact methods.

5.2.4. Adding nonwetting particles to liquid metals. Because metal-metal bonds of LMs have high cohesive forces, mixing nonmetallic species into LMs is difficult. The native oxide can facilitate mixing by interfacing LM with other materials (101). Likewise, decorating carbon species with noble metallic particles facilitates wetting with LMs (107). The addition of particles to LM can change the rheological (101, 107), magnetic (102), and thermal (101) properties of LMs.

5.2.5. Particle phase behavior of Ga. Ga particles have interesting phase behavior. When Ga freezes, it has a number of polymorphs at lower temperatures but forms an α phase at room

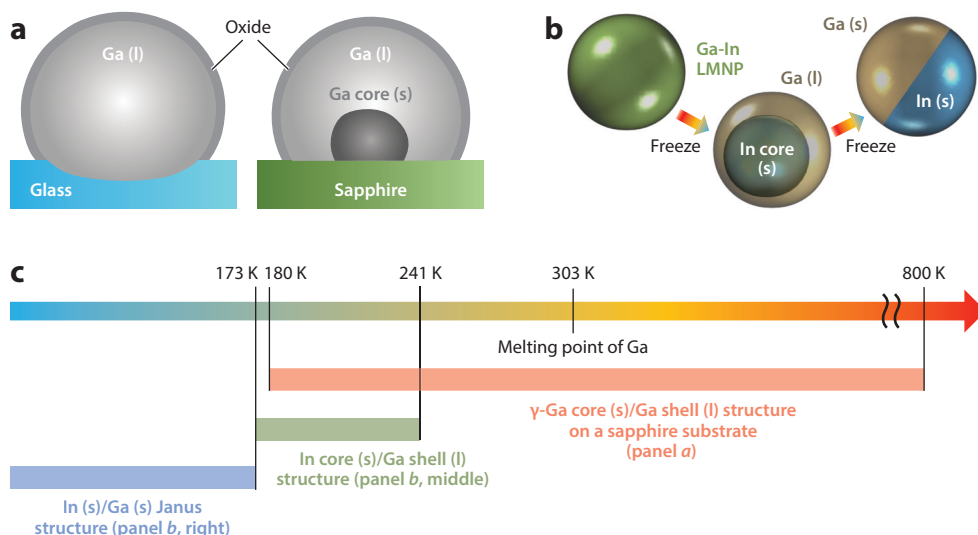


Figure 7

Phase properties of LMNPs. (a) Formation of a solid core when a Ga LMNP rests on a sapphire substrate. When resting on a glass substrate at elevated temperatures, no solid core forms, and the Ga LMNP deforms the glass. (b) Cooling alloys of Ga can cause constituent species to solidify and induce phase separation. (c) LM can exhibit extreme supercooling and superheating (indicated by the colored arrow), depending on the size scale, composition, and materials with which it interfaces. Abbreviations: l, liquid; LMNPs, liquid metal nanoparticles; s, solid.

temperature. However, by placing a Ga particle on a sapphire substrate, a new γ phase that would not normally occur can form during cooling (108) (**Figure 7a**). Remarkably, when formed on sapphire, these solid particles remain stable within a liquid Ga environment over an enormous temperature range from 180 to 800 K (**Figure 7c**). Recall that Ga normally melts near 30°C. The left side of **Figure 7a** shows the deformed equilibrium shape of the nanoparticle on the glass substrate after thermal cycling. The shape is the result of interfacial forces. Interestingly, Ga nanoparticles show extreme supercooling behavior (down to 90 K), and at certain sizes termed magic clusters, they can also exhibit superheating (see Reference 108 and references therein).

Cooling alloys of Ga can cause constituent species to solidify and phase separate in ways that depend on the composition of the alloy. For example, **Figure 7b** shows an example of Ga and In. As the metal cools, the In solidifies, while the Ga remains liquid. Interestingly, by further lowering the temperature, the particles form Janus-like structures in which a portion of the particle is solid In and a portion is solid Ga (109).

5.3. Fabrication and Mechanical Properties of Composites

Here, we refer to LM composites as materials consisting of molten or solidified LM particles dispersed in a continuous matrix (16, 19). In principle, such composites could also consist of LM as the continuous phase with non-LM fillers, although we discuss these types of materials in Section 5.2. LM elastomers (LMEs) are a common subclass of composites that consist of LM particles dispersed in elastomer (110, 111). Although many types of composites are possible, LMEs take advantage of the soft nature of the LM filler and the elastomeric matrix.

Liquid metal elastomer (LME): a binary mixture of liquid metal particles within an elastomeric (often silicone) rubbery network

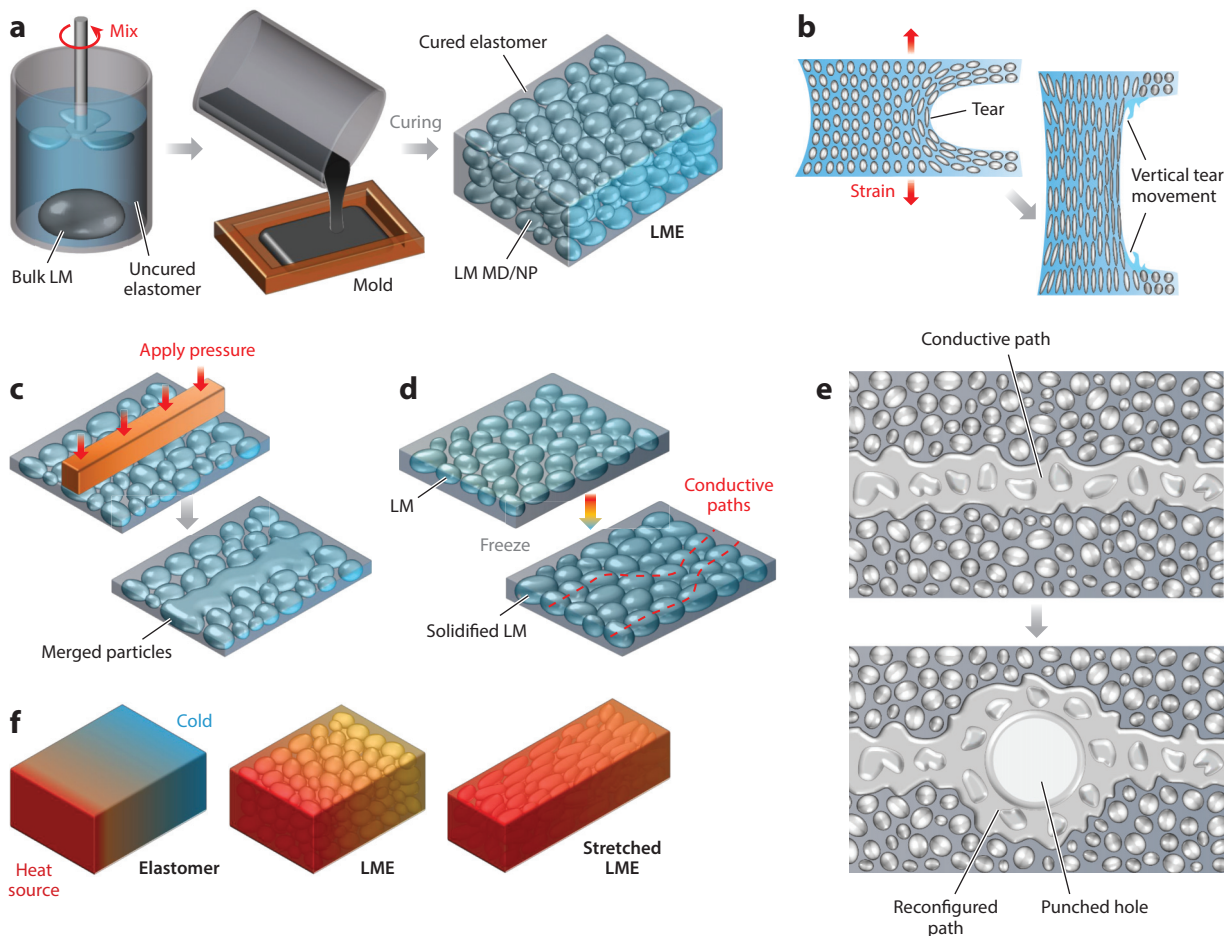


Figure 8

LME composites. (a) General process for fabricating LMEs. The shear from mixing breaks LM into micro- to nanosized particles, and curing the elastomer results in LMEs. (b) Redirecting tears along the direction of strain (red arrows) with the help of LM inclusions, thereby increasing the toughness. (c) Generation of conductive networks by mechanical sintering of LM particles. (d) Formation of a percolated network of particles by using thermal expansion allows composites to switch from an insulator to a conductor. (e) Autonomous self-healing of conductive routes after cutting or puncture. (f) Enhancement of thermal conductivity with the inclusion of LM droplets and the application of strain. Abbreviations: LM, liquid metal; LME, liquid metal elastomer; MD, microdroplet; NP, nanoparticle.

