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Full paper

Bionic intelligent hydrogel actuators with multimodal deformation and locomotion

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ABSTRACT

Inspired by common patterns of biological movement, a series of near-infrared (NIR) laser-driven bionic intelligent hydrogel actuators were successfully fabricated via the combination of infiltration method (entirety infiltration and locality infiltration), structure design and material characteristics. The controllable distribution of graphene oxide (GO) particles in a hydrogel matrix resulting from the infiltration method led to abundant bionic deformations with multiple degrees of freedom and flexibility. In addition to the highest response speed of 187.7°/s, the hydrogel actuators underwent accurate and repeatable static deformations and efficient and stabilized dynamic locomotion, demonstrated, for example, through the assembly of origami, the deformation of fingers and palm, the closing and blooming of a chrysanthemum and the crawling of an inchworm. This study opens a new path for practical bionic applications of NIR-triggered hydrogel actuators in self-assembly biosensor, targeted drug delivery and minimally invasive surgery.

1. Introduction

With the development of intelligent soft materials, the design and fabrication of soft actuators that can realize bionic movements, including swimming, jumping, capturing and crawling [1–4], have received growing research interest. Compared with traditional actuators, which have complicated mechanical structures, soft actuators can achieve complex movements with relatively simple and effective structures due to the intelligent response of their soft material components. Under external stimulations such as temperature [5], light [6], electricity [7] and humidity [8], soft materials can perform intelligent mechanical deformations, which makes them ideal materials for the design and fabrication of soft actuators, soft robots and artificial muscles [9–16]. As one of the most widely used soft materials, hydrogel has been applied in medical science [17–20], soft electronics [21] and soft robotics [22–25] due to its biocompatibility and intelligent water absorption. The swelling and deswelling behaviours initiated by such external stimulations are the basis of hydrogel actuation. Due to the advantages of non-invasive safety [26] and non-contact control [1,6], near-infrared (NIR) laser-driven hydrogel actuators can be easily controlled with higher spatial and temporal resolution than other stimuli [27,28] and have attracted much attention for the design and fabrication of novel light-driven hydrogel actuators. Carbon nanomaterials, including graphene oxide (GO) [29–31], with the remarkable ability to undergo photothermal energy transformations have been considered as candidate materials for light-driven hydrogel actuators. Through a forced resonance vibration process, NIR laser interacts with GO and increases the corresponding disordering degree, and converts kinetic vibrational energy to heat [29]. Therefore, many kinds of hydrogel actuators based on GO [32–34] have been fabricated and exhibited excellent performance. However, as a result of the crosslinking patterns and the layering methods, the existing hydrogel actuators normally exhibit slow response to stimuli, and cannot produce rapid continuous movement based on their reciprocating swelling/deswelling property. In our previous studies [35], we fabricated a NIR-triggered hydrogel that was suitable for moulding and three-dimensional printing. Although the layered and anisotropic structure of this hydrogel greatly facilitated its shape morphing, its performance was limited to simple bending/unbending deformation with low response speed.

Modulation of the hydrogel pore structures is essential for tailoring their mechanical strength, response speed, and motion behaviour [27]. Hydrogel actuators with a bilayer structure [30,35] composed of homogeneous hydrogels exhibit high mechanical strength, but were limited to slow response speed and simple motion patterns. Therefore, hydrogel actuators with a bilayer structure cannot reliably imitate motions of living beings in nature or efficiently achieve bionic movement. The successful fabrication of a NIR-driven hydrogel with high response speed via manipulation in material components and

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anisotropic structure could help achieve bionic deformations and movements. Luo and coworkers [27] synthesized a responsive hydrogel with a well-defined gradient pore construction via a heterobifunctional crosslinker that enabled the hydrothermal process. By incorporating polypyrrole nanoparticles as photothermal transducers, the photo/thermo-triggered hydrogel exhibited a rapid response to external stimuli. However, the relatively low mechanical strength, one-fold locomotion patterns under stimulation and complex fabrication method of polypyrrole nanoparticles restricted its applications. Therefore, the design and fabrication of hydrogel actuators capable of generating multi-modal movements with high response speed under NIR laser stimulation still remains a challenge in the field.

In this paper, inspired by biological motions of human fingers, palms, chrysanthemums and inchworms etc., a series of NIR-driven bionic intelligent hydrogel actuators was constructed using hydrogel synthesized from N-isopropylacrylamide (NIPAm) and 4-hydroxybutyl acrylate (4HBA). Graphene oxide (GO) was used as the photothermal transducer, and we proposed a novel method for the entirety and locality infiltration of GO to achieve controllable and remarkable actuation performances. Combined with specific structural design, the fabricated hydrogel actuators achieved versatile complex bionic motions with good response speed. These intriguing results revealed the promising prospects of this intelligent hydrogel material in the fields of 4D printing, bionic soft robots and advanced actuators.

