



Liquid Metal Fibers

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Abstract

Liquid metal (LM) is a type of metal or alloy that has a low melting point near room temperature and exhibits the properties of both liquids and metals. Such unconventional materials have been gaining increasing attention within the scientific and industrial communities. Recently, fiber-shaped LM and its composites have especially attracted diverse interest owing to their unique merits, such as excellent conductivity, intrinsic stretchability, facile phase transition, and the ability to be woven or knitted into smart fabrics. This review is dedicated to summarizing different aspects of LM-based fibers, such as their material components, fabrication and design strategies, and remarkable applications by way of their representative properties. Typical fabrication approaches, such as 3D printing of pure LM wire, coating the LM shell on the surface of the fiber, injecting a LM core into hollow fibers, and spinning of LM and polymer hybrids have been comparatively illustrated. Moreover, emerging applications that primarily utilize LM fibers have been demonstrated. Finally, future directions and opportunities in the field are discussed. This categorization of LM fibers is expected to facilitate further investigation and practice in the coming society.

Keywords Liquid metal · Stretchable and conductive fibers · Smart fabrics · Wearable electronics · Health monitoring

Introduction

Fibers have played an indispensable role in the development of human civilization and are utilized almost everywhere in our daily life [1–4]. Typical fibers, such as cotton, silk, hemp, hair, and nylon, which have unique properties, are highly relevant in society. Broadly, one-dimensional materials with diameters ranging from tens to hundreds of micrometers can be defined as fiber materials [1]. The use of conventional fibers alone cannot meet the increasing demands for fiber materials in modern society. Smart fibers with promising and irreplaceable properties, including conductivity, high stretchability, color tunability, self-cleaning, self-healing and shape memory have therefore attracted

increasing attention recently [5–10]. The development of smart fibers primarily depends on the discovery and exploitation of intelligent materials.

As newly emerging important functional materials with both liquid and metallic properties near room temperature, liquid metal (LM) and alloys are promising candidates for fabricating conductive and intrinsically stretchable fibers as they perfectly address the dilemma of conductivity and transformability trade-off [11–14]. Fiber-shaped liquid metals and their composites possess unique properties, such as excellent electrical conductivity (approximately 10^6 S/m), intrinsic stretchability, self-healing, chemical activity and knittability, thus presenting great potential for different areas [15–21]. Among various liquid metal materials, mercury is well-known LM; it has a low melting point (m.p. = -38.9 °C) and is widely applied in sphygmomanometers and thermometers. However, its considerably high toxicity prevents its widespread utilization. In contrast, gallium (Ga) and its alloys, such as eutectic gallium indium (EGaIn) and gallium indium tin (Galinstan), which are liquids at room temperature, are nontoxic and biocompatible, and they have been recognized as promising materials for diverse applications [22–25]. Through regulation of metal

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type and proportion, LMs with different melting points can be obtained [26]. It is well known that, owing to high surface tension (e.g., Ga: 700 mN/m), the LM forms a spherical structure to reduce the surface energy [27, 28].

Several key technologies have been reported for fabricating fiber-shaped (one-dimensional) liquid metal, which is generally termed a liquid metal fiber (LMF), as shown in Fig. 1. First, a liquid metal ink was 3D-printed into desirable LM wires [29, 30]. The commercial desktop LM printer was then manufactured in 2014, making the printing of LM wires rather easy and precise [31]. In addition, LM ink can easily adhere to the surface of conventional fibers with a specially designed coating layer to form fibers with LM coatings [32–34], which is suitable for various fibers, such as cotton, human hair, and polyurethane fibers. The development of coating methods relies on advancements in the interface science of LMs [35]. In addition to coating the fibers, LM can be injected into the hollows of fiber tubes or thermally fused fiber to form fibers with LM cores [15, 36]. Combining microfluidic [37] and vacuum suction [38] technologies can make the injection method more precise and efficient. Additionally, liquid metal can be dispersed evenly into an uncured polymer through mechanical mixing or ultrasonic dispersion to form smart materials with properties such as high stretchability, adjustable electrical and thermal conductivity in response to different stimuli, and self-healing [39–43]. More importantly, uncured polymers with liquid metal microdroplets inside can be fabricated into LM-embedded fibers by coaxial wet spinning or printing, which is quite efficient for producing a mass of fibers [44, 45]. More details regarding fabrication technologies will be further discussed in “Fabrication Technologies for Liquid Metal Fibers”.

The fabricated LMF can be woven into smart fabrics or functional wearable textiles. Based on the unique characteristics of LM, LMFs show great application prospects in inherently stretchable circuits for wearable devices, soft sensors, soft switches, variable stiffness electrodes, and shape



Fig. 1 Schematic illustration of a liquid metal fiber and its fabrication technologies

memory fibers [10, 12, 17, 46–50]. Significant efforts are being made to explore more characteristics of liquid metal fibers apart from conductivity and stretchability to enrich their versatility, thus providing plenty of ground to study and utilize LMFs.

To date, there has been no systematic review on the growth and influence of LM fibers, impeding the rapid development and delaying the applicability of their benefits to society. This article aims to provide a timely, systematic summary of LM fibers, elaborating on aspects ranging from their material components and fabrication technology to emerging applications and future directions. It is expected to stimulate the development of LMF and revoke attention, and promote their utilization in daily life.

Unique Features of Liquid Metals

According to the definition in numerous studies, room-temperature LM is a type of metal or alloy that exists as a liquid near room-temperature [51–53]. Mercury is a well-known LM that has been used for many years. However, toxic mercury vapor is easily generated owing to its high vapor pressure and low boiling point, posing a threat to living organisms. In addition, rubidium (Rb, m.p. = 38.9 °C, cesium (Cs, m.p. = 28.5 °C), and francium (Fr, m.p. = 27 °C) also have melting points at approximately room temperature, but their instability makes them liable to explode on reaction with water [26, 54]. In contrast, gallium is a safe metal with a low melting point (m.p. = 29.8 °C). It has been reported that the interatomic distance between adjacent Ga atoms is larger than that in most metals, which causes weak metallic binding of Ga atoms [26]. Hence, the crystal structure (solid) of gallium is vulnerable to breakage by heat, causing it to melt into a liquid. Ga has an extremely high boiling point of 2205 °C due to the reactive and unpaired valence electrons in the p shell. The low melting point and high boiling point of Ga make it quite stable and relatively safe for practical utilization. Ga can also be alloyed with other metals to form a LM with a lower melting point. For example, eutectic gallium indium (EGaIn) and gallium indium tin (Galinstan) are two typical LM materials. In addition, a Field alloy containing Bi, In, and Sn, with a melting point of approximately 62 °C, is also defined as an LM. Figure 2 shows a schematic of LMs and their most common elemental ingredients: Ga, In, Sn, and Bi. LMs have numerous unique features and potential applications in various fields, such as soft robotics, soft and stretchable electronics, and chip cooling, because they exhibit the properties of both liquids and metals [55–62].

In an ambient environment, the gallium surface automatically forms an oxide layer that can easily adhere to various surfaces [63, 64]. Vigorous stirring can generate more

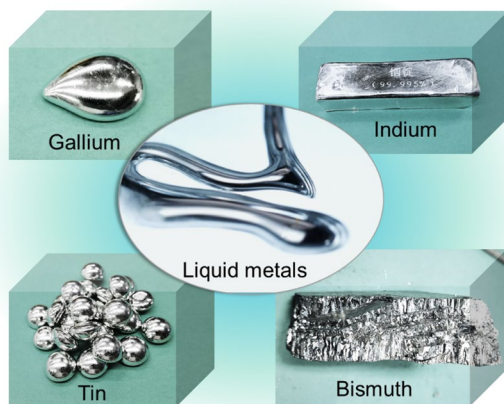


Fig. 2 Liquid metals and their most common elemental ingredients: Gallium; Indium; Tin; Bismuth

gallium oxide in the LM, and the oxidized LM has a high viscosity and is sticky, enabling it to coat the surface of other materials [65]. By stirring gallium-based LM with different elements, LM composites can be fabricated. In addition, it has been reported that gallium-based LM can “swallow” other metals via endocytosis in an environment with electrolytes such as hydrochloric acid and sodium hydroxide solutions [66]. The versatility of LMs lies in their ability to form composites with other functional materials by swallowing and mixing. Any material with desirable merits, such as high thermal conductivity, magnetism, and photothermal properties, can be loaded into LM composites through this general process [67–71]. For example, while swallowing iron particles endows the liquid metal with magnetic responsiveness, mixing tungsten endows the LM with high thermal conductivity [69]. In addition, LM can be dispersed as micro-sized droplets and mixed with various soft materials, such as uncured polymers and hydrogels, which exhibit properties different from those of LM bulk [72–74]. LMs also contain many electrons, and their liquid surface is riddled with specific chemical activity, which shows great potential in self-fueled robots, synthesis and catalysis [75–80].

