

Recent progress in gallium-based liquid metal particle composites for soft electronic applications

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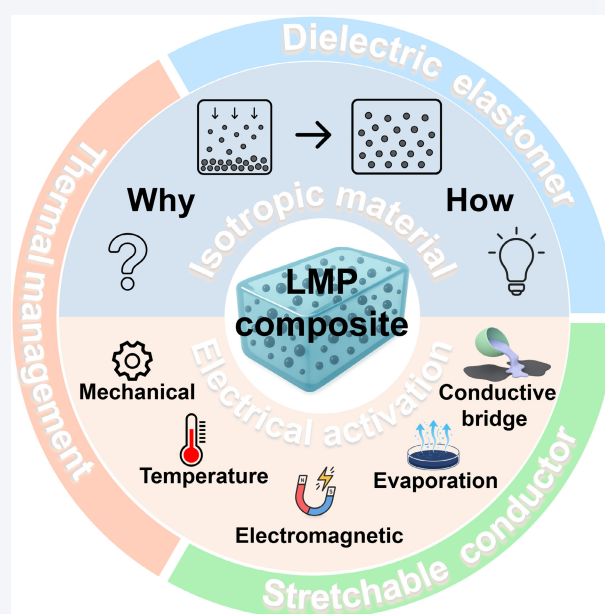
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ABSTRACT: Gallium-based liquid metals are emerging materials for soft electronics with their fluidic compliance, low toxicity, metallic conductivity, and outstanding thermal conductivity. Especially, liquid metal particles (LMPs) mixed with polymeric matrix have demonstrated promising application potentials for soft and stretchable electronic applications including stretchable integrated circuits, capacitive sensors, dielectric elastomer actuators, and thermal management materials. This review highlights methods to prepare LMPs, control and manipulate LMPs within the polymeric matrix to meet the demands of different functionalities. Furthermore, we summarized recent advances by utilizing LMP composites to enable different stretchable electronic applications. It is believed that LMP composites will serve as important enabling materials for the development of electronic skins, stretchable electronics, smart textiles, and next-generation human-machine interfaces. A comprehensive and timely review of the recent progress in LMP composites is expected to inspire further exploration and innovation in stretchable and multifunctional electronic systems.

KEYWORDS: liquid metal particle, composite, stretchable electronics, sensor and actuator, thermal management



1 Introduction

Flexible and stretchable electronics have imposed stringent requirements on the functional materials to maintain stable performance under diverse mechanical deformations, enabling their seamless adaptation to dynamic operating conditions [1–3]. Ga-based liquid metals (LMs) have emerged as highly promising materials due to their exceptional electrical conductivity, excellent thermal conductivity, low toxicity, outstanding fluidity, and self-healing properties [4, 5]. Beyond their wide applications as stretchable conductors [5–7], these materials have also demonstrated remarkable performance in forming important

electronic components such as resistors [8], inductors [8–10], capacitors [10, 11], memristors [12, 13], diodes [14, 15], and transistors [16–18].

Ga-based LMs are generally required to be shape-manipulated or patterned for stretchable and flexible electronic devices featuring high integration and multiple functionalities. However, the high surface tension, low viscosity and inherent fluidity of LMs prevent them from obtaining controllable and stable electronic architectures. One commonly adopted strategy is to fill LM into microchannels of the host elastomers [19–22]. Compared to direct injection, vacuum filling eliminates residual air inside the channels, allowing more uniform LM filling and facilitating the fabrication of complex patterns [20]. However, the approach imposes difficulties in effectively integrating LM with other functional electronic components. Another viable strategy involves patterning LM by leveraging its reactive wetting on other metallic surfaces, which requires pre-depositing metal layers on flexible substrates using evaporation, photolithography, or sputtering [23, 24]. Typically,

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acid treatment will be used to remove metal oxides and promote wetting. Despite its potential for large-scale and high-resolution fabrication, this approach is time-consuming and economically inefficient.

Recently, effective approaches were developed by incorporating micro/nano scale materials such as Cu [25–27], Ni [28], carbon nanotube (CNT) [29], Fe [30], SiO₂ [31], or MXene [32] into the continuous phase of LM to enhance its patterning, shaping and printing control. The complexity of rheological control and the high sensitivity to printing parameters hinder the scalable printing of these conductive inks, while the inherent oxidizability of LMs necessitates post-printing encapsulation to prevent material degradation. Additionally, the exceptionally high surface energy (> 0.5 N·m⁻¹) and low viscosity (~ 2 mPa·s) of Ga-based LM make it difficult to maintain uniform conductive ink [32, 33]. Low-viscosity LM is easier to extrude from the print nozzle [4], while the enrichment of solid particles may clog the nozzle, posing challenges for continuous printing.

In a different strategy, dispersing LM into particles and mixing them with polymeric materials has attracted extensive attentions. The resulting liquid metal particle (LMP) composites have shown promising performance, highly tailorable functionalities, and versatile fabrication approaches. Compared to rigid metallic fillers, the inherent fluidity and mechanical compliance of LMs reduce the effect on the mechanical properties of the polymeric materials [34]. With their multifunctionality and processability, LMP composites have found extensive applications in thermal management [35, 36], flexible circuits [37, 38], dielectric elastomer actuators [39], and capacitive sensors [40, 41]. The polymeric materials can be a viscous polymer solution, uncured elastomer mixture, or pre-gel solution which serve as the composite matrix mixing with LMPs [4]. LMPs within composite are stabilized by the polymeric material with the naturally formed oxide film on the surfaces of the LMPs. Due to the non-conductive oxide film on their surfaces, the embedded LMPs are generally isolated from each other without effective percolating networks established within the composites. For the isolated LMPs within the composite material to achieve highly conducting pathways, they must overcome the insulating barriers from the polymeric matrix and the oxide layer on the particle surface. This necessitates approach to establish electrical interconnect between the LMPs through LMP sintering. By appropriate selection of materials and sintering techniques, the composites can achieve metallic conductivity, offering a promising solution to prepare high-performance stretchable conductors [42, 43]. Conversely, dielectric materials and thermal conducting materials prefer that LMPs are homogeneously dispersed in the composites. And in many cases, electrical interconnected LMPs should be avoided to prevent unintended conduction in the composite. However, due to the much higher density of the LMPs, they can easily precipitate during the curing process of the composite, resulting in anisotropy in the materials. This hinders the performance of stretchable dielectric elastomers and thermal management materials. Consequently, strategies are required to achieve stable dispersion of LMPs in the composites.

In recent years, numbers of new fabrication strategies, mechanism insights, and innovative applications have emerged, driving the LMP composites toward practical and multifunctional integrations. However, recent review articles [44–47] on LMP composites have primarily focused on their conduction performances or have been limited in the analysis and summary of

their boarder application scopes. In light of this, this review article systematically reviews and summarizes recent research progress and functional implementation strategies for LMP composites in three important applications, stretchable conductor, dielectric elastomers, and thermal management (Fig. 1). We aim to provide a timely reference for future material designs, performance regulation, and the development of multifunctional devices based on LMP composites, stimulating continued theoretical exploration and technological innovation in soft electronics.

2 Ga-based LMPs

The fluidic nature of Ga-based LM fillers contributes exceptional conformability to the host material and induces minimized effects on the composite's mechanical properties, conferring distinct advantages for Ga-based LMP composites over conventional composites using rigid fillers. Meanwhile, leveraging the inherent metallic properties of LM, Ga-based LMP composites can be easily designed with high electrical conductivity, exceptional thermal conductivity, or superior dielectric properties. Here, we focus on some key properties of Ga-based LM, as well as several highly applicable methods for preparing LMPs.

2.1 Properties of Ga-based LM and their particles

Gallium LM exhibits a low melting point of 29.8 °C [55], which can be further tuned by alloying with other metals [56, 57]. For example, eutectic gallium-indium alloy (EGaIn, 75 wt.% and 25 wt.% for gallium and indium, respectively) melts at ~ 15.5 °C [57], while gallium-indium-tin alloy (Galinstan or EGaInSn, 68.5 wt.%, 21.5 wt.%, and 10 wt.% for gallium, indium, and tin, respectively) exhibits a much lower melting point of -19 °C [5]. LM can be processed into rod and sheet forms at low temperatures (Fig. 2(a)), enabling the fabrication of precise three-dimensional (3D) conductive structures [6]. However, the solid-liquid phase transition process of Ga-based LM under temperature changes is complex. During solid nucleation, the system must possess sufficient energy to overcome the nucleation energy barrier for the nuclei to stably exist and continuously grow. The nucleation energy barrier ΔG^* is defined as

$$\Delta G^* = 16\pi\gamma^3/3(\Delta G_v)^2 \quad (1)$$

where γ is the solid-liquid interfacial energy and ΔG_v is the volumetric Gibbs free energy difference. For Ga-based LM, ΔG_v is defined as [58]

$$\Delta G_v = -L\Delta T/T_m \quad (2)$$

where L , ΔT , and T_m represent the latent heat of melting, the degree of supercooling, and the melting point, respectively. Combining Eqs. (1) and (2) reveals that Ga-based LMs exhibit notable supercooling behavior with their large solid-liquid interfacial energy that prevent the formation of solid nucleus, together with the presence of a stable gallium oxide layer that passivates the liquid surface and suppresses heterogeneous nucleation, resulting in a high nucleation energy barrier. Consequently, these materials remain in a liquid state even when the temperature falls below their equilibrium melting point [6, 59]. For instance, bulk EGaIn has a freezing and melting temperatures at -11 ± 6 and 15.5 ± 3 °C, respectively [56]. Compared to bulk LMs, LMPs with high surface-to-volume ratio show greater supercooling effect. The fundamental

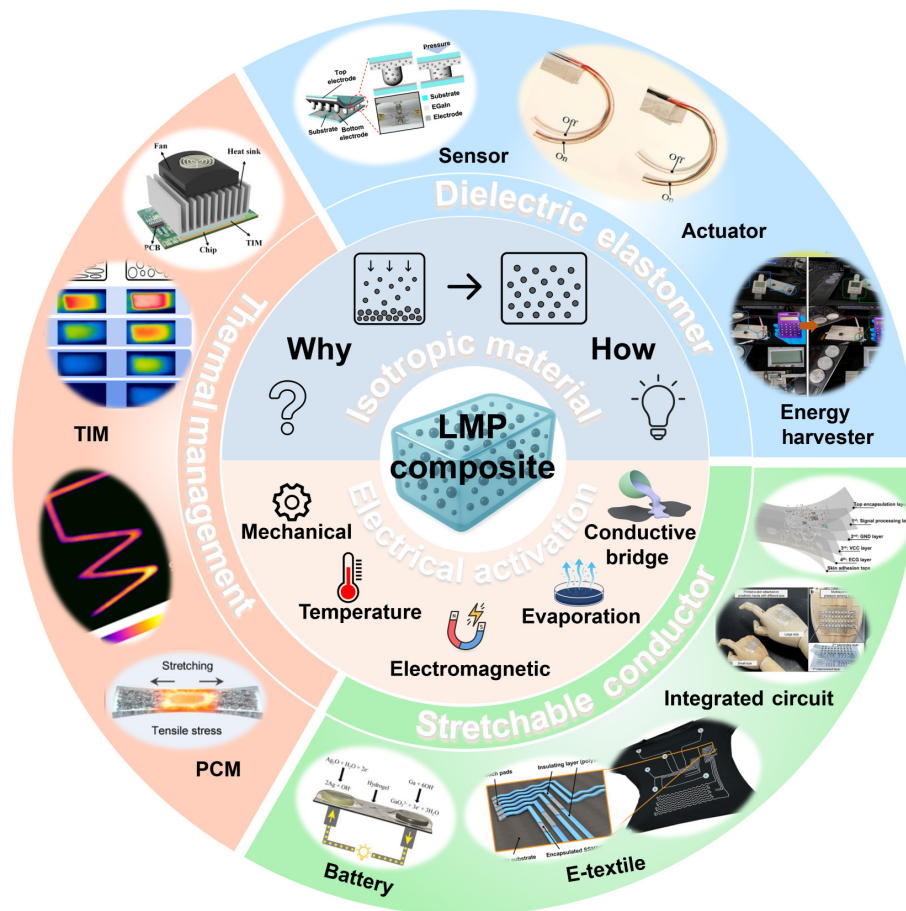


Figure 1 Schematic illustrating the multifunctional applications of LMP composites for stretchable conductors, dielectric elastomers, and thermal management materials. Reproduced with permission from Ref. [48], © Lee, G. H. et al. 2022; Ref. [37], © Elsevier Ltd. 2025; Ref. [38], © Wiley-VCH GmbH 2022; Ref. [49], © American Chemical Society 2019; Ref. [50], © American Chemical Society 2025; Ref. [39], © WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim 2019; Ref. [51], © Elsevier Ltd. 2023; Ref. [52], © American Chemical Society 2025; Ref. [53], © American Chemical Society 2025; Ref. [54], © Wang, H. Z. et al. 2024.

reason is that the reduction in particle volume sharply decreases the number of nucleation impurities, forcing solidification to occur only through homogeneous nucleation, which requires a much lower temperature. For example, stabilized EGaIn micro-/nanoparticles have significantly lower freezing temperature (down to -84.1 from -5.9 °C) and melting point (down to -25.6 from $+17.8$ °C) [56]. As shown in Fig. 2(b), the phase transition suppression is largely independent of the host material and synthesis process, allowing for wider operating temperature window for LMP composite-based electronic devices. It is worth noting that through mechanical vibration, electromagnetic excitation, acoustic cavitation, or the introduction of external particles [54, 60, 61], high-density heterogeneous nucleation sites can be instantaneously induced within the bulk and at the phase boundaries of supercooled LM, triggering rapid relaxation of the metastable supercooled state and controlled solidification. This programmable liquid-solid phase transition of LM exhibits significant application potential in reconfigurable circuit printing, reversible phase-change thermal management, and stiffness control in soft robotics [58, 61].

Gallium-based LMs have a low viscosity of approximately 1.99×10^{-3} Pa·s, roughly twice that of water [62]. Their electrical and thermal conductivities are comparable to those of pure gallium, with an electrical conductivity of around 3×10^6 S·m $^{-1}$, only one order of magnitude lower than copper [56, 63]. Their thermal conductivity is around 26 W·m $^{-1}$ ·K $^{-1}$ (@310 K), similar to stainless

steel [64, 65]. The combination of high electrical and thermal conductivity together with intrinsic fluidity makes LMs ideal candidates for soft electronic applications. Ga-based LMs, with their extremely low vapor pressure at room temperature and their negligible solubility in water [5], have low toxicity and relatively stable properties compared to other metals with melting points below ambient temperature, such as mercury (Hg), cesium (Cs), and francium (Fr). As a result, gallium alloys have emerged as the most extensively explored LMs.

Without oxidation, Ga-based LM droplets will aggregate into a bead with its surface tension exceeding 500 mN·m $^{-1}$ [57]. Under ambient conditions, Ga-based LMs will easily form a thin oxide film on their surfaces upon contact with oxygen. For instance, in the presence of 0.8% oxygen concentration, the oxidation reaction will proceed rapidly (< 0.25 s) [66]. The preferential formation of surface oxides of LMPs can be explained by the Gibbs free energy (ΔG) [67]. Due to the much larger negative ΔG of Ga compared to In and Sn (Fig. 2(c)), the oxide layers of LMPs are primarily composed of Ga_2O_3 . The presence of oxide layers enhances the wettability of Ga-based LMs, enabling LM droplets to be manipulated into non-equilibrium shapes (Fig. 2(d)) [68]. It is noteworthy that Ga-based LMs possess remarkable self-healing properties. Following physical damage, LMs can quickly restore their surface by regenerating a protective oxide film [69]. This characteristic is particularly important to fabricate electronic devices

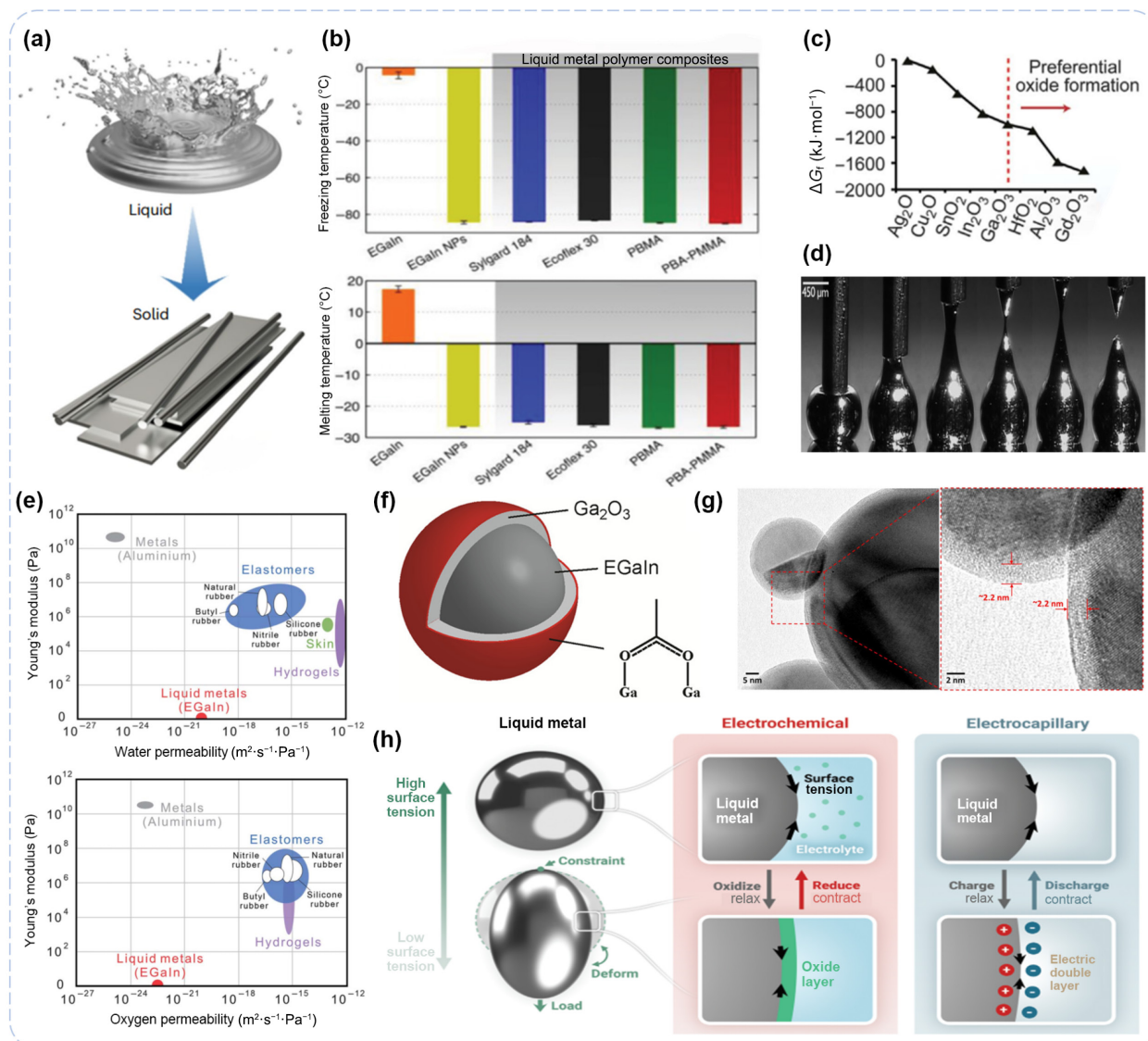


Figure 2 Characteristics of LM and LMP. (a) Schematic diagram illustrating the transformation of EGaln from liquid to rods and sheets. Reproduced with permission from Ref. [6], © Li, G. Q. 2023. (b) Freezing and melting temperatures (peak value) of EGaln LM, EGaln LMPs (≈ 500 nm diameter), and embedded EGaln LMPs in different polymer matrices (shaded background indicates LMP composites). Reproduced with permission from Ref. [56], © WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim 2019. (c) Gibbs free energy for some selected metal oxides. Metal oxides to the right of the red dashed line are expected to dominate the interface. Reproduced with permission from Ref. [67], © Zavabeti, A. et al. 2017. (d) A series of photographs illustrating the formation process of a conical tip of EGaln LM due to the presence of an oxide layer. Reproduced with permission from Ref. [68], © Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim 2008. (e) Young's modulus versus water permeability for various stretchable materials (top) and young's modulus versus oxygen permeability for various stretchable materials (bottom). Reproduced with permission from Ref. [71], © Shen, Q. C et al. 2023. (f) Illustration of the core-shell structure of Ga-based LMP. Reproduced with permission from Ref. [81], © American Chemical Society 2014. (g) STEM bright field images for LMPs with different sizes (@173 K). The thickness of the oxide layer is very similar (~ 2.2 nm) despite differences in the size of LMPs. Reproduced with permission from Ref. [72], © Elsevier Inc. 2019. (h) Schematic showing the electrochemical and electrocapillary regulation of LMP surface tension. Reproduced with permission from Ref. [77], © Liao, J. H. et al. 2023.

with self-healing capabilities [70]. The hermetic performance of LMs was also investigated by Shen et al. [71]. As shown in Fig. 2(e), compared to aluminum (Al), a commonly used barrier material with high performance, EGaln LM exhibits comparable permeability of oxygen but relatively higher permeability of water, while significantly outperforming conventional elastomers and hydrogels in both aspects. This demonstrates the strong potential of LMs as a sealant for stretchable electronic devices to effectively block the permeation of gases and vapors, which is critical to ensure long-term device stability in practical applications.

