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TOPICAL REVIEW

Stretchable composites with electrical conductivity

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Abstract

Recently, the development of flexible and stretchable electronics has resulted in extensive studies focused on the investigation of elastic composites with various functions using stretchable materials, such as rubber. Among these materials, conductive composites using stretchable conductive materials can withstand larger deformations than solid metals and simultaneously retain their conductivity. Furthermore, such conductive composite materials can be imparted with various physical properties, such as thermal and magnetic properties (in addition to conductive properties), by changing the properties of the base and filler materials. Owing to these properties, conductive stretchable composites are expected to have various areas of applications, such as sensors, heaters, and magnetic barrier films, beyond electrical wiring on flexible and stretchable substrates with large deformations. This study reviewed flexible materials exhibiting both elasticity and conductivity with a particular focus on the internal filler material and functionality.

1. Introduction

The development of wearable devices and soft robots has facilitated extensive studies on the fabrication of electronic substrates, sensors, actuators, and other components using stretchable materials. Electronic components composed of solid metals that have been used on rigid substrates are damaged by the expansion and contraction of the device itself. Therefore, stretchable conductive materials that can withstand large deformations are required.

Stretchable conductive materials, including solid metals with optimized wiring structures and liquid materials, have been proposed. Moreover, many 'stretchable composites,' wherein conductive materials are added through mixing with dielectric stretchable materials such as rubber and gel, have been proposed. Composite materials can exhibit various electrical and physical properties based on the selection of the base materials and fillers. Consequently, various combinations of these materials have been designed according to the required properties. In particular, the selection of the filler material primarily affects the properties of the composite. Many conductive filler materials have been used in composites, including metals, carbon, and conductive polymers. This study focused on presenting an overview of the types of filler materials used and their properties as conductive, stretchable composite materials. The filler materials mixed into the composite materials were divided into the following three categories (figure 1).

(i) Solid metal materials: solid metal-based micro powders such as Fe and Cu powders and Ag nanowires.
(ii) Solid nonmetal materials: nonmetallic conductive materials such as C nanotubes (CNT) and nanoparticles and conductive polymers.
(iii) Liquid or liquid–solid hybrid materials: conductive liquids such as liquid metals or hybrid materials of liquid materials such as gels and solid conductive materials.

2. Solid metal materials

Solid metals are widely used as fillers for conductive composites. Metals have high electrical conductivities and are used in composites for the same purposes

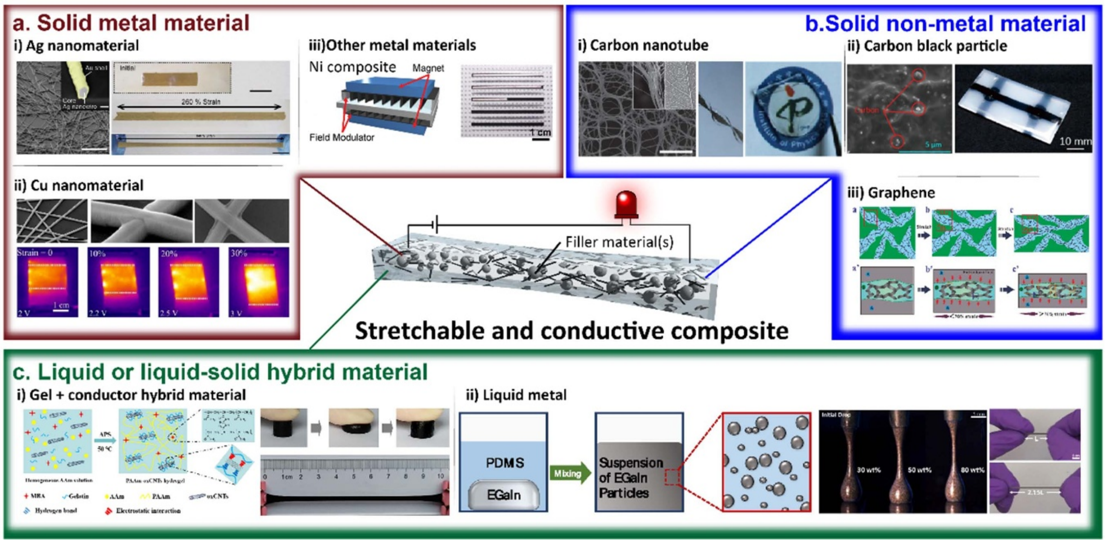

Figure 1. Conductive filler types in a stretchable and conductive composite material: (a) solid metal materials: (i) Ag nanomaterial. Reproduced from [1], with permission from Springer Nature, (ii) Cu nanomaterial. Reproduced from [2] with permission from the Royal Society of Chemistry, and (iii) Ni nanomaterial [3]. John Wiley & Sons. © 2014 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (b) Solid non-metal materials: (i) carbon nanotube (CNT) [4]. John Wiley & Sons. © 2012 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim, (ii) carbon black (CB). Reproduced from [5]. CC BY 4.0, and (iii) Graphene. Reproduced from [6] with permission from the Royal Society of Chemistry. (c) Liquid or liquid–solid hybrid materials: (i) gel and conductor hybrid materials. Reprinted from [7], Copyright 2019, with permission from Elsevier and (ii) liquid metal. Reproduced from [8] with permission from the Royal Society of Chemistry.

as electronic metal wires. In addition to their high electrical conductivity, metals often exhibit physical properties other than conductivity, such as magnetic and thermal properties. Therefore, they are often used in applications other than electrical wiring, such as magnetic actuators, heaters, and heat exchangers. This section summarizes stretchable conductive composites made of solid metal and stretchable rubber-based materials.

2.1. Silver

Ag has been widely used as a filler material for composites because of its high electrical conductivity and ease of fabricating micro- and nanostructures. Ag nanowires start sintering at a relatively low temperature of approximately 70 °C [9], and studies have been conducted to achieve high electrical conductivity by mixing Ag with a base material and followed by sintering of the mixture (figure 2(a)) [10]. In this case, each nanowire is connected by a myriad of electrical paths as it is integrated through sintering dissolution, and the entire composite is highly conductive. However, these joints often break during tensile deformation, which results in gradual disconnection and an increase in electrical resistance upon the application of multiple tensile deformations. To solve this problem, one study reported the use of Ag microparticles to ensure conductivity only through physical contact without the sintering process (figure 2(b)) [11]. In this study, a material with high conductivity and elasticity of 1000–3000 S cm^{-1} was prepared by mixing tens of micrometers of Ag flakes with polystyrene-block-polybutadiene-block-polystyrene (SBS). This material maintained conductivity even under approximately 30% elongation owing to flake-to-flake contact. However, when the composite was elongated by more than 40%, the material lost its conductivity owing to the disruption of the internal flake alignment, and the conductivity was not restored even after the composite was restored to its original length. These results indicate that although Ag-based stretchable materials exhibit high conductivity at low elongation rates, they lose conductivity at elongations exceeding 50% because of the breakdown of Ag-to-Ag connections.

