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Editorial

Innovative self-healing conductive organogel: Pioneering the future of electronics

Self-healing hydrogels utilize inherent intermolecular forces to autonomously heal physical damage resulting from excessive strain, pressure, or tearing. Applying these materials in soft robotics and tissue engineering could be beneficial. On the other hand, their efficacy in stretchable and mechanically resistant circuits is hindered by their limited electrical conductivity. In a recent publication in *Nature Electronic*, Majidi and his team [1] synthesized novel organogel, which is composed of silver micro flakes and gallium (Ga)-based liquid metal (LM) alloy droplets within a poly(vinyl alcohol)-sodium borate (PVA-Borax) gel and having self-healing and conduct electricity properties. This composite exhibits impressive self-healing capabilities and a 7×10^4 S/m electrical conductivity. Its performance is better than that of other organogels and soft conductive hydrogels. The composite material has excellent mechanical properties, including a low Young's modulus (~20 kPa) and outstanding stretchability (with a strain limit of 400%), which can be attributed to its high liquid content. Ethylene glycol (EG) as a solvent prevents drying, ensuring that the weight of the composite remains constant when exposed to room temperature and humidity. The organized composite has been tested for various applications, including bioelectrodes for electromyography (EMG) sensing, reconfigurable soft circuits, and a connective strip in a soft robot.

Flexible, stretchable, and electrically conductive self-healing materials can replicate the mechanical properties of natural biological tissue. These materials have significant potential for soft electronics, robotics, and medical devices [2]. Self-healing conductive organogels represent a promising class of materials for applications in flexible electronics, soft robotics, and wearable devices. The conductivity and self-healing ability combination enables prolonged device lifetimes, improved reliability, and reduced maintenance. Future research could focus on enhancing mechanical properties, conductivity, and healing efficiency while ensuring biocompatibility and scalability for broader practical applications. Self-healing hydrogels offer high mechanical flexibility (up to 2000%), low stiffness, recyclability, and biocompatibility, making them a promising biomaterial soft tissue. For example, an ionic gel rich in fluorine has been designed to withstand strains up to 2000% and quickly achieve mechanical and electrical self-healing in various aqueous conditions using ion-dipole interactions. Meanwhile, a polymeric organogel composed of acrylate has exhibited strong adhesion and exceptional optical transparency. Despite their low mechanical properties, rapid self-healing capabilities, and good stretchability, these materials lack the electrical conductivity needed for power electronics and soft digital circuits. To solve this problem, hydrogels were augmented with conductive fillers such as graphene, carbon nanotubes, metallic micro/nanoparticles, and conductive polymers. The development of electrically percolating channels can weaken the mechanical properties of these composites by increasing the elastic modulus, reducing the strain limit, and producing loading/unloading hysteresis [3]. This is typically achieved by utilizing a large volume fraction of conductive particles. A novel partial dehydration technique was recently designed to produce composites that preserve the hydrogel's mechanical properties and show excellent electrical conductivity. A polyacrylamide–alginate matrix generates a percolating network by incorporating Ag particles at a low concentration (~6 vol%) and eliminating some water from the hydrogel scaffold. The resultant composite has a strain limit of 250%, a low elastic modulus of 10 kPa, and an electrical conductivity of 350 S/cm. However, one of the most important disadvantages of partially dehydratable Ag-hydrogel composites is that they do not exhibit mechanical, electrical, and self-healing properties. This finding can be attributed to the choice of the matrix of hydrogel material, which cannot repair itself by spontaneous ion-dipole bonding, and the use of rigid conductive filler particles that cannot merge or hydrate upon reestablishing contact.

Majidi and his team reported a novel composite material called organogel, which is electrically conductive and self-healing properties. This composite was composed of a gel made of polyvinyl alcohol and sodium borate (PVA-Borax), which contains a network of silver micro flakes and droplets of Ga-based LM, as shown in Fig. 1. The material shows a high level of conductivity, measuring at 7×10^4 S/m, and can quickly repair itself. Due to its high liquid content, the material possesses considerable stretchability (>400% strain) and a low Young's modulus (~20 kPa). Using ethylene glycol as a solvent inhibits drying and ensures that the properties remain constant for >24 h The results demonstrate the capabilities of the organogel composite by utilizing it as a connecting layer between the battery and motor of a detached soft robot inspired by snails, a programmable soft circuit, and a bioelectrode for EMG sensing.

Electrolyte elastomers have been developed due to recent developments in integrating Ga-based LM alloys into hydrogels and soft polymers [4]. These alloys include eutectic gallium-indium (EGaIn) and gallium-indium-tin (Galinstan). Examples of hydrogel matrices used to highlight these composites include polyacrylamide, poly(ethylene glycol) diacrylate, poly(*N*-isopropyl acrylamide), polyacrylic acid, polyacrylamide, biopolymers, and PVA.