LMPs exhibit a distinctive core-shell structure (Fig. 2(f)), in

which the oxide layer is typically a few nanometers thick and does not change much with the size of the LMPs (Fig. 2(g)) [72, 73]. At room temperature, oxygen cannot permeate the oxide film at a perceptible rate, thereby preventing further oxidation of the core materials. The oxide film has negligible influence on charge transport at the interfaces of LM bulks or macroscopic LM droplets [5, 74]. When LMs of this scale come into contact, stable conductive pathways can be established. However, micro- and nanoscale LMPs exhibit ultra-low conductivity or even insulation, arising from the accumulation of stacked oxide films between particles [75]. When surface stress remains below the critical yield

value of approximately $0.2\text{--}0.6\text{ N}\cdot\text{m}^{-1}$, the LMPs' surface layer exhibits elasticity and remains structural stability [62, 76]. Once the critical yield stress is exceeded, the oxide surface fractures and the LM metals begin to flow. This is crucial for functional materials composed of LMPs, enabling the material to create interconnected networks with LM. In addition, the LMPs' surface can also be significantly modulated by electric bias (Fig. 2(h)). For instance, electrical bias can precisely control the LMPs' surface by inducing electrochemical reactions to reduce or increase the oxide layers. The LMPs' surface tension can be regulated through charging and discharging of the electric double layer at the electrolyte-LMP interface [77]. The effects of electric fields on the LMPs' surface can be further leveraged to fabricate LMPs or induce electrical interconnections between LMPs [78–80].

2.2 Preparation methods

LMPs can be typically formed from bulk LM under strong shear forces in solution [81]. By controlling the solution composition, the size distribution and oxide layer structure of LMP can be effectively adjusted. An inert and oxygen-isolated environment can suppress excessive oxide layer growth on the surfaces of LMPs. Additives can be introduced into the solution to form physical or chemical barriers between particles, maintaining particle stability and size uniformity [52, 82, 83]. Other methods apply disruptive shear forces to overcome the high surface tension and produce LMPs, employing mechanical processes such as high-speed rotor mixing, ultrasonic treatment, microfluidic techniques, and ball-milling. Electro-hydrodynamic and sieve-filtration methods are also employed to produce LMPs with narrower size distributions after adjusting the surface tension of LM via electric fields. Table 1 summarizes the basic principles, particle size range, and advantages and disadvantages of these methods for producing LMP.

2.2.1 Electro-hydrodynamic process

The electro-hydrodynamic method combines the synergistic effects of electric and flow fields to continuously produce LMPs with controllable size. A bias voltage can be applied to Ga-based LM, enabling the regulation of surface tension through electrochemistry or electrocapillarity [84, 85]. Jeong et al. investigated the electro-hydrodynamic behavior of Ga-based LM by applying a high DC voltage between a needle with an inner diameter of $60\text{ }\mu\text{m}$ and a circular copper coil electrode positioned at a specific vertical distance [86]. They demonstrated that as the electric field strength or flow rate increases, the volume of the generated droplets decreases while the number of droplets increases. It is noteworthy that the initiation of electrocapillary flow (also known as Marangoni flow) can be achieved under lower current conditions, compared to electrochemical reactions. Song et al. investigated the mechanism of electrocapillary flow to generate LMPs. A micro-syringe pump was used to inject a mixture of sodium hydroxide (NaOH) solution (which removes the oxide layer and keeps the LM surface pristine) containing polyvinyl alcohol (PVA) and EGaIn through a capillary tube into a petri dish filled with electrolyte, while an electric field was simultaneously applied between the capillary tube and petri dish [85]. Specifically, the LM fills the entire channel under the combined effects of pressure and surface tension, while the electrolyte acts as a sheath around the LM near the tube wall. When an electric field is applied, the interface tension becomes non-uniform, and its tangential gradient induces electrocapillary flow within the tube. This hurricane-like flow generates a shear force, which is sufficient to pinch off the LM and form microdroplets (see Fig. 3(a)). By increasing the current and controlling the flow rate and outlet orifice, the size of LMPs can be further reduced. Existing reports on this technology still struggle to achieve LMPs smaller than $50\text{ }\mu\text{m}$, with the resulting LMP diameters typically exceeding $100\text{ }\mu\text{m}$ [79, 84, 85].

Table 1 Comparison of methods for producing LMP

Method	Principle	Size range	Advantages	Disadvantages
Electro-hydrodynamic process	Application of the electrocapillary effect	Tens to hundreds of micrometers	Precisely controllable particle size. Excellent dispersibility. Carried out at high throughput with low energy consumption.	Carried out in an electrolyte solution. Sensitive to electrodes and electrical parameters. Relatively complex equipment system.
Sieve-filtering	Modulating surface tension by electric field	A few micrometers	Controllable particle size. Narrow size distribution. Suitable for large-scale production.	Dependence on the hole size of the filter sieve. Performed in an electrolyte environment. Requires repeated voltage reversal. High risk of LMP aggregation.
Ball-milling	Using the mechanical energy of grinding media to break up bulk LM	Hundreds of nanometers	Low-cost and proven technology. Suitable for large-scale production. Directly dispersible in polymer melts or high-viscosity media.	Relatively broad particle size distribution. Possible contamination caused by wear of grinding media. Excessive oxidation of the particles by prolonged grinding.
Microfluidic production	Shear forces generated by a two-phase fluid flowing through microchannels	Tens to hundreds of micrometers	Highly uniform particle size distribution with extremely low polydispersity. Precise and controllable particle size.	Low throughput with single-channel systems. Complex fabricating system and prone to channel blockages. Strict requirements regarding fluid viscosity.
Shearing	Shear forces generated by high-speed stirring	Tens of nanometers	High yield and simple process. Suitable for <i>in-situ</i> dispersion in high-viscosity polymer matrices. A one-pot fabrication process for LMP composites.	Broad particle size distribution. Limited minimum particle size. Possible polymer degradation by high shearing.
Ultrasonication	Ultrasonic disruption of bulk LM in a solvent containing surfactants	A few micrometers to nanometers	Simple and easy operation. Suitable for small-scale preparation in laboratories. Directly dispersible in a variety of solvents.	Relatively broad particle size distribution. Damage of the surface coating at high power levels. Localized overheating that can promote the formation of oxides.

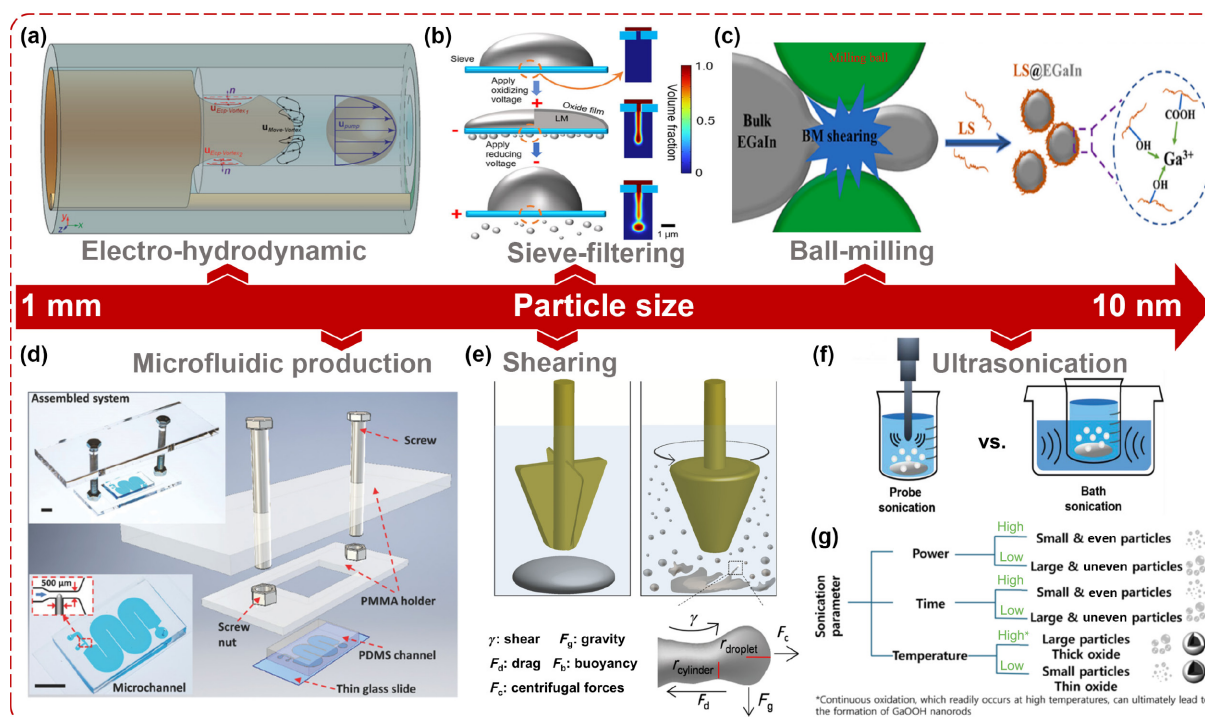


Figure 3 Fabrication methods for Ga-based LMP. (a) Schematic diagram for producing LMPs via electro-hydrodynamic method. Reproduced with permission from Ref. [79], © Wiley-VCH GmbH 2024. (b) Schematic diagram of the working mechanism for preparing LMPs via sieve-filtering and simulation results of the process. Reproduced with permission from Ref. [80], © Zhang, Q. T et al. 2025. (c) Schematic of the mechanism for LMP preparation via ball-milling. Reproduced with permission from Ref. [88], © Elsevier B.V. 2023. (d) Schematic representation of the platform for on-chip production of LM nanoparticles. Reproduced with permission from Ref. [89], © WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim 2018. (e) Schematic diagram of the LMP formation under combined forces in shear-assisted method. Reproduced with permission from Ref. [81], © American Chemical Society 2014. (f) Illustration of LMP preparation via ultrasonication. Reproduced with permission from Ref. [93], © WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim 2019. (g) Schematic flowchart of ultrasonication parameters and their effects on final particle properties. Reproduced with permission from Ref. [95], © Hwang, H. et al. 2025.

2.2.2 Sieve-filtering

Electrochemically regulated Ga-based LMs with low surface tension possess the capability to penetrate micro- and nanoscale pores [87]. Zhang et al. developed a miniaturized filter-sieve generator (MFSG) that utilizes an electric field in an electrolyte environment to produce LMPs with high productivity and tunable size distribution in a cost-effective manner [80]. As shown in Fig. 3(b), LM maintains an elliptical shape on the surface of a filter sieve with a hole size of $0.6\ \mu\text{m}$ due to its high surface tension in the NaOH environment. Upon application of an oxidizing bias, the significant decrease in surface tension allows LM to penetrate the sieve holes, forming pendant LM droplets. Reversing the bias polarity triggers a reduction reaction that removes the oxide layer. The rapid increase in surface tension then forces the LM droplets that have passed through the sieve holes to separate from the large LM bulk, forming LMPs in the underneath solution. Surfactants must be added to prevent the aggregation of LMPs. Size control was achieved by adjusting the oxidation voltage cycle. Specifically, shorter cycles cause suspended LM droplets to detach before further growth under gravity, enabling the average diameter of the resulting LMPs to be as low as $2.39 \pm 0.90\ \mu\text{m}$ [80]. Appropriately increasing the oxidation voltage can promote more efficient LMP production.

2.2.3 Ball-milling

Ga-based LM can be broken down into small particles by the impact and shear forces generated during ball-milling. Recent studies have predominantly reported LMP preparation based on

solutions. Ball-milling methods may offer an environmentally friendly alternative to these solution-based approaches. Although the use of suitable grinding materials can reduce LMPs' diameter, the prepared LMPs tend to agglomerate under mechanical stress, posing challenges for LMP preparation with the ball-milling method. To address the issues, Qiao et al. successfully synthesized nanoscale LMP using lignosulfonate (LS) as the grinding materials in their latest research [88]. As shown in Fig. 3(c), LMPs were formed by the shearing action of the milling balls. Ga_2O_3 on the particle surfaces can build hydrogen bonding with hydroxyl groups of the biomacromolecule LS. This induces LS molecules to continuously cover the LMPs surface, preventing the coalescence of individual particles. Due to the reduction characteristic provided by the abundant phenol hydroxyl groups in LS, the LS layer also prevents the further oxidation of LMPs. The scale of LMPs produced by this method is typically on the order of hundreds of nanometers. By increasing the content of the grinding materials, as well as the frequency and duration of grinding, the size of LMPs can be reduced to $180\ \text{nm}$ [88].

2.2.4 Microfluidic production

Microfluidic production technology provides a large-scale, high-throughput approach for producing LMPs with a narrow distribution of sizes. In this process, the dispersed phase (LM) and immiscible continuous phase (carrier fluid) are required to be simultaneously pumped into microchannels within a capillary-based coaxial flow chip at specific flow rates. The microfluidic approach generates particles by subjecting the LM to shear forces

within intersecting microchannels as these two phases pass through a constriction (Fig. 3(d)). The carrier fluids can be silicone oil or an aqueous solution containing surfactants. By adjusting flow rates or microchannel geometry, the microfluidic production enables precise control over particle size, morphology, and distribution, facilitating high-quality LMP production [84, 89, 90]. The relative flow rates of these two phases are important to the size of LMP. Specifically, the continuous phase with a higher flow rate can impose a greater shear force, leading to more frequent breakage and smaller microsphere sizes for LM. LMPs produced using this method are often several tens to hundreds of micrometers in size (with a minimum achievable size of 60–80 μm) [84, 90], which is larger than those prepared using shear-assisted methods (which can achieve particle size below 20 μm) [91]. Tang et al. placed the entire microfluidic fabrication platform in water and further reduced the size of LMPs generated through narrow channels to the nanoscale using ultrasonication [89].

2.2.5 Shearing

Shear-assisted methods utilize rotor-induced shear to break LM bulk into particles. When LM bulks are suspended in solution driven by the high-speed rotor, shear force predominates and drags the LM bulk into a cylindrical shape. As shown in Fig. 3(e), once the Rayleigh–Plateau limit (where radius $r_{\text{droplet}} > 1.5 r_{\text{cylinder}}$) is reached, the cylindrical LM bulk breaks into individual droplets. Within the colloidal system, the combined action of forces, including shear (γ), gravity (F_g), drag (F_d), centrifugal forces (F_c), and buoyancy (F_b), cause LM droplets to break down into smaller particles [81]. Subsequently, oxidations will stabilize the particles, yielding a solution containing LMPs of uniform size. Parameters such as solution composition, rotation speed, and mixing time enable controls of particle size and morphology. For instance, Ford et al. achieved tunable LMP diameters ranging from 2 to 100 μm by controlling mixing speed and duration [92]. The distribution of LMP obtained through shear mixing is relatively uniform, exhibiting a spherical geometry due to surface energy minimization during mixing. To achieve more uniform fragmentation and elongation of LM, Chin et al. found that periodic pauses during stirring process and addition of LM in small batches allow LMPs to settle and be impacted more frequently by the rotating stirrer [52]. Leveraging the strong affinity of polyvinylpyrrolidone (PVP) to LM, a stable polymer shell can form via hydrogen bonding on the LM surfaces, yielding elongated LMPs with a stable spindle-shaped morphology. Moreover, this method enables direct formation of LMP-polymer composite colloids in solution, highlighting a one-pot fabrication process.

2.2.6 Ultrasonication

Ultrasonication can be implemented with probe and bath ultrasonic systems (Fig. 3(f)) [93]. Nor-Azman et al. utilized high-speed imaging to reveal the formation mechanism of LMPs during ultrasonic processing [94]. The generation of particles from the surfaces of LMs is attributed to interfacial waves formed by the collapse of randomly generated cavities near the surfaces of LMs, which propels particles to be expelled and emulsified into the surrounding solutions. Simultaneously, vacuum cavities also generate close to the interface and within the LM medium, subsequently triggering perturbations on the LM surface that collapse to form craters, releasing more nanoparticles. The parameter settings of the ultrasonic process markedly influence the

LMP synthesis, affecting particle size, particle uniformity, and the growth of the surface oxide layer (Fig. 3(g)) [95]. Dispersant determines whether Ga_2O_3 or gallium oxide hydroxide (GaOOH) layers are formed on the LMP surfaces. Generally, organic solvents induce Ga_2O_3 formation on the particle surfaces with dissolved oxygen in the solutions, yielding spherical LMPs. On the other hand, aqueous solutions promote GaOOH formation with the decomposition of water molecules into H^+ and OH^- , resulting in diverse morphologies such as spherical nanoparticles, spindle-like nanoparticles, and lamellar nanorods [57]. Tang et al. demonstrated a strategy to generate stable and highly controllable LM nanoparticles ($\sim 100 \text{ nm}$) by precisely controlling the applied voltage magnitude of an ultrasonic mist maker equipped with a lead zirconate titanate (PZT) transducer [96]. This method first uses acoustic waves to generate a large number of water vapor cavities at the bottom of the LM, which then migrate upward to the LM-medium interface and collapse, thereby inducing the generation of LM nanoparticles. The nebulization allowed better control over size distribution and can produce particles approximately half the size of those generated by conventional ultrasonication. Due to the tendency of LMPs to reaggregate and the difficulty in controlling the oxide layer, the size distribution of LMPs is polydisperse, ranging from tens of nanometers to tens of micrometers. The oxidation and size of LMP can be controlled by introducing surfactants. For example, the introduction of thiol-containing compounds can reduce the thickness of the oxide layer and promote the formation of a core-shell structure, enabling uniform and stable LMPs [82, 83].

3 LMP composites

This chapter focuses on two important aspects of LMP composites: i) the effect of LMPs on the mechanical properties of the composites; ii) the critical role of isotropic LMP composites for achieving multifunctionality, as well as strategies for their fabrication.