Another composite, which incorporates silver microflakes mixed with fluorine rubber [12], maintains a high electrical conductivity of 935 S m^{-1} at 400% elongation owing to the in-situ formation of nanoparticles from crushed microflakes. This conductive composite was used as wiring on a glove to connect temperature and pressure sensors, enabling temperature and pressure sensing.

There have also been reports on the fabrication of composites using Ag nanowires and other metals. For example, a study was conducted using Ag nanowires coated with Au in a core-shell structure to prevent Ag oxidation and maintain conductivity (figure 2(c)) [1]. Another study proposed a low-cost composite with both high conductivity and elasticity by coating Ag nanowires with Cu [13].

Silver has garnered significant attention as a filler for stretchable conductive composites owing to its high electrical conductivity, low sintering

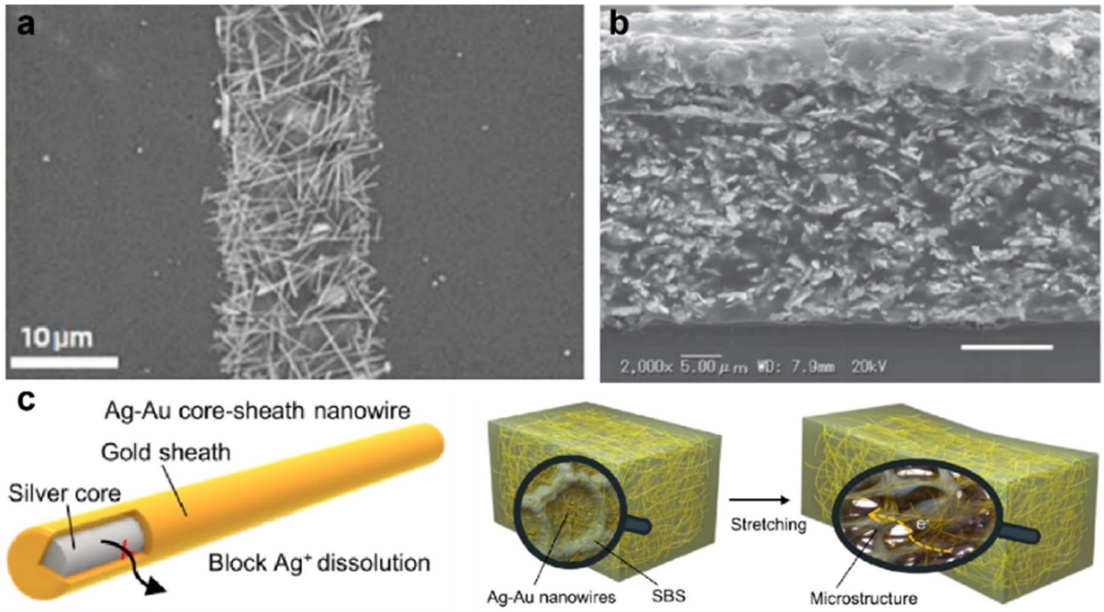

Figure 2. Ag-based composite: (a) SEM image of sintered Ag nanowire composite. Reprinted with permission from [10] Copyright 2015 American Chemical Society. (b) SEM image of sinter-free Ag micro flake composite. Reproduced from [11]. © 2019 The Japan Society of Applied Physics. All rights reserved. (c) Schematic of Ag-Au core shell nanofiber composite. Reproduced from [1], with permission from Springer Nature.

temperature, and the ability to maintain conductivity without sintering, depending on the base material. However, achieving both high electrical conductivity and elasticity with silver requires special base materials and fabrication methods, as it relies on sintering or percolation for electrical connections. Additionally, silver's performance can deteriorate under high-temperature, high-pressure conditions and exposure to sunlight owing to its metallic nature. Furthermore, silver is relatively expensive (about \$30/oz), which makes it less suitable for large-area device fabrication. Improving electrical conductivity, elasticity, and environmental resistance with more accessible processing methods could lead to more advanced applications in stretchable functional devices.

2.2. Copper

Cu nanowires have attracted attention as a low-cost material for fabricating conductive stretchable composites owing to it being less expensive than other conductive metals. Moreover, it has an electrical conductivity comparable to that of Au and Ag. Composites created by post-polymerization of polydimethylsiloxane (PDMS) and annealed copper nanowires [14] achieve a stable elongation of approximately 80% and a conductivity of 7 Ω sq^{-1}. Additionally, post-treatment enhances the chemical stability of copper, enabling reliable use for up to 50 d.

Composite material prepared by mixing SBS and Cu nanowires exhibits a high electrical conductivity of 1858 S cm^{-1} and retains this conductivity even under 150% elongation (figure 3) [15]. In addition, the composite exists in the liquid state before curing and is easily printable onto the substrate surface. In addition to its electrical conductivity, Cu has a high thermal conductivity. Consequently, it is often used as a composite material for electrical connections and heat transfer. In the fabrication of a stretchable transparent heater using Cu nanowire composites [2], the composite exhibited 91.4% light transmittance and maintained electrical conductivity at 40% strain. At 30% strain, the composite generated heat exceeding 45 °C for an applied voltage of 3 V.

Copper is among the least expensive metals (about \$0.3/oz) and is advantageous for fabricating large-capacity composites. It is an excellent conductor of both heat and electricity, making it suitable for thermal diffusion and heat exchange applications as well as electrical uses. However, copper's surface is highly prone to oxidation [16] and its electrical properties can be unstable in air. Additionally, copper requires a higher sintering temperature compared with silver [17], necessitating the use of base materials that can endure high temperatures to enhance electrical conductivity through sintering.

2.3. Other solid metals

In addition to the aforementioned metals, many other electrically conductive composite materials use metallic particles. Because Fe and Ni powders exhibit magnetic properties in addition to electrical conductivity, composite materials with both properties have been proposed. $MnFe_2O_4$ in PDMS offers multiple functionalities: electrical conductivity of 24.03 S cm^{-1} and suppression of electromagnetic disturbance (EMI-SE of −21 dB corresponds to 99.21%