2. Experimental section

2.1. Materials

N-isopropylacrylamide (NIPAm, $C_6H_{11}NO$, Aladdin, Shanghai, China, 2% stabilizer) and 4-hydroxybutyl acrylate (4HBA, $C_7H_{12}O_3$, J&K Scientific, Ltd., Beijing, China, 98%) were treated as the monomer and crosslinking agent, respectively. The initiator ammonium persulfate (APS, $(NH_4)_2S_2O_8$, Sinopharm Chemical Reagent Co., Ltd., Shanghai, China, Analytical reagent) and graphene oxide (GO, Suzhou Hengqiu Graphene Technology Co., Ltd., Suzhou, China, 95%) were used as received directly. Pure water was obtained by deionization and filtration with a Millipore purification apparatus (resistivity $\geq$ 18.2 MΩ cm).

2.2. Preparation of the hydrogel materials

To obtain hydrogels with the appropriate thickness for the fabrication of bionic intelligent actuators, hydrogel materials with 10 mL

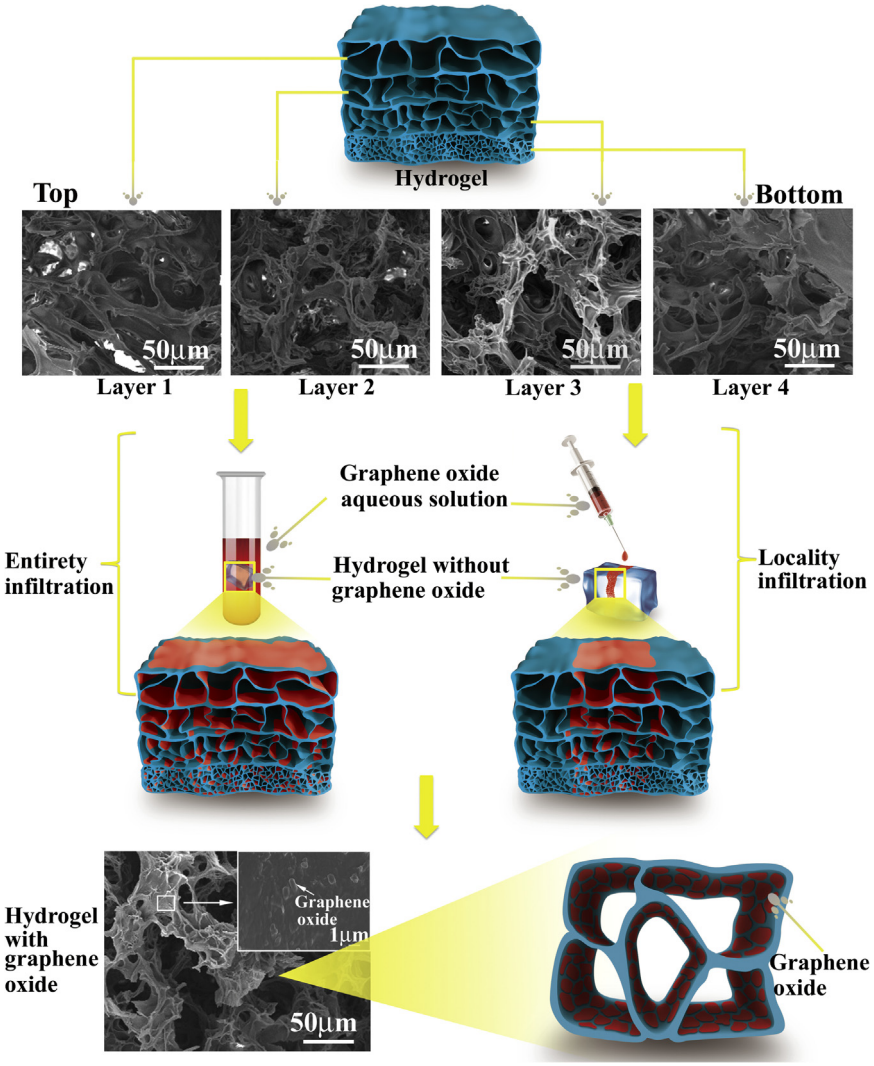

Fig. 1. Schematic of infiltration methods. According to the distribution of pore size along the thickness direction, the integrated hydrogel matrix can be qualitatively divided into four layers. The water was completely removed when the hydrogel was in the fully swollen state. Thereafter, an aqueous graphene oxide solution can be absorbed into the hydrogel. Entirety and locality infiltrations controlled the distribution of the graphene oxide particles. Graphene oxide particles adhered tightly to the pore surface.