3.1 Mechanical properties

Although LMPs are highly deformable, their native oxide layer provides a certain degree of stiffness. Consequently, the core-shell structure of LMP introduces complicate effect on the mechanical properties of LMP composites. Based on Eshelby's theory of composites, Bartlett et al. [97] found that as the loading of LMP increases, the modulus of LMP composites gradually shifts from the behavior with liquid inclusions to that with the stiff inclusions (Fig. 4(a)). Nevertheless, the increase of composite modulus by LMPs is much lower than that by solid particles. The modulus of LMP composites can maintain similar to that of biological tissues. The elastic modulus of LMP composites also exhibits significant size dependence on the LMPs. When the particle size is relatively large, LMP composites tend to soften. For example, Pan et al. [39] incorporated 10 μm LMPs with a volume fraction of 40% into PDMS, and the material's modulus decreased from 1.515 to 0.882 MPa. As LMPs approach the nanoscale, the modulus of LMP composites increases significantly. This is the result of a combined effect of increased surface tension, enhanced confinement of the liquid metal by the oxide film, and increased shielding of stress transmission by the matrix surrounding these small particles [39, 98, 99]. Composites containing LMPs of smaller size ($< 10 \mu\text{m}$) can maintain the mechanical tensile properties of the unfilled

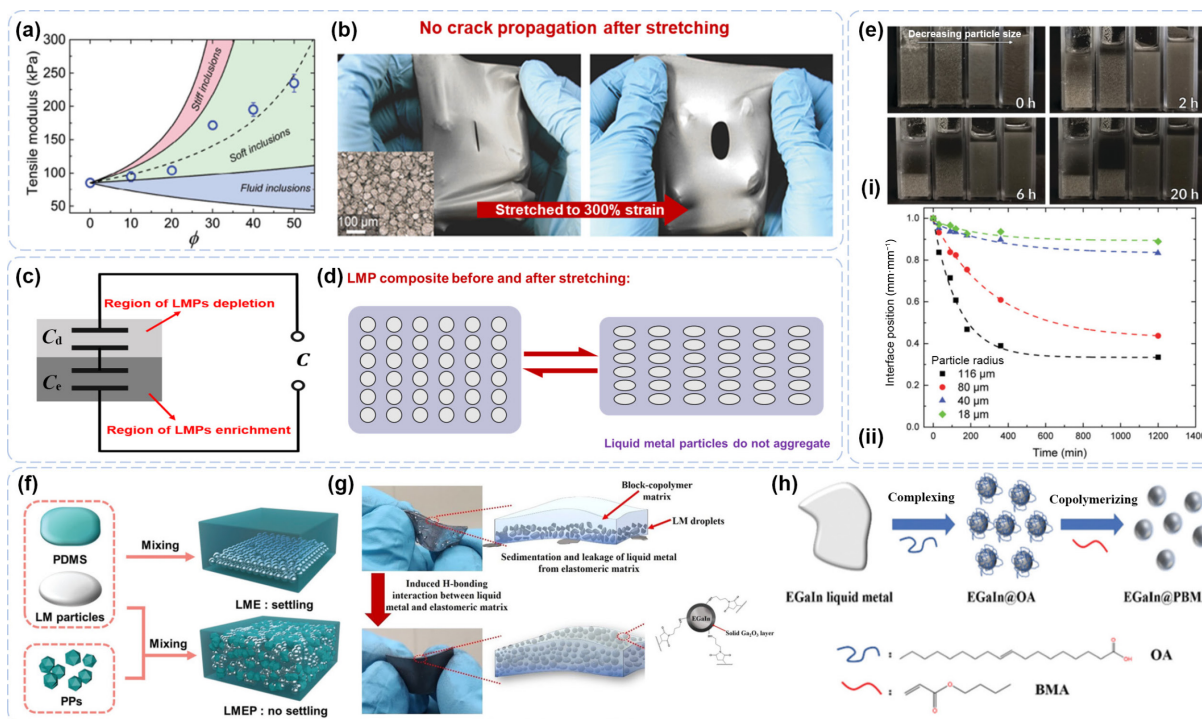


Figure 4 Mechanical properties and isotropy of LMP composites. (a) Plot of tensile modulus as a function of the volume fraction loading of LMP. Reproduced with permission from Ref. [97], © WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim 2016. (b) Photos of a notched LMP composite showing that it can be easily deformed without crack propagation. Reproduced with permission from Ref. [102], © WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim 2018. (c) Schematic diagram of the equivalent electrical model for the Janus structure formed by LMP sedimentation. (d) Schematic illustration of morphology changes of LMPs applied in stretchable dielectrics. (e) Sedimentation of LMPs with different diameters: (i) images of the settling behavior of LMPs of different sizes with respect to time. (ii) Plot of the position of the interface between LMPs rich and depleted zones, measured from the bottom of the container and normalized by the total height of the sample. Dashed lines are best-fits of the data. Reproduced with permission from Ref. [115], © The Royal Society of Chemistry 2020. (f) Schematic of the inhibition of LMPs sedimentation following the addition of PPs. Reproduced with permission from Ref. [105], © Wiley-VCH GmbH 2022. (g) Introducing hydrogen bonds between LM and elastomeric matrix to prevent LMP settling. Reproduced with permission from Ref. [118], © Banerjee, P. S. et al. 2023. (h) The mechanism of synthesizing EGaIn LMPs coated with OA for obtaining isotropic LMPs-PBMA. Reproduced with permission from Ref. [120], © The Royal Society of Chemistry 2020.

elastomer, whereas composites with larger LMPs fracture at lower stress levels [39, 100]. Lee et al. [101] applied an acoustic field to LMP composites, inducing the formation of nanoscale particles from the existed LMPs. These small particles produced a hardening effect on the matrix, and the fracture strain of the composites increased from ~420% to 560%.

Furthermore, the incorporation of LMPs can produce a toughening effect on the fracture behavior of the polymer matrix. When LMPs with a volume fraction of approximately 50% are dispersed in a silicone elastomer, the fracture energy of the composite material increases significantly from approximately $250 \text{ J}\cdot\text{m}^{-2}$ for the pure polymer to approximately $11,900 \text{ J}\cdot\text{m}^{-2}$, representing a toughening effect of nearly 50 times. As shown in Fig. 4(b), this extreme toughening effect stems from the synergistic interaction of multiple energy dissipation mechanisms [102, 103]: LMPs dissipate a large amount of energy through deformation at the crack tip region, while simultaneously guiding the crack to undergo adaptive movement. More critically, the fluid properties of the LM effectively blunt or even eliminate stress singularities at the crack tip, thereby effectively suppressing crack initiation and propagation. This ultra-high toughness does not come without a cost—the composite's rebound elasticity typically decreases significantly (i.e., increased hysteresis loss), and it exhibits a pronounced Mullins effect under cyclic loading, indicating that part of the energy dissipation is irreversible [102]. Interestingly, composites embedded with spindle-shaped LMPs can further

increase fracture energy along their long axis to four times that of composites with spherical LMPs, while significantly enhancing the composite's tensile modulus and fracture strain, thereby achieving directional modulation of mechanical response. Notably, even after tens of thousands of tensile cycles under large strains (> 70%), the LMP composites remain mechanically stable [43, 101]. This can be attributed to the reversible interface destruction and repair capabilities of the LMPs, which manifest macroscopically as a significant increase in the fatigue limit.

In summary, the regulation of polymer mechanical properties by LMPs essentially stems from the synergistic interaction between their fluid-solid bimodal behavior and the crackable oxide layer. This design concept resolves the traditional contradiction between filler reinforcement and toughening, providing a new material platform for the development of stretchable electronics, soft robotic skins, and biomimetic devices.

3.2 Isotropic LMP composites

For the rational design of LMP composites, it is generally necessary to formulate them as stable and homogeneous systems to avoid phase separation. However, the easy sedimentation of LMPs causes them to accumulate extensively at the bottom of the composite materials. Consequently, different strategies have been extensively explored to develop isotropic LMP composites, which is crucial for their applications in elastic conductors, dielectric elastomers, and thermal management materials.

3.2.1 Importance of isotropic LMP composites

Without special treatment, LMPs tend to precipitate from the composite solutions. This is the result with the combined effects of gravity, interfacial forces between the particles and polymer matrix, and the drag forces induced by the viscosity of the polymeric solutions [104, 105]. LMP deposition typically results in the formation of LMP composites with a characteristic Janus structure. Due to the localized concentration of LMPs in the Janus structure, they are more likely to form continuous conductive networks under stress. Conversely, in systems where LMP is uniformly dispersed, even with a higher overall LMP concentrations, it is difficult to effectively activate conductive pathways under the same conditions [106]. Some studies have utilized the sedimentation behavior of LMP to achieve a self-sintering effect [107, 108]. However, the isotropic LMP composites are beneficial for enabling continuous and controllable circuit patterning through various printing techniques [109–111].

In applications where the LMP composite shall remain insulated, the Janus structure may cause unintended sintering of LMPs, leading to issues of short-circuiting and degraded performance. For instance, when the LMP composites are used as dielectric materials, the equivalent electrical circuit of the LMP composite's Janus structure can be considered as two capacitors connected in series, as shown in Fig. 4(c). With C_e representing the capacitance of the LMP-enriched layer and C_d representing the LMP-depleted layer, the overall capacitance of the composite is given by

$$C = 1/(1/C_d + 1/C_e) \quad (3)$$

Clearly, the overall capacitance C can only attain the maximum when the two capacitances connected in series are equal, indicating that a highly homogeneous distribution of LMPs is important to maximize the dielectric constant. While greater loadings of LMPs lead to higher dielectric constant of the LMP composite [97], isotropic LMP composites are important to preserve their original isolated state after mechanical strains (Fig. 4(d)). Such isolation prevents the formation of conductive pathways, thereby maintaining low leakage current, reduced dielectric loss, and high breakdown strength. For thermal management materials, electrical conductivity must be carefully controlled to balance heat dissipation with electrical insulation and device safety, especially for polymeric thermal interface materials with phonon-dominated heat transport. When the weight ratios of LMPs in the matrix exceed a specific threshold ($\sim 60\%$ – 70%), the thermal conductivity of the LMP composites reaches several times that of the polymeric matrix [112, 113]. Composites with uniform-dispersed LMPs allow for higher particle loading before the formation of conductive networks within the material under external deformations. In addition, uniform dispersion of LMPs significantly enhances the dielectric properties and thermal conductivity of the composite without largely compromising the tensile strength of the host matrix [39, 97, 113].

3.2.2 Methods for obtaining isotropic LMP composites

The settling behavior of LMP can be described using the modified Stokes settling theory [106]. Assuming the LMP behaves as an ideal sphere suspended in a viscous fluid, the drag force, which determines the particle's setting velocity, can be expressed using the Stokes formula [105]. With buoyancy and drag counterbalancing the LMP's gravity, the settling velocity can be described as

$$v_s = (1 - \varphi)d^2 g(\rho_{LM} - \rho_{Matrix}) \exp(-kt) / 18\mu_{Matrix}\mu_r \quad (4)$$

where φ is LMP volume fraction; d represents the average diameter of LMP; g is the gravitational acceleration; ρ_{LM} and ρ_{Matrix} represent the densities of LM and polymeric materials, respectively; k is a rate constant for viscosity; t is curing time; μ_{Matrix} and μ_r represent the initial viscosity of the polymer and relative viscosity between the suspension and polymer, respectively. According to Eq. (4), for a specific material combination, the LMP sedimentation can be addressed by the following measures: i) adjusting the size and loading of LMP; ii) increasing the viscosity of the composite; and iii) reducing the solidification time of the composite. Among these, promoting polymer cross-linking through external conditions represents a simple and effective approach. Wang et al. [114] confirmed that accelerating composite curing through heating effectively mitigates LMP sedimentation.

Adjusting the concentration of LMP is a highly effective method for preventing LMP sedimentation [100, 106]. Increasing the concentration of LMP significantly raises the macroscopic viscosity of the composite, thereby increasing the internal friction of the system and the interactions between particles. The sedimentation effect caused by the size of LMP is even more pronounced. As can be seen in Figs. 4(e)(i), samples containing larger particles separated and accumulated at the bottom more quickly than those containing smaller particles. Figure 4(e)(ii) shows the distance from the interface of the lower LMP-enriched zone and the upper LMP-depleted zone to the container bottom over different times. All samples reached a stable state after a certain period, with no further LMP precipitation occurring. This stabilization is attributable to increased viscosity in the lower region [115]. Samples with smaller average diameters achieved stability more quickly and exhibited minimal interface changes. This confirms that reducing the size of the LMPs effectively minimizes precipitation. The elastomer incorporating small-scale LMPs demonstrated superior dispersion, with their electrical breakdown strength demonstrating a stable and linear decrease as LMP concentration increases. Under identical LMP loading conditions, the dielectric constant of elastomers containing smaller LMPs is significantly higher than that of dielectric elastomers containing larger LMPs [39]. However, high loadings combined with a small particle size result in brittle or granular materials. This is due to the large surface area-to-volume ratio of smaller particles, which prevents the elastic polymer from fully encapsulating all LMPs [91].

The jamming effect caused by the introduction of other particles such as Fe or BaTiO₃ particles can increase the viscosity of the system, thereby slowing the sedimentation of LMP [116]. It should be noted that the purpose of introducing these particles is typically to enhance the dielectric properties or thermal conductivity of LMP composites. For example, doping small amounts of low-density, high aspect ratio graphene oxide (GO) coated with polydopamine (PDA) can elevate the dielectric constant without significantly altering the elasticity of matrix, while avoiding the need for further increases in LMP concentration [117]. Naturally, careful consideration must be given to the detrimental effect that rigid fillers may have on the material's tensile properties. Certain soft particles have also been considered to stabilize the system. As shown in Fig. 4(f), incorporating polydimethylsiloxane particles (PPs) increases solution viscosity, which significantly suppress the sedimentation of smaller-scale LMPs. However, excessive filler content can compromise mechanical properties, and a volume fraction of 20% PP has been demonstrated to effectively

prevent sedimentation while maintaining good flexibility of the composite [105].

The interaction between LMPs and the polymer matrix should not be overlooked. Since most polymers lack functional groups capable of forming strong bonds with the surface of LMPs, they primarily rely on weak physical interactions to stabilize the particles, which makes it difficult to effectively suppress LMP sedimentation in the composites. For example, the contact angle of LMP droplets on SEBS films can be as high as 115°, which could cause the LMPs to detach and leak from the SEBS polymer matrix after curing. Polymers capable of forming chemical bonds with LMP surfaces have been investigated. Banerjee et al. [118] modified the maleic anhydride (MA) segment in maleic anhydride-grafted SEBS (SEBS-g-MA) block copolymers by infusing pendant hydroxyl groups along the main chain using 1-aminopropanol (AMP). The epoxy atom from the MA group was replaced by the amine nitrogen from the AMP, resulting in the formation of an imide. As shown in Fig. 4(g), formation of hydrogen bonds between the induced functional groups and the oxide layers on the LMP surface was key to mitigating LMP deposition [118]. Another approach involves inducing surfactants onto the surface of LMPs. The surface-modified LMPs can provide electrostatic repulsion to prevent them from agglomerating with each other [119]. For example, Su et al. [120] achieved stable LMP colloids by coating LMPs with oleic acid (OA). The adsorption of OA onto LMPs created a functionalized surface containing unsaturated carbon-carbon double bonds. This facilitated radical polymerisation with n-butyl methacrylate (BMA) monomers, forming highly dispersed LMP dielectric elastomers (Fig. 4(h)).

In summary, surface ligand grafting can help to suppress

agglomeration and sedimentation caused by high surface tension and density differences. Furthermore, the jamming effect can be utilized to further regulate the sedimentation behavior of LMPs. The highly dispersed isotropic LMP composites hold significant value for dielectric elastomers and thermal management materials. Table 2 summarizes the composition design, dielectric properties, and application scenarios of isotropic LMP composites from representative studies.

4 Electrical interconnections

LMP composites require treatment to become conductive due to the insulating oxide layer and surrounding polymeric matrix. Lee et al. [44] have comprehensively summarized the sintering techniques of LMP films and composites. Here, we focus on recent advances in two widely exploited strategies for the electrical interconnections of LMP composites: i) sintering under external stimulus such as stress, temperature, electric field, and evaporation; ii) interconnection with functional fillers. Table 3 provides a detailed comparison of LMP composites reported in several representative studies, covering material design, electrical performance, and electrical connection methods, as well as their merits & demerits.

4.1 LMP composites with pure LMPs

4.1.1 Mechanical sintering

Mechanical stress sintering utilizes external stress applied to LMP composites to induce deformation of the LMPs, causing the originally isolated LMPs to undergo axial elongation or radial compression. Once the local strain exceeds a critical threshold, the

Table 2 Material composition and properties of isotropic LMP composites

Materials	Mean size of the filler	Dielectric constant	Filler loading	Stretchability	Application	Ref.
PDMS/galinstan/PDMS particle (PP)	$d_{\text{LMP}} \approx 10 \mu\text{m}$ $d_{\text{PP}} < 225 \mu\text{m}$	36.7 (1 MHz)	$\varphi_{\text{LMP}} = 40\%$ $\varphi_{\text{PP}} = 30\%$	NM	Capacitive stress sensor	[105]
PDMS/EGaIn	$d_{\text{LMP}} = 80 \mu\text{m}$	60 (90 kHz)	$\varphi_{\text{LMP}} = 80 \%$	~ 400%	Capacitive strain sensor	[91]
PDMS/EGaIn/graphene oxide (GO)	$d_{\text{LMP}} = 9.5 \mu\text{m}$	60 (1 kHz)	$\varphi_{\text{LMP}} = 50\%$ $\omega_{\text{GO}} = 4\%$	~ 80%	Capacitive strain sensor	[117]
Poly(n-butyl methacrylate)/EGaIn	$d_{\text{LMP}} = 300 \text{ nm}$	22 (1 kHz)	$\varphi_{\text{LMP}} = 40\%$	~ 450%	NM	[120]
PDMS/galinstan/Fe	NM	~ 40 (1 kHz)	$\varphi_{\text{LMP}} = 30\%$ $\varphi_{\text{Fe}} = 30\%$	NM	Capacitive pressure sensor	[116]
PDMS/galinstan/BaTiO ₃ (BTO)	NM	10–11 (1 kHz)	$\varphi_{\text{LMP}} = 30\%$ $\varphi_{\text{BTO}} = 20\%$	NM		
PDMS/EGaIn	$d_{\text{LMP}} = 10 \mu\text{m}$	10–12 (1 kHz)	$\varphi_{\text{LMP}} = 40\%$	110%–120%	Dielectric elastomer actuator Dielectric elastomer generator	[39]
PDMS/EGaIn		4–5 (1 kHz)		110%–120%		
Ecoflex/EGaIn	$d_{\text{LMP}} = 10 \mu\text{m}$	5–6 (1 kHz)	$\varphi_{\text{LMP}} = 10\%$	NM		
PU/EGaIn		12–14 (1 kHz)		NM		
PVDF-HFP/EGaIn		15–16 (1 kHz)		NM		
Ecoflex/EGaIn	$d_{\text{LMP}} = 4\text{--}15 \mu\text{m}$	17–18 (1–200 kHz)	$\varphi_{\text{LMP}} = 50\%$	~ 600%	Capacitive strain sensor	[97]
PU/EGaIn	$d_{\text{LMP}} < 20 \mu\text{m}$	40–45 (1–200 kHz)	$\varphi_{\text{LMP}} = 50\%$	182%		
Silicone dielectric gel/EGaIn	$d_{\text{LMP}} = 23.64 \pm 6.12 \mu\text{m}$	~ 14 (0.01–100 kHz)	$\varphi_{\text{LMP}} = 50\%$	> 150%	Waste energy harvester	[51]
PU/EGaIn	$d_{\text{LMP}} = 0.3 \mu\text{m}$	~ 76.8 (100 kHz)	$\varphi_{\text{LMP}} = 50\%$	> 100%	NM	[121]
Maleic anhydride grafted SEBS/EGaIn	$d_{\text{LMP}} = 1.0\text{--}1.5 \mu\text{m}$	15–16 (500 Hz–1 MHz)	$\varphi_{\text{LMP}} = 40\%$	~ 1500%	Thermal management	[118]
Natural rubber/EGaIn	NM	2.75 (100 Hz)	50 phr (parts per hundred of rubber by weight)	~ 650%	NM	[122]

Table 3 LMP composites based on different methods for achieving electrical interconnection