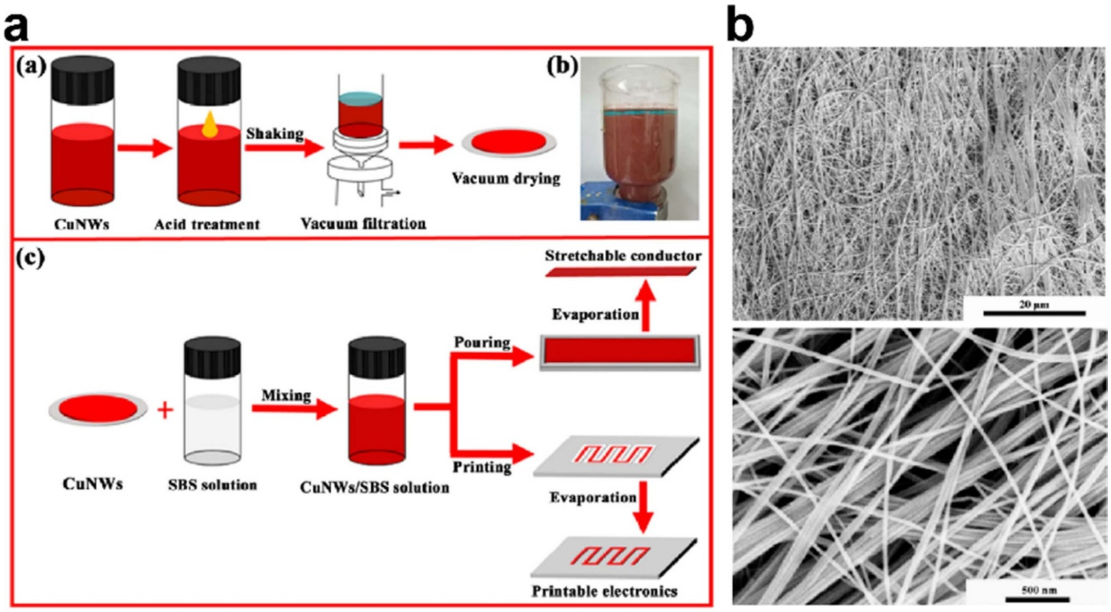

Figure 3. Cu-based composite [15]: (a) Fabrication method of Cu nanowire and SBS composite. (b) SEM image of Cu nanowire. Reprinted from [15], Copyright 2017, with permission from Elsevier.

shielding efficiency) [18]. In addition, all these properties were maintained even when the composite was elongated by more than 350%.

Composite materials using μm-order Ni powder are highly magnetic and can be patterned by magnetic force (figure 4) [3]. Furthermore, composites using large-diameter filler materials have been reported to exhibit positive piezoelectric properties, that is, decreased electrical resistance in response to tensile deformation. The Ni powder composite fabricated in this study was capable of 100% elongation, and its electrical resistance reduced to approximately 1/10 under strain. By covering this composite material with Ag nanoparticles, an approach is being developed to ensure electrical conductivity with Ag at low elongation and Ni at high elongation.

Iron, the most cost-effective metal because of its abundance as iron ore, can be used as both an electrical conductor and a magnetic shield because of its magnetic properties. However, producing nanostructured materials from pure iron is challenging owing to its tendency to oxidize. Consequently, iron oxide is typically used in combination with manganese, zinc, and other elements to create ferrites. While manganese ferrite remains relatively inexpensive (approximately \$10/oz) compared to silver and other metals, it has lower electrical conductivity compared to copper and silver.

Nickel, like iron and copper, is also relatively inexpensive (approximately \$0.6/oz), and its magnetic properties allow for processing techniques, such as magnetic patterning, that are difficult with copper and silver. Nonetheless, nickel has lower electrical conductivity than both silver and copper. Additionally, nickel has recently been identified as a carcinogenic hazard [19], prompting ongoing research to find suitable alternatives.

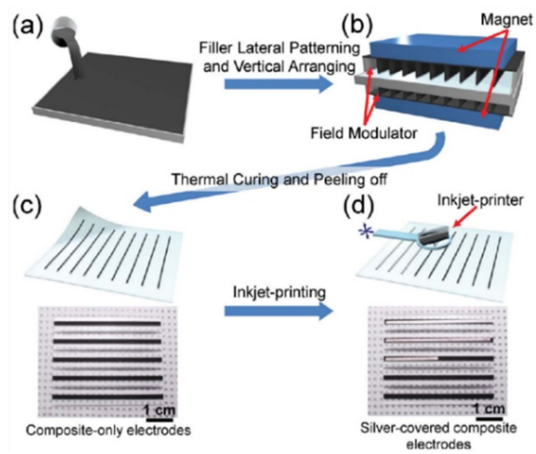

Figure 4. Magnetic patterning of Ni powder composites. [3] John Wiley & Sons. © 2014 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

The following table 1 compares the performance of conductive composites with various metal materials:

3. Solid non-metal materials

In addition to metals, C materials have free electrons owing to their atomic arrangement and can achieve electronic conductivity similar to that of metals despite being non-metallic. These materials are often used in applications that cannot be achieved with metal composites because they are easy to obtain and process, and the fine nanostructure of C facilitates the

Table 1. Comparison of composites with metal materials.

References	Material type	Base material	Fabrication process	Conductivity	Stretchability	Application
[1]	Ag nanowire	SBS	Casting	72 600 S cm^{-1}	840%	Voltage sensing electrodes
[10]	Ag nanowire	PDMS	Mixing and sintering	0.6 Ω sq^{-1}	50%	Strain sensor
[11]	Ag microflakes	SBS	Mixing	3525 ± 145 S cm^{-1}	40%	—
[12]	Ag microflakes	Fluorine rubber	Mixing	6168–935 S m^{-1}	400%	Wiring
[13]	Ag nanowire	PDMS	Annealing	1000 S cm^{-1}	300%	Wiring
[14]	Cu nanowire	PDMS	Annealing	7 Ω sq^{-1}	80%	Wiring
[2]	Cu nanowire	PDMS	Casting	8 Ω sq^{-1}	30%	Heater
[15]	Cu nanowire	SBS	Casting	1858 S cm^{-1}	920%	Wiring
[18]	$MnFe_2O_4$ nanoparticles	PDMS	Casting	18.69 S cm^{-1}	400%	Electromagnetic interference shield and strain sensor
[3]	Ni powder + Ag nanoparticle	PDMS	Mixing and magnetic patterning	Below 20 Ω	100%	Wiring

fabrication of electric wires with high optical transparency. In contrast, composites made from these materials often have lower electronic conductivities than metal composites and are mainly used for sensing applications based on the reading of changes in current values rather than for applications that require extremely low resistance. This section summarizes stretchable conductive composites that use non-metallic solid materials.

3.1. CNT

CNT are C materials that can be obtained in reaction chambers through arcing between graphite electrodes. Many composites with electrical conductivity have been proposed by mixing CNTs with rubber materials. In addition, CNT are generally lighter than metals, and the microstructure fabrication method is relatively easy; thus, composites made of fine CNTs exhibit high light transmittance. The PDMS and multi-walled CNT composite [20] can be analyzed approximately 40% elongation by direct measurement of electrical resistance and features low hysteresis (figure 5). The composite can withstand 1000 tensile tests and exhibits high sensitivity (300%–500%). Moreover, it has been successfully applied to wearable devices to detect wrist and fingertip flexion. Stretchable conductive materials comprising SWCNT films and PDMS composites [4] maintain sufficient electronic conductivity (7–53 Ω sq^{-1}) for use as wiring in LED lighting circuits even under 50% elongation. In addition, they achieve an optical transparency of approximately 60%.

