Investigation of thermal conductivity coefficient of aqueous suspension with carbon nanotubes

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Abstract. The dependence of thermal conductivity of suspension on the concentration of single-walled carbon nanotubes was investigated. Mass concentration of nanotubes ranged from 0.05 to 0.5%. Sodium dodecyl sulfate was used as a surfactant. The enhancement of 34.8% were achieved for thermal conductivity coefficient of suspensions at 0.5wt.% of CNT.

1. Introduction

Multi-walled carbon nanotubes (MWCNT) were first discovered by Radushkevich and Lukyanovich in 1952 [1], and a single-walled nanotube was obtained by Iijima in 1993 [2]. Interest in carbon nanotubes is caused by their unique electrical, thermal, optical, and mechanical properties.

The thermal conductivity of MWCNT is ~3000 Wm⁻¹K⁻¹ [3], and for SWCNT it is ~6600 Wm⁻¹K⁻¹ [4]. This fact suggests that suspensions prepared on the basis of CNTs can have high thermal conductivity. In this work, investigation of thermal conductivity of aqueous suspension with single-wall carbon nanotubes was carried out.

2. Materials and methods

2.1. Materials

Single-walled carbon nanotubes (powder) TUBALL™ was purchased from OOO “OCSiAl”, the manufacturer LLC “Plasma-chemical technology”. The main parameters of the SWCNT are presented in table 1.

| Specification               | Unit | Value | Evaluation method |
|----------------------------|------|-------|-------------------|
| Carbon content             | wt.% | 86±1  | TGA, EDX          |
| SWCNT content              | wt.% | 75±1  | TEM, TGA          |
| Metal impurities           | wt.% | 14±1  | EDX, TGA          |
| Number of layers CNT       | unit | 1     | TEM               |
| Length of CNT              | μm   | >5    | AFM               |
| Mean diameter CNT          | nm   | 1.6±0.5 | Raman, TEM      |
| G/D intensity ratio        | unit | 67    | Raman             |

Table 1. The main parameters of SWCNT

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TUBALL™ powder is black. The density of SWCNT is 1.7-1.9 g cm\(^{-3}\), and the bulk density is 0.1 g cm\(^{-3}\). The high-resolution TEM image of SWCNT is shown in figure 1. TUBALL powder contains SWCNT with a mean diameter of 1.6 nm and length of more than 5 μm. Figure 2 shows the Raman spectrum. The G/D ratio is 67. This suggests that SWCNT is of high quality.

2.2. Preparation of CNT-water suspension
For the preparation of suspensions with CNT, the so-called two-step method was used. The required amount of nanotube powder was weighed using a METTLER TOLEDO laboratory balance. Then, nanotubes were added to the base fluid containing 1 wt.% surfactant. Sodium dodecyl sulfate was used as a surfactant. Then the suspension was first thoroughly mixed mechanically and then processed by ultrasonic apparatus “Volna” UZTA-0.4/22-OM (22±1.65 kHz, 400 W). The power control range was 30-100%. Ultrasound treatment time was 90 min.

An analysis of the colloidal stability of a suspension with carbon nanotubes was made using the Turbiscan LAB. Turbiscan LAB is based on the multiple light scattering method (MLS). The test sample (20 ml) is carefully placed in a cylindrical glass cell so the meniscus is neat. The light source is an electroluminescent diode in the near infrared region (880 nm). Two synchronous optical sensors receive light transmitted through the sample (180° from the incident light, transmission sensor), and light backscattered by the sample (45° from the incident radiation, backscattering detector), respectively. The Turbiscan LAB works in scanning mode: the optical reading head scans the length of the sample (up to 55 mm), acquiring transmission T and backscattering BS data every 40 μm. This is the most complete analysis mode enabling the detection of the migration phenomena. These curves provide the transmitted and backscattered light flux in % relative to standards (suspension of monodisperse spheres and silicone oil) as a function of the sample height (in mm). These profiles build up a macroscopic fingerprint of the sample at a given time. Transmission is used to analyze clear and turbid dispersions and backscattering is used to analyze opaque and concentrated dispersions. Measurement of repeated profiles at different times allows characterizing the changes occurring in the test sample.