Materials	Filler specification	Conductivity	Method	Merits and demerits	Ref.
PDMS/Galinstan	$d_{\text{LMP}} = 2\text{--}30\ \mu\text{m}$ $\phi_{\text{LMP}} = 50\%$	$1.05 \times 10^4\ \text{S}\cdot\text{m}^{-1}$	Pressure	Simple and rapid process. Suitable for various matrices. Easily adjustable conductivity and robust electrical stability. Capable of patterning conductive pathways	[34]
PDMS/EGaIn	$\phi_{\text{LMP}} = 80\%$	$5 \times 10^4\ \text{S}\cdot\text{m}^{-1}$		Hard contact may damage fine patterns or substrate. Not suitable for curved surfaces. Risk of unintended sintering. Leakage of LM	[126]
TPU/EGaIn	$d_{\text{LMP}} \approx 1.4\ \mu\text{m}$ $\phi_{\text{LMP}} = 85\%$	$24,130\ \text{S}\cdot\text{m}^{-1}$	Stretching	Simple operation. Highly sensitive to tensile strain. Suitable for one-dimensional stretchable conductive materials	[121]
Carbopol hydrogel/EGaIn	$d_{\text{LMP}} \approx 35\ \mu\text{m}$ $\phi_{\text{LMP}} = 82.5\%$	$2.6 \times 10^5\ \text{S}\cdot\text{m}^{-1}$		Requires matrices with good elasticity. Strict requirements for LMP size (smaller sizes cannot be sintered). Stress-adaptive period of the uniform conductive network and significant drift in the initial resistance change	[78]
Ecoflex/EGaIn	$d_{\text{LMP}} \approx 5\ \mu\text{m}$	$8 \times 10^3\ \text{S}\cdot\text{m}^{-1}$	Bending	Easy to use. Capable of creating gradient conductive structures. Suitable for thin-film conductive materials	[132]
Epoxy resin/EGaIn	$d_{\text{LMP}} < 5\ \mu\text{m}$ $\phi_{\text{LMP}} = 50\%$	NM		The conductive pathways may be primarily confined to the outer tension zone. Limited bending deformation requires high LMP loading. Prone to causing LM leakage	[131]
PU/EGaIn	$d_{\text{LMP}} = 2\text{--}3\ \mu\text{m}$ $\phi_{\text{LMP}} = 74.1\%$	$2.10 \times 10^6\ \text{S}\cdot\text{m}^{-1}$	Ultrasonication	Non-contact/remote sintering. No damages to the conductive patterns and no risk of LM leakages. Compatible with a wide range of matrices. Uniform conductive penetration network. High-throughput process	[101]
PVP/EGaIn	$d_{\text{LMP}} \approx 4.21\ \mu\text{m}$ $\omega_{\text{LMP}} \approx 83.3\%$	$\sim 2 \times 10^5\ \text{S}\cdot\text{m}^{-1}$		Relatively high equipment costs. Strict optimization parameters are required Thermal effects or cavitation damage may occur	[134]
PDMS/Galinstan	$d_{\text{LMP}} = 1.91 \pm 1.21\ \mu\text{m}$ $\omega_{\text{LMP}} \approx 98.4\%$	$7.7 \times 10^5\ \text{S}\cdot\text{m}^{-1}$	High temperature induction	Fast, efficient, and controllable. High patterning accuracy. High conductivity and stable performances. No restrictions on matrices or LMP size	[141]
PAA/EGaIn	$d_{\text{LMP}} < 5\ \mu\text{m}$	NM		High risk of thermal damage, which may ruin intricate patterns or the substrate. Expensive equipment and complex process. May accelerate the oxidation of LM due to overheating	[142]
PVDF-HFP-TFE/EGaIn	$d_{\text{LMP}} \approx 3.6\ \mu\text{m}$	$4.35 \times 10^4\ \text{S}\cdot\text{m}^{-1}$	Low temperature induction	Reversible electrical properties. High requirements for LMP (smaller particles increase supercooling, raising the sintering temperature limit)	[143]
Pt-catalyzed silicone polymer/EGaIn	$d_{\text{LMP}} = 12\text{--}140\ \mu\text{m}$ $\phi_{\text{LMP}} = 50\%$	NM		Require elastic substrates with good freeze resistance Freezing may damage the substrate material. The process relies on extremely low temperatures, such as those achieved with liquid nitrogen, which increases equipment costs and safety requirements	[114]
Carbopol hydrogel/EGaIn	$d_{\text{LMP}} \approx 35\ \mu\text{m}$ $\phi_{\text{LMP}} = 82.5\%$	NM	Electrochemistry	No complex equipment required. Easy to operate and low energy consumption. Depends on electrochemical reactions. Specific requirements for the material systems. Sensitive to applied voltages. Limited conductivity. The matrix is limited to hydrogels	[78]
PDMS/EGaIn	$d_{\text{LMP}} = 150\text{--}200\ \mu\text{m}$ $\omega_{\text{LMP}} = 10\%$	NM	Dielectrophoresis	High-precision assembly and precise LMP control. Low LMP loading requirements. Assembled and sintered directly within the polymer matrix The formation of conductive pathways depends on the configuration of the auxiliary electrodes. Limited LMP sintering capability. Precise control of electric field parameters is required. Specific requirements for the dielectric properties of the matrix	[146]
PSS/EGaIn	$d_{\text{LMP}} < 2.5\ \mu\text{m}$	$2.4 \times 10^6\ \text{S}\cdot\text{m}^{-1}$	Self-sintering	No additional energy input and post-processing are required. Compatible with a wide range of matrices. High conductivity comparable to that of bulk LM. Capable of forming a Janus structure with self-encapsulating properties	[153]
PVA/Galinstan	$d_{\text{LMP}} < 10\ \mu\text{m}$ $\phi_{\text{LMP}} = 75\%$	$8.75 \times 10^6\ \text{S}\cdot\text{m}^{-1}$		Relies on controlled evaporation conditions. Specific requirements for the formulation of the dispersion. Lacks process stability and long-term reliability in large-scale production	[108]
WPU/EGaIn	$d_{\text{LMP}} \approx 7\ \mu\text{m}$ $\omega_{\text{LMP}} = 93.75\%$	$3 \times 10^6\ \text{S}\cdot\text{m}^{-1}$			[43]
PVA-Borax gel/EGaIn/Ag	$d_{\text{Ag}} = 3\text{--}10\ \mu\text{m}$ $d_{\text{LMP}} = 5\text{--}50\ \mu\text{m}$	$7 \times 10^4\ \text{S}\cdot\text{m}^{-1}$			[162]
SIS/EGaIn/Ag	$d_{\text{LMP}} < 5\ \mu\text{m}$ $\omega_{\text{LMP-Ag}} = 97\%$	$7.02 \times 10^5\ \text{S}\cdot\text{m}^{-1}$	Conductive bridge	Intrinsically conductive. High electrical conductivity and robust electromechanical properties. Tolerant of material selection. Simple material synthesis	[42]
SIS/Galinstan/Cu	$d_{\text{LMP}} \approx 5\ \mu\text{m}$ $\omega_{\text{Cu}} = 8.8\%$ $\omega_{\text{LMP}} = 88\%$	$> 10^4\ \text{S}\cdot\text{m}^{-1}$		The wettability of liquid metals and conductive bridge materials needs to be addressed. A trade-off between tensile strength and electrical conductivity must be considered. The use of precious metals such as silver results in a significant increase in cost	[166]
SBS/GaInSn/MXene	$d_{\text{MXene}} \approx 10\ \mu\text{m}$ $d_{\text{LMP}} \approx 20\ \mu\text{m}$ $\omega_{\text{MXene}} = 0.9\%$ $\omega_{\text{LMP}} \approx 90\%$	$833.407\ \text{S}\cdot\text{m}^{-1}$			[163]
Ecoflex/EGaIn/Cu@Fe	$d_{\text{Cu@Fe}} = 10\ \mu\text{m}$ $\omega_{\text{LMP}} = 80\%$	NM		Low LMP loading requirements. Controllable sintering pathways. Self-sealing properties prevent liquid metal leakage. Suitable for most substrate materials	[170]
Ecoflex/EGaIn/Ag@Ni	$d_{\text{Ag@Ni}} = 31\ \mu\text{m}$ $\omega_{\text{LMP}} = 35.7\%$ $\omega_{\text{Ag@Ni}} = 3.6\%$	$2.55 \times 10^5\ \text{S}\cdot\text{m}^{-1}$	Internalized magnetic particles	The LMP requires magnetization treatment. Complex and inefficient process. The unsintered areas are almost entirely wasted. Relies on precise control of the magnetic field, resulting in a narrow process window. Severely limited conductivity	[172]

brittle gallium oxide shell fractures, exposing the high-surface-tension LM core, which coalesces to form a three-dimensional conductive network within the polymeric matrix. Mechanical deformations are the key triggers for the electrical transition. Thus, different forms of mechanical inputs yield significant differences in the resulting effects.

As shown in Fig. 5(a), the use of a pressure mold or pen will enable controllable patterning of the conductive pathways [34, 123, 124]. Pressure-induced sintering relies on normal compressive stress directly overcoming the critical fracture strength of the oxide shell and the barrier of the polymer layer (Fig. 5(b)), resulting in a sudden drop in resistance and permanent change in the electrical properties of the LMP composites. For instance, Andrew Fessler and Carmel Majidi [34] produced conductive elastomers by incorporating LMPs into PDMS. Applied surface pressure permanently altered both the material structure and bulk properties, transforming it from non-conductive materials to conductive materials with a bulk conductivity of approximately $\sigma = 1.05 \times 10^4 \text{ S}\cdot\text{m}^{-1}$. However, the pressure-activation approach also introduces stress concentration in unintended areas due to

dissipation forces, leading to unexpected sintering. This often requires the material's surface to be coated with an additional elastomer for protection [34, 123, 125]. Alternatively, patterning the LMP composites onto substrates and applying pressure-activated sintering represent a reliable method for achieving desired conductive pathways [3, 34, 126, 127].

Tensile deformation primarily applies radial compressive stress to the particles through the transverse contraction caused by the Poisson effect (Fig. 5(c)), prompting the particles to align along the tensile axis and fuse to form conductive fiber bundles [49, 78, 128, 129]. Unlike the switch-type activation observed in pressure-induced sintering, the change in resistance during tensile sintering typically follows a monotonically decreasing trend and eventually reaches a steady state. This method imposes stringent requirements on the size of LMP [121]. Smaller particles cannot be effectively activated due to increased surface tension and enhanced stress shielding from the substrate. Cyclic loading on the LMP composite also revealed a significant tensile stress-adaptive period in its electrical conductivity, showing drift in the composite's resistance change [121, 128]. As the number of loading cycles increased, the

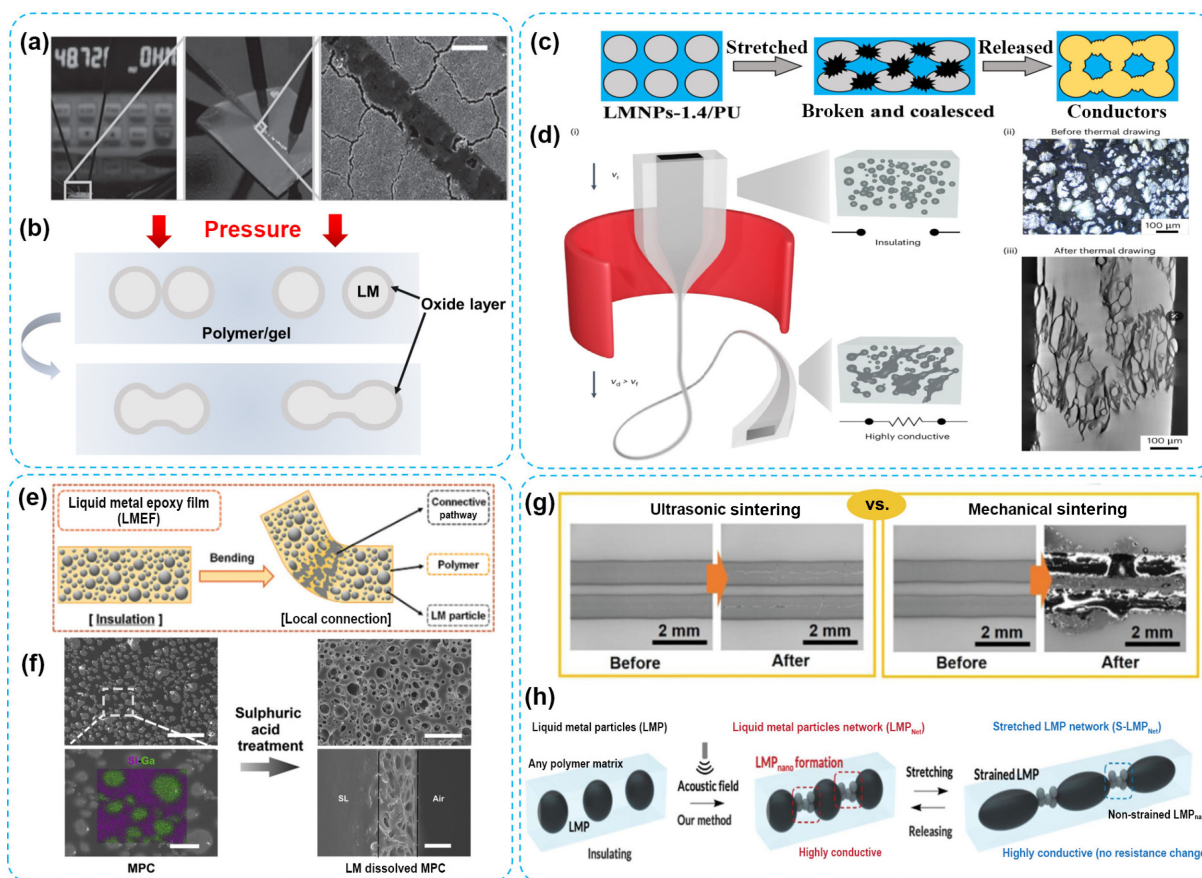


Figure 5 Mechanical sintering for LMP composites. (a) Representative SEM image showing the contact area morphology transitioning from a nanoparticle network to a liquid fusion line. Scale bar length: 20 μm. Reproduced with permission from Ref. [75], © WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim 2015. (b) The formation of conductive permeation networks within LMP composite under pressure. (c) Schematic of electrical activation of LMPs during tensile deformation. Reproduced with permission from Ref. [121], © Liu, Y. et al. 2021. (d) Schematic diagram of the thermal drawing process and the microstructure of LMP before and after thermal drawing. Reproduced with permission from Ref. [130], © Laperrousaz, S. et al. 2025. (e) The mechanism of the mechanically induced anisotropic conductivity of HED LM-epoxy resin under bending stress. Reproduced with permission from Ref. [131], © WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim 2020. (f) SEM and elemental analysis of LMP composite surface. Scale bars: top image 30 μm; bottom image 5 μm (left). SEM of LMP composite after dissolution of LM. Top image: surface morphology, scale bar 20 μm; bottom image: cross-sectional morphology, scale bar 10 μm (right). Reproduced with permission from Ref. [132], © Tang, L. X. et al. 2018. (g) Comparison of the LMP composite patterns treated by ultrasonic sintering (left) and mechanical sintering (right). Reproduced with permission from Ref. [134], © Liu, S. H. et al. 2023. (h) Schematic illustration of the formation of a highly conductive LMP network via ultrasonic sintering. Reproduced with permission from Ref. [101], © Lee, W. et al. 2022.

resistance changes converged. This is attributed to the gradual penetration of the LM under repeated tensile stress, resulting in the formation of a highly uniform conductive network. Interestingly, leveraging the tensile sintering principle in combination with thermal drawing processes makes it possible to design stretchable conductive fibers that do not require subsequent sintering treatment (Fig. 5(d)). The shear stress generated during thermal drawing has been shown to be sufficient to form conductive domains within the fibers [130].

Bending deformation creates a gradient strain field in the thickness direction of the material, causing the LMPs to fracture and coalesce under compressive stress, establishing electrical conduction pathways. As shown in Fig. 5(e), the non-uniform stress distribution results in electrical pathways forming primarily in the tensile surface layer, exhibiting overall gradient electro-activation characteristics along the bending direction [131]. Tang et al. [132] achieved circuit formation through sustained bending by directly peeling the LMP composite from a rigid substrate. Exposure of the sintered LMP composite to sulfuric acid solution resulted in complete dissolution of the LM, leaving behind an insoluble polymer with a continuous and hollow structure (Fig. 5(f)). This result provides microscopic confirmation of the formation of a continuous conductive network using this method.

Mechanical vibration sintering, also known as ultrasonic sintering, applies energy in a non-contact manner to break down particle shells [133]. Unlike mechanical stress sintering discussed above, this technique enables LMP interconnection without damaging circuit patterns or substrates (Fig. 5(g)), thereby resolving issues such as circuit failure and short circuits caused by LM leakage due to stress overload [101, 134]. Furthermore, this method can be applied to complex surfaces that are inaccessible to conventional mechanical stress sintering methods, and it typically offers higher and more uniform sintering efficiency [134]. Lee et al. [101] provided a comprehensive explanation of the formation of conductive networks via ultrasonic sintering (Fig. 5(h)). Firstly, acoustic energy is initially transmitted from the ultrasonic probe through the aqueous medium into the composite material, accumulating at the LMP-polymer-LMP interface. This accumulation arises from the significant difference in acoustic impedance between the polymer and LMP: The polymer is acoustically permeable, while the LMP has high acoustic impedance and reflects most of the wave energy. Subsequently, the aggregated acoustic energy initiates the formation of nanobubbles (cavitation) at the LMP-polymer-LMP interface. Ultimately, the collapse of these nanobubbles fractures the natural oxide shell on the LMP surfaces, generating nanoscale LMPs. These newly formed particles effectively interconnect the LMPs, enabling the materials to maintain good electrical conductivity even under high strains [101].

4.1.2 Temperature-regulated induction

The liquid core within the core-shell structure of LM has a larger coefficient of thermal expansion compared to the oxide shell [135]. Under the thermal stress, LMPs undergo morphological changes and eventually interconnect to form conductive pathways (Fig. 6(a)). Controlled high-temperature induction for LMP electrical interconnections can be achieved using lasers with high energy density. The absorption of these focused high-energy laser beams enables rapid localized heating, generating elevated thermal stresses for LMP sintering (see Fig. 6(b)) [136–139]. The width of sintered lines can be effectively controlled by adjusting laser

parameters such as the power, pulse frequency, spot size, and wavelength [138, 140, 141]. This facilitates the high-volume production and widespread application of conductive LMP composites. Processing an LMP-PDMS composite through this method enabled the high-resolution patterning of circuits on material films, yielding individual conductive traces as narrow as 37 μm and a conductivity of $7.7 \times 10^5 \text{ S}\cdot\text{m}^{-1}$ [141]. Figure 6(c) shows the activated and non-activated regions within the LMP-PDMS composite material. Laser ablation removed part of the top PDMS insulating barrier, while the gallium oxide shell fractured due to substantial thermal stresses induced by the rapid expansion of the LM core. This enabled the liquid to escape and form a continuous network. The formation of the conductive structure may also have been facilitated by the fracturing of other PDMS interface layers not directly ablated by the laser [135, 141]. Compared to mechanical sintering, laser sintering requires a higher LM loading (> 98%) within the LMP composites [141, 142]. This may potentially induce unintended mechanical activation in non-targeted areas during utilization of the materials.

The key mechanism underlying low-temperature induction for LMP electrical interconnections is the differential volume change between the LMPs and the insulating polymer matrix [3, 114, 143, 144]. Under cryogenic conditions, the polymer matrix hardens and contracts due to vitrification, while the LMPs expand as they solidify. This disparity helps to rupture the surface oxide barrier layer of the isolated LMPs, thereby interconnecting them to form conductive pathways. The freezing-activation mechanism enables a reversible transition between insulator and conductor (TIC) [114, 144]. As shown in Fig. 6(d), the rigid and expanded LMPs protrude from the rigid polymer matrix and interconnect to establish conductive pathways after a cryogenic treatment over 212 K. Upon recovery to room temperature, the rigid LMPs soften and contract into a liquid state with reduced volume whilst the rigid silicone regains its flexibility and encapsulates the LMPs internally. This restores the TIC material from its conductive state to insulating state. As shown in Fig. 6(e), the low temperature-induced change in electrical properties spans over nine orders of magnitude [114]. The resistance of TIC materials at a temperature of 212 K rapidly decreases from an almost infinite value (beyond the measurement range) to an ultra-low value ($1.78 \times 10^{-5} \Omega\cdot\text{m}$). Figure 6(f) shows that, after over 100 temperature cycles, there was no degradation or damage to the material's electrical properties, demonstrating the reliability of this cycling mechanism. Due to the significantly lower solidification temperature of micro-scale LMPs compared to bulk LMs [73, 114], the low-temperature environment may greatly limit their practical applications. Nevertheless, it shows promise for autonomous electrical connection and conversion in extraterrestrial exploration scenarios involving abrupt environmental temperature fluctuations.

4.1.3 Electric field induction

Recently, the electrical interconnection between LMPs induced by electric fields within hydrogel systems has been demonstrated [78]. Lin et al. designed a liquid metal high internal phase emulsion gel (LM-HIPEG) ink comprising EGaIn LM as the dispersed phase and carbopol hydrogel as the continuous phase. Carbopol molecules form strong adsorptive interactions with the LM oxide layer to maintain colloidal stability. Applying a bias voltage exceeding the threshold voltage (9 V) to the as-printed line ($d = 210 \mu\text{m}$, $L = 3 \text{ cm}$) resulted in a rapid decrease in resistance after a certain

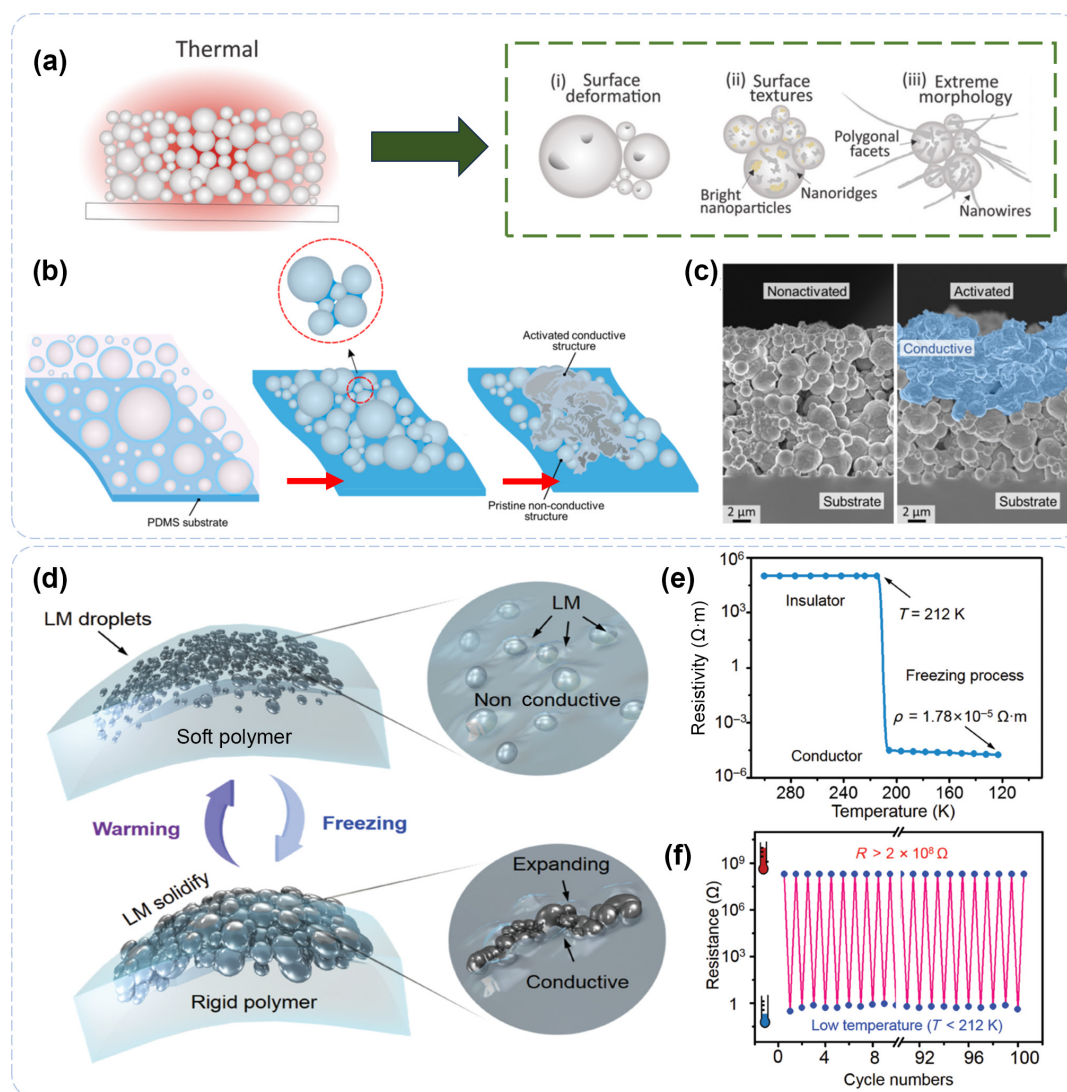


Figure 6 Temperature-regulated induction for electrical interconnection of LMP composites. (a) Schematic diagram illustrating the mechanism of thermal sintering for LMPs films. Reproduced with permission from Ref. [135], © The Royal Society of Chemistry 2019. (b) Laser activation transforms the pristine, non-conductive structure into a complex, continuous network with conductive properties. (c) SEM images showing cross-sectional views of the LMP-PDMS composite before and after laser activation. Reproduced with permission from Ref. [141], © American Chemical Society 2021. (d) Schematic illustration of mechanism in electrical transition between insulated and conductive TIC materials in response to temperature change. (e) The resistivity of TIC materials as a function of temperature. (f) Resistance change of TIC materials for 100 cycles under the temperature-regulated conditions. Reproduced with permission from Ref. [114], © WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim 2019.

period. The response time diminishes with increased voltages (Fig. 7(a)), a phenomenon attributable to the electrocapillary effect [78, 145]. As illustrated in Fig. 7(b), an electric double layer (EDL) forms at the interface between the LMPs and the electrolyte gel. The applied electric field redistributes charges and ions at the interfaces, creating a charge density gradient aligned with the electric field direction. The process will induce an interfacial tension gradient, causing the LM to flow towards regions of higher interfacial tension. Consequently, the shape of the LMP is altered, leading to the rupture of the oxide layer and the formation of conductive pathways [78].

