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An electrically conductive silver–polyacrylamide–alginate hydrogel composite for soft electronics

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Hydrogels offer tissue-like compliance, stretchability, fracture toughness, ionic conductivity and compatibility with biological tissues. However, their electrical conductivity (<100 S cm^{-1}) is inadequate for digital circuits and applications in bioelectronics. Furthermore, efforts to increase conductivity by using hydrogel composites with conductive fillers have led to compromises in compliance and deformability. Here, we report a hydrogel composite that has a high electrical conductivity (>350 S cm^{-1}) and is capable of delivering direct current while maintaining soft compliance (Young's modulus < 1 kPa) and deformability. Micrometre-sized silver flakes are suspended in a polyacrylamide–alginate hydrogel matrix and, after going through a partial dehydration process, the flakes form percolating networks that are electrically conductive and robust to mechanical deformations. To illustrate the capabilities of our silver–hydrogel composite, we use the material in a stingray-inspired swimmer and a neuromuscular electrical stimulation electrode.

Soft electronics that exhibit high electrical conductivity and match the compliance of biological tissue are important in the development of wearable computing[1,2], soft sensors[3,4] and actuators[5], energy storage/generation devices[6,7] and stretchable displays[8,9]. A variety of material architectures have been used to create soft and stretchable electronics, including deterministic (such as wavy or serpentine) structures[10,11], soft microfluidic channels[12,13] and conductive composites or polymers[14–16]. However, these conductive materials have intrinsic limitations, such as relatively high Young's modulus (≫1 MPa in some cases) or limited deformability, and are not ideally suited for applications related to bioelectronic systems (such as those that require interfacing with biological tissues). Recently, researchers have demonstrated conductive elastomers with enhanced stretchability and compliance by incorporating microdroplets of liquid metal alloys such as eutectic gallium indium (EGaIn)[17,18]. In particular, a highly stretchable and conductive polymer composite has been developed using silver and EGaIn particles embedded in an ethylene vinyl acetate copolymer[18]. Although EGaIn-based polymer composites exhibit an encouraging combination of high conductivity, stretchability and compliance, they require a large volume fraction of metallic filler and their Young's modulus (~0.1–1 MPa) is greater than the modulus of soft gels and biological materials (roughly 1–10 kPa), such as adipose (body fat) tissue[19].

Hydrogels are a promising candidate for soft electronics since they have similar mechanical properties to a range of biological materials and soft tissues[20,21], including epidermal skin[22], brain[23], spinal cord[24] and cardiac tissue[25]. Recent research has highlighted various aspects of hydrogels, including high fracture toughness, tissue-like Young's modulus (<10^2 kPa), high water content (>75%), ionic conductivity, bioactivity and biocompatibility[21,26]. These properties enable unique applications in bioelectronics[27] and soft robotics[28], including soft-matter sensors[9,29] and actuators[30]. However, hydrogels have an intrinsic ionic conductivity (10^{-5} to 10^{-1} S cm^{-1}; refs. [31–33]) that is six to nine orders of magnitude lower than the conductivity of metals, and is inadequate for digital and power electronics[34].

To improve their electrical properties, hydrogel matrices have been filled with conductive materials such as metallic fillers (for example, nanowires or micro/nanoparticles)[35–38], carbon-based conductive materials (carbon nanotubes or graphene)[39,40] and intrinsically conducting polymers (for example, poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS) or polyaniline)[3,34,41,42]. These composites demonstrate the potential for engineering hydrogels that are both electrically conductive (~10^{-5}–10^{1} S cm^{-1}) and have tissue-like mechanical compliance. However, there is a trade-off between improved electrical conductivity and lowered compliance and deformability in these conductive hydrogel composites. For example, a pure PEDOT:PSS hydrogel[34] has been developed with electrical conductivity of 40 S cm^{-1} but high Young's modulus (~2 MPa) and low maximum strain limit (<35% strain), while a soft graphene hydrogel[40] has been synthesized with favourable mechanical properties (Young's modulus of 50 kPa) but low electrical conductivity (~10^{-4} S cm^{-1}).

In this Article, we report an electrically conductive hydrogel composite that has high electrical conductivity (374 S cm^{-1}), a low Young's modulus (<10 kPa) matching that of soft biomaterials, such as adipose tissue[19], and high stretchability (250% strain). We use a polyacrylamide (PAAm)–alginate hydrogel that is embedded with a low concentration of silver (Ag) flakes. Electrical conductivity is created via a partial dehydration process[34] in which a moderate portion of water is removed to induce percolation and create electrically conductive pathways (Fig. 1a,b). Because the composite has a low concentration of metallic filler, it exhibits only modest hysteresis between loading and unloading cycles. The Ag–hydrogel composite's high conductivity, low Young's modulus, high electrical stability and high stretchability make it a suitable material for applications in soft robotics, bioelectronics and wearable electronics (Fig. 1c, Supplementary Fig. 1 and Supplementary Table 1). We demonstrate the potential applications of this soft conductor by using it in a light-emitting diode circuit that shows high mechanical compliance (Fig. 1d and Supplementary Fig. 2), a stingray-inspired

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