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Original Research Paper

Cellulose nanofiber/nanodiamond composite films: Thermal conductivity enhancement achieved by a tuned nanostructure

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ABSTRACT

Thermally conductive and electrically insulating composite materials are required for thermal management in advanced electronic industry. The present work aimed at creating a composite film of cellulose nanofiber (CNF) and nanodiamond (ND) with superior thermal conductivity. The thermal conductivity of the prepared nanocomposite film was $\sim 2.7 \text{ Wm}^{-1} \text{ K}^{-1}$, which corresponds to triple of usual CNF/ND composites with similar composition. The distinct thermal conductivity is attributed to a unique nanostructure we made out in the nanocomposite film. The nanostructure can be characterized by CNF fibrils which are densely covered with ND particles.

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1. Introduction

Electronic products such as communication devices, lighting apparatuses and energy storage instruments are getting denser and more powerful evermore. To ensure proper device operation, unwanted heat generated by the internal electronic components must be removed efficiently. A technical key point is to use electrically insulating materials that have higher thermal conductivities for heat dissipation [1]. Inorganic/polymer composite materials with superior thermal transport properties have been actively studied due to great application potential in microelectronics [2–7]. As for portable and foldable digital gadgets, lightweight property and mechanical flexibility are vital as well [8]. But the materials currently employed for heat dissipation can hardly satisfy the all requirements. There is an urgent need to develop novel materials for heat dissipation.

Cellulose is one of the most ubiquitous and abundant polymers on the earth. Due to its abundance, biodegradability and renewability, derivatives of cellulose have been studied as utilizable materials. Especially, much attention has been paid to preparation and application of cellulose nanofiber (CNF) due to its outstanding properties such as high strength combined with low weight [9]. In addition to the mechanical properties, its remarkable thermal transport property has been reported recently [10]. Thermal conductivity of CNF films is an order of magnitude greater than that

of conventional papers, which is usually less than $0.1 \text{ Wm}^{-1} \text{ K}^{-1}$. The favorable thermal conductivity should be ascribed to phonon propagation along the CNF fibrils. Although polymers are generally regarded as thermal insulators, thermal conductivity of polymer chains with fewer crystalline defects is much higher than that of bulky polymers [11]. Forces or high pressures impressed for disintegration in CNF manufacturing process might yield higher crystallinity. Nanodiamond (ND) particles usually produced by detonation is a member of nanocarbons [12]. Diamond is an outstanding material in many respects and it boasts quite high thermal conductivity ($\sim 2000 \text{ Wm}^{-1} \text{ K}^{-1}$). ND should inherit the superior property of bulk diamond to a degree. Furthermore, ND is less toxic than other nanocarbons [13]. Needless to say, both CNF and ND are electrically insulating. Considering the above stated properties of the nanomaterials, we attempted to prepare nanocomposites of CNF and ND to obtain tractable and reliable materials available for heat dissipation.

Plenty of works on polymer nanocomposites containing ND particles have been reported so far and the most of them focusing on improved mechanical properties [14]. Instances reporting enhanced thermal conductivity are scarce. Introduction of nano-sized particles in polymer composites usually brings larger hetero-interfaces accompanied by thermal resistance, which are not beneficial for thermal conduction. Merely mixing the two constituents cannot be free from the nuisance. In the present study, we aimed at a nanostructure illustrated in Fig. 1. The structure can be characterized by a CNF fibril whose surface is densely covered with ND particles. The dense distribution of ND particles should

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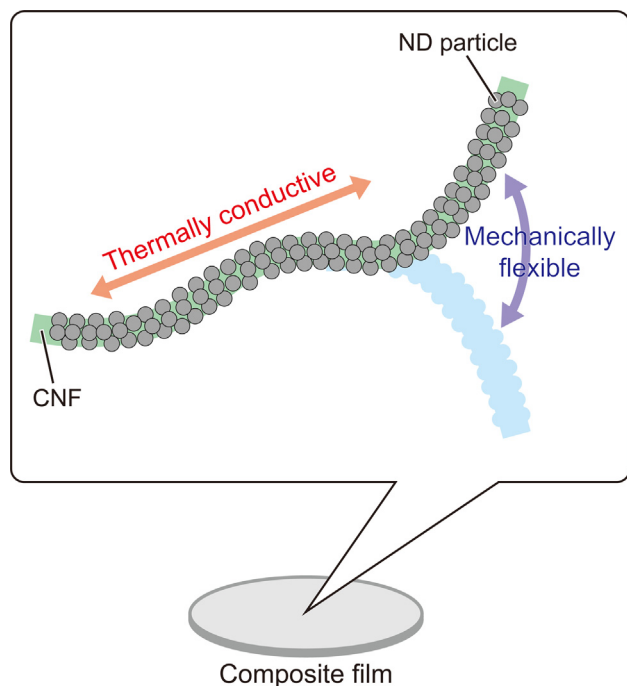


Fig. 1. Schematic to show the nanostructure consisting of CNF and ND which propagates heat efficiently.

increase the frequency of contingences among ND particles, resulting in prominent lift of thermal propagation along the CNF axis. The exterior is not a single solid layer but composed of separate ND particles. Hence, the mechanical flexibility of CNF might be maintained even after the combining process.