CNTs have a very high aspect ratio among conductive materials and can easily form conductive paths within a resin matrix. Consequently, a relatively small amount of CNTs can achieve high conductivity. However, owing to their elongated shape, CNTs tend to agglomerate, resulting in insufficient dispersion within the resin and poor conductivity stability. Several surfactants have been proposed [21] to disperse CNTs more effectively, and their effective use is expected to further improve the stability of CNTs' electrical conductivity.

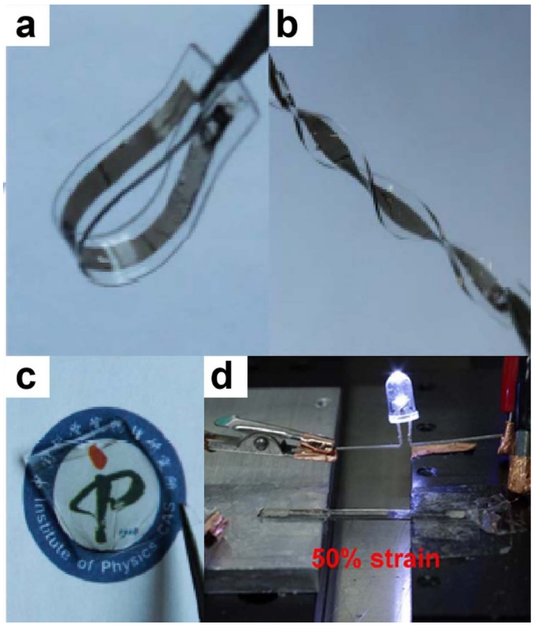

Figure 5. CNT composite [4]: (a) CNT composite wiring on a PDMS substrate. (b) Stretched and twisted substrate. (c) Transparency demonstration of the CNT network. (d) LED lighting demonstration under 50% strain. [4] John Wiley & Sons. © 2012 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

3.2. Carbon nanoparticle (carbon black (CB))

CB differs from nanotubes in that it is a micropowder with a spherical shape rather than a tubular shape. CB

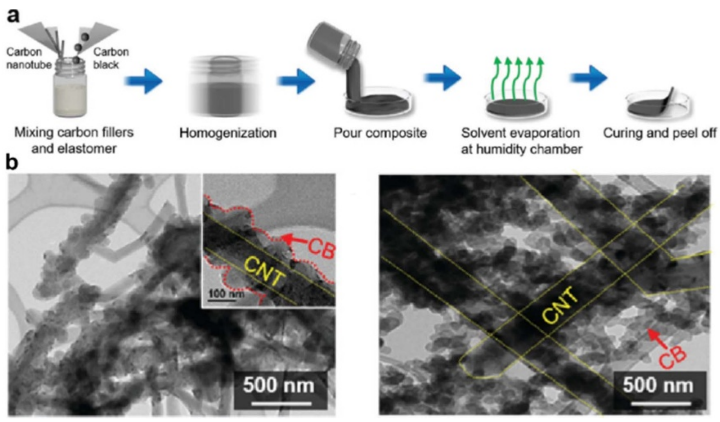

Figure 6. CB composite [23]: (a) Fabrication method of CB-CNT hybrid composite (b) TEM image of the fabricated composite. [23] John Wiley & Sons. © 2018 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

is generally mixed with rubber materials to increase their mechanical stiffness; however, it has an electronic conductivity similar to that of nanotubes, and many applications have been proposed as conductive composites. In a study on conductive composites prepared by mixing nanosized CB with Ecoflex [5], a silicone rubber material, the composites remained electrically conductive under 50% strain and were used as strain sensors. Similarly, a study used a composite of CB powder and Ecoflex as an electrode for a capacitive strain sensor. In this study, a CB-Ecoflex composite was used as ink and placed on a substrate made of a barium–titanium-Ecoflex composite [22]. The CB composite reported in this study can be used as an electrode even under 100% elongation and has succeeded in stable strain measurement even after 1000 times elongation tests.

CB is often used in CNT composites. For example, a conductive composite of CB and CNT mixed in Ecoflex has been proposed (figure 6) [23]. This study showed that CB fills the gaps between CNTs to obtain more stable electronic conductivity under elongation and that the change in electrical resistance was smaller than that of composites with non-mixed CB or CNT, even at 200% elongation. The electrical properties of this composite were stable even after 1000 cycles of repeated tensile testing. Other composites have been proposed wherein the CB surface is activated by 3-(trimethoxysilyl)propyl methacrylateon to increase its affinity with silicone resin, and CNTs are added to the surface to obtain high electrical conductivity and stability against tensile deformation [24]. This composite can be elongated by 211% and exhibited a conductivity of 248.8 S m^{-1}. The composite can be used as a highly sensitive strain sensor in a wearable device to determine throat behavior during drinking and finger bending.

CB has a simple spherical shape, which allows for high dispersibility and stable conductivity when composited. However, owing to its low aspect ratio, a relatively large amount of CB must be mixed to achieve sufficient conductivity, which may reduce the elasticity of the entire composite rubber. CB can be produced with different characteristics depending on the production method, such as activated carbon, acetylene black, and Ketjen black [25]. By optimizing the use of these various properties, it is expected to enhance the functionality of composite materials.

3.3. Graphene

Graphene is a material comprising C atoms bonded in a sheet-like structure. It has attracted attention for its useful properties, such as higher electrical conductivity than Ag and higher thermal conductivity than Cu [26, 27]. Graphene composites require fewer particles in the composite than CNT or CB composites, thereby facilitating the fabrication of electrically conductive composites without significantly compromising the physical properties of rubber (e.g. flexibility and stiffness) [28–30]. Composites of graphene mixed with natural rubber (NR) and butadiene styrene rubber (SBR) [6] were conductive at a low mixing ratio of 0.3 vol % and remained conductive after 300 times applications of 100% elongation (figure 7). The composite has been applied as a strain sensor, and a strain of approximately 120% has been successfully measured by measuring electrical resistance. A study has also been proposed to mix graphene oxide (GO), reduced GO (rGO), and graphene nanoplates (G-NPLs) in styrene–ethylene–butylene–styrene (SEBS) [31]. In this study, GO and rGO exhibited conductivity at a mixing ratio of approximately 2 wt %. In contrast, G-NPLs did not exhibit conductivity at concentrations greater than 6 wt %. The composite of GO and rGO remained conductive under 10% strain and was used as a strain sensor to detect finger flexion.