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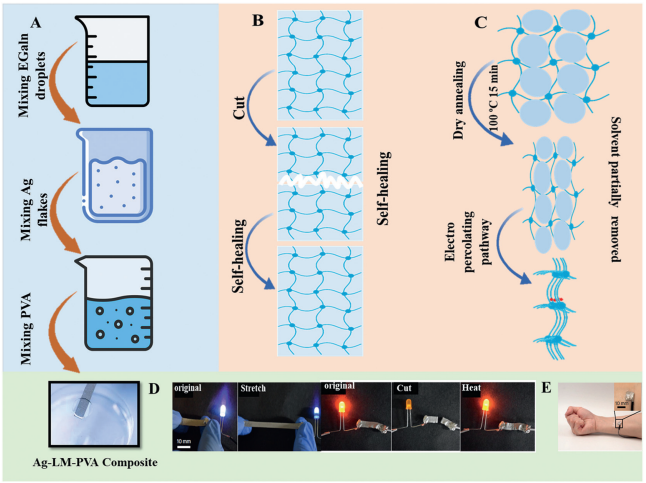

Fig. 1. (A) Synthesis of the Ag-LM-PVA organogel composite. (B) Self-healing mechanism and (C) dry annealing. (D) Demonstration of composite stretchability, self-healing and electrical conductivity. (E) The organogel composite is applied as an electrode for EMG.

However, many systems lack essential features such as self-healing capacity, hydrogel-like compliance, and high electrical conductivity. Majidi *et al.* integrated the partially dehydrated Ag-hydrogel and LM-hydrogel structures with a nontoxic, biocompatible, and self-healing PVA-Borax formulation to meet all criteria. The PVA-Borax gel's hydrogen bonding makes it highly viscoelastic and easily deformable. The gel exhibits a high level of viscoelasticity and can be easily deformed due to its crosslinking *via* hydrogen bonding. Utilizing a freeze-thaw technique improves the mechanical properties of the PVA-Borax gel, specifically its viscoelasticity. EG was used instead of water as a solvent to slow dehydration. The composite achieves high electrical conductivity using a dry annealing technique, including Ag micro flakes with EGaIn microdroplets. The results showed that the damaged composites could restore electrical conductivity and strain limits of up to 95% and 96%, respectively.

Furthermore, Majidi *et al.* synthesized and explored the mechanical property of the Ag-LM-PVA organogel composite by first combining EGaIn with an EG-based Borax solution to make LM microdroplets, then adding Ag micro flakes and an EG-based PVA solution, and finally mixing at high temperatures to gel. Following molding and dry annealing, a freeze-thaw procedure was used to improve the composite's rheological properties, yielding a highly viscoelastic material with solid-like qualities. The Ag-LM-PVA composite must exhibit a high degree of flexibility and be readily bent to be suitable for use with soft biological tissues. The strain-stress behavior of the biomaterial was affected by the duration of freezing. Longer freezing times result in more physical entanglements, increasing the tensile modulus (17.1 kPa to 42.3 kPa) but decreasing the stretchability (439.9% to 319.8%). Despite metal fillers, the composite exhibits high flexibility and minimal elasticity (~23 vol%). Cyclic performance tests reveal typical hysteresis loops, which suggest that the physical connection of hydrogen bonds in the gel was disrupted during repetitive loading and unloading cycling. The composite's ability to sustain multiple loading cycles with observed hysteresis loops suggests that it can withstand repeated use without significant degradation. It is suitable for long-term applications in soft robotics, wearable electronics, and other fields where repeated mechanical stress is common.

The Ag-LM-PVA composite initially has low ionic conductivity (0.082 S/m) due to Borax and metal fillers but lacks efficient electrical conduction channels. To overcome this limitation, Majidi *et al.* reported a dry annealing technique at 100 °C used to remove EG partially, reducing the gap between Ag flakes and EGaIn droplets and resulting in continuous conductive networks. The technique greatly increases electrical conductivity to 7.3×10^4 S/m. However, the EGaIn droplets were coated with an oxide layer, which inhibits their ability to interact with Ag flakes. The conductivity of the composite remains consistent for more than 24 h, exhibiting a change of <3%. This is due to the slow evaporation of EG at 25 °C, which effectively maintains the position of the conductive fillers. This is in contrast to water-based hydrogels, which experience a drop in conductivity over time because of water evaporation, which maintains stability.

Additionally, the polymer matrix was softer after the dry annealing process, which permits the metal particles to settle more readily, especially in thicker samples. This settling results in a decreased concentration of metal fillers near the top surface. Additionally, the composite's electrical conductivity decreases from the top to the bottom as EG evaporates more rapidly from the surface than from the center or bottom. Despite this, the composite has an average volumetric conductivity. Differences in conductivity readings might occur when sample thickness varies (0.5 mm *vs.* 1.5 mm). Thicker samples may exhibit more apparent metal filler settling and uneven EG evaporation, impacting the sample's electrical property homogeneity.