Additional auxiliary electrodes are required to apply an electric field to elastic polymer systems. Krisnadi et al. [146] reported a dielectrophoresis (DEP)-based method for aligning and assembling LMPs to form micrometer-scale conductive wires. As illustrated in Fig. 7(c), Au/Cu electrodes were deposited onto a substrate. When

the LMP-PDMS composite was placed on the electrodes, the LMPs became polarised and aggregated at the edges of the electrodes, forming electrically continuous wires where the electric field gradient was maximal. The formation of wires correlates with the content of LMPs and the strength of the electric field. Following PDMS solidification, the preserved conductive pathways exhibited an unchanged resistance under 100% tensile strain (see Fig. 7(d)). The electro-induced sintering process enables on-demand circuit formation, which can be achieved by selectively creating conductive pathways at targeted locations through the application of voltages on designed electrode patterns [146].

4.1.4 Self-sintering

Surfaces of the LMPs can be modified using different surface modifiers in the solutions, which increases the interactions of LMPs with various polymeric materials and facilitates the formation of

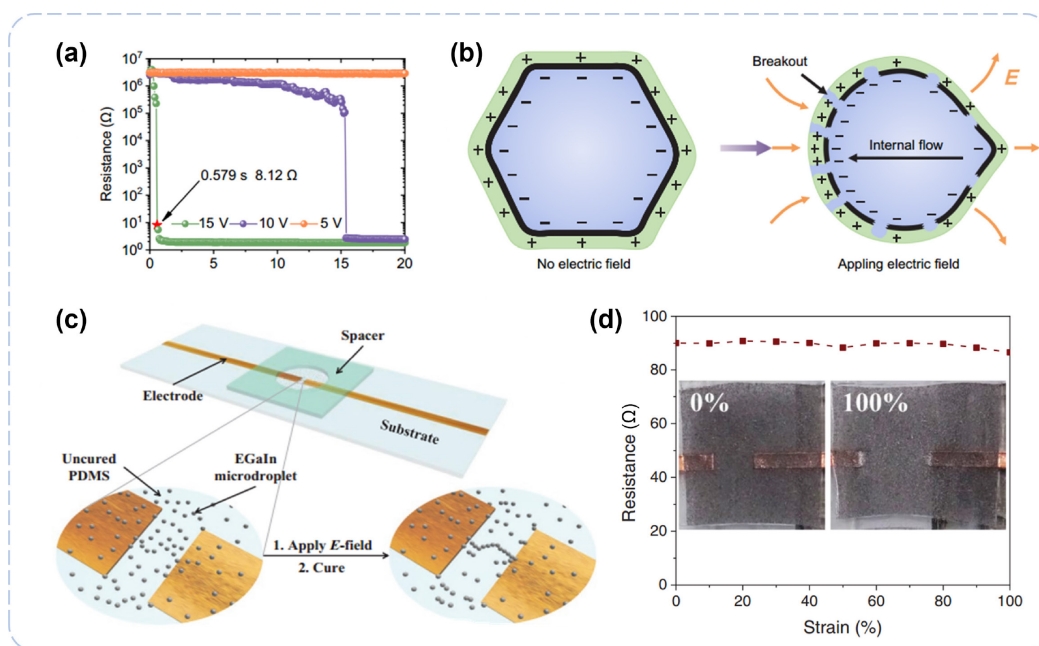


Figure 7 Electric field induction for electrical interconnection of LMP composites. (a) Comparison of activation times for the as-printed line under different bias voltages. (b) Schematic illustration of the voltage-induced capillary effect of EGaIn LMPs in LM-HIPEG. Reproduced with permission from Ref. [78], © Lin, Z. W. et al. 2024. (c) Schematic illustration of DEP-assisted assembly of EGaIn LM microwires inside PDMS matrix. (d) Resistance of a 5 mm-long, DEP-aligned EGaIn microwire versus strain. The inset shows the sample in an unstretched state and under 100% tensile strain. Reproduced with permission from Ref. [146], © WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim 2020.

homogeneous LMP colloids suitable for different applications [147]. Capillary forces generated during solvent evaporation have been shown to encourage the fusion of LMPs [107, 147, 148]. For example, using carefully selected surface modification materials, such as cellulose nanofibrils (CNFs), not only enhances the colloidal stability of LMPs but also promotes capillary forces and split liquid bridges between them (see Fig. 8(a)). This generates high tensile forces on the oxide shell, enabling evaporation-guided self-sintering [107]. LMPs without CNFs were deposited randomly on the substrate after drying, showing no significant aggregation or connection. In contrast, particles with CNFs formed a continuous liquid metal phase (Fig. 8(b)).

Further studies [149–151, 108, 152] have reached similar conclusions: Colloidal LMP solutions evaporate at room temperature or heated temperature (which disrupts the coordination between the LMPs and the additives), enabling the LMPs to self-assemble vertically under the force of gravity. This is followed by sintering via capillary forces, which typically yields a conductive layer rich in LM that is overlaid by a self-sealing layer [108, 152]. When preparing LMPs by sonicating LM in an N-methyl-2-pyrrolidone (NMP) solvent, the NMP undergoes a ring-opening reaction during thermal activation to form ring-opened NMP (roNMP). Upon binding to LMPs as a ligand, roNMP stabilises the dispersion of LMPs and significantly reduces the thickness of the oxide layer. Compared to bare LMPs, the oxide layer thickness on ligand-bound LMP surfaces decreases by approximately 42.8% [108], which is advantageous for sintering to form conductive pathways. As illustrated in Fig. 8(c), ligand detachment during thermal evaporation enables LMP deposition (an indispensable step in capillary sintering), resulting in a bilayer structure with top encapsulating polymer and bottom LMPs.

Interestingly, unlike the aforementioned approach which relied on gravity for the self-assembly of LMPs, Li et al. [153] used poly

(sodium 4-styrenesulfonate) (PSS) to functionalize the LMP surfaces and impart a negative surface charge. They then modified the target substrate with poly (diallyldimethylammonium chloride) (PDMA) to impart positive charges. The strong electrostatic attractions between the oppositely charged LMPs trigger the LMPs to self-assemble rapidly and densely upon the substrate. After removing the LMP-coated substrate from the solution and drying, capillary forces establish electrical connectivity within the LMPs, achieving a conductivity of up to $2.4 \times 10^6 \text{ S}\cdot\text{m}^{-1}$.

Electrostatic interactions between LMPs and polymers during evaporation have been recently reported to create conductive pathways [43, 154]. Molecular dynamics simulations of polymer aggregation reveal that electrostatic interactions between functional groups in polymers and gallium oxide are decisive in enabling LM to spontaneously permeate the polymer network [154]. The bulk LM can be sheared within a water polyurethane (WPU) dispersant to obtain LMPs uniformly coated with WPU by exploiting the electrostatic attraction between Ga from Ga^{3+} oxides on the LMP surface and hydrophilic carboxyl groups on the exterior of polyurethane nanoparticles. During drying, the polymer chains within the dispersion and those attracted to the LM surface progressively approach and entangle one another, resulting in a three-dimensional polymer network between the LMPs (see Fig. 8(d)) [43]. Compressive stresses arising from volumetric shrinkage force the LMPs to interconnect, thereby establishing conductive pathways throughout the entire volume. The elastic, hydrogel-inspired metalgel exhibits exceptional electrical properties and maintains a stable conductivity of $3 \times 10^6 \text{ S}\cdot\text{m}^{-1}$ even after 1,000,000 tensile cycles. The durable and free-standing metalgel film has a smooth and uniform appearance with a silvery-white lustre and exhibits no cracks even under high biaxial stretching (Fig. 8(e)). Within just a few months, Tian et al. [155] reported a highly conductive LMP composite by a similar evaporation-sintering

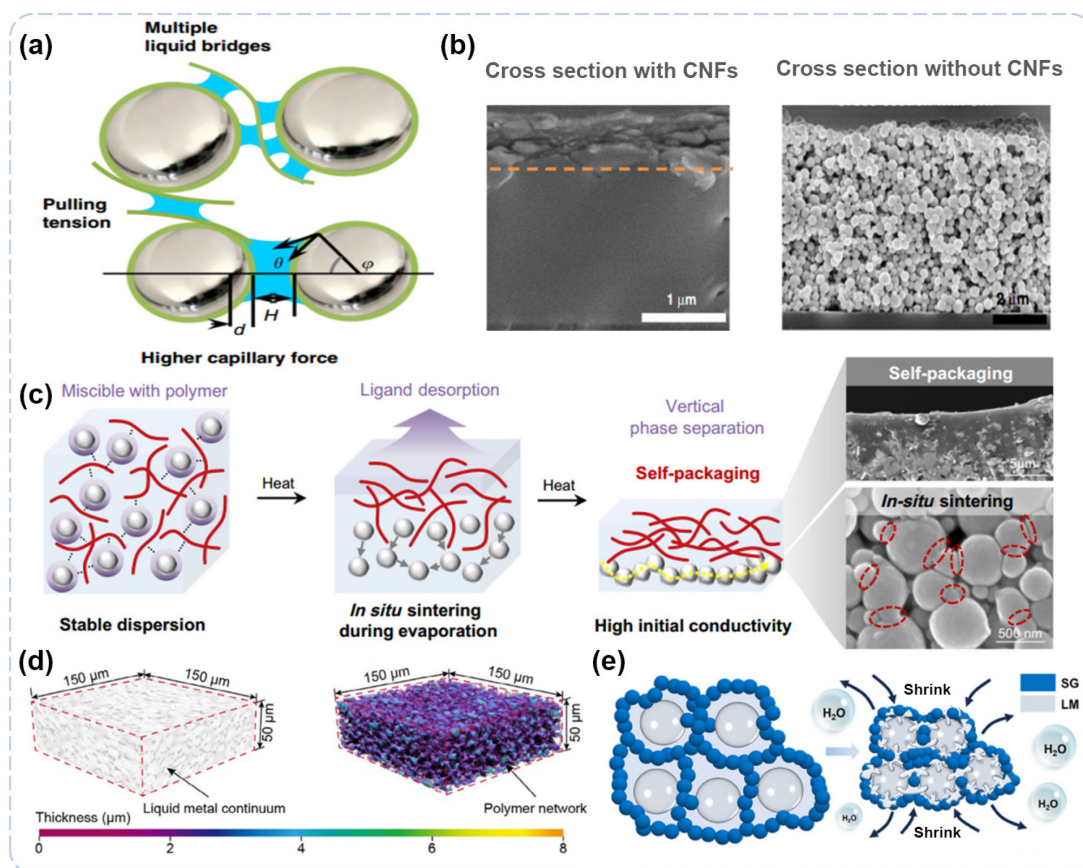


Figure 8 Self-sintering for LMP composites. (a) Schematic illustration of sintering mechanism: capillary force promoted by biological NFs, local pulling tension induced by biological NFs and splitting into multiple liquid bridges. (b) Cross sectional SEM images of LMPs layer with (Top) and without CNFs (Bottom). Reproduced with permission from Ref. [107], © Li, X. K. et al. 2019. (c) Schematic illustration and SEM images showing the *in situ* sintering and self-packaging of LB-LMPs in a polymer matrix. Reproduced with permission from Ref. [108], © Seo, H. et al. 2025. (d) Micro-CT image of the durable metalgel (left) and its porous polymer network (right). Reproduced with permission from Ref. [43], © Wiley-VCH GmbH 2025. (e) Mechanism for the electrical conductivity of LMPs-SG composite inks. Reproduced with permission from Ref. [155], © Wiley-VCH GmbH 2025.

mechanism. The formation of conductive pathways can also be attributed to the Ga–O–Si interfacial bonding arising from electrostatic interactions between LMPs and silica gel (SG), as well as the generation of small microcracks within the composite due to volumetric shrinkage of the SG host material during evaporation. Although exhibiting only one-third the conductivity of the aforementioned metalgel ($\sim 1.1 \times 10^6 \text{ S}\cdot\text{m}^{-1}$), this composite can be easily patterned into circuits via screen-printing techniques.

4.2 LMP composites with functional additives

4.2.1 Conductive bridges

Conductive and rigid fillers can act as bridges, connecting isolated LMPs and forming conductive pathways. This strategy enables the LMP composites to exhibit electrical conductivity without the need for additional post-processing sinter [42, 156–158]. As illustrated in Fig. 9(a), the most notable feature of this approach is the stable electrical resistance achieved with LMPs anchoring the conductive bridges during large deformations of the material. Specifically, the fracture of LMPs under stress releases LM, which reconnects displaced rigid fillers under deformations. The highly dynamic electrical interconnection provides a robust conductive network under mechanical strains [157, 159]. Here, we provide a brief overview of several common conductive bridge materials.

Ag is considered the ideal conductive filler with its high electrical conductivity, stable chemical properties and excellent wetting effect with LMP. Compared to Ag particles, Ag flakes with a high aspect ratio have been demonstrated to be the optimal conductive bridge material for LMP interconnects [159]. Conductive composites formed using Ag flakes with a high aspect ratio exhibited a resistance change of only 6.3% under 900% tensile strain, with an initial conductivity of $8331 \text{ S}\cdot\text{cm}^{-1}$ (Fig. 9(b)). Some studies have shown that LM and Ag can form a metallic compound AgIn_2 , which improves the adhesion of the Ag flakes to LMPs [42, 156]. Electrical properties of the conductive materials formed from the aforementioned material frameworks were further investigated when incorporating several different types of fillers: Ag, Ag-coated Ni or Fe particles, Ni, and Fe [156]. Compared to the other conductive fillers, the composite formed using Ag flakes demonstrated significant advantages in terms of conductivity, smearing tendencies and robustness. These properties stem from the unique organic bonding between the polymer and three distinct microstructures: core-shell Ga-In particles; newly formed Ag-In microparticles surrounding the LM droplets; and Ag flakes completely enveloping the particles (see Fig. 9(c)) [42]. The strategy of using Ag flakes as the electrical bridge is applicable to many polymer/gel system, including EVA [159], SIS [42, 156, 160], TPU [161], SIS-EVA [157], epoxy matrix [158], and PVA [162].

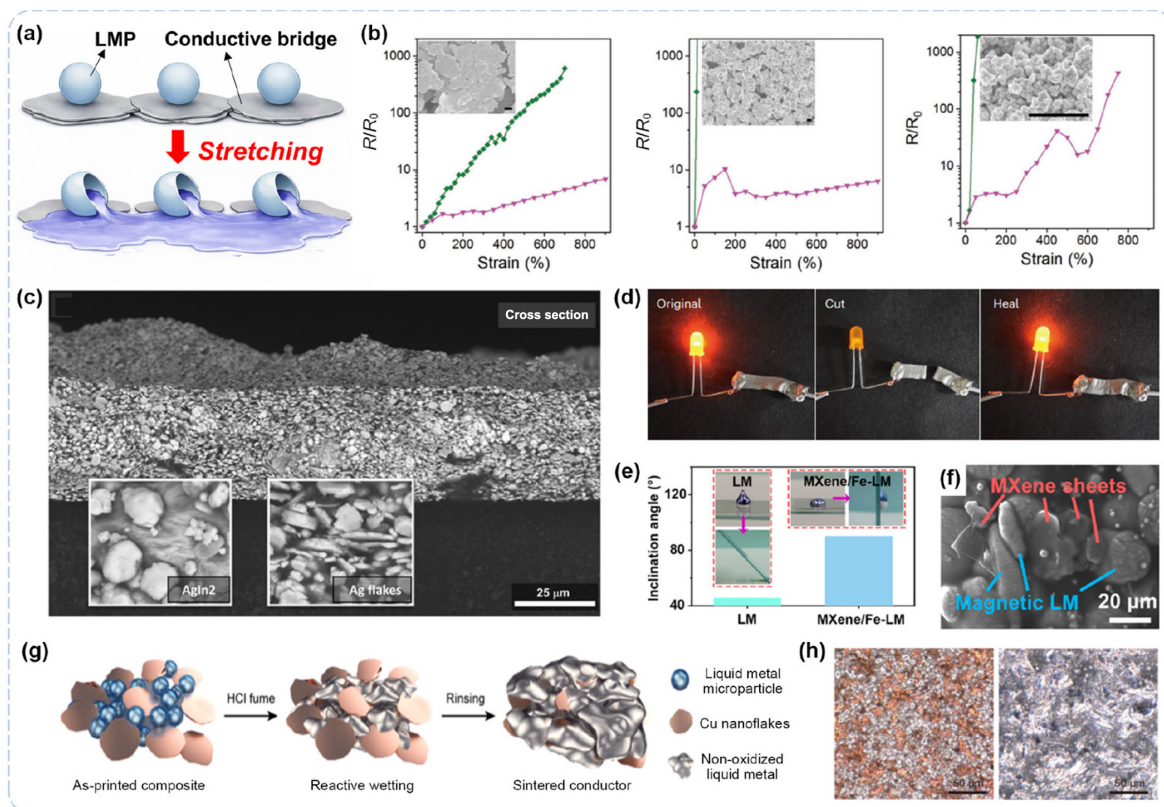


Figure 9 Conductive bridges for electrical interconnection of LMP composites. (a) Schematic of the elastic conductor with (top) and without (bottom) electrical bridges. (b) Resistance change of the elastic conductors with (pink) and without (green) EGaln LMPs under stretching strains by using different conductive fillers: Ag flakes (left), Ag microparticles (middle), and Ag nanoparticles (right), respectively. The inset figures are the SEM images of the corresponding conductive fillers, scale bar 1 μm . Reproduced with permission from Ref. [159], © WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim 2018. (c) Cross section of a sample. LMPs, Ag flakes, and newly formed AgIn₂ microparticles are visible. Reproduced with permission from Ref. [42], © American Chemical Society 2021. (d) Demonstration of composite's self-healing performance. Reproduced with permission from Ref. [162], © Zhao, Y. Y. et al. 2023. (e) Optical images of rolling angles for LM droplet and MXene-modified magnetic LM droplet on inclined plates covered with SBS films. (f) SEM image of the distribution of MXene sheets and magnetic LM droplets. Reproduced with permission from Ref. [163], © American Chemical Society 2022. (g) Schematic illustration of the mechanism for establishing conductive pathways with the assistance of HCl fume. (h) Optical microscopy images of the Cu-LMP-SIS composite before (left) and after (right) the HCl treatment. Reproduced with permission from Ref. [166], © American Chemical Society 2020.

Embedding the interconnecting LMPs and Ag flakes into a PVA-borax gel creates a conductive composite with self-healing capabilities. The self-healing property originates from the reversible, dynamic hydrogen bonding between the PVA and the borax [162]. While maintaining a high electrical conductivity of $7 \times 10^4 \text{ S}\cdot\text{m}^{-1}$, the composite exhibits rapid and effective self-healing (Fig. 9(d)), as well as favourable mechanical properties such as a low Young's modulus ($\sim 20 \text{ kPa}$) and high extensibility ($> 400\%$ strain).