In order to materialize the above stated unique nanostructure, we evolved a novel colloidal processing method. In a polar liquid, such as water, the CNF surface is prone to be charged negatively [15], while the ND surface charged positively [16]. The oppositely-charged constituent nanomaterials should feel an electrostatic attraction in aqueous solutions. With smaller separations between ND and CNF, van der Waals attraction emerges [17], which should boost fixing of the ND particles onto the CNF fibrils. We attempted to obtain the nanostructure leveraging the colloidal property. Fig. 2 gives a schema of the evolved processing method. Aqueous suspensions of CNF and ND prepared separately were introduced dropwisely into a combining vessel, which is a deep container half-filled with ultrapure water. An aqueous suspension newly developed in the combining vessel should contain CNF fibrils covered with ND particles. We obtained composite films via filtration of the resultant suspension. Composite films with and without the nano-structural tuning were prepared and characterized respectively.

2. Experimental

A commercially available CNF material (WMa-10,002, Sugino Machine Ltd., Japan) was used to prepare the nanocomposites.

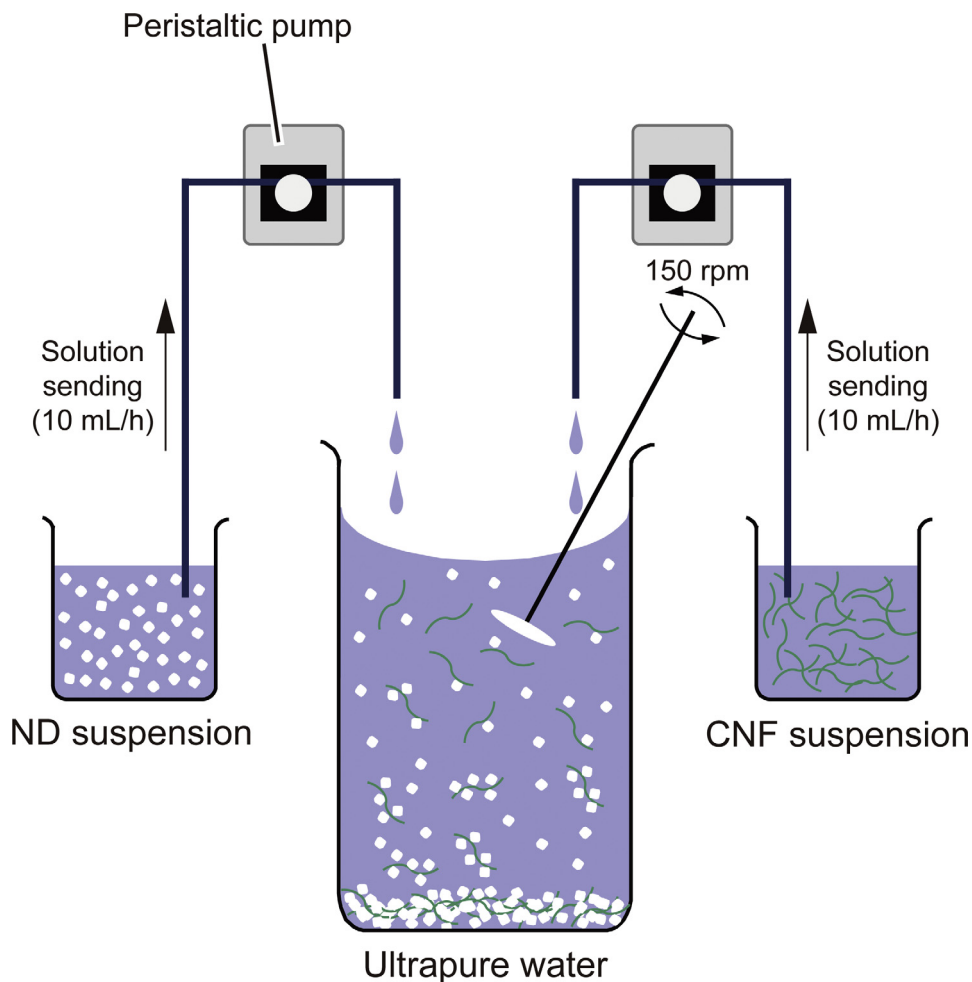


Fig. 2. Schematic of the apparatus for combining CNF and ND.

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