2.3. Thermal conductivity of suspension
The thermal conductivity coefficient of suspensions was measured using the hot-wire method. Description of the experimental setup and methods are presented in [5]. The measurement was carried out at a constant temperature of suspension 25°C. Each measurement was repeated 5 times, and then the average value was determined. The total measurement error of thermal conductivity coefficient is 3%.
3. Results and discussions

Colloidal stability of suspension was controlled by Turbiscan LAB. Figure 3 shows the backscattering profiles of the suspension with a mass concentration of 0.05% CNT without ultrasound treatment and after 90 minutes of ultrasound treatment. Backscattering profiles (figure 3) show that suspension without ultrasound treatment loses stability very quickly. Ultrasound treatment allows obtaining a stable suspension.

![Figure 3. Backscattering of the suspension with 0.05% CNT](image)

Adding CNTs significantly increases the coefficient of thermal conductivity. The dependence of the effective thermal conductivity of CNT-suspensions on various concentrations is shown in Figure 4. Thermal conductivity coefficient of suspension increases by 12.7% and 34.8% at a mass concentration of 0.25% and 0.5%, respectively.

The ratio between thermal conductivity coefficient of suspension $k_{eff}$ and particle concentration $\phi$ was first obtained by Maxwell [6]. He considered suspensions with spherical particles non-interacting with each other. Later, Hamilton and Crosser generalized the Maxwell model for non-spherical particles [7]:

$$k_{eff} = k_f \times \frac{k_p + (n-1)k_f + (n-1)\phi(k_p-k_f)}{k_p + (n-1)k_f - \phi(k_p-k_f)}$$

(1)

where $k_f, k_p$ is thermal conductivity of the based fluid and particles, respectively; $n$ is the empirical shape factor.

The empirical shape factor was correlated by Hamilton and Crosser (H-C), which has a simple expression as [7]:

$$n = 3\psi^{-g}$$

(2)

where $\psi$ is sphericity, $g$ is an empirical parameter. In the original H-C model, it was set as $g = 1$. However, in the later studies [8], it was found that $g = 1.55$ was more suitable for CNT nanofluids.

Xue [9] considered the very large axial ratio and the space distribution of the CNTs:
Murshed et al. [10] proposed model, which considers particle size, interfacial layer:

\[
    k_{eff} = k_f \left( 1 - \phi + 2\phi \frac{k_p}{k_p - k_f} \ln \frac{k_p + k_f}{2k_f} \right) \left[ 1 - \phi + 2\phi \frac{k_f}{k_p - k_f} \ln \frac{k_p + k_f}{2k_f} \right]
\]

where \( \gamma_1 = 1 + t/R \), \( \gamma_2 = 1 + t/(2R) \), in which \( t \) and \( k_f \) are the thickness and thermal conductivity of the interfacial layer at the CNT-liquid interface.

In Murshed et al. models [10], the interfacial layer thickness is defined as 2 nm for nanotube. Also in Jiang et al. model for CNTs nanofluids [8], the interfacial layer thickness is also set as 2 nm.

Figure 4 shows a comparison of present experimental data with above models. We used the following parameters in the models: \( k_p = 6600 \text{ Wm}^{-1}\text{K}^{-1} \), \( k_f = 0.608 \text{ Wm}^{-1}\text{K}^{-1} \), \( g = 1 \) and \( g = 1.55 \), \( R = 1.6 \text{ nm} \), \( t = 2 \text{ nm} \), \( k_o = 3k_f \). The CNT length of 5 µm was used in the calculation of sphericity. Figure 4 shows that experimental data are much higher than theoretical models. Good agreement was obtained with H-C model at \( g = 1.55 \).

**Conclusion**

An experimental study of thermal conductivity of water-based SWCNT was presented. The effective thermal conductivity of CNT suspension was found to increase significantly with particle concentration. Good agreement was obtained with H-C model at \( g = 1.55 \).

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