Compared to CB and CNTs, graphene exhibits superior conductivity even in small quantities due

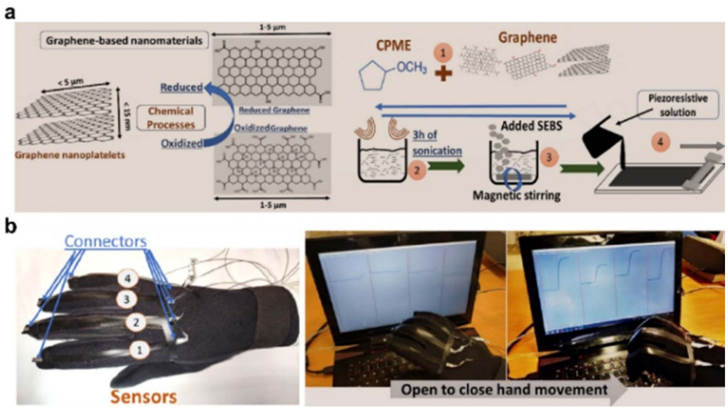

Figure 7. Graphene composite [31]: (a) schematic of graphene structure and fabrication method of graphene composite sheet. (b) Demonstration of the graphene-based conductive sheet as a finger-bending sensor. Reprinted with permission from [31]. Copyright 2019 American Chemical Society.

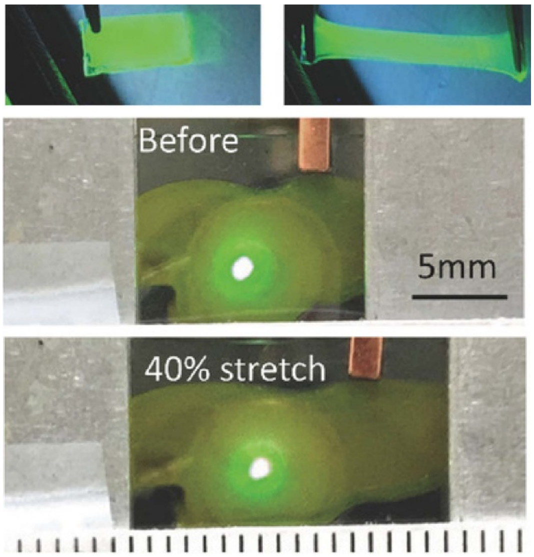

Figure 8. Stretchable light-emitting composite with PEDOT/PSS as an electrode and stretchable LEDs made of Perovskite. [36] John Wiley & Sons. © 2017 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

to its well-developed crystalline structure. However, synthesizing large quantities of graphene monolayers is more challenging than producing CNTs or CB, making it less suitable for large-scale composite fabrication. Research is ongoing to improve the synthesis of monolayer graphene [32–34]. Advancements in graphene synthesis technology are anticipated to enhance the functionality of composites.

3.4. Conductive polymer

Conductive polymers are generally termed as polymers with electronic conductivity. They are widely used for transparent electrodes and other applications, particularly because of their high transparency compared with metals and carbon materials. Poly(3,4-EthyleneDiOxyThiophene)/Poly(4-StyreneSulfonate) (PEDOT/PSS) is a typical conductive polymer and is widely used because of its excellent heat resistance and chemical stability. The sensor had a high optical transparency of 77%, could be extended by 50%, and is used as a sensor to measure finger and wrist flexion [35]. A stretchable light-emitting composite was also proposed (figure 8) [36] that used PEDOT/PSS and poly(ethylene oxide) (PEO) as an electrode on which a stretchable LED made mainly of perovskite was mounted. The stretchable substrate equipped with this LED continued to emit light even at 40% elongation, and no wire breakage was observed after 50 cycles of repeated elongation and contraction.

Polyaniline (PANI) is a representative conductive polymer. A composite of PANI and a poly(vinyl acetate) latex [37] showed approximately 5% elongation while maintaining conductivity, and a composite of polyacrylic acid (PAA) and phytic acid (PA) with PANI [38] indicating 0.12 S cm^{-1} conductivity and

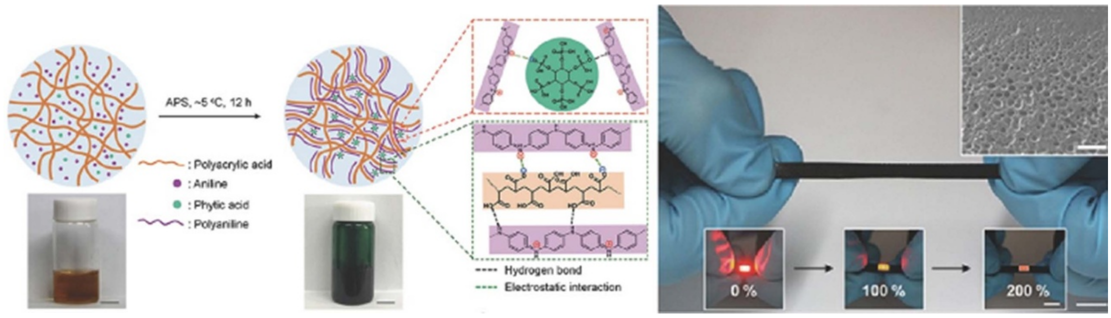

Figure 9. Self-healing conductive composites using polyaniline. [38] John Wiley & Sons. © 2017 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

460% elongation (figure 9). Further, it was used as a sensor to detect finger bending. Furthermore, this material can be cut and refused, and the change in conductivity before and after cutting is less than 1%.

Conductive polymers offer the dual benefits of transparency and conductivity, making them ideal for video output devices. Their ability to be melted allows for processing methods like roll-to-roll, and they exhibit superior processability compared to metals and carbon. However, the stretchable conductive polymers currently available have low electronic conductivity, which limits their use as direct electrical wiring. Therefore, further advancements in conductivity are needed through the synthesis of new conductive polymers.

The table 2 below compares the performance of conductive composites with solid non-metal materials.

4. Liquid or liquid–solid hybrid materials

Liquid materials have Young's moduli of almost zero and can be deformed into any shape. Several studies have been conducted using liquid materials with such deformation performance to develop conductive composites with high electrical conductivities and deformation capabilities. Conductive composites with liquid materials can solve the problems associated with composites containing conductive solid materials that lose their deformability. Composites of liquid materials are also remarkable because they exhibit properties beyond electronic conductivity, such as self-healing [39–43].

Conductive composites using liquid materials as fillers can be classified into two types: those using liquid–solid hybrid materials and those using only liquid conductive materials. In the first case, water or ionic liquids are used as the liquid materials, and C materials such as CNTs, conductive polymers, metallic particles, or liquid metals are used as the conductive materials. These fluids remain in a liquid state inside the composite after curing and enhance its deformation performance. In the second case, because liquid metal is the only practical liquid material with electronic conductivity, materials wherein the liquid metal is dispersed in a matrix, such as silicone rubber, have been developed. Research is being conducted to develop materials that exploit the properties of both liquid metals and solid conductors through their composition. This section reviews stretchable conductive composites containing liquid or liquid–solid hybrid materials.

4.1. Conductive gel

In conductive gel composites, solid conductive materials are mixed with liquid-containing base materials (figure 10). The roles of liquids in gel composites can be classified into three categories for electrical functions: dispersants, plasticizers, and auxiliaries. Water is widely used as a dispersant and plasticizer because of its biocompatibility and abundance; however, gels using organic solvents, such as ethylene glycol and dimethylformamide, also exist. The secondary doping of conducting polymers with ionic liquids is widely known to enhance their electrical properties [44].