LMEs are easy to form. **Figure 8a** shows how LM and uncured liquid elastomer can be simply mixed together. The shear from mixing causes the metal to break up into particles. Curing the elastomer (typically using heat) results in a soft, stretchable composite consisting of LM particles entrapped within an elastomeric matrix.

Because LM is soft, LMEs have a modulus and elongation that are, to a first approximation, similar to the elastomer that encases the metal. Despite being soft, the LM inclusions can increase the toughness of the elastomer by redirecting tears that would otherwise lead to failure by crack propagation (tearing) in the plane perpendicular to strain (112) (**Figure 8b**).

5.3.1. Conductive composites by percolation and sintering. In most LMEs, a thin wall of elastomer remains between the particles such that they are not percolated. It is, however, possible to mechanically sinter such particles by applying sufficient force to the LME (**Figure 8c**), which causes the thin silicone layers to rupture (110). High metal loadings ensure that percolation occurs during sintering. Recent studies have shown that particles at much lower loadings can be aligned and sintered by dielectrophoresis prior to curing the elastomer (113). Whereas bulk conductors change geometry during deformation, conductors composed of both LMEs (113) and particles (55) have emergent properties wherein the resistance across a trace is seemingly invariant to strain, with up to 700% elongation.

Ga expands when it freezes, which is another unique and interesting property. This expansion can cause particles dispersed in a polymer matrix to slightly enlarge and thereby come into electrical contact (**Figure 8d**). This contact leads to a percolated network of particles and allows composites to switch from insulating to conducting by using temperature (114). When LME is cut or punctured, LM particles smear onto the walls near the cut and thereby provide a route to create self-healing, stretchable conductors (115, 116) (**Figure 8e**).

5.3.2. Deformation-responsive (piezo and anisotropic) composites. LM inclusions can elongate during deformation, resulting in unique changes in LME properties in response to deformation (i.e., piezo properties). For example, LMEs have shown enhanced thermal conductivity in the direction of strain due to the elongation of the metal particles, whereby an $\sim 50\times$ increase in thermal conductivity following strain can be achieved (117). Conversely, this elongation concomitantly narrows the diameter of the metal particles, thereby decreasing thermal conductivity in the plane normal to strain. The enhanced thermal properties of LM fillers were recently used to improve liquid crystal elastomer actuators (118).

The dielectric properties of LMEs are responsive to deformation (119). Building on this concept, LME foams can initially increase the effective dielectric constant when compressed as the air gets displaced from the foam (120) (**Figure 9a**). Upon displacing all the air, additional compression begins to deform the elastomer and the LM particles, resulting in a decrease in dielectric properties. Thus, both positive and negative piezopermittivity (changes in dielectric with deformation) can be achieved.

The conductivity of certain LMEs changes with deformation. Mixing LM particles and certain solid metal particles into elastomer can produce composites with positive piezoconductivity. Normally conductive composites—consisting of solid conductive particles in elastomer—become more resistive when elongated because the particles move further apart and become less percolated. However, LM particles elongate with the elastomer, which contributes to the enhanced conductivity. **Figure 9b** shows a conceptual composite along with a plot that indicates that resistivity (ρ) decreases by orders of magnitude with small amounts of strain (121). This property is very useful for sensors. It is possible to magnetically prealign the solid particles prior to curing, which results in a composite with anisotropic properties; that is, the properties are different in different directions (122). **Figure 9c** shows an example of such a composite in which the conductivity decreases in the direction of alignment with tensile strain, whereas the conductivity increases perpendicular to this direction.

5.4. Interfacial Phenomena, Reactions, and Applications

The interface of bare LM is interesting because it is highly reactive and metallically conductive and has an enormous interfacial energy. Reactivity is emerging as a useful property for catalyzing

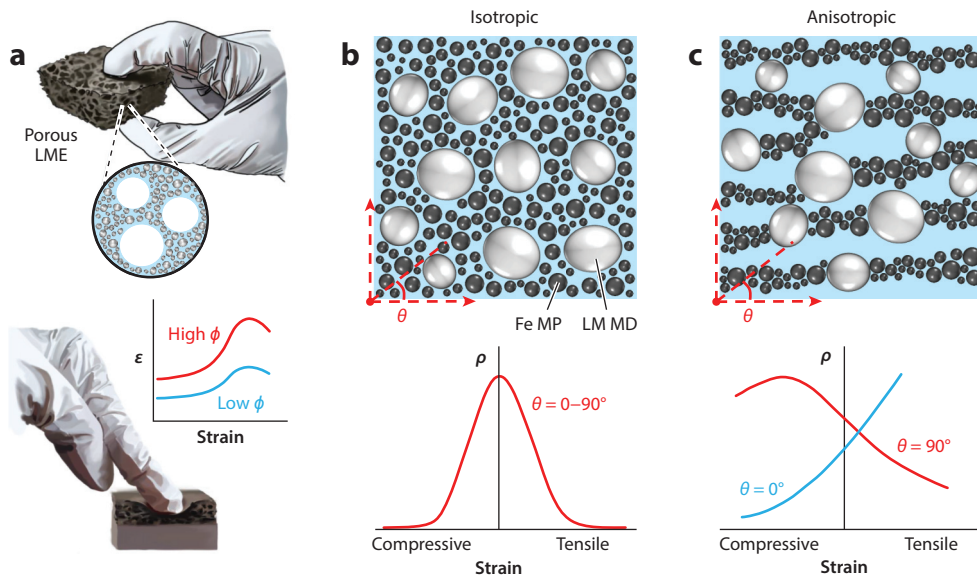


Figure 9

Deformation-responsive LME composites. (a) (Top) Piezopermittivity effect exhibited for porous LME foams. (Bottom) Permittivity, ϵ , initially increases when compressed as the air gets displaced and eventually decreases as the LM filler deforms. The higher the LM content (ϕ), the higher is permittivity. (b) (Top) Embedding solid iron (Fe) particles into LME results in composites with positive piezoconductivity in which the resistivity, ρ , decreases exponentially upon the application of either compressive or tensile strain. (Bottom) The plot shows a typical resistivity-versus-strain curve. (c) (Top) Magnetically prealigning the Fe particles prior to curing results in a composite with anisotropic properties. (Bottom) The plots show typical resistivity-versus-strain curves at different measurement angles. The material is piezoconductive along the direction of particle alignment (blue data) and is piezoresistive perpendicular to this direction (red data). Abbreviations: LM, liquid metal; LME, liquid metal elastomer; MD, microdroplet; MP, microparticle.

diverse reactions. Conductivity also allows the interface to be manipulated electrochemically to tune the interfacial tension over an unprecedented range.

5.4.1. Reactive liquid metal interfaces. One of the most exciting aspects of Ga LM is its reactivity. **Figure 10a** shows that LMs can react with ambient molecules to form a self-limiting (passivating) film that is only a few nanometers thick. The most obvious example is Ga oxide, yet the dissolution of highly reactive atomic species (such as Al) into LM can change the composition of the surface oxide (123). Nonoxide species can also be deposited; one example is graphene, although it requires elevated temperatures (124). Once formed, such species can be transferred from the LM surface onto other substrates, offering a simple way to form 2D or 2D-like materials (123, 125) (**Figure 10b**). Once transferred, the Ga oxide or other surface-generated materials can be further modified into other materials, such as chalcogenides (e.g., semiconducting Ga sulfide) (125).

Ga is inherently reactive but is typically protected by the passivating oxide. However, increasing the temperature above 600°C can induce the fracture of the self-limiting Ga oxide and—in the case of Ga-In alloy—can cause the leakage and oxidation of the In layer underneath to form In-rich oxides on the surface (126) (**Figure 10c**). The oxide can be mechanically broken with ease, which can briefly expose the metal to the surrounding ambient. The exposed metal can, for example, directly initiate free radical polymerization of monomers (37) (**Figure 10d**).

