



Cellulose/graphene bioplastic for thermal management: Enhanced isotropic thermally conductive property by three-dimensional interconnected graphene aerogel



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ABSTRACT

Environment-friendly cellulose bioplastic is a promising material to replace fossil-fuel plastic for the more sustainable future. In present study, we reported a functional cellulose bioplastic that can be used for thermal management. The cellulose bioplastic was easily fabricated by impregnating three-dimensional graphene aerogel (GA) into the cellulose solution and then hot-pressed. The cellulose/GA composites exhibited enhanced isotropic thermal conductivity and good mechanical performance. The thermal conductivity of the cellulose/GA composite was $0.67 \text{ W m}^{-1} \text{ K}^{-1}$ in vertical direction increasing by 219% and $0.72 \text{ W m}^{-1} \text{ K}^{-1}$ in parallel direction, increasing by 44% respectively. Meanwhile, the hardness of the composites reached to 148 MPa and the Young's modulus is 2.3 GPa, superior to those of most common plastics, such as Nylon and Polymethyl methacrylate (PMMA). The integrated performance of the composites could be attributed to the formation of efficient thermally conductive network and the good adhesion between the graphene cell wall and the cellulose.

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1. Introduction

Cellulose with the fascinating structures and properties has been used to fabricate various novel materials, such as papers [1,2], fibers [3], membranes [4,5], aerogels [6,7], and hydrogels [8]. However, due to the intra- and inter-chain hydrogen bonding network, the cellulose cannot be directly used to fabricate plastic until Zhang et al. [9] constructed a bioplastic from a cellulose hydrogel. After that, great efforts have been devoted to functionalize cellulose because of the potential usage in the replacement of fossil-fuel plastics for the more sustainable future [10–12].

Cellulose materials with high thermal conductive properties have a particular appeal since rapid development of the electrical equipments and electronic devices poses a higher demand on thermal management. The most common approach to improve the thermal conductivity (λ) is to introduce high thermally conductive fillers into the matrix, such as graphene [13–18], boron nitride (BN) [19–22], silicon carbide [23–25] and carbon nanotubes [26]. Because of their intrinsic structures, inorganic fillers with high-aspect ratios tend to align in the lateral direction and the oriented cellulose fibers in planar direction, contributing to the

high anisotropic thermal conductive property with remarkably enhanced in-plane thermal conductivity (λ_l). For example, Zhu [20] fabricated BN composite paper by a filtration of nanosheets BN and nanofibrillated cellulose (NFC) suspensions and obtained $\lambda_l = 26.2 \text{ W m}^{-1} \text{ K}^{-1}$ of the composite with only 5% BN loading. Our group conducted several studies on the thermal conductivity of cellulose materials previously [27,28], for example, though the aligned RGO nanosheets in the cellulose matrix could enhance λ_l to $6.17 \text{ W m}^{-1} \text{ K}^{-1}$ [27], the through-plane thermal conductivity (λ_T) was hardly changed due to the existence of large interfacial thermal resistance between the cellulose fibers surfaces [29] as well as the filler-matrix interface. Recently, the functionalized cellulose/graphene plastic chips also show the same results [30]. Apparently, the λ_l of the two-dimensional cellulose films can be well improved to a high level but the λ_T is not. The thin films and the chips utilize the intrinsic structures and the properties of fillers and the cellulose to conduct the heat along through-plane direction while the hardship of enhancing λ_T still needs to be conquered. Unfortunately, it is well known that plastics often used in three-dimensional bulk forms, which is generally expected to be isotropic. That is, the cellulose bioplastic with excellent λ_l as well as high λ_T is required [31]. The vertical alignment of fillers is one of the means proposed to enhance the λ_T of the composite via the consecutive pathway along the through-plane direction.

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Nevertheless, this method will sacrifice λ_L and the difficulties of controlling the alignment of fillers, particularly along the vertical direction are hard to overcome [32]. The compromise between the λ_L and the λ_T makes it a critical challenge to enhance λ in both directions.

In this study, we reported a functional cellulose bioplastic with isotropic thermal properties in which their thermal conductive properties were enhanced by three-dimensional interconnected graphene aerogels (GA). The bioplastic was prepared by a facile vacuum-assisted impregnating method followed by hot-pressing process. The graphene sheets are interconnected with each other to serve as continuous network. The heat flux flow along the graphene sheets and gather at the joints then it was transported to the adjoining graphene sheets to form consecutive pathway. As a result, the three-dimensional interconnected GA endows the cellulose bioplastic with high thermal conductivity and, in turn, the cellulose bioplastic strengthens the fragile GA, resulting in the better mechanical performance of the composites. The combined thermal and mechanical performances make the composite desirable for thermal management.