The processing of electronically conductive gels involves the dispersion of a large amount of conductive material in a solvent. Conductive gels can be prepared by the simple process of mixing and casting monomers, conductors, and agents. The proposed gel composite with conductive materials, such as C materials (e.g. CNTs, graphenes) [7, 46–48], conductive polymers [43, 49], metallic materials [50–53], or a combination thereof [45, 54, 55] exhibit high elongation properties of 400%–1200%. However, these conductive gels have low conductivity (10^0–10^{-4} S cm^{-1}), and the conductivity changes sensitively to strain. Therefore, these conductive gels have been applied as strain sensors [7, 43, 45, 51] and bioelectrodes [45, 46] rather than as wiring materials.

To fabricate conductive gels with high electronic conductivities, a sufficient amount of conductive material must be dispersed to construct a percolation network. However, simply increasing the amount of conductive material is challenging because an increase in the conductive material causes mechanical property problems, such as degradation of the

Table 2. Comparison of composites with solid non-metal materials.

References	Material type	Base material	Fabrication process	Conductivity	Stretchability	Application
[4]	SWCNT	PDMS	Sheet Pasting	7–53 Ω sq^{-1}	50%	Wiring
[20]	MWCNT	PDMS	Mixing	0.175–0.341 kΩ	40%	Strain sensor
[5]	CB	Ecoflex	Mixing	5–10 kΩ	50%	Strain sensor
[22]	CB	Ecoflex	Mixing	Not mentioned	100%	Capacitive sensor electrodes
[23]	CB and CNT	Ecoflex	Mixing	150 Ω sq^{-1}	200%	Current collector in a stretchable lithium-ion battery
[24]	CB and CNT	Methylvinylsiloxane silicone rubber	Mixing	248.8 S m^{-1}	211%	Strain sensor
[6]	Graphene	NR and SBR	Mixing	1.04×10^{-5} S m^{-1}	100%–350%	Strain sensor
[31]	GO, rGO, G-NPLs	SEBS	Mixing	1^{-11}–1^{-5} S m^{-1}	10%	Strain sensor
[35]	PEDOT:PSS and MWCNT	PVA	Mixing	2–5 MΩ	50%	Strain sensor
[36]	PEDOT:PSS	PEO	Mixing	10×10^3–50×10^3 S m^{-1}	40%	LED device electrode
[37]	Polyaniline	Poly(vinyl acetate) latex	Mixing	2.676 S m^{-1}	5%	Strain sensor
[38]	Polyaniline and PA	PAA	Mixing	0.12 S cm^{-1}	460%	Strain sensor, pressure sensor

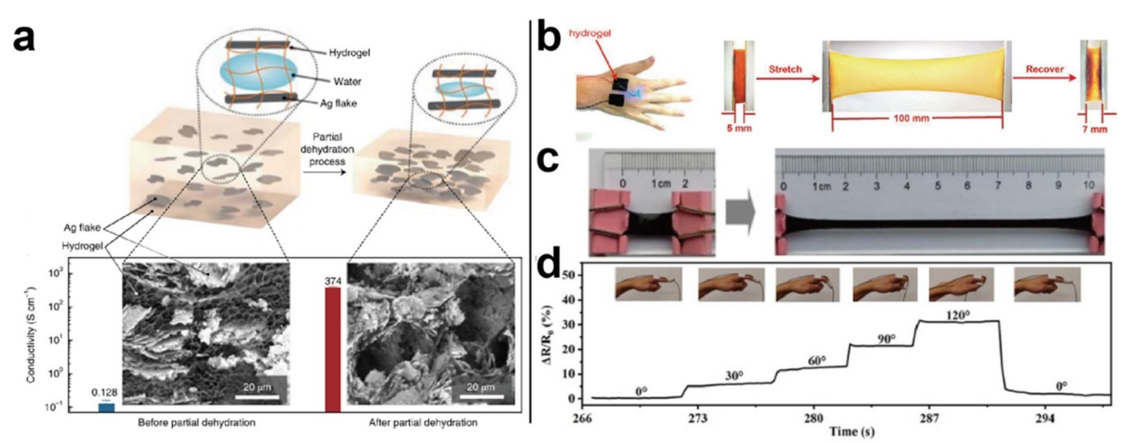

Figure 10. Liquid–solid hybrid gel composite: (a) polyacrylamide-silver flake composite with high conductivity by increasing conductor density by post-dehydration. Reproduced from [42], with permission from Springer Nature. (b) Conductive polymer gel with improved elasticity owing to conductive polymer-PNIPAAm double network structure [45]. John Wiley & Sons. © 2019 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (c) Composites with CNTs as conductors [7]. (d) Bending sensing using CNT gel composites. Reprinted from [7], Copyright 2019, with permission from Elsevier.

gel, dispersion failure owing to self-aggregation of the conductive material [56], and inhibition of gel cross-link formation owing to excessive dispersion of the material. On the other hand, there are conductive gels that solve these problems and achieve high conductivity. By partially dehydrating a gel containing dispersed silver flakes, a hydrogel with an elongation of up to 250% and conductivity as high as 374 S cm^{-1} has been proposed [42]. In this study, the gel was dehydrated following composite fabrication to increase the conductor concentration and obtain a percolation connection to suppress the inhibition of gel formation owing to the increase in conductive materials.

Another method to form an electron-conductive gel involves the formation of a matrix from a conductive material (figure 11). In this method, conductive polymers, such as PEDOT:PSS [57] and PANI [58], were used as the matrix. Pure conductive polymer hydrogels composed of PEDOT:PSS have been reported [57], exhibiting a conductivity of 40 S cm^{-1}. The plasticization and secondary doping