As LM possesses a melting point that is not far from room temperature, it can transition between the liquid and solid-states [81, 82]. The solidified LM is very stiff even at approximately 10 GPa, while it becomes liquid upon melting [83]. Researchers have utilized the dramatic change in stiffness of LMs during phase transition to fabricate materials with tunable stiffness for robots, smart electronics, and shape memory devices [84–86]. Finally, it is also widely reported that LM has good biocompatibility, making it safe for biomedical applications [40, 87–89]. The above merits of LM will also apply to the fiber-shaped LM, namely LMF, which will be further discussed in the section of the application.

Fabrication Technologies for Liquid Metal Fibers

As mentioned above, one-dimensional liquid metal wire and its composites are defined as LMFs in this paper, where the liquid metal plays a fundamental role in anticipated functions. Before the emerging applications of LMFs are introduced, the fabrication technology for fiber-shaped LM and its composites should be fully illustrated. Based on the different mechanisms, LMF fabrication can be classified as follows.

3D Printing of One-Dimensional Liquid Metal Wires

Three-dimensional (3D) printing is recognized as a promising and efficient manufacturing technology and is widely applied. In contrast to organic ink, most metals with a high melting point are difficult to melt before printing [90]. Room-temperature LMs, particularly Ga and Bi-based alloys, provide a desirable and prospective solution. By adopting LM as ink, subtle 2D electronic circuits and functional devices can be printed on various soft substrates. The printer structure scheme and printing process are schematically shown in Fig. 3a, which is driven by pneumatic or LM gravity. Its relative printing position on the substrate is controlled precisely by a computer program. Owing to the surface tension and oxide layer of LM inks, the continuous deposition of materials, printing accuracy, and structural stability can be guaranteed. Based on linear direct writing technology, researchers have also proposed handwritten liquid–metal circuit technology [91]. For convenience, users can personalize and write stable electronic circuit patterns on a soft substrate via a pen-like device. This handwritten 3D printing technology facilitates the fabrication of LMF, which has been extensively used so far. High-resolution printing of three-dimensional LMF through fine nozzles and was reported by Park et al. [92] Fig. 3b shows a scanning electron microscopy (SEM) image of printed LM lines with kinked, bent, and 3D structures on the substrate. The minimum width of LM lines even reached 1.9 μm and they can be applied to develop superfine, soft and conductive fibers for highly integrated and stretchable devices.

Overall, integrating LM with 3D printing technology not only decreases energy consumption by lowering the melting temperature but also makes it possible to print metal on target substrates to make functional elements at room temperature [31, 93]. Additionally, printing electronics eliminates the etching and cleaning processes usually needed during the fabrication of complex structures, significantly reducing the waste of raw materials. 3D printing

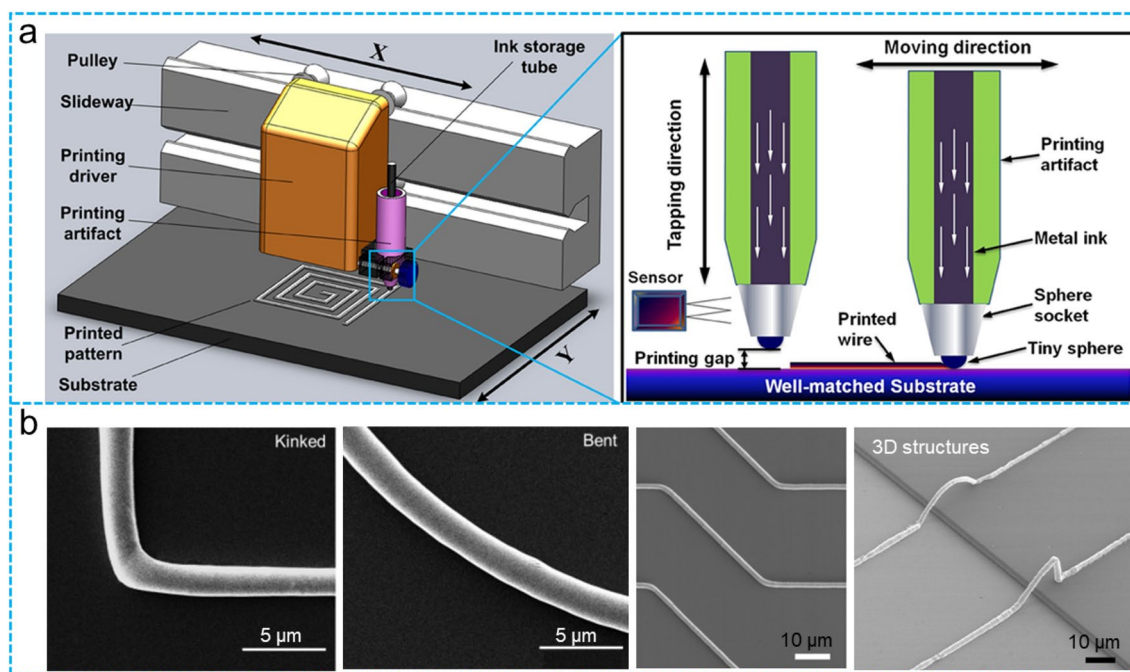


Fig. 3 Principle of printing liquid metal wire. Schematic diagram of **a** linear direct writing technology using an LM ink [31]. Reproduced with permission of Ref. 31, Copyright of © 2014 Sci. Rep. **b** SEM

images of LM ultrafine wires with kinked, bent patterns, and 3D structures [92]. Reproduced with permission of Ref. 92, Copyright of © 2019 Sci. Adv

technology is expected to promote rapid prototyping, and automatic manufacturing for ultrafine LM fibers, which will open a completely new electronic circuit manufacturing schema. Although promising, the printed LM wire cannot be applied in stretchable electronics without proper substrate and packing. Printing the wire directly on different substrates, especially fabric, is essential for extending its applications in smart clothes. To satisfy the demands of printing on a variety of substrates and achieve multiple functionalities, LM ink can be pretreated by various methods such as oxidation, adding functional particles, and increasing viscosity. Thus, LMs can be printed in viscoelastic media to achieve freestanding structures, such as hydrogels. To achieve a stable structure, the melting point of LM can be increased above room temperature, enabling automatic solidification, rapid prototyping, and efficient packaging of printed materials at room temperature [94, 95].

Coating LM on Surface of Fiber

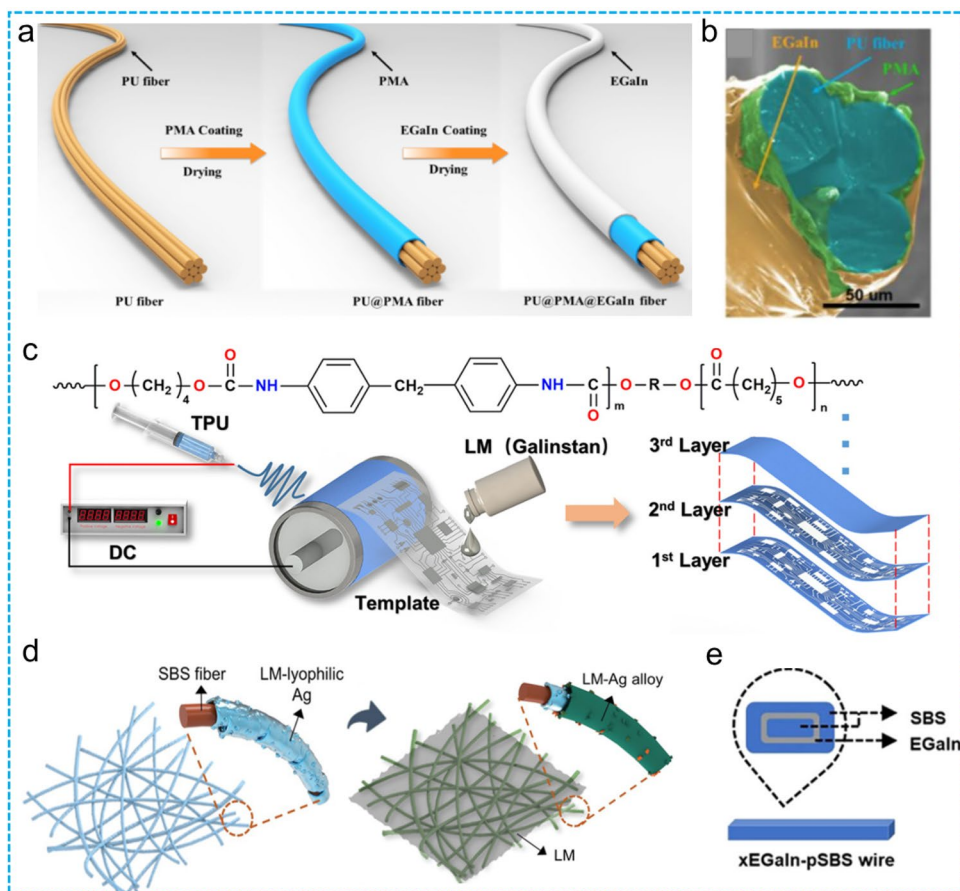
In general, it is difficult to apply LM directly onto the surface of fibers, because of its high surface tension [27]. Previous studies have indicated that LM autonomously forms a thin oxide layer [63], which can easily adhere to different surfaces. Thus, LMFs can be fabricated by immersing traditional fibers such as cotton, silk, and nylon into oxidized

LM. The surface adhesion of LM enables strong coating of the fiber, forming LM coated fibers. To increase the adhesion force, fibers can be coated with sticky materials that have a high bonding affinity to LM before immersing them in the LM. A typical study using this method was performed by Chen et al. [33]. In this study, polymethacrylate (PMA) was used as an intermediate substance connecting the EGaIn LM and polyurethane (PU) fiber, and the preparation process used in this method is shown in Fig. 4a. The micrograph of the resulting conductive fiber shows an obvious hierarchical structure (Fig. 4b), showing the effectiveness of this method. In addition, it has been reported that adding metallic particles such as nickel, iron, and copper through sufficient stirring generates more oxide within the LM, thus significantly enhancing the viscosity and adhesion to the substrates. Based on this, an LM doped with copper particles can be used to directly coat the fiber surface, and the resulting fiber displays desirable mechanical properties and liquid welding characteristics [34].

