Carbon-nanotube nanofluid thermophysical properties and heat transfer by natural convection

Y Li[1], S Suzuki[2], T Inagaki[1] and N Yamauchi[3]
[1] Ibaraki University, 4-12-1 Nakanarusawa, Hitachi, Ibaraki 316-8511, Japan
[2] Graduate School of Science and Engineering, Ibaraki University, Japan
[3] Fukushima National College of Technology, Japan

E-mail: yanrong@mx.ibaraki.ac.jp
Abstract. We measured the thermophysical properties of suspensions of carbon nanotubes in water as a type of nanofluid, and experimentally investigated their heat transfer characteristics in a horizontal, closed rectangular vessel. Using a previously constructed system for high-reliability measurement, we quantitatively determined their thermophysical properties and the temperature dependence of these properties. We also investigated the as yet unexplained mechanism of heat transport in carbon-nanotube nanofluids and their flow properties from a thermal perspective. The results indicated that these nanofluids are non-Newtonian fluids, whose high viscosity impedes convection and leads to a low heat transfer coefficient under natural convection, despite their high thermal conductivity.

1. Introduction
The present study was performed for quantitative evaluation of the previously undetermined properties of CNT nanofluids as a heat transport medium, from a materials engineering viewpoint. The CNT (MWNT; Meijo Nano Carbon Co., Ltd.) nanofluids (CNFs) were composed of 5 wt% and 1 wt% CNTs in pure water, under atmospheric pressure. The typical physical properties of CNFs are 1 - 5 μm in length, purity > 95%, content of carbon nanotube are 5 wt % and 1 wt %, and the solvent is water. The diameter of the CNTs as determined from SEM and TEM images was approximately 20-30 nm and their thickness was a few nanometers.

2. Experimental apparatus and measurement methods

2.1. CNF thermophysical properties
The method for measurement of density with a hydrometer, follows the JIS Z 8804 (JIS, 2001) and (JIS K 2249 JIS, 2001) standards. The viscosity was measured with a cone-and-plate viscometer, which is also used to determine whether the working CNF is a Newtonian fluid. The viscosity μ_R [Pa·s] of the CNF sample was obtained from the shear rate D [1/s] and the shear stress of the CNF sample placed between the rotating cone rotor and the cup plate, as follows.

$$D = 2\pi\omega r/60 \text{ x } 1/r\varphi = 2\pi\omega/60\varphi \text{ [1/s]} \quad s_c = 3M/2\pi R^3,\ M = F \text{ x } \theta/100 \text{ [Pa]} \quad \mu_R = s_c / D \text{ [mPa・s]} \qquad (1)$$

where ω [rpm] is the rotor's rate of rotation, θ [%] is the viscometer index, φ is the cone angle (1°34′), R [m] is the rotor radius (24 mm), r [m] is the rotor radial coordinate, M [N·m] is the viscous drag torque, and F [N·m] is the spring full-scale torque (67.4 μN·m). In this measurement system, the Newtonian fluid properties were thus obtained from the relationship between the shear rate and shear stress. The viscosity was also measured with a tuning-fork vibration viscometer. The adiabatic method was used to determine the specific heat. Its construction was based on a reported specific heat measurement method (Shimizu, 1994; Inagaki et al., 2011, 2012, 2013). The specific heat C_p [J/(kg·K)] of the CNF mass m [kg] in the insulated vessel was obtained from the measured temperature rise ΔT [K] and the heating time τ [s] while heating at a constant electric power Q_e [W] using

$$C_p = Q_e\tau / (m\Delta T) - W / m \text{ [J/(kg·K)]} \tag{2}$$

where W [J/K] is the heat capacity of the apparatus obtained from the known specific heat C_{pstd} (J/(kg·K)] of a standard heating medium using

$$W = Q_e\tau/\Delta T - C_{pstd}m \text{ [J/K]} \tag{3}$$

where $W = 3.796T + 25.10$ [J/K], which was the value used in Eq. (2).
The thermal conductivity was determined by a transient hot-wire method (JSTP, 1990; Inagaki et al., 2012, 2013). The thermal conductivity λ [W/(m·K)] of the CNF in the vessel was obtained from the measured temperature rise ΔT_w [K] at the surface of the resistance wire held taut in the CNF, between elapsed times t_1 and t_2 [s] from initiation of the uniform resistance wire heating, and the CNF thermal conductivity λ [W/(m·K)] was calculated using

