Thermophysical properties of ethylene glycol mixture based CNT nanofluids

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Abstract. Nanofluids are produced by dispersing nanometer-scale solid particles into base liquids such as water, ethylene glycol, etc. The thermal quadrupole method is utilized to determine the thermophysical properties of materials. By this technique, the thermal diffusivity and conductivity of different nanofluids containing surfactants (humic acid, sodium salt of humic acid and sodium carboxymethyl cellulose and multi-wall carbon nanotubes) were evaluated at room temperature and at 75 °C. Values of thermal diffusivity varying in the range from 9.60x10⁻⁸ m²s⁻¹ to 1.46x10⁻⁷ m²s⁻¹ and thermal conductivity from 0.26 Wm⁻¹K⁻¹ to 0.41 Wm⁻¹K⁻¹ were obtained. As main conclusions, it was noted that nanofluids exhibit superior heat transfer characteristics than the conventional heat transfer fluid and the thermal conductivity is enhanced by 50 % for the nanofluid containing 0.0275 mg/mL of sodium salt of humic acid + ethylene glycol, at the temperature of 25 °C.

1. Introduction
Dispersions of colloidal particles of nanometric dimensions (below 100 nm), added to conventional heat transfer fluids, including oil, water and ethylene glycol, have received great attention in constituting a new class of engineering fluids, the nanofluids. Nanoparticles can be as nanopowders, such as Al, Cu, SiC and CuO, or carbon nanotubes (also known as CNT). The excellent thermal, chemical and electrical properties of nanomaterials point these materials as the main additives for the production of high performance and multifunctional composite systems [1,2].

Preparation of nanofluids is the first key step in applying nanophase particles to changing the heat transfer performance of conventional fluids. The nanofluid does not simply refer to a liquid/solid mixture and some requirements are necessary, such as stable suspension, low agglomeration of particles and no chemical change of the fluid [3].

The aim of the present work is to investigate the thermal diffusivity and thermal conductivity of ethylene glycol with additions of multi-wall carbon nanotubes (MWCNT) and surfactants. For this purpose, the thermal quadrupole method was applied, and the tests were realized at room temperature and at 75 °C.

2. Materials and methods

2.1. Preparation of nanofluid
For the preparation of nanofluids was used ethylene glycol (EG) as a base fluid with addition of surfactants and nanomaterials. As surfactants were used humic acid (HA), sodium salt of humic acid...
(HAS) and sodium salt of carboxymethyl cellulose (CMC), all from Sigma-Aldrich, and as nanomaterials multi-wall carbon nanotubes (MWCNT), from Nanocyl (3100 series). Six different samples with surfactants and MWCNT were studied. Table 1 shows information of each sample in detail. The preparation method of nanofluids has been implemented and conducted by the Laboratory of Carbon Nanostructures Chemistry (LQN) of CDTN [4].

The preparation of nanofluids was as follows: first, the surfactant and the carbon nanotubes were weighed and placed in a beaker. After that, ethylene glycol was added, and then the samples were placed in ultrasonic bath for 3 hours, to ensure the homogenization and dispersion stability. After 1 month of preparation, no visually observable sedimentation was found, indicating good stability of the mixture.

| ID in the graphs | Nanofluids and base fluid                        |
|-----------------|-----------------------------------------------|
| 1               | EG (base fluid)                               |
| 2               | 0.0275 mg/mL HA + EG                         |
| 3               | 0.0275 mg/mL HAS + EG                        |
| 4               | 0.0275 mg/mL HA + 0.05 mg/mL MWCNT + EG      |
| 5               | 0.0275 mg/mL HAS + 0.05 mg/mL MWCNT + EG      |
| 6               | 0.03 mg/mL HAS + 0.03 mg/mL MWCNT + EG        |
| 7               | 0.0275 mg/L CMC + 0.03 mg/mL MWCNT + EG       |

2.2. Density determination of the alloys
The densities $\rho$ of the nanofluids were determined by weighing a known volume of the fluid in a glass graduated cylinder with an analytical balance Ohaus AV264CP at room temperature. The procedure was repeated three times, and the collected data are averaged. The density was determined using the equation (1):

$$\rho = \frac{m}{v}$$

Where $m$ and $v$ are the mass and the volume of the fluid, respectively. The densities of the nanofluids at 75 °C were estimated according to the variation of the density of ethylene glycol.

2.3. Determination of Thermal diffusivity – Thermal Quadrupole Method
The thermal quadrupole method was developed by Degiovanni [5] and can be seen as an extension of the concept of thermal resistance at steady state to transient conditions. It is an analytical method whose resolution is approximated by the least squares method, leading to identification of the thermal diffusivity. The sample gets a pulse of light on one side, raising its temperature. From the temperature field over the distance, it is possible to compare the theoretical curve and the experimental curve by the method of least squares. The modeling for the method is illustrated in figure 1 [5,6].