In addition, to achieve large-scale and quick preparation of liquid metal on commercial cloth, Gui et al. [32] developed spray printing technology to coat LM droplets onto various fabrics with strong adhesion.

Researchers have successfully printed LM on electrospun thermoplastic polyurethane (TPU) films using electrospinning, which is an efficient and rapid technology for preparing micro- and nanofibers. The parametric manufacturing

Fig. 4 **a** Schematic for the preparation of LM-coated fiber. **b** Cross-sectional micrograph of LM-coated fiber (picture colored) [33]. Reproduced with permission of Ref.33, Copyright of © 2020 ACS Appl. Mater. Interfaces. Schematic for the **c** design of a flexible electronic system [47]. Reproduced with permission of Ref.47, Copyright of © 2021 ACS Nano. **d** The fabrication process of LM based superelastic conductors [96]. Reproduced with permission of Ref. 96, Copyright of © 2021 Adv. Funct. Mater. and **e** multilayer LM-coated fiber [60]. Reproduced with permission of Ref.60, Copyright of © 2021 ACS Appl. Mater. Interfaces



of flexible electronic devices, such as capacitors, resistors, inductors, and multilayer circuits, can be realized using this strategy, as schematically shown in Fig. 4c [47]. In addition, researchers have prepared an LMF mat by simply coating or printing LM on an electrospun elastic fiber [97]. As the LM suspended between elastic fibers self-organizes into a transverse network and vertical bending structures, the fiber mat possesses a variety of properties, including high permeability, extensibility, conductivity, and electrical stability. Furthermore, as shown in Fig. 4d, a highly stretchable LM/superlyophilic fibrous scaffold was fabricated by coating LM onto styrene–butadiene–styrene (SBS) fiber with Ag coating [96]. Owing to the adequate wettability caused by reactive wetting between the LM and Ag, LM-based superelastic conductors have a smart conductivity-strain-enhancing feature by forming bicontinuous phases with LM. However, when the LM-based conductive coating of fibers is exposed to the ambient environment, the coating easily adheres to any other contacted objects by the same mechanism, leading to the contamination of neighboring objects and the loss of the LM coating. To overcome this challenge, the LM-coated fiber needs to be packed by coating with an additional membrane. Such a multilayer conductive fiber based on EGaIn LM has been developed as shown in Fig. 4e [60]. In short,

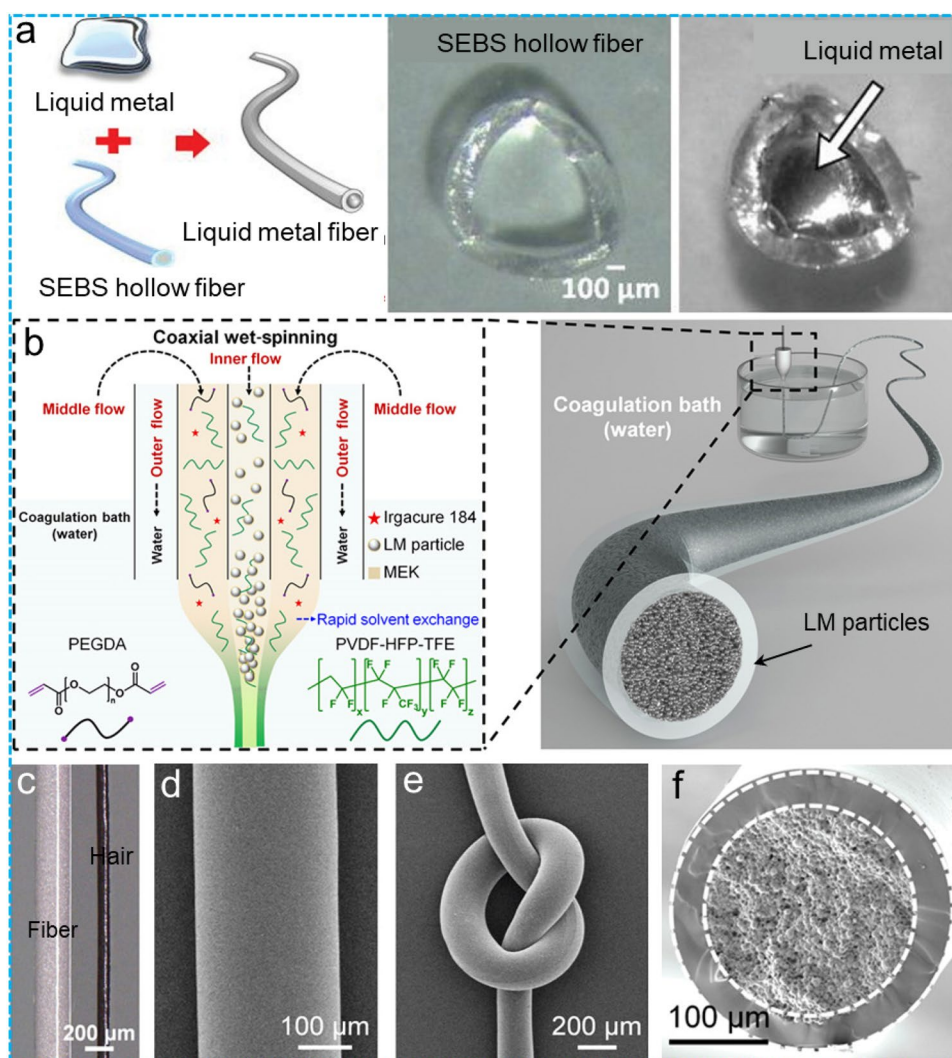
directly coating LM on the fiber surface is an excellent and facile method with broad application prospects.

In addition, the abrasion resistance and stability of LM-coated fibers during practical applications should be considered. After stretching, the highly oxidized LM would risk fracturing, leading to electrical failure, making it unable to heal automatically due to its semi-liquid nature. Direct injection of LM into the hollow fiber can avoid this issue to some extent. Significant follow-up studies need to be conducted to further this research direction.

Injecting LM Core into Hollow Fiber

A simple method of mass-producing LM fiber with a controlled diameter is through the injection of LM into hollow fiber [15]. First, commercially available melt-processing approaches are applied to fabricate stretchable hollow fibers as a shell. Then, the liquid metals are injected into the hollow fibers using a syringe [48], and the LMF is fabricated after sealing, as shown in Fig. 5a. Owing to the liquid state of the LM core, the prepared LMF can be stretched like the hollow fiber shell and maintain metallic conductivity simultaneously through the deformation of the LM. These kinds of fibers are often applied as sensors based on the

Fig. 5 Preparation of LMFs using injection and coaxial wet spinning. **a** Schematic of the fabrication process of LMFs by injecting the liquid metal into hollow fibers [48]. Reproduced with permission of Ref. 48, Copyright of © 2021 Adv. Energy. Mater. **b** Schematic diagram for the coaxial wet spinning process for fabricating the LM composite core-shell fibers. **c** Microscopic images of LMF and human hair. **d** SEM images of the external surface of LMF and **e** Knotted micro-fiber. **f** The cross-sectional SEM image of LMF. [44] Reproduced with permission of Ref. 44, Copyright of © 2021 Sci. Adv



deformation of LM and resistance change in response to external stimuli. Similarly, the liquid metal polymer composite can also be injected into elastic tubes to obtain the LMF. First, the LM is suspended in the uncured polymer and then injected into a hollow shell. Upon heating, the curing LMF is prepared by stripping the outer tube. Recently, numerous attributes of liquid metal polymer composites have been discovered, including self-healing, transitioning between an insulator and a conductor, stable conductivity during stretching, and extreme toughening. Accordingly, liquid metal polymer-based fibers also have these capabilities, making them worthy of being called smart fibers.