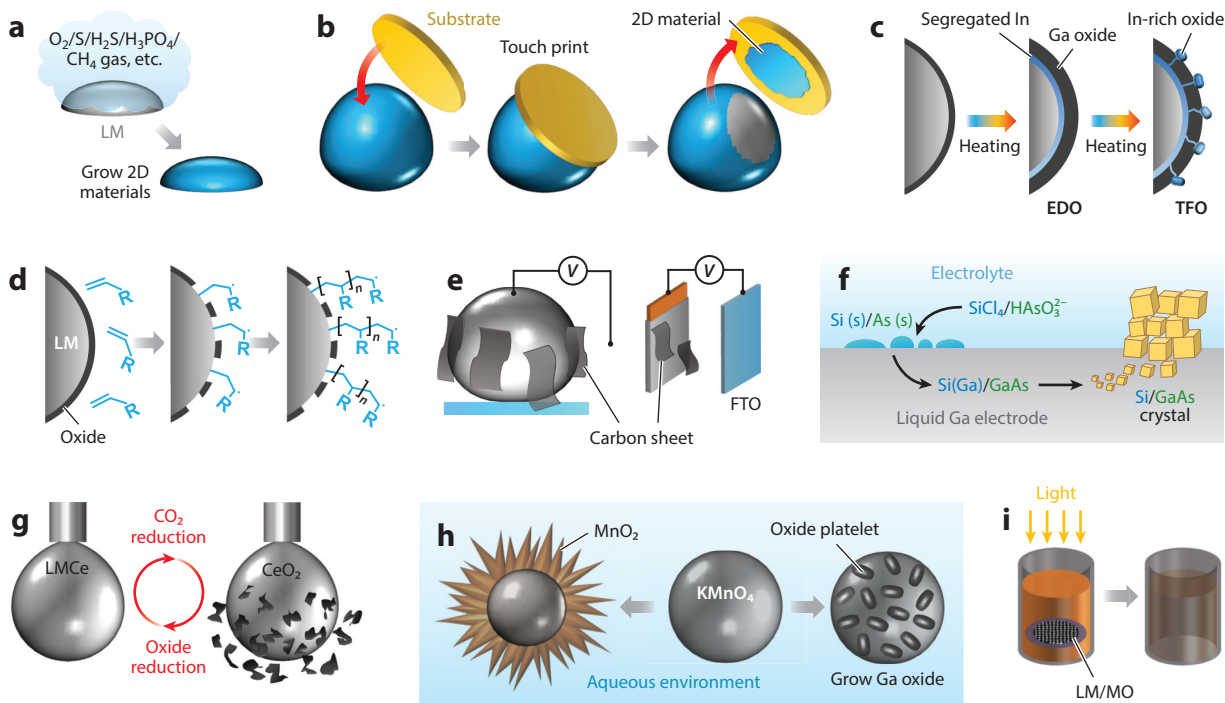


Figure 10

The reactive surface of LM. (a) Formation of 2D materials by reacting LM with ambient molecules. (b) Delaminating and transferring surface films by contacting the surface of LM with a substrate. (c) Oxide growth of LM via EDO and TFO. (d) Initiating polymerization after mechanically breaking the oxide and exposing the metal to the surrounding ambient. (e) Creation of graphitic species using LM droplets or LM-coated Cu electrodes, while FTO-coated glass is used as the counterelectrode. (f) Electrochemical liquid-liquid-solid process for producing crystalline Si or GaAs in Ga at low temperature. (g) Adding reactive metal of Ce to Ga (LMCe) to form reactive species at the interface for reducing CO_2 into solid carbon species. (h) Growth of monolayers of MnO_2 spikes and Ga oxide platelets by oxidation. (i) The use of LM/MO structures for photocatalytic degradation of organic dyes. Abbreviations: EDO, expansion-induced diffusion-limited oxidation; FTO, fluorine-doped tin oxide; LM, liquid metal; MO, metal oxide; TFO, thermomechanical fracture leakage and oxidation.

Ga has also been used as an electrode for electrochemistry. For example, it can reduce carbon species to form graphitic flakes (127) (Figure 10e). LM electrodes have also been elegantly used to electrochemically create semiconductors such as Ge, Si, and GaAs in which precursor salts reduce, dissolve in the metal, and then crystallize when the LM saturates (128, 129) (Figure 10f).

The addition of reactive metals (such as Ce) to LM allows for the formation of reactive species at the interface that can reduce CO_2 into solid carbon species (38) (Figure 10g). Importantly, the solubility of metals in LM opens the possibility to dissolve expensive or highly reactive catalysts inside LM and have them present themselves safely in high surface-to-volume ratios. The poor adhesion of most solid products to the smooth molten surface avoids coking, which is the contamination of electrodes with carbon species. These nonfouling surfaces have also proven beneficial by mixing Ga and Pd to create a catalyst that combines the advantages of heterogeneous and molecular catalysts for alkane dehydrogenation (130).

Oxidizers such as $KMnO_4$ can accelerate oxidation and lead to the formation of monolayers of MnO_2 spikes on the surface of the metal (131) (Figure 10h). Alternatively, Ga oxide rods can be synthesized on the surface of LM particles via heating (98). These processes produce LM/metal

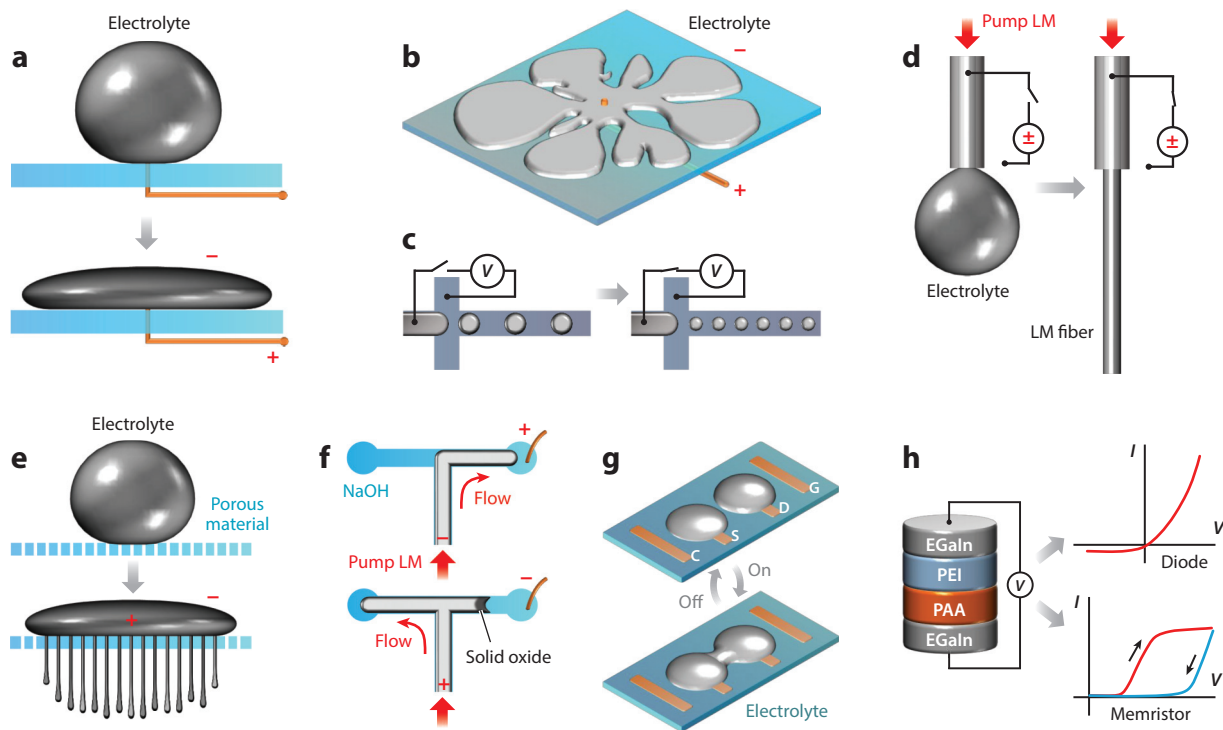


Figure 11

Electrochemically responsive interfacial tension of LM. (a) The effective interfacial tension of a LM droplet decreases by using electrochemical oxidation. (b) LM spreads as fractals after further lowering the interfacial tension to near zero. (c) Variation in tension can cause changes in the size of droplets formed in flow-focusing microfluidic devices. (d) A LM wire is formed using electrochemical oxidation to lower the tension. (e) The decreased interfacial tension can allow the metal to flow through small pores. (f) Electrochemical control of the flow direction of LM in microchannels is shown. (g) A soft LM switch is created by merging and isolating adjacent drops of LM by controlling interfacial tension. (h) Electrochemical oxidation and reduction of LM electrodes that interface with hydrogels can create entirely soft diodes and memristors. Abbreviations: C, counter; D, drain; G, ground; LM, liquid metal; PAA, polyacrylic acid; PEI, polyethyleneimine; S, source.

oxide (LM/MO) structures that can act as catalysts, as reviewed recently (132). **Figure 10i** shows LM with oxide surface species that act as photocatalysts for degrading an organic dye such as Congo Red in response to light (133). The formation of the LM/MO structure enhances photocatalytic efficiency due to factors such as increased surface porosity/area, altered energy band structure, and increased trap state density between the Fermi level and valence band (133).

