A comparison of nanofluid thermal conductivity measurements by flash and hot disk techniques

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Abstract. The conversion into nanofluids is considered a suitable solution to increase the heat transfer efficiency of such fluids. Several theories with an emphasis on different thermal nanofluid mechanisms have appeared to predict enhanced conductivity measurements. There are many ways to measure the thermal conductivity of fluids. Some researchers argued that the anomalous $k$ enhancement data are caused by inaccuracies of thermal measurement methods.

In this paper, measurements on thermal conductivities of nanofluid mixtures (alumina/water) by means of two different methods are accomplished, i.e. the flash and the hot disk technique. In the first method, a NETZSCH model LFA 447 NanoFlash is employed, while in the second one a Hot Disk model TPS 2500 S is used. A comparison between the results obtained from the different measurement techniques is done. Two-step method is used to prepare nanofluids with a nanoparticles volumetric concentration from 0.1% to 4%. Each mixture, at assigned volumetric concentration, is treated with a sonicator for different times and thermal conductivity is measured in the range of temperature from 20°C to 50°C. Moreover, for assigned volumetric concentration and sonication, the stability analysis is performed and thermal conductivity measurements are carried out to determine the effect of sonication time. Results show the thermal conductivity dependence on sonication time, and an asymptotic value is evaluated for each volumetric concentration.

1. Introduction

Heat transfer fluids, such as water, mineral oil, and ethylene glycol, always have an important role in many industrial processes. The poor heat transfer properties of these common fluids hamper their effectiveness as heat exchangers. The conversion into nanofluids is considered a suitable solution to increase the heat transfer efficiency of such fluids. Ever since the report of the abnormal thermal conductivity $k$ enhancement of nanofluids by Choi [1], many researchers have tried to explain the mechanisms leading to extraordinarily high thermal conductivity. Accordingly, several theories with an emphasis on different thermal nanofluid mechanisms have appeared to predict enhanced conductivity measurements. The predictive models were developed using the experimentally nanofluid measured thermal conductivity data, provided by many research groups.
There are many ways to measure the thermal conductivity of fluids, such as the cylindrical cell method, temperature oscillation method, steady-state parallel-plate method, \( 3\omega \) method, thermal constants analyser method, thermal comparator method, and transient hot-wire or hot disk method. However, the experimental findings have been controversial and theories do not fully explain the mechanisms of elevated thermal conductivity. Some researchers argued that the anomalous \( k \) enhancement data are caused by inaccuracies of thermal measurement methods. Nanofluids are composed of a common fluid, such as water, oil or glycols and solid particles of nanometric dimensions made of various materials. Nanofluids attract the scientific and industrial interest because of their particular properties, above all the thermal conductivity that is the most studied property.

Since 1995, when Choi [1] advanced the concept of nanofluid, showing substantial increase of heat transported in suspensions of copper or aluminum nanoparticles in water and other liquids, many studies have been done on nanofluids [2,3]. The large application of nanofluids has determined the interest in the numerical simulations and experimental investigations, in both laminar and turbulent regimes, as reviewed in Terekhov et al. [4], Sarkar [5], Chandrasekar et al. [6] and Hussein et al. [7]. These new suspensions are very complicated fluids, difficult to product, handle and study. Many parameters affect nanofluid behaviour amongst which nanoparticles size, shape and concentration, the presence of chemical agents often necessary to ensure stability, and, certainly, the method of preparation. One method frequently used to obtain a stable dispersion of nanoparticles in the fluid consists in breaking particle-particle interactions using an ultrasonic probe. The effectiveness of this method is confirmed by several works [8,9,10,11,12], which often indicate an optimal sonication time. Due to the difficulties in replicating equivalent nanofluids and controlling all the preparation parameters and the test conditions, the wide literature results are often controversial. For example, Buongiorno et al. [13] and Utomo et al. [14] do not observed anomalously high thermal conductivity enhancement.

In this study, water-based nanofluid containing \( \text{Al}_2\text{O}_3 \) nanoparticles was analysed at a volume fraction ranging between 0.1% and 4%. All the concentrations were sonicated, for a time range from 30 min to 180 min, in order to found an optimal sonication time. Diameter of nanoparticles in suspension was measured using the Dynamic Light Scattering (DLS) technique. Sonication time is effective in the reduction of nanoparticles size up to 120 min. Further investigations has allowed to assess the stability of properties such as the viscosity and the thermal conductivity over time, in the presence of different concentrations of nanoparticles [15-19]. Thermal conductivity of nanofluids at all the concentrations were measured using laser flash and hot disk methods, in a temperature range between 25°C and 65°C, and a comparison between the two sets of data was done.