Majidi reported electromechanical coupling, in which a strip of Ag-LM PVA composite's electrical resistance increases when the composite stretches. Additional research is necessary to gain a comprehensive understanding of electromechanical behavior. However, they attribute it to the limited volume of conductive particles and the sparse percolating network. Surprisingly, a minor drop in resistance was found with small strains (0%–5%), most likely due to the rupture of the EGaIn oxide shell. This causes Ag and indium to react, producing a more conductive Ag-In alloy. The material must remain electrically conductive under repeated deformation for many applications. Resistance increased by 228% after 10 cycles and 700% after 405 cycles because of irreversible elongation and electrical route degradation. Despite this, the conductivity after 405 cycles was 8.5×10^3 S/m, which is suitable for digital circuits.

Majidi investigated the electrical self-healing and mechanical properties of the Ag-LM-PVA organogel composite. Due to its flexible and reversible hydrogen bonding, the PVA-Borax organogel matrix enables rapid and efficient self-repair (up to 100%). The results demonstrated the strain and stress properties of both pristine and healed samples after 5 min at room temperature. The freeze-thaw procedure considerably affects self-healing efficiency. Regarding mechanical self-healing efficiency of frozen samples for ten minutes had the highest level (96.4%). Additionally, extended freezing times decreased efficiency (42.0% for 20 min and 6.3% for 30 min) due to increasing physical entanglement, which impedes the formation of hydrogen bonds. The finest self-healing results were obtained without freeze-thaw therapy.

The Ag-LM-PVA composite achieves electrical conductivity by establishing connections between Ag micro flakes and EGaIn microdroplets. When the material is cut and reconnected, these particles restore the electrical network, allowing for fast self-healing. After three cycles, the electrical self-healing efficiency averages 86%. Proper alignment of the split surfaces was critical for optimum efficiency since misalignment lowers conductivity. However, it has lesser electromechanical coupling than the clean composite. When activated EGaIn stretched, it bridges particle cracks while preserving significant electromechanical connection.

To show the potential of organogel, Majidi *et al.* developed Ag-LM-PVA composite having many applications in soft-matter robotics and electronics that highlighted its mechanical compliance, self-healing, material reconfigurability, and high electrical conductivity. One example is an untethered crawling robot inspired by snails, which is not connected to any other device and

is equipped with an electrical motor and internal battery enclosed in a flexible silicone covering [5]. The motor was linked to the battery using an Ag-LM-PVA strip that can be repaired when cut. At first, the robot moves at a velocity of 4.19 mm per second. However, when the strip was halfway cut, the speed decreased by >50% to a value below 2 mm per second. After manually reconnecting the strip, the robot regains 68% of its original speed.

Furthermore, the Ag-LM-PVA strips were reconfigurable. The results demonstrated the Ag-LM-PVA organogel composite as a versatile bioelectrode for EMG. The device provided precise measurements with a favorable noise-to-signal ratio and could adapt its dimensions and shape. At first, it could reliably detect muscle activity on the forearm, but it was not large enough to detect action on the calf. Combining two electrodes increased signal quality. This composite improves the bioelectronics interface because of its conductivity, biocompatibility, and self-healing capabilities, enabling accurate muscle activity detection.

The Ag-LM-PVA composite demonstrates excellent electrical conductivity, measuring 7×10^4 S/m. Additionally, it possesses rapid self-healing capabilities, recovering 96% of its strain limit within 5 min and almost instantly restoring 95% of its conductivity. The material possesses Young's modulus of around 20 kPa, exhibits high stretchability with a strain capacity of 400%, and remains stable under normal environmental conditions. The mechanical self-healing of the composite is attributed to the reversible hydrogen bonds present in the PVA-Borax hydrogel. In contrast, LM coating Ag particles allows electrical self-healing, resulting in fast recovery.

The results illustrated the Ag-LM-PVA organogel's potential by building a snail-inspired crawling robot with self-healing interconnects that allowed for sustained functioning even after injury. The composite also functioned as a modular component in reconfigurable circuits as a bioelectrode for EMG sensing, recording impulses from several body sites. These examples demonstrate its potential uses in electronics, soft robotics, and healthcare.

Declaration of competing interest

The authors declare that any known conflicting financial interests or personal relationships could have appeared to influence the work reported in this study.

CRediT authorship contribution statement

Salim Ullah: Writing – review & editing, Writing – original draft, Data curation, Conceptualization. **Jianliang Shen:** Writing – review & editing, Supervision. **Hong-Tao Xu:** Writing – review & editing, Supervision.

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