Ga can also undergo galvanic replacement reactions in which Ga oxidizes while reducing a nearby metal salt such as Au^+ and Ag^+ ions (134, 135). Electrons from Ga can tunnel through the oxide to react with the metal salts to form nanoparticles.

5.4.2. Electrochemically responsive interfacial tension. Applying a potential to Hg in electrolyte can lower the tension modestly due to the formation of electrical double layers at the interface. This effect is termed electrocapillarity, and the drop in tension is predicted by the capacitive energy resulting from the charged double layer. However, the decrease in tension of Ga-based LM depicted in **Figure 11a** goes well beyond what is possible via electrocapillarity (25). The drop in tension begins when the oxide starts to deposit electrochemically (a process termed anodization or electrochemical capillarity). Although the mechanism by which anodization lowers tension is

Eutectic gallium indium (EGaIn): a popular LM formed by the combination of Ga and In (75-wt% Ga, 25-wt% In) with the lowest melting point, 15.7°C

poorly understood, it is thought that the deposited species act as a surfactant for the metal and create stresses that oppose the interfacial tension. Remarkably, applying only 1 V in electrolyte (1 M NaOH) can cause the metal to behave as if the tension was zero or near zero (136), producing a phenomenon whereby the LM can start spreading as fractals (137) (**Figure 11b**). Variation in interfacial tension can change the size of droplets formed in microchannels, since the size represents a balance of shear and interfacial forces (33) (**Figure 11c**). Since interfacial forces are dominant at submillimeter length scales, the ability to manipulate them is a useful way to move LM.

The enormous drop in tension has interesting fluid implications. Liquids normally exit a nozzle by forming droplets, but when the tension is low (by applying ~ 1 V), the LM exits as a stable stream [a wire or fiber (**Figure 11d**)] with a diameter similar to that of a human hair (100 μm) (138). The lowered tension can allow the metal to flow through small pores (139) (**Figure 11e**). Interestingly, switching the potential can remove the oxide and return the metal to a state of high tension (140). The so-called beating heart can be created by using LM droplets that oscillate between high and low tension as they detach and touch, respectively, an electrode that drives oxidation (141). In a microchannel, voltage can be employed like a valve to direct the pathway of LM at junctions without the need for mechanical moving parts. The metal follows the path with the lowest tension, which is dictated by the location of the counterelectrode (142) (**Figure 11f**). Alternatively, the voltage can electrochemically oxidize the metal to form a mechanical impediment that redirects the movement of the LM toward alternative pathways (**Figure 11f**). The spreading process is dynamic, and consequently the shapes that form are not stable. However, freezing the metal causes it to solidify and maintain its shape at room temperature.

These changes in surface state have been used to create soft devices that use both ionic and electronic principles. For example, **Figure 11g** shows a LM switch (143). In this example, a source and drain consist of Cu pads wetted with LM. By toggling the potential applied to the electrodes, including the counter and ground, the metal can spread and merge together (to form a conductive path) or break apart due to capillary forces (to form a resistive state). Likewise, electrochemical oxidation and reduction can be used to create entirely soft memory devices termed memristors (**Figure 11b**). Sandwiching hydrogels between LM [eutectic gallium indium (EGaIn)] electrodes creates a conductive pathway. One hydrogel, polyethyleneimine, is chosen to have a pH that keeps the metal-gel interface oxide free. The other hydrogel, polyacrylic acid, has a pH such that the oxide can form (a resistive state) or reduce (a conductive state). This principle can be used to create a resistive memory (memristive) device in which the resistive and conductive states represent 1 and 0, respectively (144). With a small tweak, the same device can be used as a diode (145). Such devices do not work as well as commercial electronics but are interesting because they are built entirely from liquid-like materials (similar mechanically to the human brain).

5.5. Emerging Directions

Figure 12 captures some of the exciting directions of LM research. Due to space constraints, we elaborate on the promise of LM for biomedical applications (23, 26). Although Ga has no natural function in the body, Ga salts have been FDA approved for a number of applications such as MRI contrast agents. Ga^{3+} interacts in the body similarly to Fe^{3+} since they have similar ionic radius and oxidation state, although unlike iron, the 2+ oxidation state of Ga is energetically unfavorable. This property has enabled pharmaceutical (treating disorders of calcium and bone metabolism) (146), antibacterial (8, 103), and anticancer (96, 105) applications. **Figure 12a** depicts biomedical applications ranging from those discussed above (23) to direct injection of LM into vascular networks for contrast agents (147) and into the bone to serve as bone cement (148). These applications are only recently being explored and will likely develop significantly over the next decade.

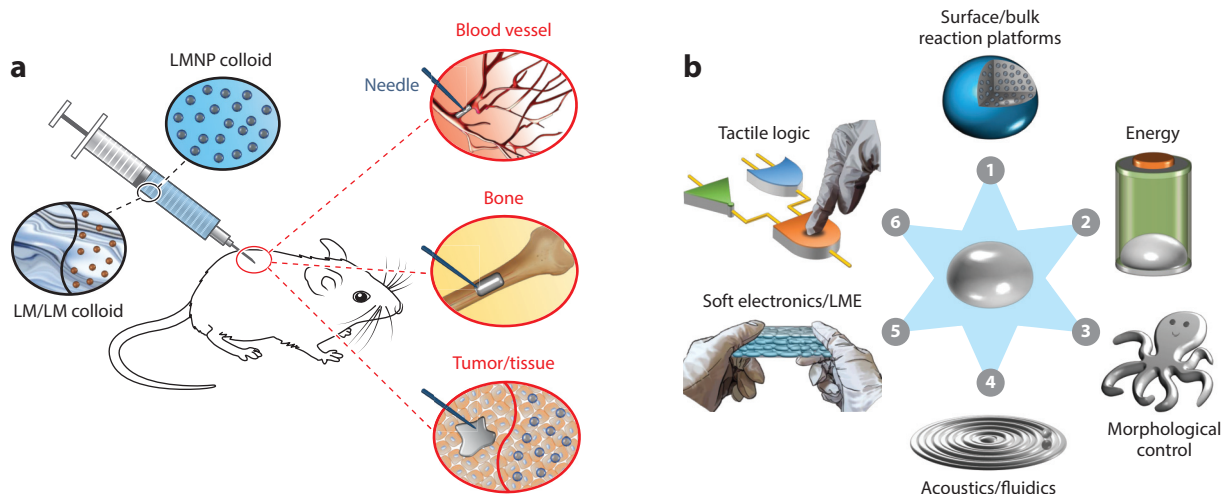


Figure 12

Emerging directions for future LM research. (a) Biomedical applications in the fields of medical imaging, cancer therapy, nanomedicine, orthopedic treatment, etc. (b) Applications in materials science, electronics, energy, chemistry, and fluidics. (❶) Utilizing the reactivity of Ga to carry out reactions in which the LM is a catalyst, a solvent, an electrode, a source of electrons, or an initiator. (❷) Utilizing LM for energy storage (batteries) or generation. Due to fluidic and deformable features, anodes made from LMs are intrinsically dendrite free and have self-healing properties. Furthermore, LMs do not need extra energy input to maintain the liquid state, which endows them with numerous advantages in energy storage devices compared to high-temperature LMs (149). (❸,❹) Taking advantage of the liquid properties to reconfigure the shape of the metal on demand for reconfigurable devices (optics, electromagnetic devices, circuits) to explore novel acoustic and fluidic properties and to understand and harness the huge changes in tension made possible by electrochemistry (136). (❺) The use of LMs allowing for soft and flexible electronics that can be built into wearable sensors and textile-based electronics as well as enabling new human-machine interfaces and teaming with biosensors, electrically functioning nerve connectors, and implantable electrodes. (❻) Soft logic and mechanological systems (150). Abbreviations: LM, liquid metal; LME, liquid metal elastomer; LMNP, liquid metal nanoparticle.

In addition to applications in the biomedical sciences, a range of LM applications are emerging in areas such as surface/bulk reaction platforms, energy, morphological control, acoustics/fluidics, soft electronics/LMEs, and tactile logic (**Figure 12b**). For an example of a tactile logic demonstration, please see the sidebar titled Logic Using Soft Materials.