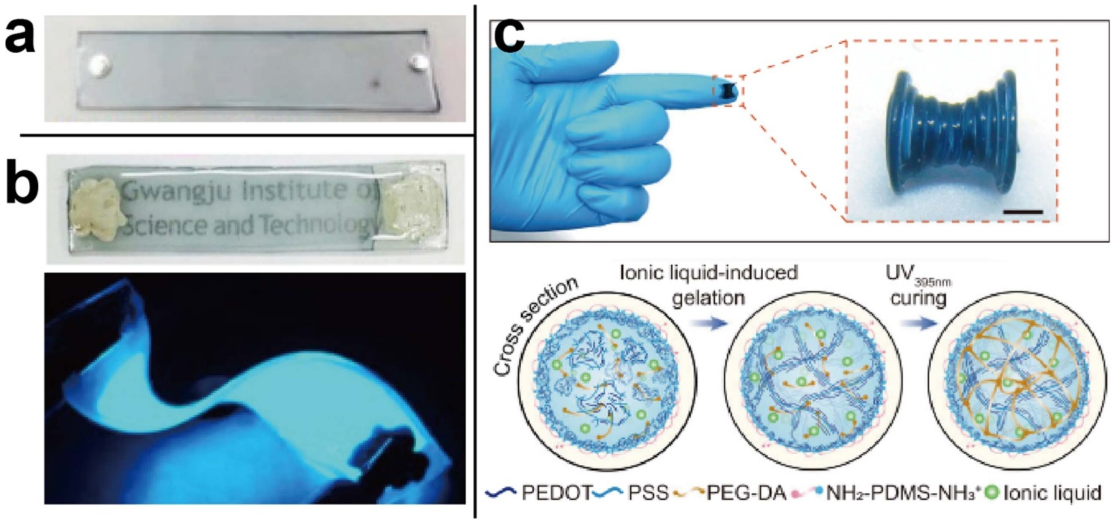

Figure 11. Conductive gel composites formed by conductive matrices. (a) PEDOT:PSS-Ionic-liquid-fluorosulfactant mixed conductive composite on PDMS substrate. [59] John Wiley & Sons. © 2020 Wiley-VCH GmbH. (b) Electroluminescent device using PEDOT:PSS-Ionic liquid transparent conductive gel as electrode. Reprinted with permission from [60]. Copyright 2017 American Chemical Society. (c) Conductive gel using PEDOT:PSS-PEG dual network. Reproduced from [62]. CC BY 4.0.

of PEDOT:PSS networks with ionic liquids have also been used to produce gels with conductivities ranging from 600–1200 S cm^{-1} [59, 60]. However, gels comprising solely conductive polymers exhibit high stiffness of up to several MPa and low tensile resistance of up to 50% [57, 59, 60]. Therefore, gels with dual-network structures incorporating conductive polymers and networks of stretchable materials, such as poly (N-isopropylacrylamide) (PNIPAAm) [58], polyvinyl alcohol (PVA) [61], and polyethylene glycol (PEG) [62], have been widely developed. In these gels, the introduction of nonconductive networks reduced the conductivity to approximately 10–10^{-2} S cm^{-1}. On the other hand, mechanical properties have been greatly improved, achieving elongation exceeding 100% in addition to stiffness of less than a few hundred kPa, which is equivalent to that of dispersed conductive gels.

Conductive composites, wherein conductive solids are supported by gel materials, have been extensively studied; however, they are still plagued by multiple challenges. The tradeoff between conductivity and deformation performance is the most important issue for improving the performance of composite materials. In particular, hydrogels have limitations such as changes in properties owing to solvent volatilization and voltage limitations owing to their narrow potential windows. In addition, the complete elimination of the influence of ionic conduction in hydrogels and ion gels is completely challenging. In general, the performance of conductive gel composites as pure electron conductors is inferior to that of solid composites. The use of these materials requires applications that can adapt to the unique properties of their liquid content, such as their performance as mixed-ion electron conductors and very high elongation performance.

Conductive gels exhibit both solid and liquid properties and have garnered attention for their low Young's modulus and high elasticity. However, they are primarily valued for their ionic conductivity, which poses challenges for electronic conductivity. For instance, percolation issues can affect conductivity, but gel network composites made from conductive polymers offer exceptionally high electronic conductivity at the expense of elasticity compared to conventional dielectric gels. Future developments are anticipated to leverage the unique properties of gels, such as creating ion-electron conductors that combine their ionic and electronic conductivity and developing implantable electrodes that utilize the gels' liquid properties.

4.2. Liquid metal

Liquid metals, which remain in a liquid state at room temperature, are the only liquid materials currently in practical use that exhibit electrical conductivity through electronic conduction. Among these, Ga-alloy-based liquid metals like EGaIn and Galinstan are widely employed in stretchable electronics due to their high biological safety and extremely low vapor pressure. Their ability to combine electronic conductivity with the inherent deformability of liquids allows composite materials to overcome the trade-off between conductivity and flexibility.

Liquid metal composites are attracting attention as materials that combine high electrical conductivity and elasticity despite the challenge of requiring an activation process. During the preparation of the composite materials, a strong mixing force is applied using ultrasonic waves or shear mixing to disperse the liquid metal throughout the polymer. The surface of the dispersed liquid metal was immediately covered with a gallium oxide film, which inhibited the electronic conduction between the liquid metal particles; thus, a mechanical activation process was generally required for the liquid metal composite to become conductive. The magnitude of force required for the mechanical activation process is influenced by the liquid metal content and particle size within the composite (figure 12). Therefore, precise control of these properties can facilitate the fabrication of composites with desirable localized electrical characteristics [63]. In an activated composite, liquid metal particles connect with each other to form conductive pathways, thus yielding high electrical conductivity and resistance to deformation [39–41, 64–66]. The SBS-matrix composite exhibited a conductivity of 12 000 S cm^{-1} following mechanical activation. The composite maintained a conductivity of 2000 S cm^{-1} after 500% elongation and also exhibited self-healing properties [40]. The PDMS composite matrix [8] exhibited a conductivity of 0.7 Ω cm^{-1} following activation by local compression or delamination.

Mechanical activation can cause undesirable damage to the composite materials and substrates, which can complicate the manufacturing process. Therefore, composite materials that exhibit conductivity without additional activation processes are under development (figure 13). For example, a process has been reported to simultaneously cure and activate polymers through self-activation using contractile forces associated with solvent volatilization. A conductivity of 2179 S cm^{-1} was reported for this composite without additional activation [67]. Another study reported conductivity without mechanical activation using precipitation to control the density of the liquid metal inside the composite [68].

Combining liquid metals with other conductive materials can improve the electrical properties of the composite materials. Mixed composites of graphene [69], Ag [70–72], Fe [73], and Ni powders [74] with liquid metals and polymers have been reported to achieve highly conductive wiring that does not require sintering, conductive property control using magnetic filler orientation, or recyclable stretchable wiring. Gels composed by mixing ionic liquids, liquid metals, and Ni powder [75] exhibited its performance as mixed conductors with both the high ionic conductivity (2.1 mS cm^{-1}) characteristic of ionic liquids and the electronic conductivity (25 S cm^{-1}) from metal. Composites of liquid metal and solid metal particles offset each other with the disadvantages of solid materials, such as low conductivity at large elongations and the need for an activation process, which is a disadvantage of liquid metals.

The following table 3 compares the performance of conductive composites that incorporate the liquid materials discussed in this section.

Liquid metal composites are gaining attention because they do not compromise the stretchability of the base material, offering both high elasticity and electrical conductivity. However, conventional liquid metal composites require post-activation to establish connections between liquid metal particles, and their processable resolution is limited by particle diameter, resulting in lower conductivity and resolution compared to pure liquid metal wiring. Conversely, composites do not need post-filling of liquid metal and can be printed in three dimensions—an option not feasible with pure liquid metal—facilitating applications such as large-area 3D interconnects that leverage these properties.

