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## Effect of magnetic field and surfactant on dispersion of Graphene/water nanofluid during solidification

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### Abstract

Sodium dodecyl sulfonate (SDS) and carboxyl methyl cellulose (CMC) were used as surfactants to prepare Graphene/water nanofluids. These nanofluids as well as Graphene/water nanofluid without surfactant solidified with a static magnetic field, and then melted at room temperature. The zeta potentials, particle sizes, absorbencies, thermal conductivities, solidification and melting photographs were applied to evaluate the dispersion of Graphene/water nanofluids during solidification. It was found that the dispersion of the nanofluid without surfactant was severely destructed during solidification, while the other nanofluids with surfactant obtained relatively good dispersion by magnetic field. But sediments were observed to accumulate gradually over time in the subsequent melting Graphene/SDS/water nanofluid. We thought that CMC and SDS probably improved the dispersion of graphenes in the solidified water by decreasing the contact angle of graphene by water and thus increasing the nucleation rate of Graphene/water nanofluid. External electromagnetic field could further enhance the nanoparticle dispersion in the solidification of nanofluids by driving graphenes to move towards the solidification interface. This dispersion effect of magnetic field was strongly dependent on the amount of surfactant adsorbed on the graphenes. With increasing adsorption amount of surfactant, the dispersion of surfactant-coated graphenes in water became better.

*Keywords:* Nanofluid; Magnetic field; Solidification; Dispersion stability; Surfactant

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### 1. Introduction

Nanofluids, which are solid-liquid composite materials consisting of nanometer sized solid particles, fibers, rods or tubes suspended in different fluids [1], provide a promising technical selection for thermal energy storage because of its many advantages such as anomalously high thermal conductivity and relatively low supercooling degree [2, 3]. Although recent researches have covered the experimental studies on subcooling degree, solidification rate and enthalpy of nanofluids, little has been focused on the dispersion stability of nanofluids during solidification, which directly affects the useful life of nanofluids. The research of X.F. Li et al. [3] showed that the temperature difference between wall and middle of container during solidification of Al<sub>2</sub>O<sub>3</sub>/water nanofluids monitored using an infrared imaging experimental system even exceeded 10°C. We therefore speculated that ice nucleation might not occur simultaneously in nanofluids even though they had very high thermal conductivities, and the container wall would probably be the first place where the formation of ice nuclei occurred. If this speculation was

valid, then there was a possibility that those nanoparticles on which ice nuclei couldn't form successfully would be pushed away by the solidification front and finally cluster together somewhere in the container. As a result, the advantages of nanofluids over traditional working fluids in thermal storage applications declined or even disappeared. Magnetic field is an effective method to control the trajectory path of charged particles. As is well known, the charges of nanoparticles could be increased by adsorbing ionic surfactants onto their surfaces. Therefore, we proposed that exerting a magnetic force on surfactant-coated nanoparticles to make them move towards the solidification interface might help to obtain good distribution of nanoparticles in the solidified nanofluids, and thus solve the failure problem of nanofluids. In this study, we chose SDS and CMC as surfactants to disperse the graphenes of high thermal conductivity in the deionized water. The effects of magnetic field and surfactant on the nanoparticle distributions of Graphene/water nanofluids during solidification were investigated. Preliminary discussions on the solidification mechanisms concerning surfactant adsorption and magnetic field were conducted to explain how the two factors affected the dispersion of nanofluids during solidification.

## 2. Experimental methods

### 2.1. Preparation of Graphene/water nanofluids

Graphenes (ShangHai ChaoWei Nanotechnology Co., Ltd., China) were used as the additives. They were nearly spherical and had a relatively uniform size of 100 nm. Deionized water was utilized as the base fluid, with the pH pre-adjusted to  $10 \pm 0.5$  by adding NaOH. SDS and CMC were chosen as surfactants. The graphenes and the surfactants were added into water successively, and then ultrasonicated for 2 hours to prepare Graphene/water nanofluids. Graphene/water nanofluid without surfactant was also prepared for comparison. The nanoparticle concentrations of all three nanofluids were controlled at about 0.1 wt%. The mass ratio of graphenes to surfactants was 1:1. All test nanofluids were characterized by absorbance, zeta potential, particle size and thermal conductivity measurements.

### 2.2. Experimental apparatus and procedure

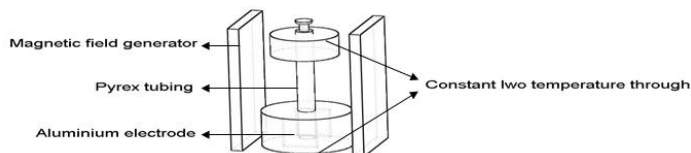


Figure 1 Schematic diagram of the experimental set up

The experimental setup used consisted of a magnetic field generator, an adjustable voltage regulator, two constant low temperature troughs, a temperature data logger and a computer. Figure 1 was the schematic diagram of the system used for solidification. The cell was constructed from inner diameter of 1 cm, double-walled, cylindrical, pyrex tubing. When filled with sample, the cell was lowered into an alcohol bath (cold side). The upper part of the cell system was heated by a water reservoir (hot side), kept at some temperature elevated to provide a rough control of the temperature gradient in the nanofluid of the cell itself. During the growth of several centimeters of ice, the temperature of the bath had to be lowered as time progressed; and for each new alcohol bath temperature, there was doubtless a new gradient in ice. Therefore, trapping or rejection tests for graphene nanoparticles were made at several points along a length of many centimeters in this study. Solidification experiment of each nanofluid was

repeated at least three times under the same experimental conditions in order to ensure reliability. The thermal conductivities of the subsequent melting nanofluids were measured in the study.