LOGIC USING SOFT MATERIALS

An octopus is a remarkable creature that inspires new types of soft sensors, actuators, and robotics. Interestingly, more than half of its neurons are found in its arms, whereas vertebrates have most of their neurons centralized in the brain. Decentralized neurons raise the possibility of carrying out distributed logic, in which materials themselves make decisions at the local level. This concept could eliminate the complex wiring required to send signals between sensor arrays and a centralized computer. LM offers the intriguing possibility of making entirely soft tactile logic elements that can make decisions (albeit, very simple decisions) based on the way they are touched. **Figure 12b** shows a conceptual example. LM circuits embedded in an elastomer can increase or decrease conductivity and/or capacitance, depending on how they are designed and touched. This property has been utilized to redirect electricity to do work, cause color changes, and change thermal properties of a material (150). Whereas most smart materials simply produce a single output response to a single input stimulus, truly smart materials should be able to respond appropriately, depending on the state of multiple inputs.

DISCLOSURE STATEMENT

The authors are not aware of any affiliations, memberships, funding, or financial holdings that might be perceived as affecting the objectivity of this review.

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LITERATURE CITED

1. Sacks O. 2003. Gallium. *Chem. Eng. News Arch.* 81(36):88
2. Jacoby M. 2016. Liquid metals went to work. *C&EN Glob. Enterp.* 94(49):26–26
3. Woo M. 2020. Liquid metal renaissance points to wearables, soft robots, and new materials. *PNAS* 117(10):5088–91
4. Dickey MD. 2020. EML webinar overview: liquid metals at the extreme. *Extreme Mech. Lett.* 40:100863
5. Spells KE. 1936. The determination of the viscosity of liquid gallium over an extended range of temperature. *Proc. Phys. Soc.* 48(2):299
6. Züger O, Dürig U. 1992. Atomic structure of the α -Ga(001) surface investigated by scanning tunneling microscopy: direct evidence for the existence of Ga₂ molecules in solid gallium. *Phys. Rev. B* 46(11):7319–21
7. Kim J-H, Kim S, So J-H, Kim K, Koo H-J. 2018. Cytotoxicity of gallium-indium liquid metal in an aqueous environment. *ACS Appl. Mater. Interfaces* 10(20):17448–54
8. Goss CH, Kaneko Y, Khuu L, Anderson GD, Ravishankar S, et al. 2018. Gallium disrupts bacterial iron metabolism and has therapeutic effects in mice and humans with lung infections. *Sci. Transl. Med.* 10(460):eaat7520
9. Cochran CN, Foster LM. 1962. Vapor pressure of gallium, stability of gallium suboxide vapor, and equilibria of some reactions producing gallium suboxide vapor. *J. Electrochem. Soc.* 109(2):144–48
10. Briggs LJ. 1957. Gallium: thermal conductivity; supercooling; negative pressure. *J. Chem. Phys.* 26(4):784–86
11. Giguère PA, Lamontagne D. 1954. Polarography with a dropping gallium electrode. *Science* 120(3114):390–91
12. Jacob AR, Parekh DP, Dickey MD, Hsiao LC. 2019. Interfacial rheology of gallium-based liquid metals. *Langmuir* 35(36):11774–83
13. Dickey MD. 2014. Emerging applications of liquid metals featuring surface oxides. *ACS Appl. Mater. Interfaces* 6(21):18369–79
14. Joshipura ID, Ayers HR, Majidi C, Dickey MD. 2015. Methods to pattern liquid metals. *J. Mater. Chem. C* 3:3834–41
15. Dickey MD. 2017. Stretchable and soft electronics using liquid metals. *Adv. Mater.* 29(27):1606425
16. Kazem N, Hellebrekers T, Majidi C. 2017. Soft multifunctional composites and emulsions with liquid metals. *Adv. Mater.* 29(27):1605985
17. Kalantar-Zadeh K, Tang J, Daeneke T, O'Mullane AP, Stewart LA, et al. 2019. Emergence of liquid metals in nanotechnology. *ACS Nano* 13(7):7388–95
18. Daeneke T, Khoshmanesh K, Mahmood N, de Castro IA, Esrafilzadeh D, et al. 2018. Liquid metals: fundamentals and applications in chemistry. *Chem. Soc. Rev.* 47(11):4073–111
19. Chen S, Wang H-Z, Zhao R-Q, Rao W, Liu J. 2020. Liquid metal composites. *Matter* 2(6):1446–80
20. Khoshmanesh K, Tang S-Y, Zhu JY, Schaefer S, Mitchell A, et al. 2017. Liquid metal enabled microfluidics. *Lab Chip* 17(6):974–93

21. Ren L, Xu X, Du Y, Kalantar-Zadeh K, Dou SX. 2020. Liquid metals and their hybrids as stimulus-responsive smart materials. *Mater. Today* 34:92–114
22. Khondoker MAH, Sameoto D. 2016. Fabrication methods and applications of microstructured gallium based liquid metal alloys. *Smart Mater. Struct.* 25(9):093001
23. Yan J, Lu Y, Chen G, Yang M, Gu Z. 2018. Advances in liquid metals for biomedical applications. *Chem. Soc. Rev.* 47(8):2518–33
24. Song H, Kim T, Kang S, Jin H, Lee K, Yoon HJ. 2020. Ga-based liquid metal micro/nanoparticles: recent advances and applications. *Small* 16(12):1903391
25. Eaker CB, Dickey MD. 2016. Liquid metal actuation by electrical control of interfacial tension. *Appl. Phys. Rev.* 3(3):031103
26. Sun X, Yuan B, Sheng L, Rao W, Liu J. 2020. Liquid metal enabled injectable biomedical technologies and applications. *Appl. Mater. Today* 20:100722
27. Lin Y, Genzer J, Dickey MD. 2020. Attributes, fabrication, and applications of gallium-based liquid metal particles. *Adv. Sci.* 7(12):2000192
28. Neumann TV, Dickey MD. 2020. Liquid metal direct write and 3D printing: a review. *Adv. Mater. Technol.* 5(9):2000070
29. Cheng S, Wu Z. 2012. Microfluidic electronics. *Lab Chip* 12(16):2782
30. Chiechi RC, Weiss EA, Dickey MD, Whitesides GM. 2008. Eutectic gallium-indium (EGaIn): a moldable liquid metal for electrical characterization of self-assembled monolayers. *Angew. Chem. Int. Ed.* 47(1):142–44
31. **Tang S-Y, Khoshmanesh K, Sivan V, Petersen P, O'Mullane AP, et al. 2014. Liquid metal enabled pump. *PNAS* 111(9):3304–9**
32. Thelen J, Dickey MD, Ward T. 2012. A study of the production and reversible stability of EGaIn liquid metal microspheres using flow focusing. *Lab Chip* 12(20):3961–67
33. Tang S-Y, Joshipura ID, Lin Y, Kalantar-Zadeh K, Mitchell A, et al. 2016. Liquid-metal microdroplets formed dynamically with electrical control of size and rate. *Adv. Mater.* 28(4):604–9
34. Yang T, Kwon B, Weisensee PB, Kang JG, Li X, et al. 2018. Millimeter-scale liquid metal droplet thermal switch. *Appl. Phys. Lett.* 112(6):063505
35. Cumby BL, Hayes GJ, Dickey MD, Justice RS, Tabor CE, Heikenfeld JC. 2012. Reconfigurable liquid metal circuits by Laplace pressure shaping. *Appl. Phys. Lett.* 101(17):174102
36. Jamtveit B, Meakin P. 1999. *Growth, Dissolution, and Pattern Formation in Geosystems*. Dordrecht, Neth.: Springer
37. Ma J, Lin Y, Kim Y-W, Ko Y, Kim J, et al. 2019. Liquid metal nanoparticles as initiators for radical polymerization of vinyl monomers. *ACS Macro Lett.* 8(11):1522–27
38. Esrafilzadeh D, Zavabeti A, Jalili R, Atkin P, Choi J, et al. 2019. Room temperature CO₂ reduction to solid carbon species on liquid metals featuring atomically thin ceria interfaces. *Nat. Commun.* 10:865
39. Hardy SC. 1985. The surface tension of liquid gallium. *J. Cryst. Growth* 71(3):602–6
40. Dickey MD, Chiechi RC, Larsen RJ, Weiss EA, Weitz DA, Whitesides GM. 2008. Eutectic gallium-indium (EGaIn): a liquid metal alloy for the formation of stable structures in microchannels at room temperature. *Adv. Funct. Mater.* 18(7):1097–104
41. Regan MJ, Kawamoto EH, Lee S, Pershan PS, Maskil N, et al. 1995. Surface layering in liquid gallium: an X-ray reflectivity study. *Phys. Rev. Lett.* 75(13):2498–501
42. Larsen RJ, Dickey MD, Whitesides GM, Weitz DA. 2009. Viscoelastic properties of oxide-coated liquid metals. *J. Rheol.* 53(6):1305–26
43. Doudrick K, Liu S, Mutunga EM, Klein KL, Damle V, et al. 2014. Different shades of oxide: from nanoscale wetting mechanisms to contact printing of gallium-based liquid metals. *Langmuir* 30(23):6867–77
44. Kramer RK, Boley JW, Stone HA, Weaver JC, Wood RJ. 2014. Effect of microtextured surface topography on the wetting behavior of eutectic gallium-indium alloys. *Langmuir* 30(2):533–39
45. Kadlaskar SS, Yoo JH, Abhijeet, Lee JB, Choi W. 2017. Cost-effective surface modification for Galinstan[®] lyophobicity. *J. Colloid Interface Sci.* 492:33–40
46. Khan MR, Trlica C, So J-H, Valeri M, Dickey MD. 2014. Influence of water on the interfacial behavior of gallium liquid metal alloys. *ACS Appl. Mater. Interfaces* 6(24):22467–73