It is worth noting that MXene, a novel and extensively studied two-dimensional material, can also serve as an ideal electrical bridge to connect LMPs. However, the poor wetting behavior between MXene sheets and LM with a high surface tension must be first addressed. As shown in Fig. 9(e), it has been demonstrated that mechanical stirring can embed Fe particles in liquid metal without alloying, significantly improving wettability between LM and MXene [163]. MXene sheets in poly(styrene-butadiene-styrene) (SBS) adhere tightly to the surface of LM droplets with Fe particles due to enhanced wettability and hydrogen bonding (Fig. 9(f)). MXene sheets have markedly inferior electrical conductivity to Ag flakes and therefore fail to impart high electrical properties to the composite materials. Nevertheless, the composite performs well in electromagnetic shielding applications [163].

The wettability between LM and Cu is significantly enhanced in

acidic or alkaline environments [25, 26, 164, 165], due to the removal of the passivating oxides. As shown in Fig. 9(g), Cu flakes and LMPs were evenly dispersed within the SIS matrix. The initial conductivity of the composite, which employed Cu flakes as the electrical bridge, was only $1.9 \times 10^{-7} \text{ S}\cdot\text{cm}^{-1}$ [166]. This was primarily due to that Cu flakes were unable to connect the LMPs effectively. Following the HCl fume treatment, the composite's conductivity increases to $1.1 \times 10^4 \text{ S}\cdot\text{cm}^{-1}$ and this change is permanent [166]. The color of the composite material also changes from the reddish-brown of the dispersed Cu flakes to the metallic-gray of the LM (Fig. 9(h)). This visually supports the reaction between LM and Cu, forming the intermediate metallic compounds CuGa_2 and CuGa . These compounds play a positive role in establishing a robust conductive percolating network within the composite because they exhibit higher electrical conductivity than pristine liquid metal [27, 166].

Additionally, some studies have achieved promising results by utilizing non-metallic fillers as electrical bridges [167–169]. However, compared to bridges using metallic fillers, the non-metallic bridges only confer limited conductivity to the composites. The electrical conductivity of these composite materials can be predicted using the percolation theory [156, 159]

$$\sigma = \sigma_0(V_f - V_c)^s \quad (5)$$

where σ_0 is the electrical conductivity of the filler, V_f is the volume fraction of the filler in the polymer matrix, V_c represents the volume fraction at the percolation threshold, and s denotes the fitting exponent. Pursuing high electrical conductivity requires a higher proportion of conductive fillers within the matrix. This inevitably compromises the tensile properties of the composite, demonstrating a classic trade-off effect.

4.2.2 Internalized magnetic particles

Conductivity can be achieved by manipulating conductive particles with magnetic fields [170–172]. However, LM lacks ferromagnetic properties and thus will not show magnetic response. Incorporating copper-coated iron particles into the LMPs through reactive wetting of gallium and copper can impart ferromagnetic characteristics to the materials. As shown in Fig. 10(a), applying an external magnetic field causes the ferromagnetic LMPs to aggregate and interconnect, forming conductive pathways within the material. This method actively induces the sedimentation and accumulation of magnetic LMPs through a strong external force, significantly reducing the required LM loading in polymers. For instance, Ecoflex filled with 200% mass ratio of non-magnetic LMPs is non-conductive, whereas Ecoflex filled with 80% mass ratio of magnetic LMPs becomes conductive after magnetic activation [170]. This has significant implications for the fabrication of lightweight stretchable conductors. Adjusting the mixing speed to 50, 200 and 400 rpm yielded average Feret diameters of 266.9 ± 172.1 , 144.3 ± 53.8 , and 130.7 ± 44.9 μm for magnetic LMPs filled with Ag-coated Ni particles, respectively. Following magnetic activation, materials with average Feret diameters of 144.3 ± 53.8 and 130.7 ± 44.9 μm

remained non-conductive, requiring additional mechanical stress sintering to achieve conductivity [172]. This demonstrates the importance of particle sizes for magnetic activation. Furthermore, the magnetic field activation of the LMPs must be completed before the polymer matrix cures. Post-curing results in a distinctive Janus structure comprising a light grey insulating upper layer and a dark grey conductive bottom layer containing a high concentration of magnetic liquid metal particles (Fig. 10(b)) [170]. Interestingly, magnetic field could be controlled to pattern the sintering of the magnetic LMPs, enabling a rapid and high-throughput production of conductive composites (see Fig. 10(c)). Regions depleted of LM were formed after the magnetic LMPs are attracted by the magnetic fields. These regions created insulating zones that effectively prevent unintended mechanical sintering upon curing [172].

4.2.3 Biomaterials

Research has explored achieving electrical connections in sintering-free LMPs by incorporating active biomaterials. Recently, Rezaie et al. proposed integrating dormant *Bacillus subtilis* endospores into LMP to construct electroactive LM materials [173]. The unique surface structure and abundant chemical groups of spores strongly interact with the gallium oxide layer on the LMP surface. These strong interactions induce localized rupture, enabling conductive connections between LMPs without other sintering process (Fig. 10(d)). When the spores germinate into cells with extracellular electron transfer (EET) activity under suitable conditions, the material's conductivity further increases, forming an “alive conductor” that can be activated on demand [173]. The conductivity of the LM materials embedded with spores is 1.1×10^4 $\text{S}\cdot\text{cm}^{-1}$. After the spores germinate into electrically active cells, the conductivity surges dramatically to 5.1×10^6 $\text{S}\cdot\text{cm}^{-1}$,

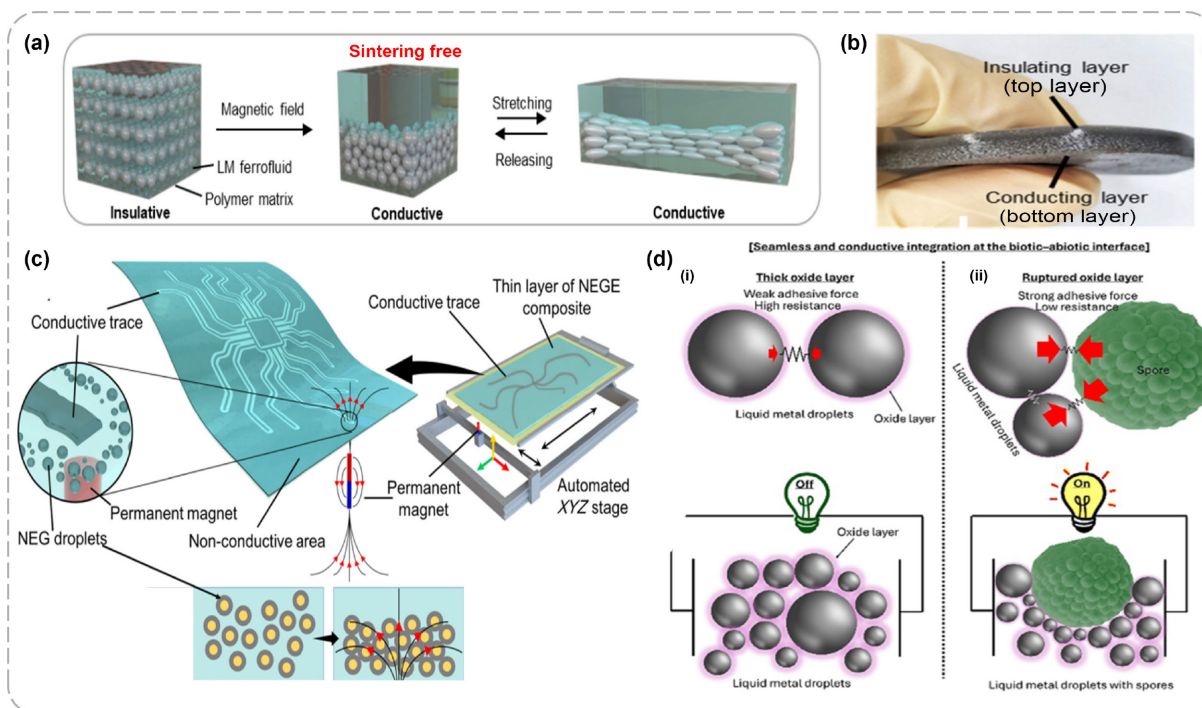


Figure 10 Internalized magnetic particles and bacterial endospores for electrical interconnection of LMP composites. (a) Schematic illustration of the magnetic aggregation to create highly stretchable and conductive magnetic-LMP composites. (b) The photograph of the conductive magnetic-LMP composite with a Janus structure. Reproduced with permission from Ref. [170], © Peng, M. Y. et al. 2023. (c) Mechanism of conductive trace patterning utilizing a miniature magnet. Reproduced with permission from Ref. [172], © Hoang, T. T. et al. 2022. (d) Schematic illustrations of LMPs without (i) and with (ii) spores. Reproduced with permission from Ref. [173], © Rezaie, M. et al. 2025.

approaching the conductivity of bulk metals. This material, acting as an electrogenic bacteria via EET, holds potential to serve as a communication conduit between biological systems and electronic devices, as well as a bio-driven electronic component. Although this strategy has not yet been applied to establishing electrical connectivity in LMP composites, it suggests a promising route toward *in vivo*-stable and electrically robust composites through the incorporation of biomaterials and LMPs.

5 Application

LMP composites combine the exceptional electrical and thermal conductivity of LMs with high tensile strength, easy processability, and good biocompatibility of polymers or gels, enabling a new generation of flexible and stretchable devices with unprecedented performances.

5.1 Stretchable conductors

Conductive networks formed by conventional rigid fillers, such as carbon nanotubes or silver nanowires, are susceptible to large resistance change when subjected to tensile strains, due to network breakage or increased filler spacing [111, 174]. These composites struggle to achieve electrical conductivity of over $104 \text{ S}\cdot\text{cm}^{-1}$ with a critical strain of more than 300% [174]. In contrast, LMP composites possess high electrical conductivity comparable to that of bulk LM. At the same time, their stretchability is largely depended on the polymer matrix. The LMP composites have minimal change in resistance ($< 5\%$) under high strain ($> 300\%$). The robust electrical performance could maintain even after thousands of tensile cycles [43, 101, 159]. Given that LMP composites possess high conductivity, excellent electrical stability, and good processability, we will focus on exploring their applications in highly integrated circuits, stretchable batteries, and electronic textiles.

5.1.1 Stretchable integrated circuits

Pure LM materials face challenges in achieving high patterning resolution, good shape fidelity, and complex spatial structures [175]. LMP composites, however, exhibit highly controllable rheological behaviour and structural stability, enabling the material to maintain stable architectures after curing. Advances in sintering technology have further propelled the development of LMPs in electrical circuit patterning. Materials printed onto flexible substrates form robust and stable conductive pathways after sintering, thereby driving the rapid advancement of stretchable integrated electronic devices. E-skin, as a stretchable and highly integrated electronic device mimicking human skin's functionalities, should be able to sense different input signals with the key characteristics of natural skin, including stretchability, self-healing properties, biocompatibility, and moisture permeability [48, 176, 177]. LMP composite, with its high conductivity, exceptional stretchability, and good biocompatibility, is one of the important materials for enabling E-skins.

LMP composites based on polymeric materials such as SIS, TPU, PVDF-HFP, and PDMS have demonstrated excellent printability [178]. To achieve stretchable and highly integrated electronic systems, it is essential to print stretchable circuits with high resolution. Lee et al. [178] introduced meniscus-guided printing (MGP) of LMP-polystyrene sulfonate (PSS) composites. The method enables patterning of LM electrodes with a resolution down

to $50 \mu\text{m}$ on various substrates such as VHB tape, PDMS, hydrogel and biogel. The self-sintering characteristics under evaporation simplifies the fabrication process and provides the capability of large-scale productions. As shown in Fig. 11(a), E-skins could be tailored for different prosthetic hand sizes, which realized multi-layer arrays for pressure and strain sensing. In addition to direct ink writing, stretchable circuits could also be fabricated via laser etching, stencil printing, or other patterning methods (Fig. 11(b)) [109]. Ma et al. [110] reported a method for fabricating continuous LM polymer microfibers using electrohydrodynamic (EHD) printing (Fig. 11(c)). The printed microfibers had a width ranging from 4 to $70 \mu\text{m}$. A minimum fiber width of $1.5 \mu\text{m}$ could also be printed. Although the low volume fraction of $3.07 \text{ vol.}\% \pm 0.01 \text{ vol.}\%$ LMPs resulted in a material conductivity of only $\sim 188.2 \pm 22.4 \text{ S}\cdot\text{cm}^{-1}$, this method offers a new insight for patterning highly stretchable sensors for integrated E-skin (see Fig. 11(d)). Zheng et al. [179] proposed a pressure-stamped method to fabricate stretchable integrated electronic devices. Unlike other methods, the approach produces nanofiber membranes from a TPU solution containing LMPs via electrospinning. Selective pressure is then applied using a rigid mold to sinter the LMP composites and form patterned conductive pathways. This process enables rapid fabrication of functional circuits with high elasticity and breathability.

With the ongoing advancement of LM circuits, stretchable electronic devices with multilayer and highly integrated circuits have made remarkable progress [180–182]. They achieve much higher integration density than single-layer architectures and enable new functionalities that are difficult to realize with traditional stretchable circuits. The 3D integrated electronics use soft vias to establish electrical connections between multiple circuit layers. Gu et al. [37] designed a printable paste with LMP composite based on evaporation sintering. The composite exhibited an initial electrical conductivity of $20,974 \text{ S}\cdot\text{cm}^{-1}$, high stability under mechanical strains ($R/R_0 \approx 21.12$ at 2200% strain), and excellent cyclic stability during tensile loading exceeding 40,000 cycles. The outstanding conductivity and stability of the composite ensures reliable electrical connections between the rigid electronic chips and enable dependable interlayer connectivity through soft electrical vias. A multilayer stretchable wireless patch for real-time electrocardiogram (ECG) monitoring was designed (Fig. 11(e)(i)). Despite incorporating 64 vias and 58 electronic chips at a density of $5 \text{ chips}\cdot\text{cm}^{-2}$, the patch maintains stable electrical signals under stretching, twisting, and bending (Fig. 11(e)(ii)). The patch retains stable ECG signal acquisition after mechanical deformation, comparable to commercial ECG acquisition systems (Fig. 11(e)(iii)) [37]. Nevertheless, vias would demand multiple processing steps and could be regarded as stress concentration points that lead to structural failure. Soft electronic vias with LM-stair structure achieved through rapid three-dimensional assembly of LMPs was reported to address this challenge [125]. Through selective curing, the stair-like structure within the photoresist serves as the foundation for the programmed settling and assembly of LMPs, forming a continuous trace of conductive composite from the top to the bottom layer of the circuits without through holes or bond lines (Fig. 11(f)(i)). After sintering, the LM-stair vias can electrically connect two distinct circuit planes. For example, a stretchable sensing circuit fabricated in Fig. 11(f)(ii) comprises two distinct layers: The top layer features nine white LEDs to indicate the magnitude of magnetic field, while the bottom layer incorporates

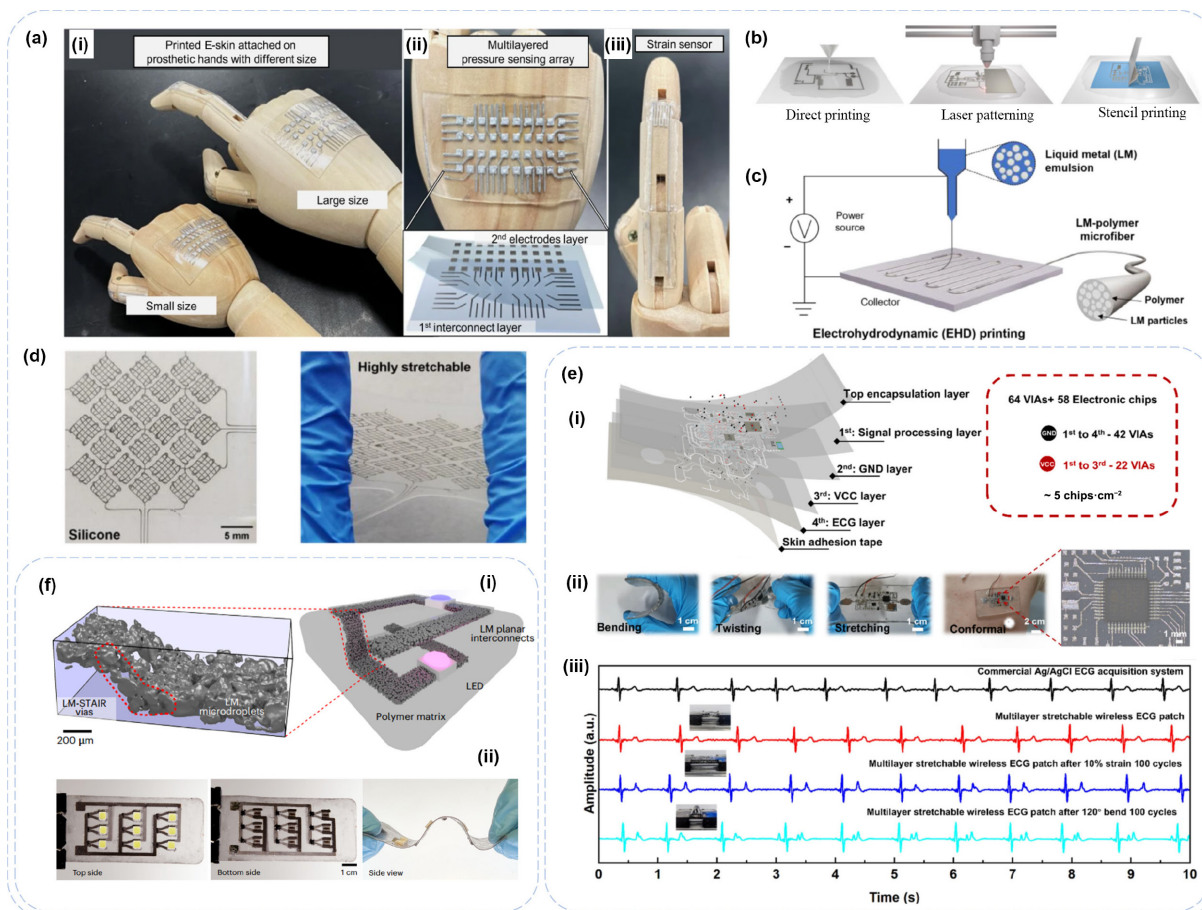


Figure 11 Application of stretchable integrated circuits. (a) Customized E-skin: (i) Photograph of customized E-skin attached to prosthetic hands with different sizes. (ii) Photograph and illustration of multilayer pressure sensing array. (iii) Photograph of strain sensor. Reproduced with permission from Ref. [48], © Lee, G. H. et al. 2022. (b) Soft circuit can be produced by ink printing, laser patterning, or stencil printing. Reproduced with permission from Ref. [109], © Lopes, P. A. et al. 2021. (c) Schematic illustration of EHD printing with LM-polymer emulsion. (d) Photographs of the assembled touch sensor before (left) and after stretching (right). Reproduced with permission from Ref. [110], © Wiley-VCH GmbH 2025. (e) A multilayer composition of multilayer stretchable wireless ECG patch: (i) structure of the wireless ECG patch; (ii) photographs of the patch under bending, twisting and stretching conditions and local magnification view; (iii) comparison of ECG signal data acquired by commercial Ag/AgCl ECG acquisition system, multilayer stretchable wireless ECG patch after mechanical deformation. Reproduced with permission from Ref. [37], © Elsevier Ltd. 2025. (f) LM-stair vias for E-skin: (i) reconstructed micro-CT scan of LM-stair vias (left) and a schematic of LM-stair vias seamlessly connecting top and bottom planar interconnects; (ii) photographs of the magnet sensing and indicating flexible circuit made through the LM-stair approach. Reproduced with permission from Ref. [125], © Ho, D. H. et al. 2024.

nine Hall effect sensors to detect the magnetic field. The sheet resistance of LM-stair via is below $0.3 \Omega\cdot\text{sq}^{-1}$, providing new enabling technology for the development of highly integrated and multifunctional stretchable electronic systems.

5.1.2 Stretchable batteries

Due to their superior flexibility and fluidity, Ga-based LMs promise various new battery configurations as a dendrite-free anode [183]. Stretchable batteries with stable structure and recyclability can benefit from the stable electrical connection and processability of LMP composites [38, 184]. By incorporating functional materials, the electrical and electrochemical properties of LMP composites can be tuned to enable stretchable batteries with superior performance [185, 186]. For example, Costa et al. [38] reported a 3D-printed stretchable liquid gallium battery, achieving a maximum capacity of $19.39 \text{ mAh}\cdot\text{cm}^{-2}$. In the design, highly conductive LMP-Ag-SIS composite was used as current collectors and electrical connections to facilitate efficient electron transport, while LMP-carbon black (CB)-SIS served as the anode (Fig. 12(a)(i)). It has been discovered

that LMP-CB-SIS composite exhibits a liquid-like behavior once in contact with the electrolyte. This leads to a self-feeding and self-aggregation properties of LMP, which facilitates more gallium exposure to the electrolyte and delays the formation of dead surfaces, thereby enhancing the total areal capacity [38, 187]. Multiple batteries integrated into the belt fabric provide continuous power for 22 h, enabling uninterrupted data collection and transmission of clinical-grade ECG data via Bluetooth at 100 Hz (Fig. 12(a)(ii)).