Based on this method, the diameter of the LMF depends on the inner diameter of the hollow shell, which ranges from several hundred micrometers to several millimeters for LMFs with good properties. With a larger diameter, the LM inside easily flows through the hollow shell and causes the nonuniform distribution of LM. Conversely, with a smaller diameter, the LM is difficult to inject into the extremely

minuscule hollow fiber as the injection resistance increases dramatically. It is urgent to develop a hands-free method of injecting LM into micro-scale hollow fibers. Researchers have successfully injected LM into microfluidics using vacuum [38], which might inspire hands-free injection of LM into hollow fibers. For example, one inlet of the hollow fiber can be covered with LM, and the structure can be placed in a vacuum chamber to remove the air inside. After restoring the atmospheric pressure, the positive pressure gradient could quickly push the metal through the fiber. If applicable, batch production of liquid-metal-filled fiber could be achieved and more applications can be explored.

Spinning of LM Composites

As we know, electrospinning is a crucial and fascinating method of fabricating fiber materials, especially ultrafine fibers, and it is composed of a power supply, a spinneret, a pump, and a conductive collector. The electrified liquid droplets can

be extruded from the spinneret and extended by electrical force to form tenuous fibers, which must solidify quickly to maintain their structure on the conductive collector [98]. Therefore, charging a liquid droplet is a crucial step in electrospinning. However, it is difficult to charge LM directly because of its high electrical conductivity. Hence, electrospinning is often applied to fabricate traditional fibers and then integrate them with LM. Direct fabrication of ultrathin LMFs using electrospinning technology remains a significant challenge.

Another spinning technique, coaxial wet spinning, enables the mass-production of LMFs that are several hundred meters in length, as schematically shown in Fig. 5b [44]. In general, coaxial needles are needed in this technique, and the peripheral part of the injector is usually filled with coating materials, such as SEBS and PDMS. In addition, the central part of the injector is usually filled with ink, which is the liquid material. In this review, we only discuss LM as the central ink in the coaxial spinning method. To manufacture LMFs by coaxial spinning, elastic materials are often used in peripheral materials to retain flexibility and stretchability. Furthermore, LM composites are used in central materials to endow the composite fiber with metallic conductivity and retain flexibility and stretchability. This method can be used to manufacture the hollow fiber and LM composite filler simultaneously, and the ink filled in the fiber can be changed freely, i.e., Changing the LM from Ga to EGaIn in this method only takes two steps: removing the initial Ga and adding EGaIn into the coaxial injector. The diameter of spinning LMF can reach approximately 270 μm , slightly larger than hair (Fig. 5c), which has a uniform and regular surface (Fig. 5d) and can easily be knotted (Fig. 5e). Figure 5f shows the core–shell structure of LMF. The two very recent studies apply pure LM as the central ink to fabricate a highly stretchable and conductive (up to $3.4 \times 10^6 \text{ S/m}$) fiber through a coaxial wet-spinning process, which can be used in wearable sensors and recoverable switches [46, 99]. These studies have demonstrated the feasibility and superiority of coaxial wet spinning in the continuous fabrication of LM fibers on a large scale, which is just the beginning of the long journey. More work still needs to be done in this area.

Emerging Applications Based on LMFs

To summarize emerging applications of LMFs, this section focuses on the applications that primarily utilize the advantageous properties of liquid metals that are not replaceable. For instance, although liquid metal (EGaIn) has a good electrical conductivity of $3.4 \times 10^6 \text{ S/m}$ (at 20 °C), it is still approximately an order of magnitude lower than those of metals such as copper, silver, and aluminum. Hence, LMFs are only utilized as a conductive fiber in applications where softness or stretchability is crucial.

The emerging applications are classified based on the different properties of LMFs.

First, based on their softness and conductivity, LMFs can be applied as soft and stretchable circuits to connect functional electronic components in wearable devices. Furthermore, based on their resistance (or capacitance) change in response to pressure, tension, and rotation stimuli, LMFs can be utilized as soft sensors for detecting finger bending, breathing frequency, and touching pressure. The LM-polymer fiber has the ability to change between insulator and conductor in response to temperature change, stretching, and pressing. It can thus be applied as a soft switch. Furthermore, LM can change its stiffness from pure liquid to a very hard state through phase transition with temperature change, which can be applied a variable stiffness electrode and shape memory fiber.

Stretchable and Conductive Circuit in Wearable Electronics

Flexible and stretchable electronics have been recognized as crucial technologies for merging electronic systems and human tissues [100]. Material innovation has greatly contributed to the advancement of stretchable electronics by developing smart materials that are more stretchable, conductive, and biocompatible. Benefiting from their intrinsic stretchability and high conductivity, liquid metals and their composites with fiber shapes provide a promising platform for the development of stretchable electronics. When there is a large stretch (over 1000%), the LMF maintains the ability to transport electrons stably, which is different from the solid conductive fiber that might suffer electrical failure after stretching [97]. Due to its fluid properties, the LMF electrically self-heals after severe structural damage. With excellent compatibility, liquid metal fibers can be adapted to different contours, such as clothes, for more versatility. In addition to clothes, they can be directly printed on human skin as an electronic circuit that can monitor real-time physiological states such as heart rate, breath rate, or even blood pressure when combined with other conventional sensors.

Guo et al. [34, 101, 102] fabricated semiliquid metal by loading LM with other metallic particles, such as Ni and Cu, in an aerobic environment. These substances can be printed or coated on stretchable substrates to form conductive and wearable electronics (Fig. 6a) [34]. In addition, LED lights, chips, and batteries can be connected by LMF-based circuits on a T-shirt. Figure 6b demonstrates an interactive circuit based on LMFs. When the finger touches the sensing chip, LED light can be illuminated. After stretching (75% strain) or bending (180°), the LED arrays connected by an LMF circuit can maintain stable states. This observation shows the good stability of the LMF circuit in terms of resistance after twisting, bending, and stretching. Additionally, an LMF

can be applied as a stretchable heating wire under an input current. As shown in Fig. 6c, an LMF strand is connected to a power source to generate current. The temperature of the cotton fabric increases from room temperature to 58 °C under a current of 0.5 A after 120 s due to the Joule heating effect. The heat increase curves of the LMF strand under different input currents are also provided. A conductive LMF can generate eddy currents and Joule heat under an alternating magnetic field. LMFs can be woven or knitted into fabrics with self-heating ability, making such clothing acceptable in cold environments. Based on electromagnetic induction, a wireless charging cloth can be achieved using the LMF, as shown in Fig. 6d.

Zheng et al. [97] fabricated a highly stretchable conductive LMF by coating LM onto electrospun fibers. With the pre-stretching process, the LM can be transformed into a porous material that hangs among the fibers. The

permeability of the fabricated fiber is higher than that of commercially available nylon cloth, allowing it to penetrate sweat or alcohol. The conductivity of this LMF remains quite stable after stretching to 1800% strain, which might have resulted from the porous structure of LM, making it promising for applications as a stretchable conductor. Multilayer electrical circuits can be integrated into one LMF to realize more functions.

Overall, due to their softness and conductivity, LMFs can provide an intrinsically stretchable electronic circuit on clothes or even on skin with seamless contact in applications such as smart clothes and biomedical monitoring. Here, the LMF only serves as a bridge to incorporate electronic functions with soft clothes or skin, making wearable devices more versatile. Smart clothing with diverse functions can be realized by utilizing LMFs and other components to achieve functions, such as illumination, Joule heating, wireless