$$\lambda = I^2 Re\,ln(t_2/t_1) / (4\pi\Delta T_W) \text{ [W/(m·K)]} \tag{4}$$

where I [A] is the applied electric current and Re [Ω/m] is the electrical resistance per submerged length of the resistance wire.
The temperature dependence of each property was estimated by a least squares approximation, and the standard error of the estimate was defined (JSME, 1987) as

$$SEE = [\sum_{k=1}^{N}(X_{meas,k} - X_{caic,k})^2 / (N - C)]^{1/2} \text{ [variable]} \tag{5}$$

where $X_{meas,k}$ is the measured value at a given temperature, $X_{caic,k}$ is the approximated value at that temperature, N [-] is the total number of measurement points, and C [-] is the number of constants in the equation for the approximation curve. For all measured values relating to the thermophysical properties, outliers were discarded by the modified Thompson τ technique.

2.2. Heat transfer by natural convection

The heat transfer vessel (HT) was a horizontal, closed rectangular vessel having the previously reported configuration (Inagaki et al., 2011, 2012, 2013).

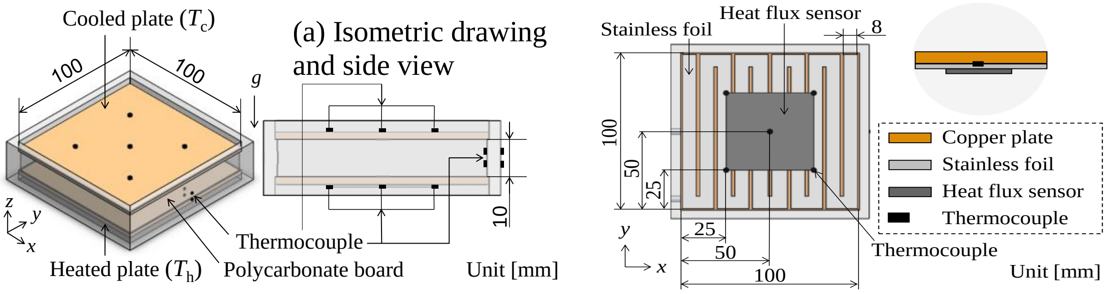


Fig. 1 Schematic illustration of experimental apparatus. (b) Detail of heating part (Right)

The HT vessel configuration is shown schematically in Figs. 1 (a) and 1 (b). The outside surface of the upper copper plate, was cooled naturally at room temperature. Although Joule heating was thus applied from the lower surface in this experimental configuration, the heat transfer surface was formed by contact of the fluid via a 5-mm-thick copper plate. To estimate the heat loss from the vessel bottom, a heat flux sensor was placed between the insulation layer and the heat transfer surface. The 50-mm-thick glass wool insulation on the bottom and sides of the vessel formed an adiabatic wall. The mean of the temperature outputs of the lower set of five was taken as the heated surface temperature T_h [K], and that of the upper set of five as the cooled surface temperature T_c [K].
In calculating the coefficient of heat transfer by natural convection, the net heat flux q' [W/m^2] was

$$q' = q_{in} - q_{loss} = VI/A - (q_b + q_s) \text{ [W/m}^2\text{]} \tag{6}$$

where q_{in} [W/m^2] is the input heat flux, q_{loss} [W/m^2] is the heat loss, V [V] is the input voltage, I [A] is the input current, A [m^2] is the wet surface area, q_b [W/m^2] is the heat loss from the vessel bottom as determined from the heat flux sensor, and q_s [W/m^2] is the heat loss from the vessel sides. In the present study, the ratio of the heat loss q_{loss} [W/m^2] to the thermal flux input q_{in} [W/m^2] was approximately 40%.

3. Results and discussion

3.1. Thermophysical properties of liquid phase state

Figure 2 (a) show the temperature dependence of the CNF density obtained with the hydrometer. As can be seen, at 300 K the density of CNF 5 wt% was ρ = 1032 kg/m^3, whereas the literature value for the water solvent was ρ = 997 kg/m^3. For CNF 1 wt% at the same temperature, the density was ρ = 1001 kg/m^3, which is 1.004 times that of water. The density for the CNF were shown to be higher than those for pure water but to approach the values for water at low concentrations. The least-squares approximation of the relationship between the CNF density as defined in this study and the temperature yielded the following results.

$$\rho = 973.73 + 0.749T - 1.848 \times 10^{-3}T^2 \text{ (293 - 343 K) [kg/m}^3\text{] (CNF: 5 wt\%)} \quad (7)$$

$$\rho = 753.85 + 1.940T - 3.722 \times 10^{-3}T^2 \text{ (293 - 343 K) [kg/m}^3\text{] (CNF: 1 wt\%)} \quad (8)$$

The standard error of the estimate (*SEE*) was 1.25 kg/m^3 for Eq. (7), and 0.737 kg/m^3 for Eq. (8).