31. Shows the use of interfacial tension gradients on liquid metal to pump and mix fluids.

47. Ilyas N, Cook A, Tabor CE. 2017. Designing liquid metal interfaces to enable next generation flexible and reconfigurable electronics. *Adv. Mater. Interfaces* 4(15):1700141
48. Lin Y, Gordon O, Khan MR, Vasquez N, Genzer J, Dickey MD. 2017. Vacuum filling of complex microchannels with liquid metal. *Lab Chip* 17(18):3043–50
49. Ladd C, So J-H, Muth J, Dickey MD. 2013. 3D printing of free standing liquid metal microstructures. *Adv. Mater.* 25(36):5081–85
50. Park Y-G, An HS, Kim J-Y, Park J-U. 2019. High-resolution, reconfigurable printing of liquid metals with three-dimensional structures. *Sci. Adv.* 5(6):eaaw2844
51. Watson AM, Cook AB, Tabor CE. 2019. Electrowetting-assisted selective printing of liquid metal. *Adv. Eng. Mater.* 21(10):1900397
52. Boley JW, White EL, Kramer RK. 2015. Mechanically sintered gallium-indium nanoparticles. *Adv. Mater.* 27(14):2355–60
53. Liu S, Yuen MC, White EL, Boley JW, Deng B, et al. 2018. Laser sintering of liquid metal nanoparticles for scalable manufacturing of soft and flexible electronics. *ACS Appl. Mater. Interfaces* 10(33):28232–41
54. Morris NJ, Farrell ZJ, Tabor CE. 2019. Chemically modifying the mechanical properties of core-shell liquid metal nanoparticles. *Nanoscale* 11(37):17308–18
55. Thrasher CJ, Farrell ZJ, Morris NJ, Willey CL, Tabor CE. 2019. Mechanoresponsive polymerized liquid metal networks. *Adv. Mater.* 31(40):1903864
56. Jeong SH, Hjort K, Wu Z. 2015. Tape transfer atomization patterning of liquid alloys for microfluidic stretchable wireless power transfer. *Sci. Rep.* 5:8419
57. Khondoker MAH, Ostashek A, Sameoto D. 2019. Direct 3D printing of stretchable circuits via liquid metal co-extrusion within thermoplastic filaments. *Adv. Eng. Mater.* 21(7):1900060
58. Lu T, Finkenauer L, Wissman J, Majidi C. 2014. Rapid prototyping for soft-matter electronics. *Adv. Funct. Mater.* 24(22):3351–56
59. Pan C, Kumar K, Li J, Markvicka EJ, Herman PR, Majidi C. 2018. Visually imperceptible liquid-metal circuits for transparent, stretchable electronics with direct laser writing. *Adv. Mater.* 30(12):1706937
60. Zheng Y, He Z-Z, Yang J, Liu J. 2014. Personal electronics printing via tapping mode composite liquid metal ink delivery and adhesion mechanism. *Sci. Rep.* 4:4588
61. Ma B, Xu C, Chi J, Chen J, Zhao C, Liu H. 2019. A versatile approach for direct patterning of liquid metal using magnetic field. *Adv. Funct. Mater.* 29(28):1901370
62. Kramer RK, Majidi C, Wood RJ. 2013. Masked deposition of gallium-indium alloys for liquid-embedded elastomer conductors. *Adv. Funct. Mater.* 23(42):5292–96
63. Jeong SH, Hagman A, Hjort K, Jobs M, Sundqvist J, Wu Z. 2012. Liquid alloy printing of microfluidic stretchable electronics. *Lab Chip* 12(22):4657–64
64. Gozen BA, Tabatabai A, Ozdoganlar OB, Majidi C. 2014. High-density soft-matter electronics with micron-scale line width. *Adv. Mater.* 26(30):5211–16
65. Kim M, Alrowais H, Pavlidis S, Brand O. 2017. Size-scalable and high-density liquid-metal-based soft electronic passive components and circuits using soft lithography. *Adv. Funct. Mater.* 27(3):1604466
66. Yan S, Li Y, Zhao Q, Yuan D, Yun G, et al. 2018. Liquid metal-based amalgamation-assisted lithography for fabrication of complex channels with diverse structures and configurations. *Lab Chip* 18(5):785–92
67. Li G, Lee D-W. 2017. An advanced selective liquid-metal plating technique for stretchable biosensor applications. *Lab Chip* 17(20):3415–21
68. Hirsch A, Michaud HO, Gerratt AP, de Mulatier S, Lacour SP. 2016. Intrinsically stretchable biphasic (solid-liquid) thin metal films. *Adv. Mater.* 28(22):4507–12
69. Nijhuis CA, Reus WF, Barber JR, Dickey MD, Whitesides GM. 2010. Charge transport and rectification in arrays of SAM-based tunneling junctions. *Nano Lett.* 10(9):3611–19
70. Andrews JB, Mondal K, Neumann TV, Cardenas JA, Wang J, et al. 2018. Patterned liquid metal contacts for printed carbon nanotube transistors. *ACS Nano* 12(6):5482–88
71. Tang S-Y, Sivan V, Petersen P, Zhang W, Morrison PD, et al. 2014. Liquid metal actuator for inducing chaotic advection. *Adv. Funct. Mater.* 24(37):5851–58
72. Zhu JY, Tang S-Y, Khoshmanesh K, Ghorbani K. 2016. An integrated liquid cooling system based on Galinstan liquid metal droplets. *ACS Appl. Mater. Interfaces* 8(3):2173–80