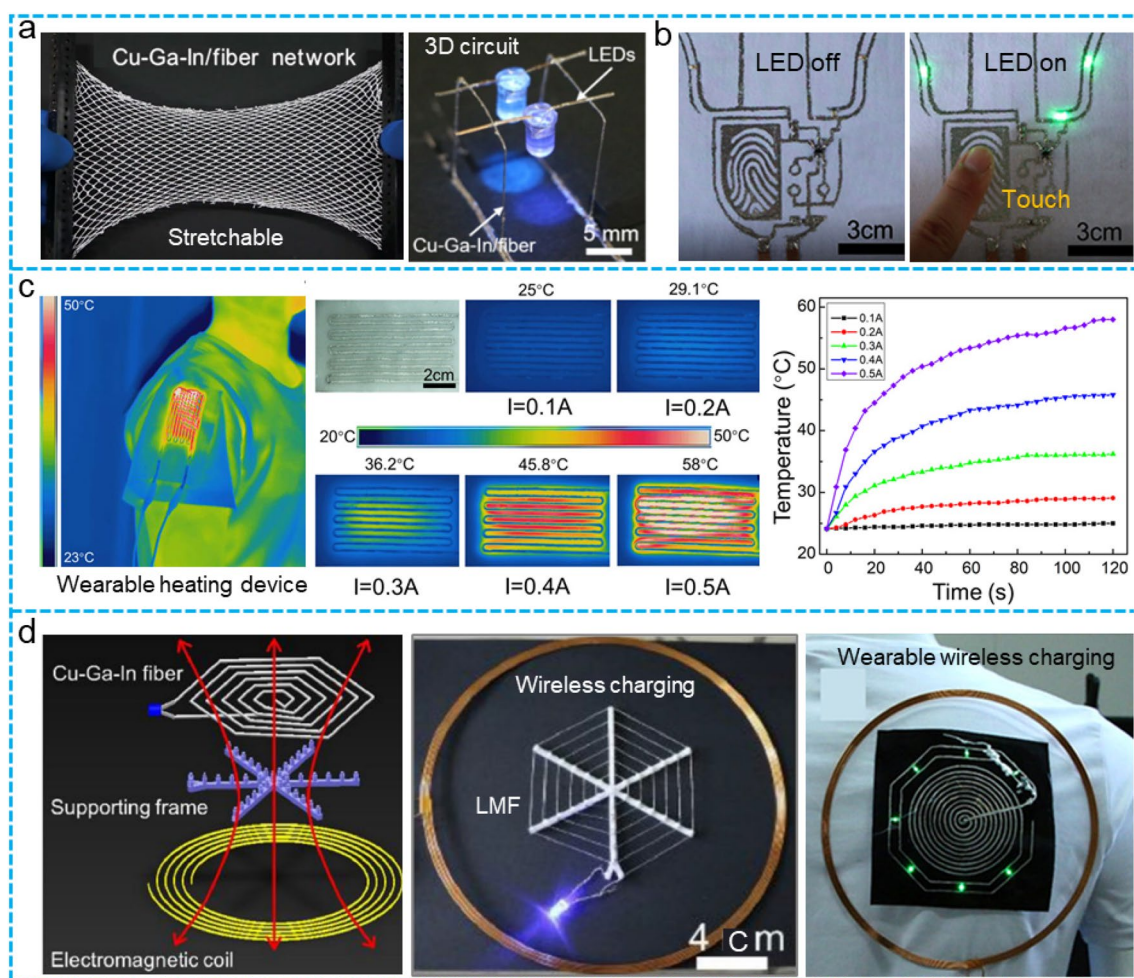


Fig. 6 Wearable electronics for smart clothes based on LMFs. **a** Stretchable and conductive LMFs for 3D electronics and interactive circuit **b**, where the LED lights can illuminate in response to selective touching. **c** Infrared temperature images of self-heating circuits on a cloth and temperature curves with heating time under different input

currents: 0.1–0.5A. **d** A stretchable circuit based on LMFs with the ability to wirelessly power LED on smart cloth. [34, 102] Reproduced with permission of Ref.34, Copyright of © 2020 Appl. Mater. Today. Reproduced with permission of Ref.102, Copyright of © 2019 ACS Appl. Mater. Interfaces

charging, color changing, transmitting, and signal receiving. These wearable electronics applications have the potential to change the concept of clothes. However, there are still several problems that need to be solved before they can be used practically in daily life. First, as a stretchable circuit, the electric stability of the LMF during movement, stretching, and bending should be guaranteed to make it reliable. Thus, the manufacture of lightweight liquid metal fiber in clothes is an important task for practical purposes [103]. When the density of the LMF decreases, the number of smart elements in one smart cloth can increase, and thus, the functionality becomes more powerful. Finally, liquid metal leakage from wearable devices and contamination must be avoided using advanced packing technologies.

Soft and Wearable Sensors Based on the Transformation of LMF

Liquid metals injected into hollow fibers can transform in response to external stimuli, and the resistance or capacitance of the LMF can change accordingly. This phenomenon is applied in the fabrication of soft and stretchable sensors. In particular, considering the merits of LMFs, such as excellent conductivity, knittability, and softness, LMF sensors show great potential in wearable and implantable devices for sensing human motion behaviors and monitoring physiological signals. Depending on the detection mechanism, LMF sensors can be classified as resistance change sensors or capacitance change sensors.

The equation describing the resistance change of an LMF can be written as follows [11]:

$$\Delta R = \rho \frac{\Delta l}{\Delta A} \quad (1)$$

where, ΔR represents the resistance change, ρ represents the resistivity of the material, Δl is the length change, and ΔA is the cross-sectional surface area change. When the LMF elongates by strain, the resistance increases owing to the increase in length, and the surface area decreases. Utilizing this basic mechanism, LMFs have been demonstrated to sense strain, pressure, and deformation [104]. In addition, they can be woven or knitted into intrinsically soft textiles and integrated with clothes or human skin for practical applications. As shown in Fig. 7a, a wearable device can monitor breathing rate by detecting the signal (or voltage) that is related to the resistance change of LMF [105]. Other stimuli such as joint bending, blood pressure, heart rate, and finger gesture can also be monitored using LMF-based wearable clothes [106–108]. Theoretically, any physiological process that results in strain or pressure changes can be monitored using an LMF resistive sensor. Compared with other materials, LMF sensors have high electrical conductivity and are mechanically compatible with human tissue, making

them promising for applications in wearable or implantable sensors.

Although promising, this type of sensor can only sense the stimulus intensity and not the position where the stimuli occur. Sorin et al. [13] proposed an LMF sensor fabricated by injecting the LM into a tubular hyperelastic channel fiber, as shown in Fig. 7b. In their sensing method, a time-domain voltage wave was applied, and it was reflected when the fiber was pressurized. By detecting the time delay, the position that caused the pressure could be determined. The time delay had a positive relationship with the pressing position. This achievement demonstrates the probability of accurate position detection by the LMF pressure sensor, which allows the application of the sensor to monitor the pace-making position of the heart or blood pressure in specific parts of the body. Moreover, the change in capacitance between the fibers can be detected to determine the torsion angle. LM can be injected into hollow flexible fibers such as polydimethylsiloxane (PDMS) fibers. Two fibers can be used to form a capacitance sensor that can detect rotation, stretching, and touch [36]. The geometry change induced by twisting or stretching leads to a capacitance change. When the fiber recovers to its original state, the capacitance returns to its initial state. Similarly, stretching and touching, which might induce geometric changes, can also be measured by detecting the dynamic capacitance. Rotation sensing has been employed as an example to illustrate the working mechanism of capacitive sensors based on LMFs. The capacitance C is given by:

$$C = \frac{\epsilon S}{d} \quad (2)$$

So:

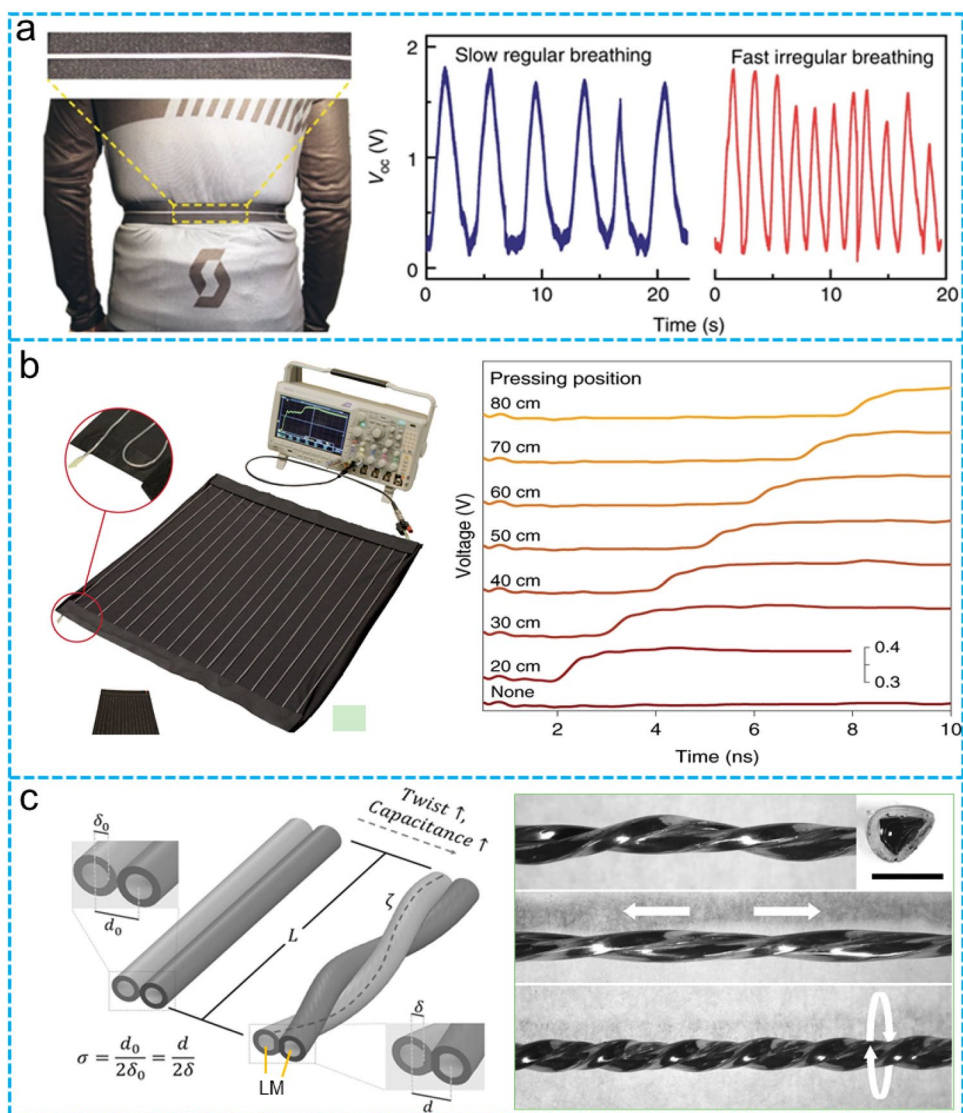
$$dC = \frac{\epsilon}{d} dS \quad (3)$$

where, ϵ represents the capacitivity, dC is a small change in capacity, d represents the distance between two terminals, and S is the surface area.

