Influence of the MWCNT Crystal Structure on the Thermal Conductivity and Viscosity of the MWCNT Suspension

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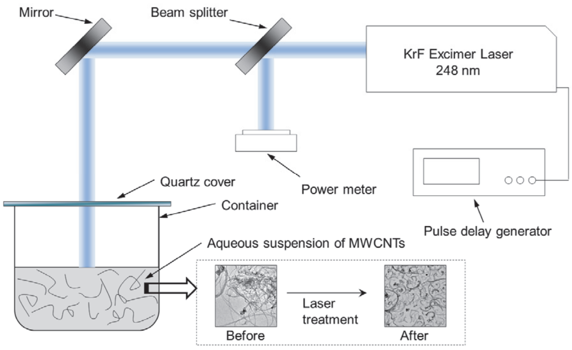

Fig. 1 Experimental setup for laser treatment of MWCNT suspensions.

1. Introduction

Fluids containing suspensions of nanometer-sized particles have been attractive as innovative coolants for nuclear systems owing to their enhanced heat transfer performance [1]. Additionally, recent studies show that suspended nanoparticles in a sodium coolant suppress sodium-water reaction [2], which is one candidate technology to improve the safety of a prototype Gen. IV sodium-cooled fast reactor. In this work, the effects of KrF excimer laser irradiation on an aqueous coolant with suspended MWCNTs (Multiwalled carbon nanotubes) were experimentally examined. The thermal conductivity and viscosity of the coolant were measured after laser treatment. This work is mainly focused on the influence of the MWCNT crystal structure on those properties of the MWCNT suspension using the TEM imaging technique and Raman spectral measurement.

2. Methods and Results

De-ionized (DI) water and MWCNTs were used to prepare aqueous suspensions. The MWCNTs were purchased from Hanwha Nanotech Corporation. The nanotubes (CM-95) had outer diameters of 10 nm to 15 nm, lengths of 10 µm to 20 µm. SDS was purchased from Sigma-Aldrich as a surfactant. MWCNTs (0.5 mass%) and SDS (0.5 mass%) were added to DI water. The suspension was ultrasonicated for 30 min at 380 W and 20 kHz to 25 kHz using an ultrasonic homogenizer (Scientz-IID, Ningbo Scientz Bio-tech Co., Ltd.). The prepared suspensions were stable with no visual sedimentation over several weeks.

A simple experimental setup (Fig. 1) was established for introducing laser irradiation to the suspensions. Twenty milliliters of the MWCNT suspension were placed in a 100 mL beaker. The beaker was covered with a quartz wafer with a thickness of 500 µm to reduce the evaporation. The MWCNT suspensions were irradiated using a KrF excimer laser (Compex 201, Lambda Physik, 248 nm wavelength). The laser fluences varied from 15 mJ·cm^{-2} to 144 mJ·cm^{-2}, and the pulse width was 24 ns. The irradiation time and repetition rate (10 Hz) were controlled by a pulse delay generator. The laser beam cross section was 10 × 20 mm^{2}.

High-resolution transmission electron microscopy (HR STEM, JEM-2200FS, JEOL) was used with a beam acceleration voltage of 200 keV to examine the size, entanglement, aggregation, and crystal structure of the MWCNTs. The samples were prepared by diluting the suspension with DI water, introducing the dilute suspension to the TEM grid, and drying the grid at room temperature. The thermal conductivity of the suspensions was measured using the 3ω method. The sensor was immersed in the suspension vertically and the thermal responses during periodic heating were acquired at heating frequencies of 1 Hz to 10 Hz. The viscosities of the suspensions were measured using a parallel-plate rheometer (MCR 101, Anton Paar). Test samples for Raman spectral measurements were collected by evaporating the aqueous MWCNT suspensions in an oven at 60 ˚C. The Raman spectra of the dried samples were measured in backscattering geometry using a Raman microscope (LabRam HR, Horiba Jobin Yvon) fitted with a liquid-nitrogen-cooled CCD detector.