73. So J-H, Dickey MD. 2011. Inherently aligned microfluidic electrodes composed of liquid metal. *Lab Chip* 11(5):905–11
74. Sciambi A, Abate AR. 2014. Generating electric fields in PDMS microfluidic devices with salt water electrodes. *Lab Chip* 14(15):2605–9
75. Hallfors N, Khan A, Dickey MD, Taylor AM. 2013. Integration of pre-aligned liquid metal electrodes for neural stimulation within a user-friendly microfluidic platform. *Lab Chip* 13(4):522–26
76. Parekh DP, Ladd C, Panich L, Moussa K, Dickey MD. 2016. 3D printing of liquid metals as fugitive inks for fabrication of 3D microfluidic channels. *Lab Chip* 16(10):1812–20
77. Yerasimou Y, Pickert V, Ji B, Song X. 2018. Liquid metal magnetohydrodynamic pump for junction temperature control of power modules. *IEEE Trans. Power Electron.* 33(12):10583–93
78. Krupenkin T, Taylor JA. 2011. Reverse electrowetting as a new approach to high-power energy harvesting. *Nat. Commun.* 2:448
79. Jeon J, Chung SK, Lee J-B, Doo SJ, Kim D. 2018. Acoustic wave-driven oxidized liquid metal-based energy harvester. *Eur. Phys. J. Appl. Phys.* 81(2):20902
80. Tang W, Jiang T, Fan FR, Yu AF, Zhang C, et al. 2015. Liquid-metal electrode for high-performance triboelectric nanogenerator at an instantaneous energy conversion efficiency of 70.6%. *Adv. Funct. Mater.* 25(24):3718–25
81. Dong C, Leber A, Das Gupta T, Chandran R, Volpi M, et al. 2020. High-efficiency super-elastic liquid metal based triboelectric fibers and textiles. *Nat. Commun.* 11:3537
82. Wu J, Tang S-Y, Fang T, Li W, Li X, Zhang S. 2018. A wheeled robot driven by a liquid-metal droplet. *Adv. Mater.* 30(51):1805039
83. Zhu S, So J-H, Mays R, Desai S, Barnes WR, et al. 2013. Ultrastretchable fibers with metallic conductivity using a liquid metal alloy core. *Adv. Funct. Mater.* 23(18):2308–14
84. Park J, Kang HS, Koo M, Park C. 2020. Autonomous surface reconciliation of a liquid-metal conductor micropatterned on a deformable hydrogel. *Adv. Mater.* 32(37):2002178–83
85. Park Y-L, Majidi C, Kramer R, Bérard P, Wood RJ. 2010. Hyperelastic pressure sensing with a liquid-embedded elastomer. *J. Micromech. Microeng.* 20(12):125029
86. **So J-H, Thelen J, Qusba A, Hayes GJ, Lazzi G, Dickey MD. 2009. Reversibly deformable and mechanically tunable fluidic antennas. *Adv. Funct. Mater.* 19(22):3632–37**
87. Cheng S, Rydberg A, Hjort K, Wu Z. 2009. Liquid metal stretchable unbalanced loop antenna. *Appl. Phys. Lett.* 94(14):144103
88. Cooper CB, Arutselvan K, Liu Y, Armstrong D, Lin Y, et al. 2017. Stretchable capacitive sensors of torsion, strain, and touch using double helix liquid metal fibers. *Adv. Funct. Mater.* 27(20):1605630
89. Palleau E, Reece S, Desai SC, Smith ME, Dickey MD. 2013. Self-healing stretchable wires for reconfigurable circuit wiring and 3D microfluidics. *Adv. Mater.* 25(11):1589–92
90. Tonazzini A, Mintchev S, Schubert B, Mazzolai B, Shintake J, Floreano D. 2016. Variable stiffness fiber with self-healing capability. *Adv. Mater.* 28(46):10142–48
91. Farrell ZJ, Tabor C. 2018. Control of gallium oxide growth on liquid metal eutectic gallium/indium nanoparticles via thiolation. *Langmuir* 34(1):234–40
92. Farrell ZJ, Thrasher CJ, Flynn AE, Tabor CE. 2020. Silanized liquid-metal nanoparticles for responsive electronics. *ACS Appl. Nano Mater.* 3(7):6297–303
93. Gan T, Shang W, Handschuh-Wang S, Zhou X. 2019. Light-induced shape morphing of liquid metal nanodroplets enabled by polydopamine coating. *Small* 15(9):1804838
94. Yan J, Malakooti MH, Lu Z, Wang Z, Kazem N, et al. 2019. Solution processable liquid metal nanodroplets by surface-initiated atom transfer radical polymerization. *Nat. Nanotechnol.* 14:684–90
95. Yu Y, Miyako E. 2017. Manipulation of biomolecule-modified liquid-metal blobs. *Angew. Chem.* 129(44):13794–99
96. **Lu Y, Hu Q, Lin Y, Pacardo DB, Wang C, et al. 2015. Transformable liquid-metal nanomedicine. *Nat. Commun.* 6:10066**
97. Zhang J, Yao Y, Sheng L, Liu J. 2015. Self-fueled biomimetic liquid metal mollusk. *Adv. Mater.* 27(16):2648–55
98. Lin Y, Liu Y, Genzer J, Dickey MD. 2017. Shape-transformable liquid metal nanoparticles in aqueous solution. *Chem. Sci.* 8(5):3832–37

86. An early demonstration of functional electronics that are stretchable and self-healing.

96. Reports on the formation of functional nanoparticles and utilization for drug delivery.

111. The first report of liquid metal elastomers that remain soft by using fillers that alter other properties.

117. Reports that the deformation of liquid metal fillers can change the properties of composites

99. Lu Y, Lin Y, Chen Z, Hu Q, Liu Y, et al. 2017. Enhanced endosomal escape by light-fueled liquid-metal transformer. *Nano Lett.* 17(4):2138–45
100. Tang J, Zhao X, Li J, Guo R, Zhou Y, Liu J. 2017. Gallium-based liquid metal amalgams: transitional-state metallic mixtures (TransM2ixes) with enhanced and tunable electrical, thermal, and mechanical properties. *ACS Appl. Mater. Interfaces* 9(41):35977–87
101. Daalkhajav U, Yirmibesoglu OD, Walker S, Mengüç Y. 2018. Rheological modification of liquid metal for additive manufacturing of stretchable electronics. *Adv. Mater. Technol.* 3(4):1700351
102. Carle F, Bai K, Casara J, Vanderlick K, Brown E. 2017. Development of magnetic liquid metal suspensions for magnetohydrodynamics. *Phys. Rev. Fluids* 2(1):013301
103. Elbourne A, Cheeseman S, Atkin P, Truong NP, Syed N, et al. 2020. Antibacterial liquid metals: biofilm treatment via magnetic activation. *ACS Nano* 14(1):802–17
104. Wang D, Gao C, Wang W, Sun M, Guo B, et al. 2018. Shape-transformable, fusible rodlike swimming liquid metal nanomachine. *ACS Nano* 12(10):10212–20
105. Chechetka SA, Yu Y, Zhen X, Pramanik M, Pu K, Miyako E. 2017. Light-driven liquid metal nanotransformers for biomedical theranostics. *Nat. Commun.* 8:15432
106. Yu Y, Miyako E. 2018. Alternating-magnetic-field-mediated wireless manipulations of a liquid metal for therapeutic bioengineering. *iScience* 3:134–48
107. Park Y-G, Min H, Kim H, Zhexembekova A, Lee CY, Park J-U. 2019. Three-dimensional, high-resolution printing of carbon nanotube/liquid metal composites with mechanical and electrical reinforcement. *Nano Lett.* 19(8):4866–72
108. Losurdo M, Suvorova A, Rubanov S, Hingerl K, Brown AS. 2016. Thermally stable coexistence of liquid and solid phases in gallium nanoparticles. *Nat. Mater.* 15(9):995–1002
109. Tang S-Y, Mitchell DRG, Zhao Q, Yuan D, Yun G, et al. 2019. Phase separation in liquid metal nanoparticles. *Matter* 1(1):192–204
110. Fassler A, Majidi C. 2015. Liquid-phase metal inclusions for a conductive polymer composite. *Adv. Mater.* 27(11):1928–32
111. Jeong SH, Chen S, Huo J, Gamstedt EK, Liu J, et al. 2015. Mechanically stretchable and electrically insulating thermal elastomer composite by liquid alloy droplet embedment. *Sci. Rep.* 5:18257
112. Kazem N, Bartlett MD, Majidi C. 2018. Extreme toughening of soft materials with liquid metal. *Adv. Mater.* 30(22):1706594
113. Krisnadi F, Nguyen LL, Ankit, Ma J, Kulkarni MR, et al. 2020. Directed assembly of liquid metal–elastomer conductors for stretchable and self-healing electronics. *Adv. Mater.* 32(30):2001642
114. Wang H, Yao Y, He Z, Rao W, Hu L, et al. 2019. A highly stretchable liquid metal polymer as reversible transitional insulator and conductor. *Adv. Mater.* 31(23):1901337
115. Blaiszik BJ, Kramer SLB, Grady ME, McIlroy DA, Moore JS, et al. 2012. Autonomic restoration of electrical conductivity. *Adv. Mater.* 24(3):398–401
116. Markvicka EJ, Bartlett MD, Huang X, Majidi C. 2018. An autonomously electrically self-healing liquid metal–elastomer composite for robust soft-matter robotics and electronics. *Nat. Mater.* 17(7):618–24
117. Bartlett MD, Kazem N, Powell-Palm MJ, Huang X, Sun W, et al. 2017. High thermal conductivity in soft elastomers with elongated liquid metal inclusions. *PNAS* 114(9):2143–48–116
118. Ford MJ, Ambulo CP, Kent TA, Markvicka EJ, Pan C, et al. 2019. A multifunctional shape-morphing elastomer with liquid metal inclusions. *PNAS* 116(43):21438–44
119. Bartlett MD, Fassler A, Kazem N, Markvicka EJ, Mandal P, Majidi C. 2016. Stretchable, high-k dielectric elastomers through liquid-metal inclusions. *Adv. Mater.* 28(19):3726–31
120. Yang J, Tang D, Ao J, Ghosh T, Neumann TV, et al. 2020. Ultrasoft liquid metal elastomer foams with positive and negative piezopermittivity for tactile sensing. *Adv. Funct. Mater.* 30(36):2002611
121. Yun G, Tang S-Y, Sun S, Yuan D, Zhao Q, et al. 2019. Liquid metal–filled magnetorheological elastomer with positive piezoconductivity. *Nat. Commun.* 10:1300
122. Yun G, Tang S-Y, Zhao Q, Zhang Y, Lu H, et al. 2020. Liquid metal composites with anisotropic and unconventional piezoconductivity. *Matter* 3(3):824–41