As shown in Fig. 7c, the end-to-end distance L should be constant. After twisting, the distance ζ increases, leading to an increase in the capacitance [36]. As reported, such sensors can detect torsion up to 800 rad/m, which is much higher (by two orders of magnitude) than the detection limit of current sensors. Similarly, other sensors can be developed according to the relationship between capacitance and different stimuli, such as strain and touch.

The LMF sensor requires an external energy supply and power. Self-powered sensors have been fabricated by combining LMFs with other technologies, such as triboelectric nanogenerators (TEGs) [48, 109, 110], which can harvest and store mechanical energy and electromagnetic energy

Fig. 7 LMF-based soft sensors. **a** Breathing frequency sensing using an LMF sensor. [105] Reproduced with permission of Ref. 105, Copyright of ©2020 Nat. Commun. **b** A triangular liquid metal stretchable fiber is integrated in a stretchable fabric, which is connected with a custom pulse generator. Pressure on the LMF can be quantified and localized simultaneously from the reflection waveforms. [13] Reproduced with permission of Ref. 13, Copyright of ©2020 Nat. Electron. **c** Two twisted fibers with LM core capable of sensing the torsion, stretching, and touching by detecting the capacitance change. [36] Reproduced with permission of Ref. 36, Copyright of © 2017 Adv. Funct. Mater



by utilizing triboelectricity and electromagnetic induction [105]. This technology guarantees the long-term use of LMF sensors without the risk of energy depletion, making it promising for the development of intelligent wearable devices.

The key advantages of LMF-based soft and wearable sensors are their conductivity, knittability, and transformability. Owing to their many merits, soft and wearable sensors based on LMFs are expected to attract increasing attention in important fields, such as the metaverse, smart clothes and intelligent health care. However, LMF sensors still have several limiting aspects that need to be addressed before their further application, including sensing sensitivity, miniaturization, fabrication cost, and packing technology. A very recent study reported a quipu-knotted LMF pressure sensor with better sensitivity than traditional LMF pressure sensor; this sensor can be applied in sensing gastrointestinal motility in multiple anatomic sites [111]. This study might inspire

future researchers to develop sensors with better sensitivity through modifying the structure of LMFs (e.g. knotted, pre-stretched) and exploring more application scenarios.

Soft Switches Based on LMFs that can Transition Between Insulator and Conductor

Fiber-shaped materials made from liquid metal-embedded elastomers have the ability to reversibly transition between insulator and conductor states in response to various stimuli, such as temperature and stretch. These properties of LMFs can be applied to soft and wearable electrical switches [112]. After the LM is mixed with a polymer, LM microdroplets can be suspended in the polymer matrix. Each LM droplet has a polymer shell, and the adjacent droplets are disconnected at room temperature, which results in a large resistance ($> 10^8 \Omega$). Freezing can induce solidification of the LM, accompanied by an abnormal expansion in volume and

glass transition of the polymer matrix, which contributes to the connection of solidified LM and forms a conductive path with a resistance of 0.05Ω [39]. The resistance recovers to its initial state upon increasing the temperature. This transition is reversible and repeatable. Such soft switches have been applied in a minicar, which started to work after deep freezing and stopped when the temperature was increased. However, the transition temperature is too low (212 K) for use in ambient environments. Zhang et al. [113] achieved a higher transition point at room temperature by utilizing gallium and polymers with a higher glass transition temperature. Consequently, these soft switches can be applied as a soft overheating protection device. When the critical temperature is reached, the resistance of the fiber in the device increases dramatically and breaks the circuit, and this behavior can be utilized for overheating protection. The stretchability and fiber shape of these materials make them feasible for incorporation into fabrics such as thermal blankets, thereby protecting humans from fires caused by the overheating of the thermal blankets. In addition, the principle of polymer rheology has been utilized to interpret the relationship between resistance change with both time and temperature. It has been established that the resistance change is not only a response to temperature but also to time, providing a theory for the switchable conductivity in LMFs.

Zhang et al. [14] also reported a fiber composed of an LM gradient dispersed in a polymer exhibiting a new transition phenomenon; the LMF was insulating at temperatures lower than the melting point of the LM and became conductive as the temperature was increased above the melting point of the LM and the glass transition point of the polymer (Fig. 8a), these results are in contrast to those of a previous study. Upon heating, the resistance of the LMF declined from over $10^8 \Omega$ to 36Ω at 28.7°C , which is completely reversible and repeatable. The LMF was woven into clothes to sense temperature changes. After heating with sunlight, the LMF transitions from insulator to conductor, along with the LED turning on, as shown in Fig. 8b.

By selecting different components of LM ($\text{Ga}_{30}\text{In}_{70}$), a new biphasic liquid metal polymer composite that exhibits temperature and stretch-driven transitions between insulator and conductor behavior has been developed [112]. As shown in Fig. 8c, the transition phenomenon depends on the stretching speed. At a tensile speed of 2 mm/min , the material remains conductive without a transition until it finally breaks. However, at a higher tensile speed of 60 mm/min , the material can change from conductor to insulator immediately. The transition phenomenon of an LMF between insulator and conductor in response to the strain can also be repeated, as shown in Fig. 8d. In addition, the biphasic LM polymer can also change between insulator and conductor in response to temperature change (Fig. 8e), making them a promising material for making multi-stimuli responsive

switches. An interesting demonstration has been performed to mimic a spider web that can sense the position and force of a mechanical shock (Fig. 8f). A smart web is woven with an LMF connected to an LED and a power source. When subjected to violent struggling, the relevant LMF transitions from the conductive state to the insulating state owing to the high deformation rate of 200 mm/min , resulting in the LED turning off. However, a gentle struggle with a low deformation rate does not turn the LED off. Therefore, the momentum and position of the web can be inferred from the state of each LED.

In brief, the LM-polymer mixture fiber can switch its conductivity between insulator and conductor in response to different stimuli, which provides the opportunity to develop soft switches, and even soft memories. The insulator state and conductive state can be converted into “0” and “1” respectively. Different information can be stored by the resistance change of the LMF. Based on this concept, smart clothes made of LMF can read and store information. However, the concept is very nascent and riddled with many unknowns, especially regarding the underlying mechanism of transition. In addition, a time delay of the transition induced by thermal stimuli is inevitable, making it difficult to achieve precise control.

Variable Stiffness Electrode

Fibers with tunable stiffness in response to external stimuli have gained increasing attention because of their practical applicability in robotics and external fixators. LMFs can also regulate their stiffness on a large scale through the melting and solidification of the LM [114]. Compared with typical tunable stiffness fibers made from shape memory polymers, LMFs have higher electrical and thermal conductivities, leading to a shorter response time and larger stiffness change ratio [115]. Specifically, at temperatures lower than the m.p. of LM, the LMF remains rigid with a high stiffness of several GPa, while it can become intrinsically flexible after LM melts at a temperature higher than its m.p. Therefore, the m.p. of LM is a vital factor that should be considered when fabricating the LMF with the desired stiffness switch. Typically, gallium and BiInSn are found to be important smart building blocks to obtain switchable stiffness for various practical applications [116].