5. Outlook and current challenges

This paper summarized composite materials that exhibit high stretchability and electrical conductivity. The mechanical and electrical properties of composite materials can be varied by varying the filler material. Composite materials using various filler materials have been proposed and are broadly classified as solid metals, solid nonmetals, and non-solid materials. Depending on the type of metal, solid metals can provide composite materials with only electrical conductivity and also secondary functions such as thermal conductivity and magnetization. Composites with solid non-metallic materials, mainly C, are expected to be applied as sensors because of their high sensitivity to deformation. In addition, the high transparency of composites owing to the miniaturization of fillers and their low percolation thresholds have garnered attention. Composite materials using liquid materials can withstand greater deformation than those filled with solid fillers owing to the extremely high deformation capacity of the liquid. Moreover, they have a specific self-healing function even if broken. These functionalities can be achieved by optimizing the filler material. Composite materials with high conductivity and elasticity are indispensable for the electrical driving of devices with large deformations, such as wearable devices and soft robots.

However, composite materials are yet to overcome the following challenges. (i) There is a trade-off between the filler material and the stretchability of the composite material itself. To maintain conductivity, the filler materials must be filled above the percolation threshold. This increases the hardness of the composite, reduces its stretchability, interferes with the bonding of the base polymer, and reduces its strength. This hinders the application of

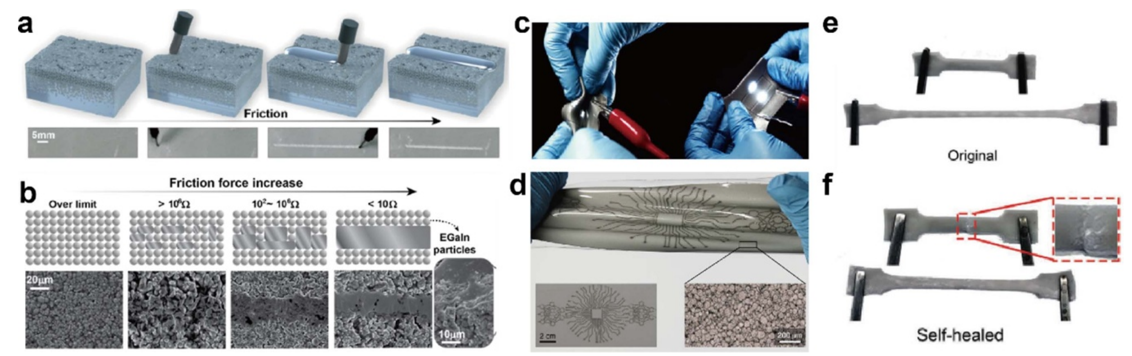

Figure 12. Liquid metal conductive composites that develop conductivity by mechanical sintering. (a) Schematic of mechanical sintering [65]. (b) Formation of conductive paths between liquid metal micro-particles during mechanical sintering. Reprinted with permission from [65]. Copyright 2019 American Chemical Society. (c) Liquid metal composite maintaining conductivity under large deformation. Reproduced from [39]. CC BY 4.0. (d) Liquid metal composite with complex conductive patterns formed by local pressure application. Reproduced from [41], with permission from Springer Nature. (e) and (f) composites that maintain elongation properties after recovery from amputation owing to self-healing properties [40]. John Wiley & Sons. © 2020 Wiley-VCH GmbH.

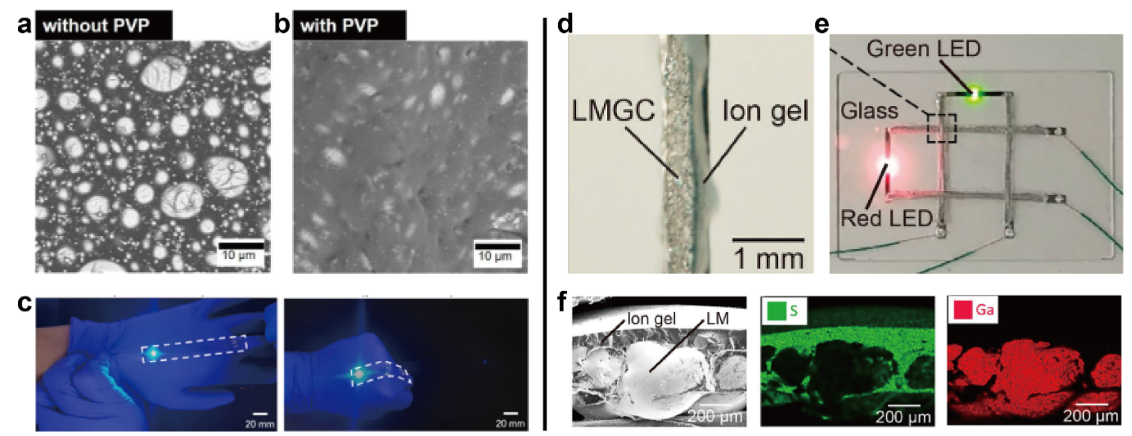

Figure 13. Liquid metal composites that develop conductivity without sintering. (a), (b) Surface SEM images with and without self-activation of conductive composite [67]. This composite is made of liquid metal and PDMS by a chemical self-activation process using PVP. (c). Demonstration of conductivity during deformation. Reprinted with permission from [67]. Copyright 2022 American Chemical Society. (d)–(f) Conductive liquid metal-ion gel composite with distribution control by precipitation of liquid metal [68]. (d) Composite with two layers of insulating ion-gel layer and conductive liquid metal, (e). 3D cross wiring utilizing insulating ion-gel layer, (f). Cross-sectional SEM-EDX image of a two-layer structure. Reproduced from [68]. CC BY 4.0.

Table 3. Performance comparison of conductive composites which contain liquid materials.

References	Conductive material	Base material	Fabrication process	Conductivity	Stretchability
[7]	CNT	PAAm hydrogel	Casting	0.067 S m^{-1}	700%
[42]	Silver flake	PAAm-Alginate hydrogel	Stencil printing	374 S cm^{-1}	250%
[60]	PEDOT:PSS/ Ionic liquid	PEDOT:PSS	Spin coating	1000 S cm^{-1}	180% (on prestretched PDMS) 28% (composite only)
[62]	PEDOT:PSS	PEDOT:PSS-PEGDA-amin terminated PDMS hydrogel	Liquid-in-Liquid 3D printing	164 S m^{-1}	200%
[8]	eGaIn	PDMS	Injection	0.7 Ω cm^{-1}	215%
[41]	eGaIn	PDMS	Casting	1.37×10^3 S cm^{-1}	50%
[64]	eGaIn	PU	Stencil Printing	22 532 S cm^{-1}	2260%
[72]	eGaIn/Ag flake	EVA	3D printing	8331 S cm^{-1}	1000%

composites in devices that require large deformations while maintaining sufficient conductivity. (ii) Filler bonding within the composite material occurs randomly, resulting in poor reproducibility during repeated deformation. As the filler material flowed within the base material, multiple cycles of tension and contraction resulted in gradual changes in the overall electrical properties. This large hysteresis poses a significant challenge to the stable operation of conductive composite materials.

Data availability statement

No new data were created or analysed in this study.

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Conflict of interest

The authors declare no conflict of interest.

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