123. Zavabeti A, Ou JZ, Carey BJ, Syed N, Orrell-Trigg R, et al. 2017. A liquid metal reaction environment for the room-temperature synthesis of atomically thin metal oxides. *Science* 358(6361):332–35
124. Fujita J, Miyazawa Y, Ueki R, Sasaki M, Saito T. 2010. Fabrication of large-area graphene using liquid gallium and its electrical properties. *Jpn. J. Appl. Phys.* 49(6S):06GC01
125. Carey BJ, Ou JZ, Clark RM, Berean KJ, Zavabeti A, et al. 2017. Wafer-scale two-dimensional semiconductors from printed oxide skin of liquid metals. *Nat. Commun.* 8:14482
126. Cutinho J, Chang BS, Oyola-Reynoso S, Chen J, Akhter SS, et al. 2018. Autonomous thermal-oxidative composition inversion and texture tuning of liquid metal surfaces. *ACS Nano* 12(5):4744–53
127. Mayyas M, Li H, Kumar P, Ghasemian MB, Yang J, et al. 2020. Liquid-metal-templated synthesis of 2D graphitic materials at room temperature. *Adv. Mater.* 32(29):2001997
128. Cheek Q, Fahrenkrug E, Hlynchuk S, Alsem DH, Salmon NJ, Maldonado S. 2020. In situ transmission electron microscopy measurements of Ge nanowire synthesis with liquid metal nanodroplets in water. *ACS Nano* 14(3):2869–79
129. Gu J, Fahrenkrug E, Maldonado S. 2013. Direct electrodeposition of crystalline silicon at low temperatures. *J. Am. Chem. Soc.* 135(5):1684–87
130. Taccardi N, Grabau M, Debuschewitz J, Distaso M, Brandl M, et al. 2017. Gallium-rich Pd-Ga phases as supported liquid metal catalysts. *Nat. Chem.* 9(9):862–67
131. Ghasemian MB, Mayyas M, Idrus-Saidi SA, Jamal MA, Yang J, et al. 2019. Self-limiting galvanic growth of MnO₂ monolayers on a liquid metal—applied to photocatalysis. *Adv. Funct. Mater.* 29(36):1901649
132. Liang S-T, Wang H-Z, Liu J. 2018. Progress, mechanisms and applications of liquid-metal catalyst systems. *Chemistry* 24(67):17616–26
133. Zhang W, Ou JZ, Tang S-Y, Sivan V, Yao DD, et al. 2014. Liquid metal/metal oxide frameworks. *Adv. Funct. Mater.* 24(24):3799–807
134. Hoshyargar F, Crawford J, O'Mullane AP. 2017. Galvanic replacement of the liquid metal Galinstan. *J. Am. Chem. Soc.* 139(4):1464–71
135. David R, Miki N. 2018. Tunable noble metal thin films on Ga alloys via galvanic replacement. *Langmuir* 34(36):10550–59
136. Khan MR, Eaker CB, Bowden EF, Dickey MD. 2014. Giant and switchable surface activity of liquid metal via surface oxidation. *PNAS* 111(39):14047–51
137. Eaker CB, Hight DC, O'Regan JD, Dickey MD, Daniels KE. 2017. Oxidation-mediated fingering in liquid metals. *Phys. Rev. Lett.* 119(17):174502
138. Song M, Kartawira K, Hillaire KD, Li C, Eaker CB, et al. 2020. Overcoming Rayleigh-Plateau instabilities: stabilizing and destabilizing liquid-metal streams via electrochemical oxidation. *PNAS* 117(32):19026–32
139. Yun FF, Yu Z, He Y, Jiang L, Wang Z, et al. 2020. Voltage-induced penetration effect in liquid metals at room temperature. *Natl. Sci. Rev.* 7(2):366–72
140. Khan MR, Trlica C, Dickey MD. 2015. Recapillarity: electrochemically controlled capillary withdrawal of a liquid metal alloy from microchannels. *Adv. Funct. Mater.* 25(5):671–78
141. Yu Z, Chen Y, Yun FF, Cortie D, Jiang L, Wang X. 2018. Discovery of a voltage-stimulated heartbeat effect in droplets of liquid gallium. *Phys. Rev. Lett.* 121(2):024302
142. Tang S-Y, Lin Y, Joshipura ID, Khoshmanesh K, Dickey MD. 2015. Steering liquid metal flow in microchannels using low voltages. *Lab Chip* 15(19):3905–11
143. Wissman J, Dickey MD, Majidi C. 2017. Field-controlled electrical switch with liquid metal. *Adv. Sci.* 4(12):1700169
144. Koo H, So J, Dickey MD, Velez OD. 2011. Towards all-soft matter circuits: prototypes of quasi-liquid devices with memristor characteristics. *Adv. Mater.* 23(31):3559–64
145. So J, Koo H, Dickey MD, Velez OD. 2012. Ionic current rectification in soft-matter diodes with liquid-metal electrodes. *Adv. Funct. Mater.* 22(3):625–31
146. Bernstein LR. 1998. Mechanisms of therapeutic activity for gallium. *Pharmacol. Rev.* 50(4):665–82
147. Wang Q, Yu Y, Pan K, Liu J. 2014. Liquid metal angiography for mega contrast X-ray visualization of vascular network in reconstructing in-vitro organ anatomy. *IEEE Trans. Biomed. Eng.* 61(7):2161–66

123. Demonstrates the ability to remove native oxide from liquid metal for synthesizing 2D-like materials at room temperature.

129. One of several papers that illustrate the use of Ga for reactions.

136. Reports that electrochemical oxidation lowers the effective interfacial tension to near zero.

148. One of the first papers to illustrate the promise of liquid metal for biomedical applications.

148. Yi L, Jin C, Wang L, Liu J. 2014. Liquid-solid phase transition alloy as reversible and rapid molding bone cement. *Biomaterials* 35(37):9789–801
149. Guo X, Zhang L, Ding Y, Goodenough JB, Yu G. 2019. Room-temperature liquid metal and alloy systems for energy storage applications. *Energy Environ. Sci.* 12(9):2605–19
150. Jin Y, Lin Y, Kiani A, Joshipura ID, Ge M, Dickey MD. 2019. Materials tactile logic via innervated soft thermochromic elastomers. *Nat. Commun.* 10:4187

RELATED RESOURCES

1. Dickey MD. 2015. *Liquid metal*. Video, TEDxNCSSM, Febr. 20. https://www.youtube.com/watch?v=IfUnK_rME8E
2. Liu J, Yi L. 2018. *Liquid Metal Biomaterials*. Berlin: Springer
3. Liu J, Sheng L, He ZZ. 2019. *Liquid Metal Soft Machines*. Singapore: Springer Nat.
4. RMIT Univ. 2014. *Liquid metal enables non-mechanical pump*. Video, Febr. 24. <https://www.youtube.com/watch?v=ehfH0s2ksPk>
5. Zhang S. 2018. *Scientists make progress in developing liquid metal robot*. Video, Oct. 25. <https://www.youtube.com/watch?v=MAJ8puKE6jA>
6. Interact Lab. 2017. *Programmable liquid matter: shape deformation of liquid metals in dynamic electric field*. Video, Oct. 2. <https://www.youtube.com/watch?v=nSFwbCjnyY4>
7. Dickey MD. 2013. *3D printing of liquid metals at room temperature*. Video, July 8. <https://www.youtube.com/watch?v=ql3pXn8-sHA&t=1s>
8. Dickey MD. 2020. *EML webinar by Michael Dickey on liquid metals at the extreme*. Video, June 24. <https://www.youtube.com/watch?v=a55SohyyKo4&t=978s>
9. Kalantar-Zadeh K. 2020. *Liquid-based electronics and optics*. Video, June 29. <https://www.youtube.com/watch?v=BcZOCazqQ1w>
10. Dickey MD. 2014. *Shape reconfigurable liquid metal*. Video, Sep. 15. <https://www.youtube.com/watch?v=mB2ZqO5E1Zo&t=43s>