Figure 1. Heat transfer through a unidirectional plane wall.
The corresponding equations are:

\[
\varphi_e = -\lambda \frac{\partial T}{\partial x} \bigg|_0 = q_e - h_e \cdot S \cdot (T_e - T_\infty) \quad (2)
\]

\[
\varphi_s = -\lambda \frac{\partial T}{\partial x} \bigg|_0 = q_s + h_s \cdot S \cdot (T_s - T_\infty) \quad (3)
\]

Where: \( \varphi \) corresponds to the density of heat flow per unit of time, \( \lambda \) is the thermal conductivity (Wm\(^{-1}\)K\(^{-1}\)), \( T \) is the temperature (K), \( S \) is the surface (m), \( h \) is the heat exchange coefficient (Wm\(^{-2}\)K\(^{-1}\)) and \( q \) is the heat flow (Wm\(^{-2}\)).

To determine the thermal diffusivity, it was used the diffusivimeter Protolab model QuadruFlash 1200 (figure 2) of Thermophysical Properties Measurement Laboratory (LMPT) of CDTN. The thermal diffusivity was measured 10 times for each sample, and the data averaged.

This diffusivimeter, manufactured in Brazil, is constituted by a xenon lamp (1200 J), responsible for the energy pulse, three K-type thermocouples of special class, an InSb infrared detector, an oven for heating the sample and a signal processing unit. A short thermal excitation is generated by the xenon lamp. The flash is directed inside a resistive furnace, allowing heating of the tested specimen from room temperature to 1200 °C and the temperature rise of the specimen is measured by three K-type thermocouples.

![Figure 2. Diffusivimeter QuadruFlash 1200.](image)

2.4. Determination of Thermal conductivity

The thermal conductivity \( k \) (Wm\(^{-1}\)K\(^{-1}\)) of each sample was calculated by the equation (4):

\[
k = \alpha \cdot \rho \cdot C_p
\]

Where: \( \alpha \) is the sample thermal diffusivity (m\(^2\)s\(^{-1}\)), \( \rho \) is the sample density (kgm\(^{-3}\)) and \( C_p \) is the specific heat of the material (Jkg\(^{-1}\)K\(^{-1}\)).

The specific heat \( C_p \) of all the fluids was assumed to be specific heat of the ethylene glycol at both temperatures.
3. Results and discussion

3.1. Density determination

Table 2 presents the densities of the nanofluids. The maximum standard deviation obtained was 0.32%. It can be seen that the density decreases with increasing temperature. As expected, the densities of nanofluids are in good accordance with the density of the base fluid.

| ID | Density (kgm⁻³) |
|----|----------------|
|    | 25 °C | 75 °C |
| 1  | 1112.43 | 1069.86 |
| 2  | 1102.13 | 1059.96 |
| 3  | 1108.63 | 1066.21 |
| 4  | 1099.77 | 1057.68 |
| 5  | 1121.80 | 1078.87 |
| 6  | 1121.33 | 1078.42 |
| 7  | 1141.37 | 1097.69 |

3.2. Thermal diffusivity and thermal conductivity

Tables 3 and 4 present the thermal diffusivity and thermal conductivity of the nanofluids.

| ID | Thermal Diffusivity (x10⁻⁷ m²s⁻¹) |
|----|-----------------------------------|
|    | 25 °C | 75 °C |
| 1  | 0.97  | 0.96  |
| 2  | 1.41  | 1.41  |
| 3  | 1.46  | 1.44  |
| 4  | 1.33  | 1.34  |
| 5  | 1.33  | 1.34  |
| 6  | 1.26  | 1.27  |
| 7  | 1.10  | 1.12  |

| ID | Thermal Conductivity (Wm⁻¹K⁻¹) |
|----|---------------------------------|
|    | 25 °C | 75 °C |
| 1  | 0.26  | 0.27  |
| 2  | 0.37  | 0.39  |
| 3  | 0.39  | 0.41  |
| 4  | 0.35  | 0.37  |
| 5  | 0.36  | 0.38  |
| 6  | 0.34  | 0.36  |
| 7  | 0.30  | 0.32  |

As expected, the pure ethylene glycol showed lower values than the fluids with additives. It can be observed that with the temperature increase the thermal diffusivity did not change significantly and the thermal conductivity increased slightly for all the studied fluids (figure 3). The maximum standard deviation obtained was 9 %. 

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Figure 3. Thermal conductivity of the nanofluids.

The fluids with only the surfactants HA and HAS (0.0275 mg/mL each) exhibited the highest increases of thermal conductivity. However, there was no substantial increase in the thermophysical properties of the nanofluids with addition of the surfactants plus MWCNT. The probable cause of this difference is due to the lower volumetric concentration of MWCNT investigated in this study in comparison with other studies [7]. The nanofluid with CMC exhibited the lowest thermal conductivity increase in comparison to the other fluids.

4. Conclusions
The present work concerns the determination of the thermophysical properties of MWCNT/EG based nanofluids. The following conclusions are made based on the experimental results.

- Nanofluids exhibit superior heat transfer characteristics than the conventional heat transfer fluid;
- The thermal conductivity is enhanced by 50% for the nanofluid containing 0.0275 mg/mL HAS + EG, at the temperature of 25 °C and 52% at 75 °C.

More studies will be done to investigate the thermophysical properties of nanofluids applying higher concentrations of MWCNT.

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