5.1.3 E-textiles

E-textiles integrate electronic functionality into wearable fabrics, providing a new platform for health management and physiological monitoring [20, 188]. LMP composites can be deposited onto fabrics via printing [49, 189] or woven into fabrics in their fibrous structures [161, 190]. Tang et al. [49] developed a highly stretchable composite ($380,000 \text{ S}\cdot\text{m}^{-1}$ at 1000% strain) using TPU as the host matrix, capable of being printed onto various substrates. Utilizing screen printing, they fabricated intricate integrated circuits on a T-



Figure 12 Application of printed batteries and E-textiles. (a) Printed battery: (i) schematic diagram of structure and working mechanism; (ii) biomonitoring wearable belt powered by multiple stretchable batteries for acquiring body temperature, respiration and heart monitoring. Reproduced with permission from Ref. [38], © Wiley-VCH GmbH 2022. (b) Integrated wearable T-shirt: (i) a schematic shows the highly integrated T-shirt; (ii) photograph of the printed T-shirt for smart wearables. Reproduced with permission from Ref. [49], © American Chemical Society 2019. (c) Universal printability of the LMPs-Ag-SISI-EVA composite to adapt to textiles with (i) different structures and (ii) various materials. Reproduced with permission from Ref. [157], © American Chemical Society 2024. (d) Wet spinning for fabricating continuous fiber. Reproduced with permission from Ref. [143], © Zheng, L. J. et al. 2021. (e) Novel passive E-textile for mechanical motion sensing. Reproduced with permission from Ref. [196], © Wiley-VCH GmbH 2025. (f) E-textile for outdoor access control operation in winter and door access of a kiln in operation under extreme temperature. Reproduced with permission from Ref. [161], © Zhang, Y. et al. 2025. (g) Photograph (left) and circuit schematic (right) of E-textile based on twisted fibers. Reproduced with permission from Ref. [190], © Lee, G. H. et al. 2025.

shirt, endowing it with multifunctional sensing and heating capabilities (Fig. 12(b)). To make the E-textile washable, an insulating layer was printed onto the circuits using commercial PU ink. Chemically stable fluoropolymer (FP) can prevent oxide layers from corroding in aqueous solutions, reducing the release of cytotoxic metal ions to enhance the biocompatibility of the composite. E-textiles fabricated with LMPs-FP composites have been demonstrated as washable, skin compatible electrodes suitable for thermotherapy and epidermal biopotential recording [189]. The composite using Ag flakes as electrical bridge not only eliminates the need for subsequent sintering but also exhibits remarkable electrical conductivity [42, 160]. At the same time, the incorporation of Ag flakes endows the composite with easily tunable rheological properties and excellent adhesion. An LMPs-Ag-SISI-EVA composite exhibits a maximum adhesion strength of up to ~ 0.6 MPa and low viscosity below 100 Pa·s [157]. It enables efficient printing on fabrics with diverse patterning structures (Fig. 12(c)).

Textiles typically exhibit intricate surface properties that pose challenges for high-precision and fine printing [191]. Meanwhile, weak adhesion between the materials and the fabric will induce detachment, leading to poor stability of the E-textiles. These issues can be effectively tackled by preparing the functional materials into fibers and weaving them into fabrics. LMP composite fibers can be prepared by dip-coating [192, 193], extrusion method [163], and wet spinning [161, 194–196]. The advantages of wet spinning (see Fig. 12(d)) for the large-scale production of continuous fibers are well recognized. The friction between composite fibers and fabrics with an outer insulating layer can form a single-electrode triboelectric nanogenerator, enabling self-powered motion sensing E-textiles (Fig. 12(e)) [143, 196]. Other studies indicate that E-textiles fabricated by LMP composite fibers hold broad applications in energy harvesting [196], electromagnetic shielding [163], heating [143, 194], and strain sensing [129, 194]. Additionally, Zhang et al. [161] observed that LMPs-Ag-TPU composite fibers enhance electrical conductivity and electromechanical stability across the

temperature range of -30 to 180 °C. Heating promotes the Ag-LM interaction following the thermal deformation and fracture of LMPs. By contrast, freezing contracts the TPU matrix, densifying the conductive filler. The release of LM triggered by mechanical activation further enhances conduction stability, achieving temperature-adaptive electromechanical stability and durability. The integration of composite fibers and chips in gloves allows for close-range wireless communication in extreme temperature environments (Fig. 12(f)), paving the way for E-textiles in low- and high-temperature work settings. Fibers usually require extra processes for encapsulation to guarantee the functional stability of E-textiles. However, Lee et al. used the sedimentation behavior of LMPs to produce self-assembled composite fibers [190]. LMP-TPU composites were printed onto a substrate. Once sedimentation and curing were completed, the fibers were obtained by peeling from the substrate. The mechanical robustness and electrical stability can be further enhanced through twisting of multiple fibers. The stable conductivity and mechanical robustness of twisted fibers, combined with their scalability in manufacturing, allow the integration of electronic components and circuits into commercial garments, resulting in multifunctional smart E-textiles with reliable performance (Fig. 12(g)).

5.2 Stretchable dielectric elastomers

The incorporation of fillers can significantly increase the dielectric constant of elastomers, with titanium dioxide and titanates being of particular interest [197]. However, this may increase the material's dielectric loss, and the stiffening effect of rigid fillers on the elastomer significantly reduces the material's flexibility. LMPs can significantly enhance the dielectric constant at a volume fraction far lower than that of rigid fillers, without significantly compromising the elastomer's inherent flexibility, elastic modulus, and dielectric breakdown strength [97]. This characteristic gives LMP composites important application potential in smart sensors, soft robotics, and wearable energy harvesting systems.

5.2.1 Capacitive strain sensors

LMP composites provide an effective solution for biomechanical sensing. The sensing principle for composite materials can be based on resistive and capacitive sensing. For resistive sensing, LMP composites typically exhibit small resistance changes under mechanical deformations [48, 49, 179], limiting their sensitivity. Conversely, insulating LMP composites demonstrate good performance with capacitive sensing owing to their superior dielectric properties [50, 198]. The capacitive sensors are typically fabricated with the composite material sandwiched between two electrode layers, responding to strains with altered capacitance.

Tutika et al. [91] investigated the response of sensors fabricated using LMP-Ecoflex composites (Fig. 13(a)) under uniaxial tensile strain. As illustrated in Fig. 13(b), the sensor response under strains was found to be linear. The capacitive strain sensors were able to detect motions from picking up balls of varying sizes (Fig. 13(c)). Unlike traditional slab-type capacitive sensors, Kang et al. [199] constructed a U-shaped strain sensor, which consists of two textile electrodes and a LMP composite (Fig. 13(d)). This effectively utilizes the external electric field shielding behavior of U-shaped capacitive sensors, addressing the trade-off between stability and integration in capacitive sensors. The U-shaped strain sensor demonstrated a strong response when integrated with a soft gripper (Fig. 13(e)).

By using LMP composites with a foam structure, the device

sensitivity can be largely improved. When the compressive force reaches a certain threshold, low-dielectric air pockets can be entirely eliminated, thereby achieving a significantly altered capacitance value and enhancing the composite's sensing sensitivity (Fig. 13(f)) [40, 41, 198]. Moreover, the porous structure with significantly reduced matrix modulus favors the suppression of unintended sintering of LMPs: i) It lowers the stress transmitted to LMPs during deformation, thereby increasing the critical strain value required to induce particle fracture and aggregation and ii) it increases the spacing between particles, thus reducing the likelihood of forming conductive pathways by the LM during deformation [200]. The porous foam structure can be achieved by dissolving sacrificing materials, such as sugar particles, embedded in the composite, offering advantages of straightforward and low-cost processing, adjustable pore size and porosity [40, 41]. Upon exceeding a certain pressure threshold, the LMP was compressed from a spherical to an elliptical shape, inducing negative piezopermittivity [40]. Under this effect, the increase in capacitance caused by the geometric change was mitigated by the reduce in dielectric properties, resulting in a constant capacitance signal. To enhance the detection of high stresses in composite foams, a dual-layer structural material has been reported [41]. This comprises a LMP composite foam layer with low elastic modulus of 7.8 kPa, and a pure elastomer layer with higher elastic modulus of 655 kPa. Its unique structure significantly enhances the pressure detection range of the foam composite (Fig. 13(g)). Recently, Han et al. [50] have reported a micropillar microstructure that enhances stress concentration effects, thereby maximizing the magnitude of change in the effective dielectric constant and electrode spacing under applied pressure (Fig. 13(h)). By regulating the laser scanning time during laser etching to control the height of the micropillar and appropriately adjusting the loading of LMPs, a high sensitivity of 2.07 kPa $^{-1}$ was achieved within the low-pressure range (< 135 Pa).

Capacitive strains sensors are required to overcome challenges of decoupling difficulties, poor linearity, and insufficient sensitivity for their wide applications. Liu [198] developed a capacitive three-axis strain sensor using LMP composite foam for human gait monitoring (Fig. 13(i)). The three-axis strain sensor employs a stacked configuration wherein capacitors with different sensing directions are arranged in tiers: One capacitor is sensitive only to normal strain perpendicular to the interface, while the other two are sensitive to both normal and shear strain (parallel to the interface). By employing a capacitor sensitive only to normal strain, the X - and Y -axis shear strain signals can be decoupled from the normal strain signals, enabling interference-free and three-axis strain measurement for human gait [198].

5.2.2 Soft actuators

Dielectric elastomer actuator (DEA) stands out as a promising candidate for artificial muscles due to its simple structure, high stretchability, rapid response speed, and large strain output [201]. The DEA works with the Maxwell stress p

$$p = \epsilon_r \epsilon_0 E^2 \quad (6)$$

where ϵ_r is the elastomer's dielectric constant, ϵ_0 is the permittivity of vacuum, and E is the applied electric field [201, 202]. Employing materials with higher dielectric constants can effectively increase the output performance and reduce the driving voltages [203]. LMP composites not only maintain the mechanical properties of the highly deformable host elastomer but also provide a high dielectric

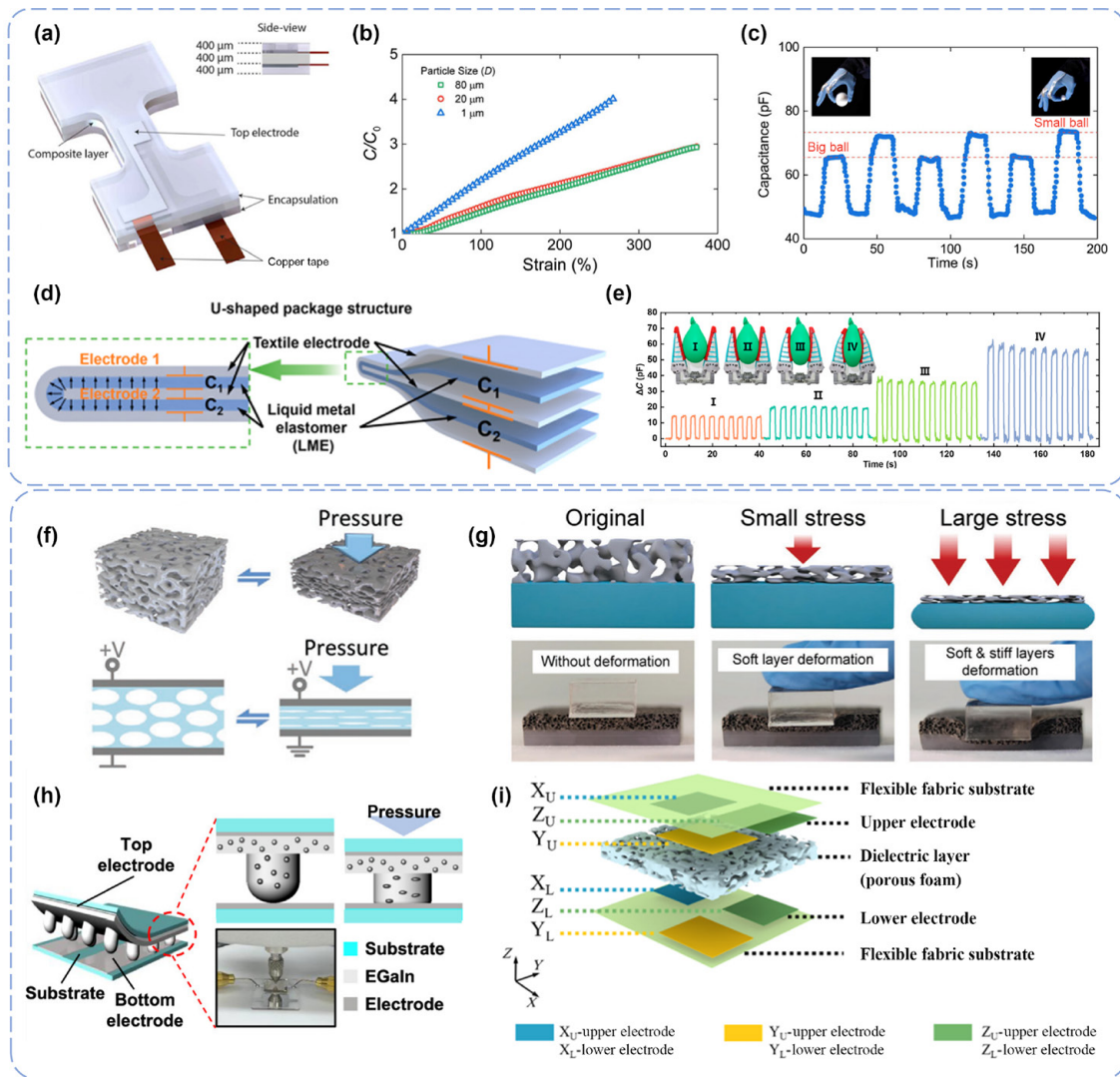


Figure 13 Application of capacitive strain sensors. (a) Schematic of the sensors used to evaluate the change in capacitance as a function of strain, side view shows thickness of individual layers. (b) Plot of the ratio (C/C_0) versus strain for composites with different LMP size. (c) Capacitance signal as a function of time for gripping differently sized balls. Reproduced with permission from Ref. [91], © American Chemical Society 2019. (d) Exploded view and design schematic of the U-shaped electrode strain sensor. (e) Cyclic capacitance variation curve of the strain sensor when grasping a deformable balloon. Reproduced with permission from Ref. [199], © American Chemical Society 2025. (f) Schematic diagram showing the principle of how permittivity increases with compression. Reproduced with permission from Ref. [40], © WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim 2020. (g) Schematic and pictures of different amounts of compressive strain of pressure sensor. Reproduced with permission from Ref. [41], © Wiley-VCH GmbH 2021. (h) Schematic of pressure sensing mechanism of LMP composite sensor. Reproduced with permission from Ref. [50], © American Chemical Society 2025. (i) Exploded view of the flexible three-axis strain sensor. Reproduced with permission from Ref. [198], © American Chemical Society 2025.

constant, making them attractive materials for the development of DEAs [91]. Pan et al. [39] demonstrated that incorporating nanoscale LMP fillers into dielectric elastomers enhance their dielectric constants and electromechanical properties. Figure 14(a) compares the performance of DEAs with and without LMPs, where the DEA with LMPs exhibits larger deformation and driving force. During multiple drive cycles, the measured blocking force of DEA with LMPs exceeded that of DEA without LMPs by 80% (Fig. 14(b)). Compared to conventional rigid fillers, the incorporation of LMPs into dielectric elastomers induces reduced impacts on their modulus, stretchability, and dielectric breakdown strength.

Incorporating LMP into an elastomer matrix can significantly enhance their thermal conductivity. This enables fast and uniform heating of thermally driven actuators during Joule heating or other

temperature activations. Wang et al. [204] introduced ethanol, a low-boiling-point phase-change initiator, into the LMP composite. Under heating conditions, the evaporation of liquid ethanol causes the composite to expand and freely deform as pressure increases. A soft robot using the composite was designed, which can be locomoted by wireless heating and cooling. LMP composites were also demonstrated to exhibit enhanced photothermal response with high photothermal conversion efficiency, which can efficiently convert light into thermal energy in the near-infrared region [205]. Tan et al. [206] incorporated LM nano-particles into carboxyl-functionalized polyurethane elastomers, endowing the dielectric elastomer with responsiveness to both electric fields and near-infrared light (Fig. 14(c)). Photothermal heating reduces the elastic modulus of the elastomer, enabling higher actuation strains under electric fields and achieving on-demand modulation without

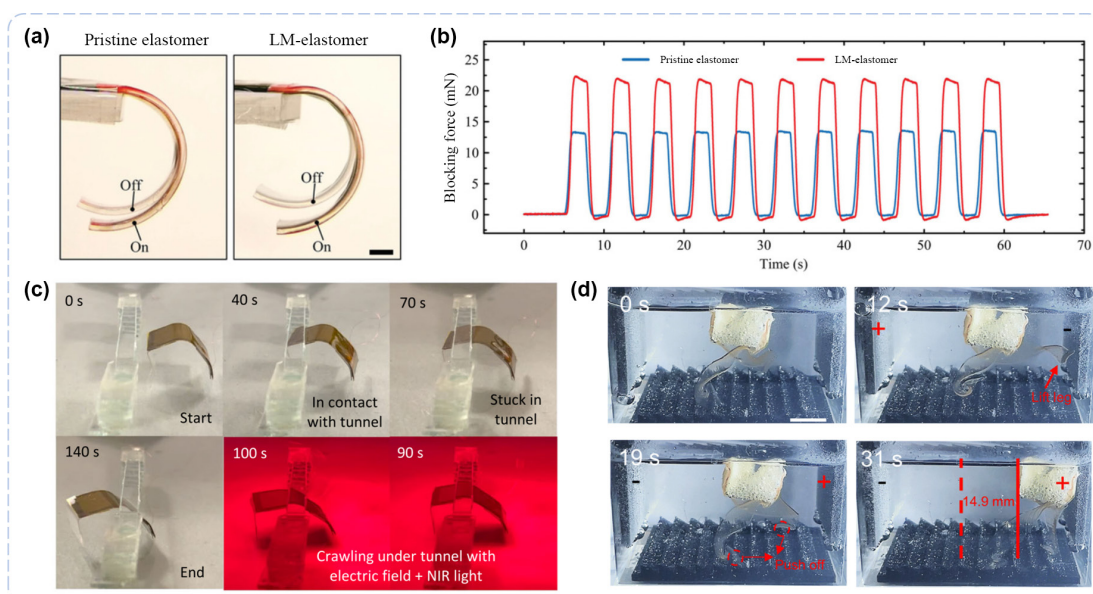


Figure 14 Application of soft actuators. (a) A driving cycle image of DEA using unfilled elastomer (left) and LM-elastomer nanocomposite (right). Scale bar is 5 mm. (b) Representative blocking force histories over 11 field on and off cycles. Reproduced with permission from Ref. [39], © WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim 2019. (c) Digital image of the crawler based on DEA with and without NIR stimulation. Reproduced with permission from Ref. [206], © Tan, M. W. M. et al. 2022. (d) Photos of an underwater soft walking robot based on two LMP-hydrogel composite legs. Reproduced with permission from Ref. [208], © The Royal Society of Chemistry 2026.

complex electrode patterns or increased driving voltages. Interestingly, shape morphing can be programmed into the LMP composite using liquid crystal elastomer as the polymeric matrix, enabling the material to undergo reversible transition between pre-programmed morphologies through electrical or thermal stimulation [207].

Electro-responsive hydrogel actuators generate deformation through ionic migration and electric field-induced osmotic pressure imbalance, exhibiting exceptional controllability and repeatability. Zhang et al. discovered that doping a small amount of LMP enhances the dielectric response of hydrogels, yielding greater deformation [208]. On one hand, the higher dielectric constant reduces ion-ion Coulomb interactions, promoting ion dissociation and migration. On the other hand, interfacial polarization around LMP microdomains promotes localized field concentration. Figure 14(d) shows an underwater soft robot using two LM-hydrogel legs with differing LMP contents. Asymmetric deformation of the legs enables directional propulsion driven by friction interactions with the substrate when the electric field polarity is alternated. This soft robot achieved 14.9 mm of forward movement within 31 s.

5.2.3 Energy harvesters

LMP composites with large mechanical compliance and high dielectric constant have emerged as promising materials to address the growing demand for self-powered electronics and distributed sensor networks. They can be used to harvest ambient electromagnetic energy and low-frequency mechanical energy, converting them into accessible electrical outputs.