Figure 2 (b) show the temperature dependence of the viscosity measured with the tuning-fork vibration viscometer. As can be seen, the viscosity for CNF 5 wt% at 300 K was μ_V = 49.82 mPa·s, and those for CNF 1 wt% was μ_V = 2.57 mPa·s. A least squares-approximation of the dependence of the CNF viscosity on temperature yielded the following results.

$$\mu_V = 591.47 - 2.940T + 3.782 \times 10^{-3}T^2 \text{ (293 - 343K) [mPa·s] (CNF: 5 wt\%)} \quad (9)$$

$$\mu_V = 6.149 \times 10^4 - 9.507 \times 10^{-2}T + 5.877T^2 - 1.815 \times 10^{-2}T^3 + 2.802 \times 10^{-5}T^4 - 1.728 \times 10^{-8}T^5 \text{ (293 - 343K) [mPa·s] (CNF: 1 wt\%)} \quad (10)$$

The *SEE* was 2.55 mPa·s for Eq. (9), and 0.125 mPa·s for Eq. (10). Thus, the viscosity for the CNF are larger than those for pure water but approach the values for water as the concentration decreases.

Figures 2 (c) and 2 (d) show the CNF viscosities as measured with the cone-and-plate viscometer. This method was employed because, unlike the tuning-fork vibration method, it permits resolution of the important question of whether the CNF is a Newtonian fluid. The measurements were performed at rotational speeds of N = 10, 20, 50, and 100 rpm.

Figures 2 (e) and 2 (f) show the relationship found between the shear rate D [1/s] and the shear stress s_c [Pa] for the CNFs at 303 K at different rotational speeds. The relationship is seen to be nonlinear in all cases, thus confirming that the present CNFs are non-Newtonian fluids.

Figure 2 (g) shows the temperature dependence of the specific heat as obtained by adiabatic specific heat measurement method. The specific heat for CNF 5 wt% at 300 K was found to be C_p = 3841 J/(kg·K). The value given in the literature for water at 330 K is C_p = 4179 J/(kg·K). For CNF 1 wt%, the specific heat was C_p = 4097 J/(kg·K), representing a ratio to that of water of 0.980. A least-squares approximation of the relationship between the CNF specific heat and the temperature yielded the following results.

$$C_p = 4672.3 - 3.859T + 3.631 \times 10^{-3}T^2 \text{ (293 - 343 K) [J/(kg·K)] (CNF: 5 wt\%)} \quad (11)$$

$$C_p = 5908.2 - 11.47T + 1.809 \times 10^{-2}T^2 \text{ (293 - 343 K) [J/(kg·K)] (CNF: 1 wt\%)} \quad (12)$$

