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A Study on Excimer Laser Treatment of Coolants with Suspended Multiwalled Carbon Nanotubes

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1. Introduction

Fluids containing suspensions of nanometer-sized particles have been attracted 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 the laser treatment. Also, the TEM imaging technique and Raman spectral measurement were conducted to inspect the irradiated MWCNTs.

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.

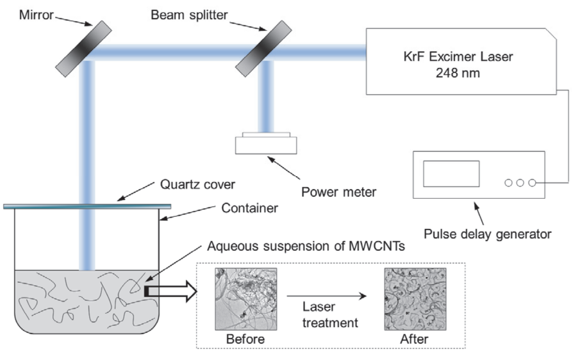

Fig. 1 Experimental setup for laser treatment of MWCNT suspensions.

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 of thickness 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 $mJ \cdot cm^{-2}$ to 144 $mJ \cdot cm^{-2}$, 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 fluence was fixed at 144 $mJ \cdot cm^{-2}$, and the effects of laser irradiation time on the thermal conductivity and viscosity of suspensions were measured as a function of irradiation time, (0, 5, 10, 30 and 60 min) (Fig. 2 and 3). As the irradiation time increased, the thermal conductivity and viscosity decreased and reached saturation. The thermal-conductivity enhancement decreased gradually over 30 min and saturated at 5 % after 30 min (Fig. 2). The results of Nan's effective medium theory (EMT) [3,4], considering the aspect ratio p of the particles, were displayed along with the experimental results. The MWCNTs were assumed to be randomly dispersed in a homogeneous medium (water) and the interfacial thermal resistance between the MWCNTs and water was neglected. Before the laser irradiation, *p* was higher than 100 and Nan's EMT overestimated the thermal conductivity enhancement by more than 100 %. This large deviation from the experimental result (~ 16 % enhancement) is estimated to be due to the assumption of ellipsoidal particles in the EMT. When *p* was high, the MWCNTs were curved and twisted unlike the ellipsoidal shape. The saturated thermal conductivity after 30 min treatment lied between the lines of $p = 10$ and 20 (Fig. 2), which agreed well with the TEM observation that *p* of the short MWCNTs ranged from 10 to 20. These results are consistent with Nan's EMT as the MWCNTs with low *p* were close to the ellipsoidal shape.

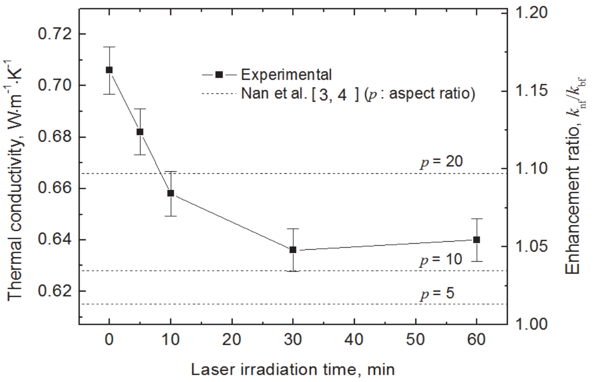

Fig. 2 Variation of the thermal conductivity of the MWCNTs suspensions with laser irradiation time.

The viscosity also decreased gradually over 10 min, then saturated at longer irradiation times (Fig. 3). At a shear rate of 0.05 s^{-1}, the viscosities of the irradiated samples reached values as low as 1/200 the viscosity of the nonirradiated sample (Fig. 3). However, at a higher shear rate of 500 s^{-1}, the viscosity was not significantly affected by irradiation.

Fig. 4 shows the Raman spectra of the MWCNTs as a function of the laser irradiation time, 0 – 60 min at a laser fluence of 144 $mJ{\cdot}cm^{-2}$. All spectra clearly showed peaks at 1350 cm^{-1} (D band) and 1583 cm^{-1} (G band), which were characteristic of amorphous and graphite carbons, respectively. The intensity ratio between the G and D bands, I_G/I_D was analyzed to evaluate the quality of the nanotubes. As the irradiation time increased, I_G/I_D decreased gradually, indicating that amorphization of the MWCNTs occurred upon laser irradiation.

3. Conclusions

This work investigated for the first time the excimer laser interaction with suspended MWCNTs in water. 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. Considering the trade-off between heat transfer properties and the pressure drop, an optimal laser fluence and irradiation time should be selected depending on the desired final characteristics.

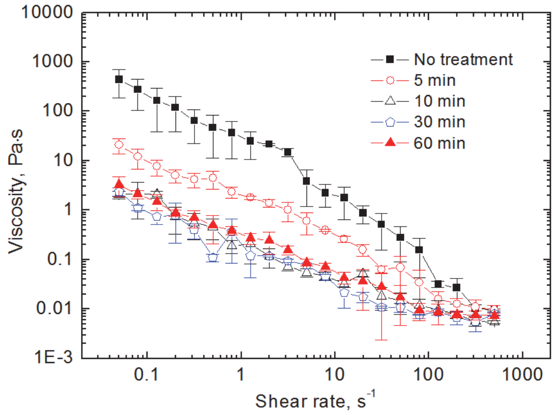

Fig. 3 Variation of the viscosity of the MWCNTs suspensions with laser irradiation time.

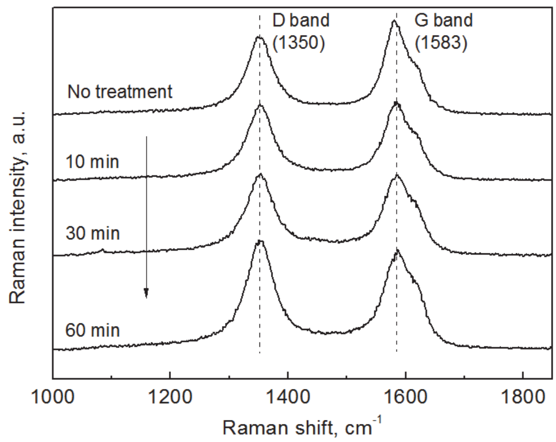

Fig. 4 Raman spectral measurements of the MWCNTs.

ACKNOWLEDGMENTS

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