Gallium features an ideal melting point (29.8°C) between room temperature and human body temperature, making it a promising candidate for biomedical applications. For instance, a gallium fiber can remain rigid off-body and become soft when it comes in contact with the human body. Integrating its conductivity, an LMF-based electrode can be implanted in a rigid state and then turned to a flexible state when warmed to human body temperature. High stiffness is beneficial for the implantation process, while flexibility is

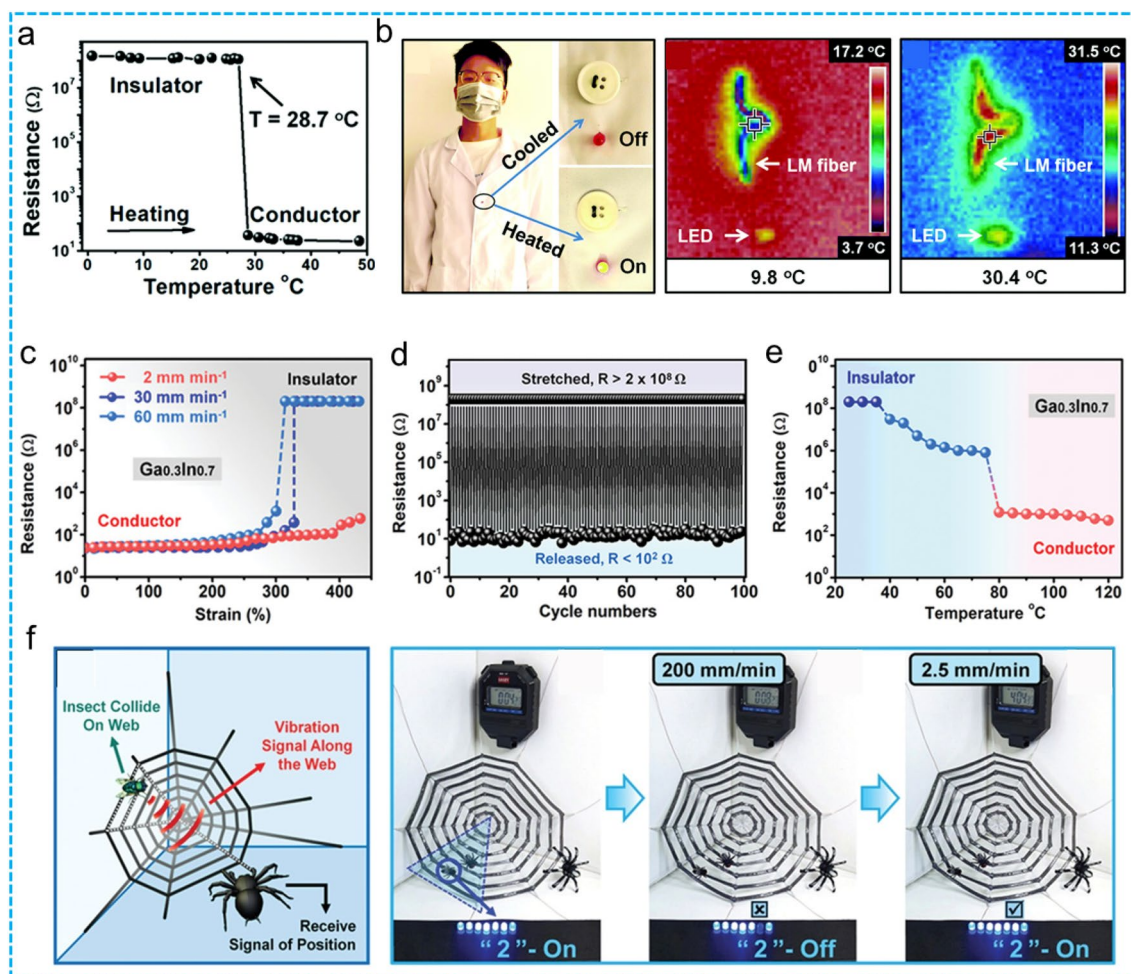


Fig. 8 LMF can transition between insulator and conductor in response to external stimuli and its applications in soft switches. **a** Resistance curves of an LMF depicting transition from insulator to conductor with temperature. **b** LMF woven into clothes for sensing the temperature change [14]. Reproduced with permission of Ref. 14, Copyright of © 2020 Mater. Horiz. **c** Resistance-strain curves of bi-phase LMF showing the variation in transition behavior with the

stretch speed. **d** Repeated transition of an LMF between insulator and conductor in response to the strain. **e** Resistance–temperature curve of LMF using Ga₃₀In₇₀. **f** A spider web-like LMF network that can detect the deformation rate through the resistance change [112]. Reproduced with permission of Ref. 112, Copyright of © 2021 Adv. Mater

elastically compatible with soft tissues, decreasing mechanical damage, and delivering long-term monitoring.

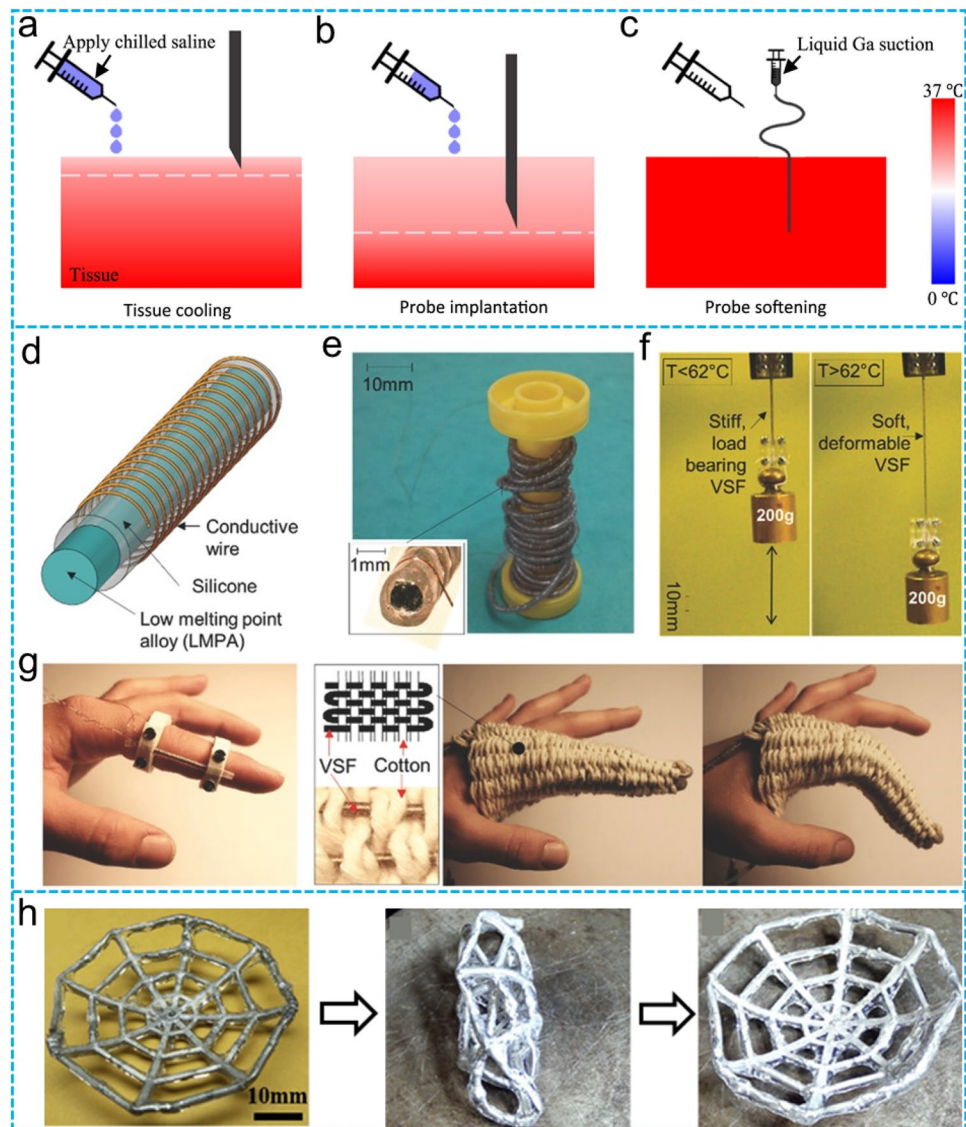
Chiou et al. [82] proposed a multifunctional neural probe with ultra-large tunable stiffness (five orders) by integrating a gallium wire. As shown in Fig. 9a, pressurized liquid gallium can be injected into microfluidic channels and then cooled to a rigid state with a stiffness of 9.8 GPa, which is convenient for implantation into the deep brain (Fig. 9b). After implantation, Ga can be melted by the body heat (at a temperature of 37 °C) and can easily be removed from the channels (Fig. 9c), making the electrode soft and elastically compatible with the brain tissues for different functions, such as chemical sensing and agent

delivery. In vivo tests indicate that stiffness-tunable electrodes reduce the immunoinflammatory response and help in long-term applications. Regarding the phase transition of gallium wires, Jeong et al. [117] reported mechanical transformable systems composed of a soft electronic layer with different functions and a gallium frame layer serving as a mechanical transformative platform. The stiffness of the system could be altered by changing the temperature.