The laser irradiation time was fixed at 30 min, and the effects of the laser fluence on the thermal conductivity and viscosity of the suspensions were measured as a function of the fluence, (0, 15, 53, 105, and 144) mJ·cm^{-2} (Fig. 2). At a laser fluence of 15 mJ·cm^{-2}, the thermal conductivity and viscosity of the suspension were not influenced by laser irradiation. As the fluence reached 53 mJ·cm^{-2}, both the thermal conductivity and the viscosity decreased. Therefore, to affect the thermal conductivity and viscosity of the MWCNT suspensions, the fluence was required to surpass a threshold of 50 mJ·cm^{-2}. As the fluence

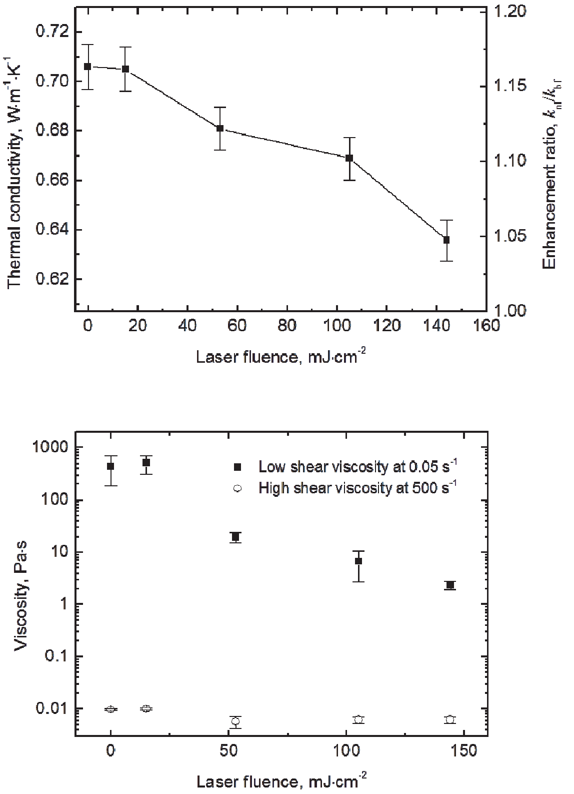

Fig. 2 Variation of the thermal conductivity (upper) and viscosity (lower) of the MWCNT suspensions with laser irradiation fluence.

increased from 53 mJ·cm^{-2} to 144 mJ·cm^{-2}, the thermal conductivity enhancement decreased from 16 % to 4.8 % and the viscosity at the low shear rate decreased dramatically.

The crystal structures of the MWCNTs could be observed in high-resolution TEM images (Fig. 3). The walls of the MWCNTs prior to laser irradiation were neat and continuous longitudinally (Fig. 3a). However, after laser irradiation, the MWCNTs contained many defects and the walls were broken and partially damaged (Figs. 3b). The irradiated samples displayed damaged crystal structures, as shown in Figs. 3b, as well as the undamaged crystal structures, as shown in Fig. 3a. The amorphization of the MWCNT was also observed by the Raman spectra measurement. As the irradiation time increased, I_G/I_D decreased gradually, indicating that amorphization of the MWCNTs occurred upon laser irradiation.

3. Conclusions

Excimer laser irradiation of a suspension of MWCNTs provided an effective way to adjust the thermal and rheological characteristics of the suspension. As the irradiation time increased, the influence of the low aspect ratio, disrupted network, and amorphization decreased both the thermal conductivity and viscosity of the MWCNT suspensions. In this work, it was revealed that the change in the MWCNT crystal structure played a significant role in the thermal and rheological properties of the MWCNT suspension.

ACKNOWLEDGMENTS

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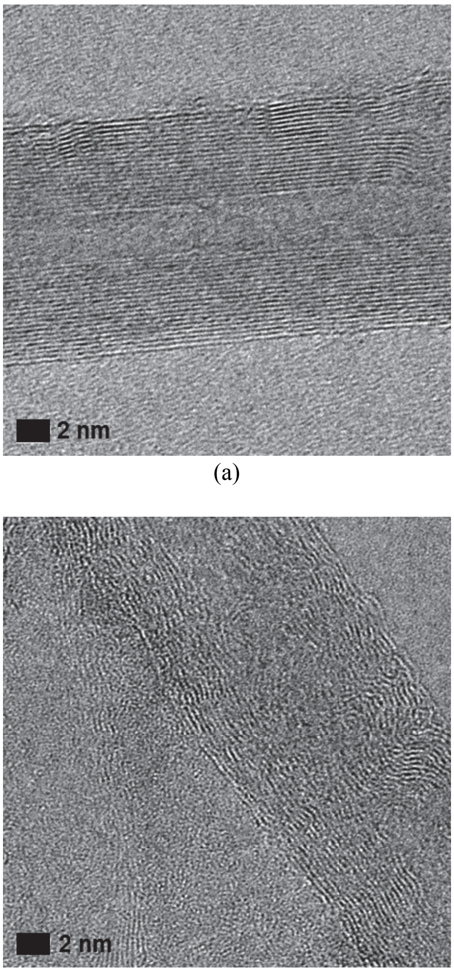

(b)

Fig. 3 TEM images of the MWCNT crystal structure; (a) before the laser irradiation, (b) after the laser irradiation.