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Stretchable and magneto-sensitive strain sensor based on silver nanowire-polyurethane sponge enhanced magnetorheological elastomer

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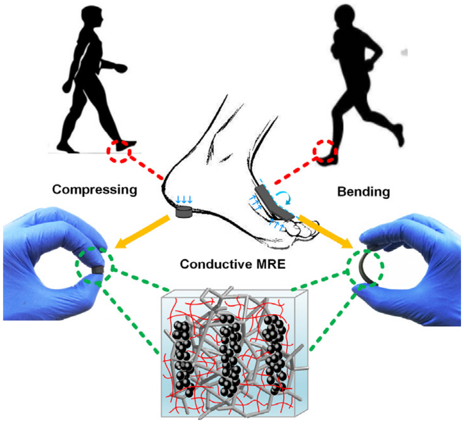
HIGHLIGHTS

- The conductive magnetorheological elastomer (MRE) can be applied as tensile, compressive and magneto-sensitive sensor.
- The stiffness of matrix and filler content significantly influence the mechanic-electric properties of conductive MRE.
- The resistance of conductive MRE increases 91.8% and 67.6% under the tensile and compressive loading.
- The relative resistance increment of conductive MRE reaches as high as 100% under 428mT magnetic field.

GRAPHICAL ABSTRACT



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ABSTRACT

This work reported a stretchable, compressible and magneto-sensitive strain sensor based on conductive magnetorheological elastomer (MRE) containing silver nanowires (AgNWs) dip-coated polyurethane sponge, carbonyl iron particles and polydimethylsiloxane (PDMS) matrix. The mechanic-electric-magnetic coupling properties of conductive MRE were investigated and they were significantly influenced by mechanical properties of PDMS matrix and the AgNWs content. By increasing curing agent weight ratio, the cross-linking density of the PDMS increased thus the magneto-induced modulus decreased. The MRE was able to be used in the strain sensors, because its relative resistance variation reached 91.8% and 67.6% when 20% tensile strain and 10% compressive strain were applied. By applying a 428 mT magnetic field, the relative resistance of MRE sensor increased to 200%. Based on the experimental results, a possible mechanism was proposed to investigate the mechanic-electric-magnetic coupling sensing characteristics. Finally, the MRE sensors can be integrated on a shoe to detect the foot motion, demonstrated this material was promising in the intelligent devices like artificial skin, composite electrodes and soft sensor.

1. Introduction

Due to the reversibly mechanical properties under magnetic field, magnetorheological elastomer (MRE) has attracted increasing

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applications in vibration control [1], radio absorbing [2] and suspension system [3,4]. It was found that the natural frequency of MRE based devices could be significantly changed by applying magnetic field. Usually, MRE is mainly consisting of two parts: polymer matrix and magnetic particles. During the curing process, the constituents are mixed homogeneously in the presence of external magnetic field. Thus, the particles form linear structures aligned in the magnetic field direction in the final MRE. Over the past decades, numerous efforts have been conducted to improve mechanical properties of MRE [5–8]. Nevertheless, the traditional MREs were unsuitable for novel electrical devices like sensors due to the poor conductivity.

In order to meet the requirement of novel electrical devices, conductive fillers such as graphene, graphene oxide, graphite, and metal particles were incorporated into polymer matrix to improve the conductivity of magnetorheological (MR) materials. Pang et al. developed a novel MR material enhanced by graphite and the enhanced magnetic field dependent conductivity provided a new way for magnetic field detection [9]. Bica et al. fabricated a magnetoresistive sensor based on graphene enhanced MRE and discussed the influence of transverse applied magnetic field and compression pressure on the resistance to prove the potential towards a strain sensor [10]. The developments in portable and foldable electrical devices have heightened the need for stretchable and flexible sensors [11–15]. Sensors integrated with MR materials could synchronously reflect the strength of stimulation applied to the devices by identifying the varied mechanical state of component, which was very important in practical application. So flexible and conductive sensors based on MR materials were fabricated.

The responsive of sensors under external stimulation like stretching and compressing is mostly dependent on the variation of electrical properties such as resistance or capacitance. Both sensitivity and stretchability should be taken into consideration to manufacture high performance sensors. To supersede the traditional rigid sensors, several stretchable and flexible sensors, for example, conductive additives deposited on flexible substrates [16–22], conductive networks or films [23–27] and other alternative fillers like conductive foams or sponges embedded in different substrates were manufactured [28–34]. Very recently, three dimensional (3D) structures enhanced flexible sensors have attracted great interests due to excellent conductivity and stretchability. After incorporating these 3D conductive structures into the elastomeric substrate such as polydimethylsiloxane (PDMS), flexible hybrids with interconnected conductive networks were achieved. Jun et al. prepared conductive graphene-PDMS composite by infiltrating 3D reduced graphene oxide foams with PDMS and it was found large graphene could significantly enhance conductivity of composite [35]. Ge et al. fabricated elastomeric and magnetic strain sensors with high conductivity and stretchability by incorporating the conductive carbon nanotubes coated polyurethane sponge (PUS) into MRE, the PUS/MRE was demonstrated to be applied as strain sensors due to the strengthening 3D topological scaffold and the stable strain dependent resistance [36]. However, these sensors would suffer from weak stretchability, low gauge factor and large hysteresis due to the brittleness and weak adhesion of conductive materials.