Shape Memory Devices

Integrating the stiffness change of the LM and elastic fibers has been recognized as a promising approach to achieve extrinsic shape memory [85, 119, 120]. For example, an LMF composed of gallium and an elastic polymer can be

Fig. 9 Variable stiffness LMF through phase change of LM. **a** Schematic illustration of (a) freezing the Ga-filled electrode through applying chilled saline, **b** implantation of the solidified Ga-based electrode, and **c** melting Ga with body heat and then removing it from the soft electrode [82]. Reproduced with permission of Ref. 82, Copyright of © 2019 Biosens. Bioelectron. **d** Schematic and **e** image of a variable-stiffness fiber (VSF) with an LM core, silicone encapsulation, and a conductive wire. **f** Tunable stiffness of a VSF controlled by temperature change. **g** Prototype of VSF-based finger splint, which can be assembled into the fabric with cotton filament. [10] Reproduced with permission of Ref. 10, Copyright of © 2016 Adv. Mater. **h** A deployable mesh antenna with programming and recovering process changing the phase of LM between liquid and solid [118]. Reproduced with permission of Ref. 118, Copyright of © 2018 Addit. Manuf



stretched, and its state can be maintained by solidifying the LM at low temperatures. Upon heating, the fibers contract to their original length after the melting of the LM. This process includes the main features of shape memory: deformation, fixing, and recovery. Although the phase transition of Ga is promising for large-scale stiffness tuning, it is difficult to solidify quickly at ambient temperature owing to the high supercooling effect of Ga [121]. Bi, In, and Sn eutectic alloys (Field alloys, m.p. = 62 °C) are typically applied as stiffness-tunable materials for a stable state in an ambient environment. After heating, melted Field alloys can be injected into hollow fibers, which can be woven into wearable fabrics with shape memory.

Floreano et al. [10] proposed a variable-stiffness fiber (VSF) with a Fields alloy core injected into a silicone shell and implemented a conductive wire for Joule heating to melt the alloy again, as shown in Fig. 9d, e. Upon heating for tens

of seconds, the LMF could achieve a remarkable change in stiffness (more than 700 times). They also demonstrated the self-healing capabilities and large-scale transformability of the fibers in the liquid state and high load-bearing performance in the stiff state (Fig. 9f). Furthermore, alloy fibers can be knotted, knitted, and bonded into different shapes, which can be implemented in other fabrics to achieve switchable stiffness. As shown in Fig. 9g, a VSF is knitted together with cotton thread to develop a variable-stiffness fabric as an adaptive splint for bone fractures. The LMF can conform seamlessly to the finger in the soft state, while it can fix and provide structural support to the finger in the stiff state. In addition, when the position of the finger is changed, Joule heating causes the melting of the LMF core which adjusts to the new finger position, and the rigid LMF can also support the finger.

Furthermore, this promising concept has been successfully utilized as a non-invasive external fixation technique in real clinical applications at the People's Hospital of Qijing, China. Based on the phase transition of low melting point alloys, the external fixation device can switch between transformable and supporting states. The operation process is similar to the aforementioned method. Compared to prevailing fixation bandages made of plaster cast and high polymer, LM bandages feature shorter operating times, higher supporting strengths, and better conformability to physiological contours, bringing more comfort to the user's experience. More importantly, this bandage can be adjusted by reheating according to the fixing requirement in the rehabilitation process without the need for removal and refixing. To date, several hundred patients with bone fractures in various body parts have benefited from this new type of LMF bandage.

Zhang et al. [118] developed several lattice shells with the LM core inside to achieve reconfigurable structures based on the shape memory effect. In particular, with the good conductivity of LM, reconfigurable antennas with deployable ability can be developed. As shown in Fig. 9h, the mesh antenna can be programmed (folded) at a high temperature (over 62 °C) and maintained at room temperature, and it can recover to its original shape by heating. Other reconfigurable structures, such as hand gestures, can also be achieved using this strategy.

Briefly, the phase transition of LM in response to a narrow temperature change can be utilized to achieve shape memory fibers. The key advantages of LMFs are their ideal melting point and significant change in stiffness. However, there are several challenges in utilizing stiffness-tunable LMFs. First, improving the heating efficiency and reducing

the time delay in the phase transition are critical for developing a stiffness-tunable LMF. Additionally, thermal shock (> 62 °C) poses a risk to the biological tissue that needs to be addressed. More advanced and safe heating methods are thus needed. Finally, the supercooling of gallium that impedes solidification also causes inaccurate control.

In summary, we listed the different kinds of liquid metal fibers, typical fabrication methods, key features, and emerging applications for the convenience of the readers, as shown in Table 1.

Summary and Perspectives

In conclusion, this review summarizes the materials, fabrication technologies, and emerging applications of liquid metal fibers. The unique advantages of LMFs include its excellent transformability, electrical conductivity, and knittability, which makes it possible to develop smart fibers and devices with the characteristics of self-healing, self-heating, self-powering, wireless charging, large-area displays, mechanical sensing, stiffness tuning, shape memory, and switchable conductivity. The concept of LMFs is still in its infancy, and there are many challenges that must be overcome; however, it also presents profound opportunities that deserve further attention.

- 1) The unique properties of LMFs make them promising candidates for the development of soft and conductive fiber on smart clothes with diverse fantastic functions. To make the demonstration a reality, the integration level of LMF based electronics should be improved

Table 1 Liquid metal fiber (LMF) types, typical fabrication methods, key features, and emerging applications

LMF types	Typical fabrication methods	Key features	Emerging applications
Pure-LM fiber	3D-printing	High resolution (< 10 μm) [92] Free-standing High conductivity (up to 3.4×10^{16} S/m) [91]	Soft circuits in soft electronics 3D structure circuit Reconfigurable antenna [92]
LM-shell fiber	Coating	Large area manufacturing σ : 1.8×10^{16} S/m High stretchability (1,800%)* [97]	Wearable sensors [97] Soft Joule heaters [12] Smart clothes [102]
LM-core fiber	Injecting	Easy operating LM easily deformed Soft-stiffness transition [10] High stretchability* σ : up to 3.4×10^{16} S/m [15]	Soft sensors for detecting pressure, stretching, and rotating [15] Variable stiffness electrode [82] Shape memory devices [10]
LM-mixture fiber	Spinning 3D printing	Scalable and rapid prototyping High stretchability* Conductive 4.35×10^{14} S/m [44] Switch in conductor and insulator [14]	Stretchable and wearable circuits in smart clothes [44] Soft switches [14] Wearable sensors [99]

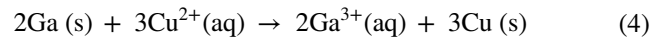
σ Electrical conductivity

*It depends on different components

significantly. Based on the current reports, the diameter of most LMFs is several hundred micrometers, and there are few reports on nano-sized LMFs. Therefore, the fabrication resolution of LMFs should be improved by applying more advanced technologies. In addition, the mechanical and electrical stability of LMFs is important in practical applications. The liquid state is advantageous in soft electronics, but the leakage of liquid metal is troublesome, and advanced LMF packaging technologies are worth exploring. Finally, the mass manufacturing technology of liquid metal fibers should be improved.

- 2) The LMF sensor has the combined properties of conductivity and flexibility. Thus, a tiny pressure or stimulus can induce the transformation of LM, and the resistance (capacitance) changes accordingly, which endows sensors with high sensitivity. Furthermore, LMFs are mechanically compatible with human tissues, and it is possible to achieve long-term detection of physiological signals in vivo. A recent study reported a sensor based on LMFs with a quipu- knotted shape (for improving the sensitivity of pressure sensing) that can be successfully applied in gastrointestinal manometry; this study shed light on the utilization of LMF sensors in vivo [111]. In the future, LMFs may have the ability to be applied in other pressure sensing applications in vivo, such as sensing intracranial pressure, tympanic pressure, bladder pressure, intraocular pressure, and spinal canal pressure. In this promising field, significant follow-up studies should be conducted.
- 3) Combining LMFs with new materials will bring new functions. As a result, the combined materials have various intelligent properties. For instance, intelligence switches, sensors, and electrical heating can be achieved by combining liquid metal and other flexible, conductive materials. Importantly, LM combined with natural biomaterials can be used to develop biocompatible and biodegradable LMFs. Ye et al. [122] reported that liquid metal particles can combine with gluten in wheat to form LM-biomaterial composites, which can be applied in electronic skin for motion sensing. According to this paper, the combination of EGaIn and gluten brings the composite several properties: self-healing ability, high elasticity, better biocompatibility, and biodegradability. This study will inspire more research on fiber-shaped LM and biomaterial (e.g. protein, starch) composites, and their biomedical applications.
- 4) Chemical properties of LM could be explored for smart fibers with more functions. For example, Kim et al. [123] recently demonstrated that the LM coating on fabric can provide nucleation sites for growing LM-Cu particles through galvanic replacement of Cu^{2+} ions

exploited for the electrical substitution reaction between gallium particles and copper ions.



It can directly realize fabrics resistant to pathogens with uniformity, easy processing complexity, and strong adhesion. Compared to the loading of copper particles alone, the LMCu-loaded fabric exhibited strong resistance to pathogens (99% of pathogens could be killed within 5 min) and maintained antimicrobial properties after repeated use and erosion by aerosols and contaminated droplets. Thus, it is a promising candidate for the production of antibacterial and antiviral fabrics. LMFs will have more properties and functions as liquid metals and their composites are continuously explored.

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Declarations

Conflict of interest Authors declare no conflicts of interest.

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