



Highly structured solvents enable integrated organogel networks with robust adhesion and mechanics

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ABSTRACT

Simultaneous achievement of tissue-like softness, robust durability, and strong adhesion in bio-integrated electronics is impeded by the micro-phase separation and structural heterogeneity inherent in traditional multi-component gels. Herein, we propose a solvent-mediated integrated structure paradigm to resolve this bottleneck. Low-field nuclear magnetic resonance analysis reveals that glycerol exists in a highly structured solvent state within the network, exhibiting highly confined solvent behavior distinct from free-flowing solvents. By leveraging this highly structured nature, this strategy transforms the solvent from a passive dispersion medium into an integral structural component anchored via dense hydrogen bonding. This solvent-polymer integration eliminates discrete phases and creates a homogeneous energy landscape, effectively resolving the intrinsic stiffness-toughness trade-off to achieve rapid photopolymerization, exceptional stretchability (>2000%), and high modulus. Consequently, the PHEAA/Gly system demonstrates robust adhesion (up to 209.6 kPa). Crucially, this strategy is validated as a universal toolkit applicable to diverse acrylate monomers. Beyond mechanical performance, the integrated gel exhibits stable electrical insulation, cytocompatibility and conformal adhesion to biological tissues. Demonstrated through nondestructive device retrieval, ultra-stable interconnects under extreme strain (>900%), and multimodal bio-sensing, this work shifts the design focus from additive complexity to intrinsic solvent-structure engineering, offering a foundational framework for next-generation bio-integrated electronics.

1. Introduction

Flexible electronic devices represent a rapidly expanding frontier in wearable health monitoring, soft robotics, and human-machine interfaces [1–3]. Their seamless integration into daily life requires materials that can simultaneously deliver tissue-like softness, robust mechanical durability, and strong, conformal adhesion to diverse surfaces [4–6]. While conventional hydrogels offer biocompatibility, they are intrinsically plagued by dehydration issues and a classic trade-off between stiffness and toughness [7–12]. To overcome these

limitations, researchers have developed extensive multi-component design strategies [13,14]. Notable approaches include constructing interpenetrating polymer networks (IPNs) to enhance tensile strength [15,16], incorporating nanofillers to provide additional energy-dissipating mechanisms [17,18], and chemically grafting adhesive moieties to improve interfacial bonding [19,20]. Parallel developments employ organic co-solvents, such as ethylene glycol (EG), glycerol, or DMSO mixed with water to create organo-hydrogels aimed at functional adaptation to complex operating environments [21–28].

However, these additive strategies introduce significant complexity

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