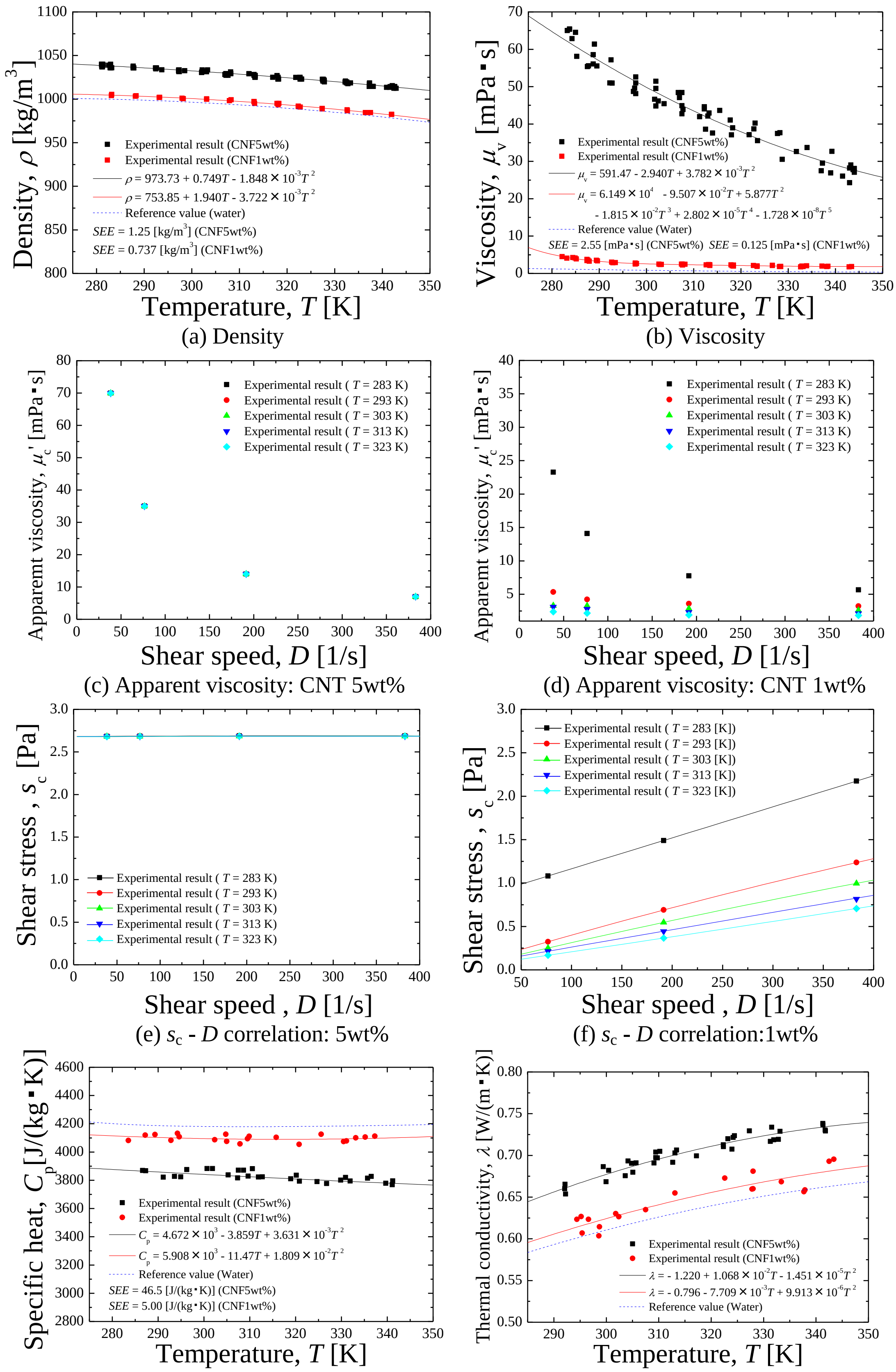
The *SEE* for Eq. (11) and (12) was 46.5 and 5.00 J/(kg·K), respectively.

Figure 2 (h) shows the dependence of the thermal conductivity on temperature, as obtained by the transient hot-wire method. The results show that the thermal conductivity for CNF 5 wt% at 300 K is λ = 0.6773 W/(m·K). The literature value for water at 330 K is λ = 0.6104 W/(m·K). For CNF 1 wt%, the thermal conductivity at 300 K was found to be λ = 0.6242 W/(m·K), representing a ratio to that of water of 1.02. A least-squares approximation of the relationship between the CNF thermal conductivity and temperature yielded the following results.

$$\lambda = -1.220 + 0.01068T - 1.451 \times 10^{-5}T^2 \text{ (293 - 343 K) [W/(m·K)] (CNF: 5 wt\%)} \quad (13)$$

$$\lambda = -0.796 - 0.007709T + 0.9913 \times 10^{-5}T^2 \text{ (293 - 343 K) [W/(m·K)] (CNF: 1 wt\%)} \quad (14)$$

The *SEE* of the estimates for Eq. (13) and (14) was 9.25 x 10^{-5} and 1.21 x 10^{-2} W/(m·K), respectively.