Because of the excellent electrical, mechanical and optical properties, silver nanowires (AgNWs) have attracted great attention for the potential application in sensors [37–43]. Amjadi et al. reported a sandwich-structured strain sensor with high stretchability based on AgNW-PDMS composite which could be integrated into a glove to detect finger motion [44]. Ge et al. fabricated a stretchable conductor based on PUS-AgNW-PDMS composite with novel interconnected binary networks, the excellent conductivity and stability enabled itself to be used for future electric devices [45]. So traditional MRE could be markedly enhanced by AgNW based 3D conductive structures and the final product will attract much attention in novel electric devices due to their excellent mechanical and electrical properties.

The previous work [36] mainly focused on the rheological properties and tensile sensing performance of MRE featuring carbon nanotubes dip-coated PUS. Meanwhile, this paper emphasized the enhancement of filler material content and curing agent ratio on rheological behaviors, and the potential of MRE as a multifunctional strain sensor. Not only tensile, but also compressive and magnetic sensing performance were investigated to understand the magnetic-mechanic-electric coupling behavior. In this paper, AgNW/PUS was immersed into carbonyl iron particles (CIPs) doped PDMS matrix to develop a novel conductive MRE. Strain sensors based on conductive MRE showed good electrical and mechanical properties because of the 3D interconnected AgNW networks coated PUS backbones and CIPs chain-like structures. The responsive of sensors under cyclic tensile, compressive and magnetic loading was investigated. Possible sensing mechanisms were proposed to discuss the influence factors on the sensing performance. Finally, a shoe integrated with MRE sensors was developed for the motion detection of foot.

2. Experimental section

2.1. Materials

The PDMS (type Sylgard 184) precursor and curing agent were purchased from Dow Corning GmbH, USA. CIPs (type CN) with an average diameter of 6 μm from BASF were used as magnetic particles. Polyvinylpyrrolidone (PVP), $AgNO_3$, glycerol, ethanol and NaCl were from Sinopharm Chemical Reagent Co., Ltd. Deionized water was self-prepared. Commercially available PUSs were used as the 3D scaffold.

2.2. Preparation

Silver nanowires (AgNWs): 5.86 g PVP was added into 190 mL glycerol, then the mixture was heated to 90 °C and cooled to 50 °C under stirring (~100 rpm) to make PVP fully dissolved. Afterward, 1.58 g $AgNO_3$ and a solution containing 58 mg NaCl, 0.5 mL deionized water and 10 mL glycerol were added and heated up to 210 °C in 20 min under slow stirring (~50 rpm). Ultimately, the heater was moved away and the gray-green solution in flask was transferred to a beaker. After a week's stably stratification, a layer of sediment containing AgNWs at the bottom of beaker was collected. The obtained AgNWs were washed by deionized water, then the solution was centrifuged under 7000 rpm for 15 min, the upper layer of solution was poured out and the sediment was washed again.

Conductive PUSs: Porous PUSs were cut into small pieces with the dimension of 20 mm in diameter and 1 mm in thickness for rheological tests, 2 mm in thickness and width by length of 10×50 mm^2 for tensile tests and 10 mm in diameter and 5 mm in thickness for compressive tests. The pieces were all cleaned by deionized water twice, immersed in acetone for 2 h, and dried at 80 °C for 2 h. Then, the pieces were immersed into the ethanol solution of AgNWs (~1.5 mg/mL), moved into oven, and dried at 80°C for 4 h. By repeating the dip-coating procedure for 3, 4, 5 and 6 times, PUSs with different conductivity (marked as 3rd, 4th, 5th and 6th dip-coated conveniently) were obtained. Copper wires were adhered to both the ends of conductive PUSs for tensile tests longitudinally, as well as the top and bottom of conductive PUSs for compressive tests by silver conductive paint to test the electrical properties (Fig. 1).

Conductive MREs: At first, magnetorheological (MR) precursor with different curing agent weight ratio (1:10, 1:20, 1:25 and 1:30) of PDMS matrix was prepared. The CIPs, PDMS matrix and curing agent were mixed in a beaker for 10 min. Then the beaker was transferred into ultrasonic machine which was kept at 80% power for 10 min to further separate. The weight fraction of CIPs was 60 wt% for all MR precursor. Conductive PUS pieces were immersed into MR precursor, then transferred into a vacuum oven and treated for