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Tough, Fatigue-Resistant Polyaniline Hydrogels with Multiple Reversible Physical Interactions for Wearable Sensors

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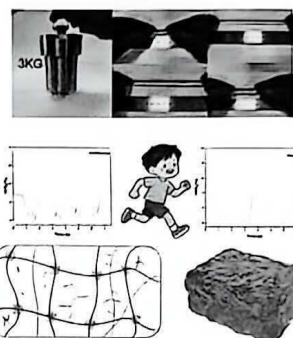
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ABSTRACT: Conductive hydrogels have emerged as promising candidates for stretchable and flexible electronic devices. However, simultaneously maintaining high electrical conductivity, sensitive strain responsiveness, and fatigue resistance under extreme deformation remains a major challenge. In this study, MnO_2 was embedded into the hydrogel matrix as an in situ oxidizing agent to initiate the in situ oxidative polymerization of aniline within the hydrogel network within 3 h. Lauryl methacrylate (LMA) and cetyltrimethylammonium bromide (CTAB) were introduced to cross-link the polyacrylic acid (PAA) hydrogel through hydrophobic association, while quaternized chitosan (QCS) was incorporated as a reinforcing component, thereby yielding the PALQMI hydrogel with integrated mechanical robustness, stretchability, and electrical conductivity. Through the rational regulation of multiple reversible physical interactions, including hydrogen bonding, electrostatic interactions, and metal–ion coordination, the optimized $\text{PALQ}_{0.5}\text{M}_{30}\text{I}_{0.5}$ hydrogel achieved a tensile strength of 1.62 MPa, an elongation at break of 2833.2%, and a toughness of 20.119 MJ m^{-3} . It also showed remarkable fatigue resistance during cyclic loading–unloading tests. The incorporation of polyaniline endowed the hydrogel with a high electrical conductivity of $161.49 \text{ mS cm}^{-1}$. The hydrogel detected a broad strain range of 10–1000% and exhibited a maximum gauge factor (GF) of 5.667 in the 600–1000% strain range. Moreover, PALQMI enabled clear and reproducible detection of human-motion-related strains and could function as a pressure sensor for encrypted information transmission. These results demonstrate the considerable potential of PALQMI for wearable strain sensors and flexible electronic devices

KEYWORDS: hydrophobic associative hydrogels, conductive polymers, polyaniline, flexible wearable sensors, flexible electronics



1. INTRODUCTION

Flexible wearable devices have attracted considerable attention in fields such as electronic skin and health monitoring because of their lightweight nature, adaptability to diverse human activities, real-time signal acquisition capability, and high monitoring efficiency.^{1–7} Owing to their three-dimensional (3D) polymeric networks, facile synthesis, favorable stretchability, tunable electrical conductivity, and good biocompatibility, hydrogels have been widely explored as promising materials for flexible wearable sensors.^{8–12} However, conventional hydrogels still suffer from insufficient electrical conductivity, mechanical degradation during long-term service, and irreversible deformation under repeated loading,^{13–16} highlighting the urgent need to develop advanced hydrogel-based sensing materials.

Hydrophobic association (HA) cross-linking has been recognized as an effective strategy for improving the toughness of hydrogels.^{17–20} In HA hydrogels, surfactant micelles facilitate the formation of physically entangled hydrophobic domains within the polymer network, thereby enhancing mechanical performance. Hydrophobic polymer chains can be entangled and coiled within micelles, and their reversible movement within individual micelles or between adjacent micelles enables effective energy dissipation, leading to a

substantial improvement in toughness.^{21–24} Nevertheless, HA networks alone are often insufficient to provide the combined mechanical robustness and electrical conductivity required for flexible wearable sensors. To further strengthen the hydrogel network, polysaccharide-based polymers have been widely introduced because of their good flexibility, mechanical tunability, biocompatibility, and abundant functional groups. These biomacromolecules, such as alginate,^{25,26} cellulose,^{27,28} and starch,^{29,30} can increase the internal network density of HA hydrogels and enhance intermolecular interactions. For example, Li et al.²⁷ incorporated HA interactions and cellulose nanofibers (CNFs) into a polyacrylamide/poly(vinyl alcohol) (PAM/PVA) double-network hydrogel. The CNFs served as bridges within the dual network and formed abundant hydrogen bonds with the polymer chains, thereby reinforcing the network structure and improving the mechanical properties of the hydrogel. Similarly, Jiang et al.³¹ developed a conductive

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