(a) Density (b) Viscosity

(c) Apparent viscosity: CNT 5wt% (d) Apparent viscosity: CNT 1wt%

(e) s_c - D correlation: 5wt% (f) s_c - D correlation:1wt%

(g) Specific heat (h) Thermal conductivity

Fig.2 Natural convection heat transfer ratio in a horizontal enclosed rectangular container

3.2. Heat transfer characteristics in natural convection

Figure 3 shows the heat transfer characteristics of the CNF 5 wt% thermal medium in a state of induced natural convection in the horizontal, closed rectangular vessel, with the mean Nusselt number $\overline{Nu}_1$ [-] as the vertical axis and the net heat flux q' [W/m^2] as the horizontal axis. Since a CNF is a non-Newtonian fluid, we investigated the heat transport mechanism of the CNF 5 wt% based on the heat transfer coefficient of the mean heat dissipation area rather than performing the nondimensionalization generally applied for Newtonian fluids. For reference, the figure also shows the heat transfer coefficient for the mean heat dissipation area with pure water as the working fluid, which was found to be in close agreement with the heat transfer correlation equation for natural convection in a fluid layer in a horizontal, closed rectangular vessel (JSME, 1986, 2009). As shown in this figure, the mean heat transfer coefficient with CNF 5 wt% as the working fluid is substantially smaller than that for pure water. In comparison with the thermophysical properties of pure water as the reference standard, the difference in the viscosity (by about 50 times) for CNF 5 wt% is far larger than the difference in the heat transfer coefficient (about 1.2 times), which can be attributed to a substantial suppression of buoyancy-induced convection. This is also clear from the results of the nondimensional analysis of the heat transfer coefficient shown in Fig. 3. In short, the Rayleigh number Ra_1 [-] for the CNF is small because of its high viscosity, and as a consequence its heat transfer capacity is reduced.

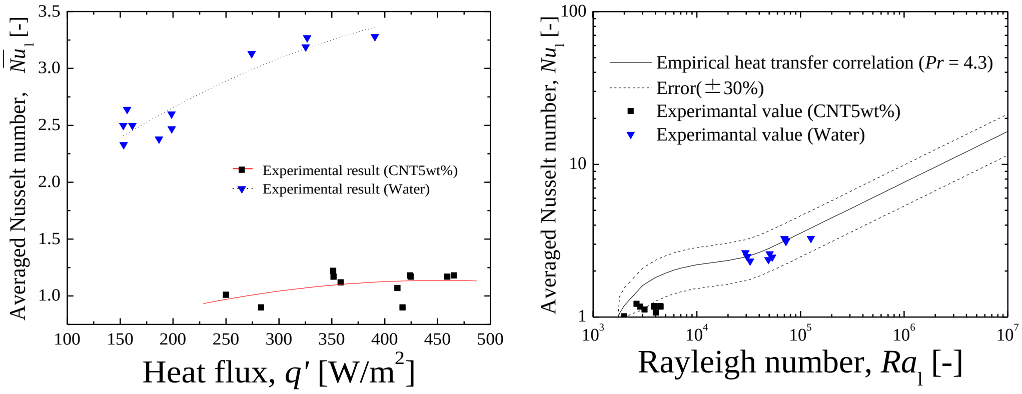


Fig.3 Natural convection heat transfer in a horizontal enclosed rectangular container

4. Conclusion

In the present study, we have investigated the thermophysical properties of CNFs, and in particular the temperature dependence of the thermal properties in CNF (1 and 5 wt%), and presented the results in the form of a heat transfer database that is essential for development of a heat transport process in a system configuration designed for actual use. The experimental results also confirm that the CNFs are non-Newtonian fluids. The effect of the 5 wt% CNF on viscosity outweighs that on thermal conduction, and suppression of buoyant convection clearly results in low heat transfer by natural convection.

References

Inagaki T and Kitazawa G 2011 *Kagakukogaku Ronbunshu* **37** 261
Inagaki T and Uezu T 2012 *Kagakukogaku Ronbunshu* **38** 263
Inagaki T and Isshiki T 2013 *Kagakukogaku Ronbunshu* **39** 33
JIS K 2249 2001 *Crude petroleum and petroleum products-determination of density and petroleum measurement tables based on a reference temperature*
JIS Z 8804 2001 *Methods of measuring specific gravity of liquid*
Meijo nano carbon 2010 *Carbon Nano-tube Fulid and its application* JP 4635103 B2
Shimizu Y 1994 *Univ. of Ryukyu, Department of Education, Bulletin* **45** 349
Japan Society of Mechanical Engineers 1986, 2009 *JSME data book: Heat Transfer 5th Edition* 56
Japan Society of Mechanical Engineers 1987 *Measurement uncertainty*
Japan Society of Thermo - Physical Properties 1990 *Thermo-physical Properties Handbook* 568