The harvesting of ambient electromagnetic energy relies on the dielectric response of materials under external electromagnetic fields. Kim et al. [209] have reported in detail on the energy harvesting mechanism, known as the energy-loss return gate (ELRG), using water as the dielectric material. Similarly, the electromagnetic energy harvester can be fabricated using LMP

dielectric elastomer coated over conductive materials. As shown in Fig. 15(a), when the energy harvester is exposed to environmental electromagnetic fields such as those generated by electrical power lines or alternating current (AC) electric systems, the high dielectric constant of the LMP dielectric elastomer facilitates significant polarization, driving displacement currents through external loads. Bhuyan et al. [51] uniformly dispersed LMP at a loading of 50% volume fraction into a silicone dielectric gel (Sylgard 527, Dow) to produce LMP dielectric elastomer with a dielectric constant of up to 15. After molding the composite into a circular shape, LM electrode was coated onto one end to form an ELRG-based electromagnetic energy harvester. As shown in Fig. 15(b), the dissipated energy recovered from a laptop by three energy harvesters connected in parallel is sufficient to power low-power electronic gadgets.

The inherent stretchability of LMP composites allows them to conform to moving surfaces and withstand large cyclic strains. The dielectric elastomer generator (DEG), which shares consistent device structure with the DEA (Fig. 15(c)), serves as an effective device for converting low-frequency mechanical work into electrical energy. The operation principle of DEG lies in that equal and opposite charges on the surfaces of the dielectric elastomer undergo significant changes in their electrostatic potential during mechanical deformations [210]. The DEG with LMP composite resulted a 53% increase in electrical energy output compared with a DEG with pure elastomer [39]. Triboelectric nanogenerator (TENG) represents another major research focus in low-frequency energy harvesting, providing advantages in simple device architecture and versatile designs. Under low-frequency mechanical stimulation, the deformation of the dielectric elastomer alters the spacing and spatial distribution of the LMPs. This morphological change significantly induces a substantial variation in the composite's intrinsic capacitance, thereby driving charges flow toward external loads via electrostatic induction and triboelectric coupling. For instance, the LMP composite was sandwiched between two Ecoflex layers (Fig. 15(d)). When incorporating 50 wt.% LMP in elastomer, the

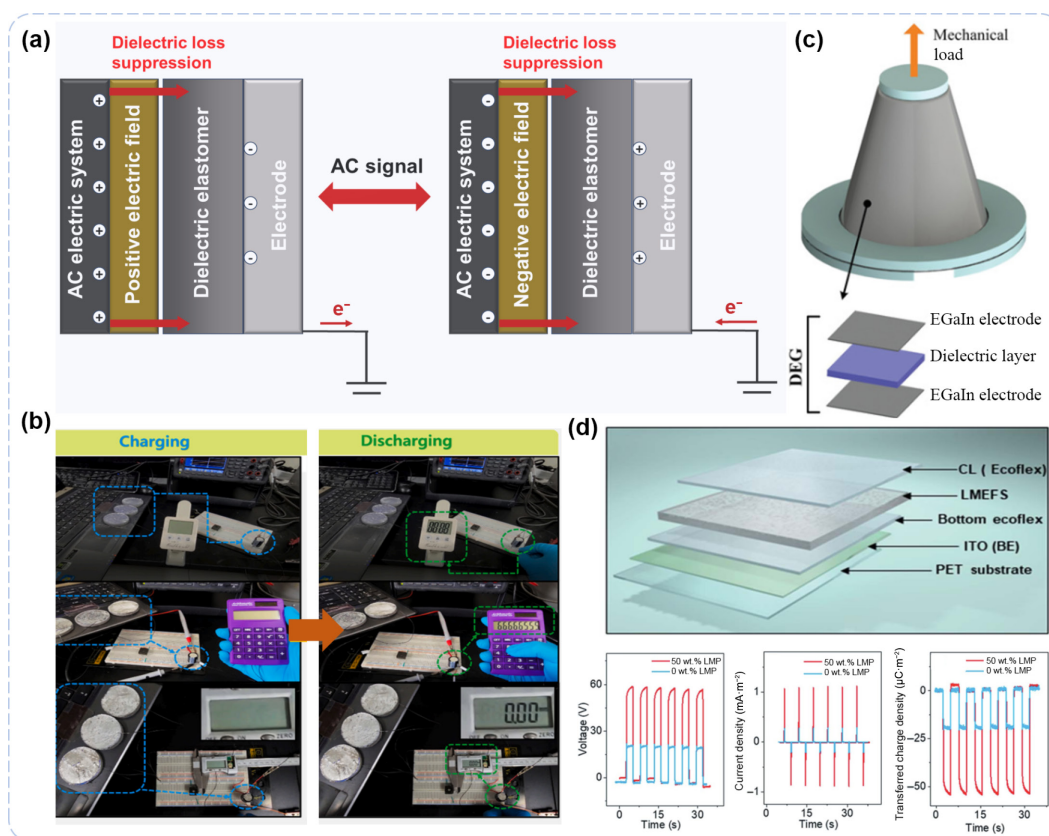


Figure 15 Application of energy harvesters. (a) Schematic diagram of the LMP composite utilizing ELRG to harvest electromagnetic energy. (b) Photos of three electromagnetic energy harvesters for powering various electronic gadgets such as a table clock, calculator, and vernier caliper. Reproduced with permission from Ref. [51], © Elsevier Ltd. 2023. (c) A schematic diagram of a DEG consisting of an LMP complex sandwiched between two layers of LM electrodes. Reproduced with permission from Ref. [39], © WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim 2019. (d) LMP composite based-TENG: 3D schematic illustration of the structure of the TENG (top) and the comparison between TENG with 0 and 50 wt.% LMPs's open-circuit voltage, short-circuit current density and short-circuit transferred charge density (bottom). Reproduced with permission from Ref. [211], © The Royal Society of Chemistry 2019.

output performance of LMP-TENG enhanced by 250% [211]. Nayak et al. [212] demonstrated that LMP composite with foam structure could enhance the capacitive charge coupling between pores and electrodes, which is beneficial for further improving the electrical outputs.

5.3 Stretchable thermal management

The application of Ga-based LM in thermal management can be categorized into heat transfer fluid (HTF), thermal interface material (TIM), and phase change material (PCM) [213]. HTF relies on the high thermal conductivity of LM fluid to transfer heat from high-temperature sources to low-temperature heat sinks within thermal management systems. Here, we focus on TIM and PCM, which represent important application areas of LMP composites.

5.3.1 TIM

TIMs are commonly used in the heat dissipation process of high-power electronic devices. As shown in Fig. 16(a), when electronic devices are integrated with heat sinks, the presence of an air gap (the thermal conductivity of air is only $0.026 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$) limits the effective contact area, thereby increasing the interfacial thermal resistance and hindering heat transfer. LMP composites, which combine high thermal conductivity with excellent mechanical compliance, can largely improve the contact interfaces. Furthermore, the LM released from the fractured LMPs at the

interface between the TIM and the device can penetrate microscopic defects on the device surface, forming an expanded contact area that displaces trapped air and maximizes the effective contact area [214].

LMP composites form a continuous LM permeation network within the matrix through sintering, achieving high electrical conductivity and outstanding thermal conductivity. The thermal conduction mechanism, dominated by free electrons within the continuous phase of LM, endows the composite with excellent thermal conducting performance. However, electrical conductivity may pose safety hazards of short-circuiting, requiring electrical insulations of the LM composites for some thermal applications. Due to the dynamics of phonon transport, electrically insulating soft materials with high thermal conductivity typically have large elastic modulus [113]. The introduction of liquid-phase LMPs with high thermal conductivity enables the creation of heat transfer pathways within elastic matrices while preserving the conformability of the elastomer, thereby facilitating efficient thermal exchange in stretchable electronics [97, 112, 113]. Bartlett et al. [113] also discovered that strain generates thermal conduction pathways through *in-situ* elongation of deformable LMPs, significantly enhancing thermal conductivity in the tensile direction. For composites filled with 50 vol.% LMPs, the thermal conductivity in the tensile direction reaches $9.8 \pm 0.8 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ at 400% tensile strain. This value approaches the thermal conductivity of certain metals such as bismuth and stainless steel. Chin et al. prepared LMP

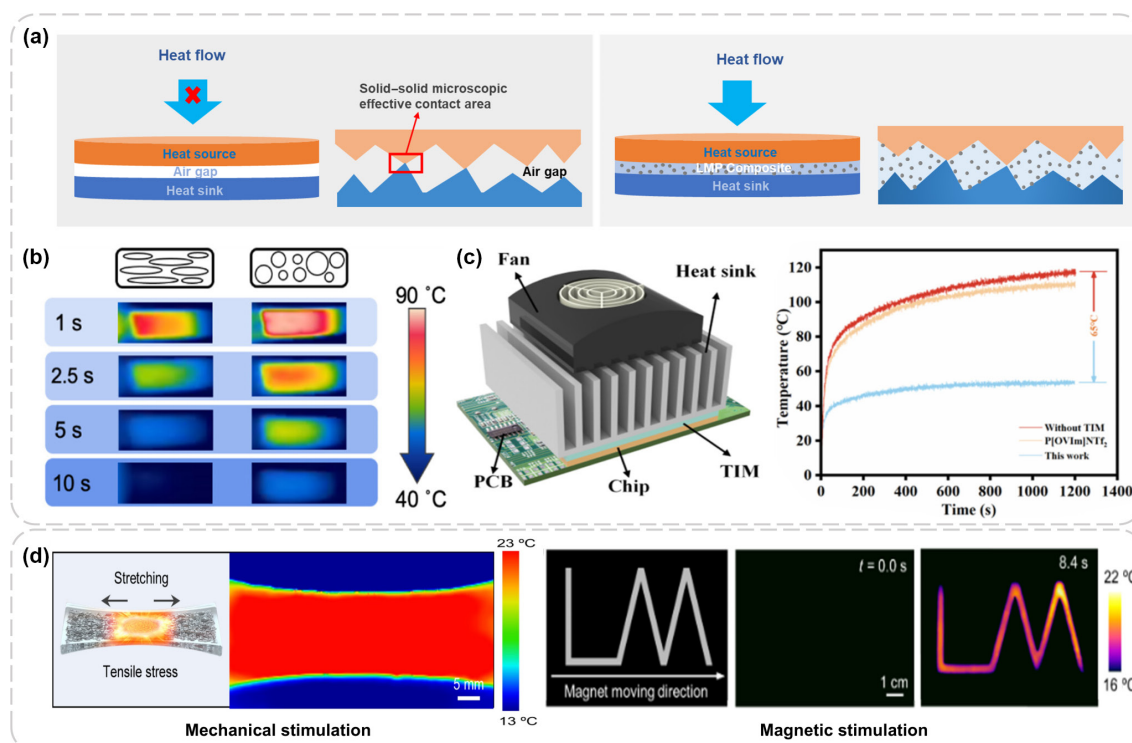


Figure 16 Thermal management materials. (a) Schematic diagram of heat dissipation without (left) and with (right) LMP composites used as TIM. (b) Thermograms comparing LMP composites with elongated LMP inclusions and spherical LMP inclusions at selected time points. Reproduced with permission from Ref. [52], © American Chemical Society 2025. (c) Heat dissipation test schematic (left) and temperature evolution comparison (right). Reproduced with permission from Ref. [53], © American Chemical Society 2025. (d) Phase change material: schematic image and infrared image showing the solidification-induced temperature increases of the LMEA triggered by mechanical stretching (left). Infrared image of predefined patterns based on magnetic field-induction (right). Reproduced with permission from Ref. [54], © Wang, H. Z. et al. 2024.

composites containing both elongated LMP inclusions with oriented alignment and spherical LMP inclusions. As shown in Fig. 16(b), the LMP composite with elongated LM inclusions exhibits superior heat dissipation capability [52].

Tutika et al. [215] prepared composites with liquid-phase, solid-phase, and liquid–solid multiphase fillers by adding LMPs, metal particles, and a combination of LM and metal particles to the polymers, respectively, to investigate their thermal and mechanical properties. Compared to composites containing only LMP, filling the composite with equal volume of LMP and spherical metal particles including Fe, Ag, Cu and Ni enhanced their thermal conductivity, though this method inevitably came at the expense of mechanical properties [215]. Due to the high electrical conductivity of metals, deformations may lead to electrical conduction in the composite [216]. Incorporating carbon materials with high thermal conductivity and lower electrical conductivity—such as graphite [217], graphene [35, 218–220], and diamond [36, 53]—into elastomers to form liquid–solid multiphase systems has been reported as an effective approach. For example, coating graphene nanoplates onto the surface of LMPs has been demonstrated as an effective method to enhance the thermal conductivity of the composite materials, achieving a thermal conductivity of $5.67 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ [218]. Without increasing the barrier density, the affinity between LMP and graphene creates more pathways for phonon transport, resulting in a corresponding increase in thermal conductivity [35]. LMP fills the micro-gaps between diamond particles via capillary action, effectively reducing interfacial thermal resistance [36, 53]. After filling poly (ionic liquid) (PIL) poly (1-octyl-3-vinylimidazole) bis (trifluoromethanesulfonyl) imide

(P[OVIm]NTf₂), the composite materials achieved a thermal conductivity of $14.2 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ and a tensile elongation of 74% [53]. After 1000 s of continuous chip operation, the composite reduced the temperature by 65 °C, significantly outperforming systems without thermal interface material and those using only pure polymeric materials (Fig. 16(c)). Table 4 provides a detailed comparison of the performance of LMP composite materials in TIM across multiple representative studies.

5.3.2 PCM

PCM based on Ga-based LM are attractive, which can be designed to achieve heating or cooling effects [221]. This is because Ga-based LMs possess a high volumetric latent heat during phase change process. For instance, liquid Ga at room temperature exhibits exceptionally high volumetric latent heat of $473 \text{ kJ}\cdot\text{L}^{-1}$, surpassing water's volumetric latent heat of $336 \text{ kJ}\cdot\text{L}^{-1}$ [54]. Each LMP in the composite materials acts as a microscopic latent heat reservoir, storing the latent heat of solidification in a deeply supercooled metastable state. For example, Tang et al. [54] incorporated magnetic LMPs into an elastomer to achieve controlled heat release driven by the phase change of LMP. The supercooling behavior of LMP suppresses spontaneous nucleation under ambient conditions, ensuring thermodynamic stability of LMP composites. When subjected to mechanical or magnetic stimulation, stress-induced rupture of the oxide layer and solid–liquid interface disturbances instantaneously introduce heterogeneous nucleation sites, driving the supercooled LMP to complete the liquid–solid phase transition within milliseconds and explosively release latent heat [59]. The low thermal conductivity of the elastomer matrix ($< 0.2 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$)

Table 4 Soft TIM based on LMP composites

Materials	Mean size	Filler loading	Thermal conductivity	Stretchability	Ref.
Ecoflex 00-30/EGaIn	$d_{\text{LMP}} = 15 \mu\text{m}$	$\phi_{\text{LMP}} = 50\%$	$4.7 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$	$> 600\%$	[113]
PDMS/Galinstan	$d_{\text{LMP}} = 2\text{--}3 \mu\text{m}$	$\omega_{\text{LMP}} = 92.5\%$	$> 2 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$	$\sim 50\%$	[112]
Ex-Sil 100/EGaIn	$d_{\text{LMP}} = 10\text{--}15 \mu\text{m}$	$\phi_{\text{LMP}} = 80\%$	$6.7 \pm 0.1 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$	$\sim 30\%$	[215]
PDMS/EGaIn	Aspect ratio > 5 Long axis length $\approx 140 \mu\text{m}$	$\phi_{\text{LMP}} = 40\%$	$1.51 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$	NM	[52]
Aramid nanofiber/EGaInSn/graphene nanosheet	$d_{\text{LMP}} = 0.4 \mu\text{m}$	$\omega_{\text{LMP}} = 70\%$	$5.67 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$	$< 4\%$	[218]
PDMS/EGaIn/graphene nano-platelet (GnP)	$d_{\text{LMP}} < 10 \mu\text{m}$	$\omega_{\text{LMP}} = 50\%$ $\omega_{\text{GnP}} = 6.3\%$	$0.9 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$	$> 100\%$	[35]
P[OVIm]NTf2/EGaIn/Diamond	$d_{\text{Diamond}} = 40 \mu\text{m}$ $d_{\text{LMP}} < 10 \mu\text{m}$	$\phi_{\text{LMP}} = 42.5\%$ $\phi_{\text{Diamond}} = 42.5\%$	$14.2 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$	74%	[53]
Epoxy /EGaIn/Diamond	$d_{\text{LMP}} = 6.4 \mu\text{m}$ $d_{\text{Diamond}} = 150 \mu\text{m}$	$\omega_{\text{LMP}} \approx 40.7\%$ $\omega_{\text{Diamond}} \approx 40.7\%$	$2.53 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$	NM	[36]

restricts heat diffusion, causing heat to be highly localized to the region where the LMP phase transition occurs and resulting in a detectable transient temperature rise (Fig. 16(d)). This programmable and sensitive heat release mechanism makes them very interesting for achieving multifunctional integration in intelligent soft systems.

6 Summary and outlook

This review summarizes recent advancements in LMP composites. By combining highly stretchable polymeric matrices with LM, the composite materials can be engineered to exhibit outstanding electrical conductivity, dielectric properties, or thermal conductivity, positioning them as key materials for the advancement of stretchable electronics. We systematically introduce methods to prevent LMP precipitation and induce electrical interconnection between LMPs, outlining the most recent research advances in stretchable conductors, dielectric elastomers, and thermal management materials based on LMP composites. Although remarkable progress has been made in the explorations and applications of LMP composites, further developments with in-depth investigation are still required in several areas to fully leverage their potential applications. Some of the important challenges that need to be addressed are discussed below.

1) Further development of LMPs should focus on efficient particle size tailoring, scalable manufacturing, eco-friendly processes, and their controllable functionalization. These technologies will help to support the preparation, patterning, and electrical activation of LMP composite to enable the fabrication of devices with sophisticated architecture.

2) The sedimentation behavior of LMP in composite materials is a key factor that adversely affects the stability of their dielectric and thermal properties. The current challenge lies in the fact that the high density and high surface tension of Ga-based LMs make them prone to sedimentation or agglomeration in low-viscosity prepolymers, while the dynamic evolution of the oxide layer further exacerbates the time-dependent nature of the dispersion state. Future research should focus on developing efficient surface modification strategies and dispersion additive systems to effectively extend storage life and processing windows while maintaining particle integrity. Concurrently, efforts should be made to further optimize the comprehensive performance and long-term service stability of LMP composites while maintaining the material's isotropy.

3) Investigation in the electrical interconnection strategies for LMPs will continue to resolve the controllability and large-scale fabrication challenges. Leveraging *in-situ* characterization techniques and multiscale theoretical simulations to deepen understanding of the kinetic mechanisms underlying the rupture of the LMP oxide layer, the exudation and fusion of LM, and the formation of conductive networks within the polymeric matrix will help mitigate the trade-off between stretchability and conductivity.

4) The LMP composite preparation and sintering remain confined to the laboratory scale. Practical pathways for scaling up for industrial production are still lacking. The large-area uniformity, batch-to-batch consistency, and process reproducibility are required to be addressed. Furthermore, long-term reliability of the device's performance remains unclear when it is subjected to complex environmental conditions (such as cyclic strains and temperature/humidity fluctuations). Future research should focus on material design compatible with scalable manufacturing processes and the establishment of standardized reliability testing methods.

5) Structural innovations and material composites for LMP dielectric elastomers will be key to enhance their electromechanical performance, advancing their applications in sensing, actuation, and energy harvesting. The introduction of high-dielectric ceramics or two-dimensional fillers can further optimize the synergistic enhancement of the dielectric constant and elastic modulus.

6) Although significant progress has been made in TIM, further research is needed in areas such as interfacial wetting behavior, phonon spectrum matching, and interlayer design. At the same time, the supercooling behavior and tunable phase transition of LMPs present a unique opportunity for developing thermal management materials that combine high thermal conductivity with thermal storage capabilities [54]. Future research should focus on temperature control strategies based on alloy composition regulation, meeting the requirements of thermal buffering and heat conduction in soft electronic devices under ambient conditions.

7) Future researches on LMP composites are expected to establish environmentally friendly technologies to reduce the potential threat that soft and wearable electronic devices, which are showing vigorous development and fast-paced production in the model digital era, pose to the ecological environment. It is also essential to explore strategies for the efficient recovery and reuse of gallium-based liquid metals from discarded devices to achieve a closed-loop recycling system for metal resources, whose acquisition are energy and chemical intensive.

Data availability

Not applicable.

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Declaration of competing interest

All the contributing authors report no conflict of interests in this work.

Author contribution statement

Y. Y.: Validation, writing manuscript. J.-W. L.: Writing manuscript. Y. H.: Validation. X.-Q. L.: Validation. H.-B. S.: Validation. Y.-Z. L.: Manuscript review. J.-X. W.: Project administration, manuscript review, funding acquisition. All the authors have approved the final manuscript.

Use of AI statement

None.

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