### 3. Results and discussion

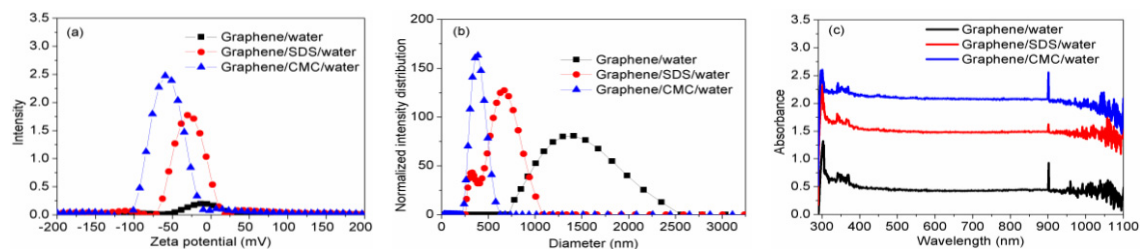


Figure 2 (a) Zeta potential distributions, (b) particle size distributions and (c) adsorption spectra of Graphene/water, Graphene/SDS/water and Graphene/CMC/water nanofluids.

The zeta potentials, particle sizes and absorbencies of Graphene/water nanofluids with and without surfactant were presented in Figure 2a-c. SDS and CMC apparently increased the absorbance of Graphene/water nanofluid, and decreased its average values of zeta potential and particle size. The observation that Graphene/CMC/water nanofluid had lower zeta potential compared to Graphene/SDS/water nanofluid indicated that the stabilization effect of CMC was much stronger. Accordingly, the CMC-stabilized nanofluid would have the best dispersion stability. In the study, the stabilization of Graphene/CMC/water nanofluid could last 1 week at least in the stationary state.

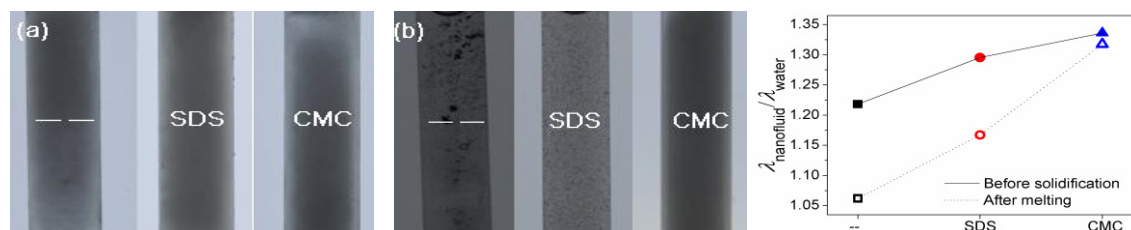


Figure 3 (a) Solidification and (b) melting photographs of Graphene/water, Graphene/SDS/water and Graphene/CMC/water nanofluids with a 2T magnetic field, (c) thermal conductivities of Graphene/water nanofluids before solidification and after melting

Figure 3a-b showed the solidification and melting photographs of Graphene/water nanofluids with and without surfactant. It was found that only the Graphene/CMC/water nanofluid still maintained good dispersion after melting, and no visible sediment appeared. As it known, graphenes were hydrophobic materials, which meant ice nuclei were hard to form on their surfaces. Accordingly, the nucleation rate of Graphene/water nanofluid might be low. Part of graphenes might not serve as ice-nucleating substrates. Instead, they were likely to be pushed upwards by the advancing solidification front, and eventually clustered together at the interfaces between ice crystals and the upper part of glass container. With regard to the SDS- and CMC-stabilized nanofluids, their relatively good dispersion stability as stated above revealed that the SDS and CMC molecules probably contacted with water through their hydrophilic head groups. This surfactant adsorption structure would reduce the contact angle of graphene by water. We therefore considered that SDS and CMC could increase the nucleation rate of Graphene/water nanofluid,

and thus improve the dispersion of nanofluids during solidification. On the other hand, surfactant-stabilized Graphene/water nanofluids possessed higher zeta potentials than that without surfactant (Figure 2a). Therefore, the downward magnetic force could be exerted on the charged SDS- and CMC-coated graphenes. These charged graphenes then could crash their way into the advancing solidification interface only if the magnetic force was sufficient. Unfortunately, this situation didn't happen to the SDS-stabilized nanofluid. The low zeta potential of this nanofluid (shown in Figure 2a) suggested that there probably weren't enough SDS molecules adsorbed onto graphenes to prevent them from accumulating at the interfaces between ice crystals. As shown in Figure 3b, visible sediment appeared in the SDS-stabilized nanofluid. Fortunately, the other CMC-stabilized nanofluid could achieve good dispersion in solidification because of its very high zeta potential. The thermal conductivity difference before solidification and after melting for this nanofluid was about 1.84%. Whereas for the Graphene/water and Graphene/SDS/water nanofluids, the thermal conductivities were obviously reduced after melting due to the severe agglomeration of graphene nanoparticles (shown in Figure 3c).

## Results and discussion

Nanoparticle distributions during solidification of Graphene/water nanofluids with and without surfactant were investigated with a magnetic field. It was found that graphenes tended to aggregate together in the solidification of Graphene/water nanofluid without surfactant. Adding surfactants SDS and CTAB could prevent the aggregation of graphenes to some extent, but couldn't eliminate graphene aggregation absolutely. Magnetic field could help SDS and CTAB to improve the dispersion of Graphene/water nanofluid during solidification. Its effect was closely related to the amount and orientation of surfactants adsorbed on nanoparticle surfaces. Thus, choosing appropriate surfactants seemed very important to stabilize nanofluids with magnetic field. In this study, only the CMC-stabilized Graphene/water nanofluid was found to maintain a relatively good dispersion during solidification.

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