2. Experiment

2.1. Materials

Expandable graphite was bought from Jinrilai Co., Ltd. (99.0%, Qingdao, China). Concentrated sulfuric acid (98.0% H_2SO_4 , AR), sodium hydroxide (NaOH), hydrogen peroxide (30% H_2O_2 , AR), potassium permanganate (KMnO_4 , SP), urea and hydrochloric acid (HCl, AR) were provided by Sinopharm Chemical Reagent Co., Ltd. Cellulose was purchased from Sigma-Aldrich Company Ltd. All the agents were used directly as bought.

2.2. Fabrication of graphene aerogel

GO sheets were prepared from the graphite powder with a modified Hummers' method. The concentrated sulfuric acid (140 mL) was slowly added and stirred in the three-neck round-bottom flask containing graphite powder (4 g). The flask was kept

in an ice-bath. Afterwards, potassium permanganate (14 g) was added quite slowly into the mixture, and the temperature was maintained at 0°C . Subsequently, the mixture was raised to 35°C and kept stirring for 2 h. Then, the mixture was poured out and added deionized water (300 mL) at first. After stirring for 15 min additional deionized water (500 mL) was then added, followed by addition of 30 wt% H_2O_2 (10 mL), turning the color of the solution from brown to yellow. The mixture was washed with diluted hydrochloric acid (10 vol%) 3 times to remove residual metal ions and then followed by repeated washing and centrifuging to make it neutral. The 6 mg/mL GO suspension was prepared by sonicating the obtained GO in water. The GO suspension (50 mL, 6 mg/mL) was ultrasonically dispersed for 1 h before transferred to a Teflon-lined autoclave and kept at 180°C for 12 h to get graphene hydrogel. Graphene aerogel was obtained by freeze-drying for 48 h.

2.3. Fabrication of cellulose/GA composite

As illustrated in Fig. 1, cellulose was dissolved in 7 wt% NaOH/12 wt% urea aqueous solution pre-cooled to -12°C to form a 6 wt% transparent cellulose solution. The cellulose solution was vacuumized for 30 min to remove the air bubble. To facilitate the infiltration, as-prepared GA was immersed into the cellulose solution under vacuum and ice-bath for 6 h for cellulose to penetrate into the aerogel. Afterwards GA and cellulose were kept under room temperature for 2 days for cellulose to gelate, followed by regeneration in ethanol for another 2 days. Then the composite was thoroughly washed with water to remove the remaining ions and formed cellulose hydrogels. The resulting cellulose hydrogels were hot-pressed at 50°C for two days and then the cellulose/GA composite was obtained. The filler content (3 wt%) of GA in the composite was determined by the weight ratio of the GA to the cellulose powder.

2.4. Characterization and measurement

The morphology and microstructure of the samples were investigated by Scanning electron microscopy (SEM, JSM-7500F, JEOL, Japan). The structure and chemical composition of cellulose, GA

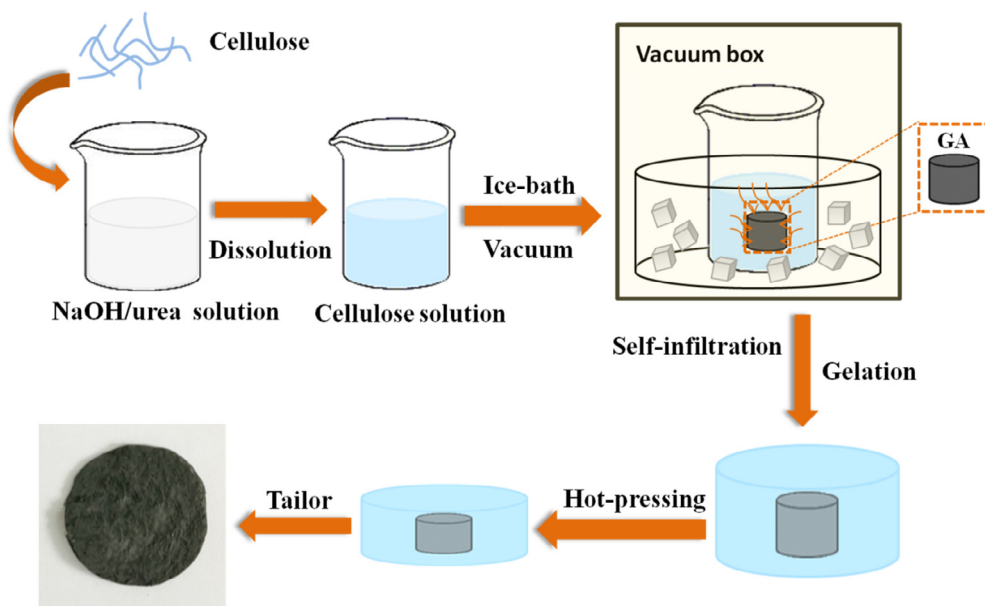


Fig. 1. Fabrication process of cellulose/GA composites. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

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