A Structural Gel Composite Sensor Enabled Underwater Mechano-sensing with High Sensitivity

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Abstract. Developing gel-based electronics for various scenarios, such as underwater and open air, is a great challenge for traditional gel composite, due to its intrinsic weakness like uncontrollable swelling induced deformation, signal distortion caused by dehydration, large hysteresis in sensing signal, etc. In this work, we proposed a structural gel composite (SGC) approach by encapsulating the conductive hydrogel/MXene with a lipid gel (Lipogel) layer through an in-situ polymerization. The hydrophobic Lipogel coating endows the SGC with strong anti-swelling underwater and dehydration feature, and enables long-term ultra-stability (over 90 days) and durability (over 2000 cycles) for SGC for underwater mechanosensation. As a result, the SGC based mechanoreceptor demonstrates a high and stable sensitivity (GF of 14.5). Moreover, several SGC based conceptual sensors with high sensitivity are developed to unveil their profound potentials in underwater monitoring of human motions, waterproof anticounterfeiting application, and tactile trajectory tracking. This work may shed light on the development of gel electronics with robust and resilient sensing performance in aquatic environments.

Introduction

Recent research on responsive hydrogel-based technologies indicates their great potential in different applications like mechanosensation [1-3], soft robots [4-6], and energy storage [7-10]. Among them, the wearable hydrogel-based sensing device enable flexible mechanical properties with dynamic interactions and high sensing sensitivity. However, under aqueous conditions, traditional hydrogel-based sensing device can be compromised due to the swelling behavior of intrinsic hydrophilic molecular chains [11,12], resulting in irregular output signals and irreversible damage to the mechanical properties.

Herein, we describe a structural gel composite (SGC) strategy by creating a hydrophobic lipid gel (Lipogel) layer in situ on the hydrogel/MXene core. The obtained SGC contains a robust Lipogel/hydrogel interface to realize the stable mechanical performances and the integral hydrogel/MXene structure, where the Lipogel layer is anchored on the hydrogel surface with an adjustable thickness (up to 200 μm). The SGC presents ultra-stability for mechanical property both in air and underwater, reliable underwater sensing capability with high accuracy and faster responsiveness. A flexible SGC based mechanoreceptor is achieved with high sensitivity (GF of 14.5). We also demonstrate a series of SGC based sensors with capability to perform instantaneously and logical tactile sensing at underwater environment.

The synthesis process of SCG is shown in figure 1.

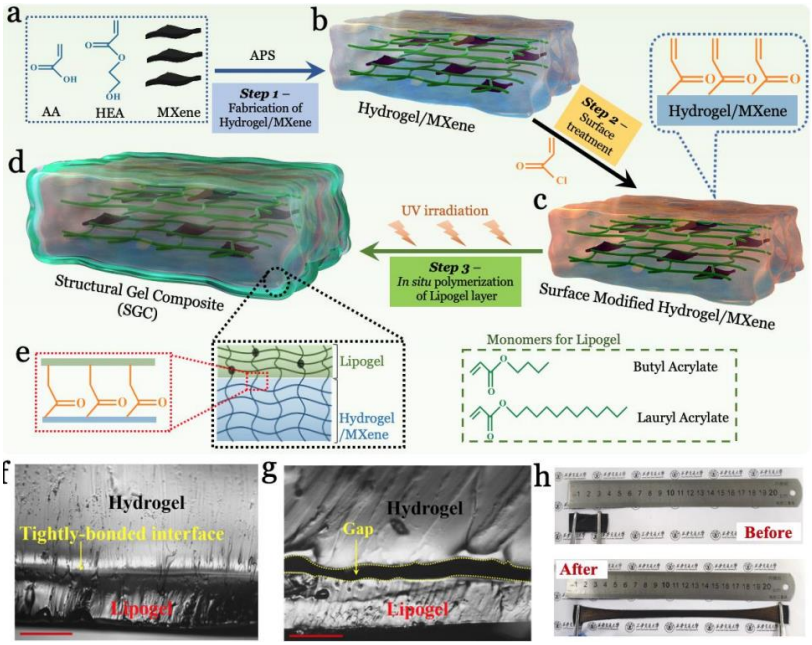

Fig. 1. Schematic illustration of the fabrication of structural gel composite. a) Structure of monomer for synthesizing PAHM hydrogel. b) Diagram of PAHM hydrogel. c) Diagram of surface modified PAHM hydrogel. d) Structures of monomer for synthesizing Structural gel composite. Diagrams of e) Structural gel composite and f) the Organo/Hydro interface.

Mechanical behaviour of the bulk hydrogel. The bulk hydrogel shows an excellent elongation (can be stretched up to 1000% strain). The optimal toughness with excellent stretchability can be achieved when the addition of MXene is 8wt%. The relation between fractured toughness and fractured strain and the ratio of different monomers is evaluated to reach a desired mechanical property. Additionally, the structural gel composite presents outstanding self-recovery capability and resilience. Benefited from the brilliant self-recovery capability and robust interfaces, the structural gel composite demonstrates good anti-fatigue and durable behavior based on the overlapping tensile loading-unloading curves from 2^{nd} to 20^{th} cycles.

Long-term stability underwater and air. The structure gel composite presents no noticeable volume change after immersing in water for 200 h. A quick tensile testing uncovers a long-term underwater mechanical stability for SGC by retaining over 80% of original elastic modulus, fracture strain, tensile strength and toughness after immersing in water for 200 h. Also, the SGC surface presents a WCA of $115.4° \pm 3.8°$ due to the hydrophobic nature of Lipogel layer. Interestingly, this hydrophobic Lipogel layer can effectively preserve the SGC in the open air from dehydration, where the SGC can sustain over 90% of its original weight after 200 h.

Long-term underwater adhesion. The SGC can adhere to different substrates both in the open air and underwter. The SGC even possesses stronger underwater adhesion on the hydrophobic substrates (PTFE, PE) than the hydrophilic ones (glass, copper, pigskin and rubber). Our SGC can retain stable adhesion strength even after 8 repeatedly pasting-peeling cycles both in the open air and underwater. Moreover, the adhesion capability has no noticeable decline whether storing in air or water for 5 hours, presenting a reliable long-term adhesion.

Stain-responsivenss of SGC flexible sensors. The electrical sensing outputs can be precisely perceived at either a low strain (0.2%-2%) or a high deformation (50%-500%). The SGC can accurately monitor and distinguish various frequency from 0.04 Hz to 0.32 Hz, and also presents prominent reversibility and good recovery during the stretching-holding procedure. Moreover, our SGC demonstrates a responsive time of 163.1 ms and a recovery time of 178.8 ms. The reliable adhesion of SGC also benefit the accuracy and responsiveness.

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