2. Experimental

2.1. Materials and preparation

The water-based nanofluid containing \( \text{Al}_2\text{O}_3 \) nanoparticles was purchased by Alfa Aesar at a mass concentration 50%. A surfactant, not specified by Alfa Aesar, was present at a concentration less than 1%. The nanofluid was diluted using bidistilled water, in order to obtain the other volume fractions (0.1%, 0.5%, 2.0%, 3.0% and 4.0%) by means of a precision balance KERN 440-45 N, with a maximum uncertainty of 0.2 g). The first step was to prepare nanofluids at 2, 3 and 4 vol.%. Each composition was sonicated for 30, 60, 120, 150 and 180 min, using a Hielscher UP 400S sonicator (frequency 24 kHz, maximum power 400 W).

Then, the nanofluid, resulting more stable, was used to obtain the other two compositions, 0.1 and 0.5 vol.%, adding bidistilled water. The maximum uncertainty on the volumetric concentration is 0.5% for the nanofluid mixture at 0.1%.

2.2. Nanofluid stability characterization

The Dynamic Light Scattering (DLS) technique was used to analyse the average size of nanoparticles in suspension within the base-fluid, by means of a Zetasizer Nano ZS (Malvern). The instrument
measures the mean diameter of nanoparticles reading the back-scattered light at 173° and all the tests were performed at 25°C. This instrument can detect particle size from 0.6 nm to 6 μm using the DLS process, with a declared accuracy better than ± 2%. A sample of fluid is put into a proper cell, which is illuminated by a laser and the particles scatter the light which is measured using a detector. The particles move randomly under Brownian motion and their speed is used to determine the particles dimension.

2.3. Instruments
In order to make a comparison, two different instruments, based on different techniques, are used for the thermal conductivity measurements on the same samples.

The first instrument is the NETZSCH model LFA 447, based on the “nano-flash” technique. A pulse of electromagnetic energy heats a sample side and the temperature increases as a function of time. It is evaluated on the opposite face of the same sample by means of an infrared detector, as shown schematically in figure 1a. The energy, released from the Xenon flash lamp (wavelength: from 150 nm to 2000 nm), can be adjusted using a software in terms of voltage and pulse length. The pulse width is adjustable between 0.06 ms and 0.3 ms. The higher the thermal diffusivity of the sample, the faster the energy increase on the other side. The flash method by means of LFA 447 was employed for fluid and pastes for the first time in [20]. The optimal volume of liquid in the sample holder was obtained by a calibration of the system. In fact, it can be filled in a wrong way as illustrated in figure 1b and 1c where the overfilled sample holder and the optimal volume of liquid in the sample holder, are schematically shown, respectively. The drawing of the sample holder employed in the measurements in the NanoFlash NETZSCH LFA 447 device is reported in figure 1d.

The calibration to estimate the fluid volume is obtained evaluating the thermal diffusivity of bidistilled water as a function of the fluid volume at 25°C, as shown in figure 2. It was found that the optimal volume is equal to 0.055 ml. This value is in agreement also with the nominal volume of the sample holder [20].

Figure 1. NanoFlash NETZSCH LFA 447 system: (a) working principle, (b) overfilled sample holder, (c) optimal volume of liquid in the sample holder (d) drawing of the sample holder, size in mm.
Figure 2  Thermal diffusivity as a function of the volume of fluid in the sample holder for bidistilled water at 25°C and comparison with the literature data.

Temperature profile as function of time, which is detected by the infrared sensor, depends on thermal diffusivity of the material, solid or liquid, as indicate in [21-23]. In figure 3a, an ideal response curve is reported. The evaluation of thermal diffusivity is carried out estimating the half maximum temperature rise and the corresponding time, $t_{50}$, as schematically shown in figure 3b. In the simplest one dimensional adiabatic model the thermal diffusivity is calculated as

$$a = 0.1388 \frac{d_s^2}{t_{50}} \tag{1}$$

where $a$ is the thermal diffusivity, $d_s$ is the sample holder diameter and $t_{50}$ is the time at which the half maximum temperature rise corresponds. The following equation

$$\lambda(T) = a(T) \rho(T) c(T) \tag{2}$$

allows to obtain the thermal conductivity of the liquid sample.

In figures 3c and 3d the effective response curves with bidistilled water at 25°C (b) and 65°C (c) are shown. The uncertainty of the "Nanoflash LFA 447" system for the thermal diffusivity measurements is 3% as indicated by the datasheet. The tests were repeated on three different samples and on each sample temperature ranged between 25°C and 65°C. For each sample and at assigned temperature the run was repeated ten times, i.e. the Nanoflash system carried out ten shoots for the assigned temperature. For each temperature the mean value was evaluated and the corresponding variance resulted less than ±3%.

The second instrument is a Hot Disk model TPS 2500 S, that uses a transient plane source method. The main part of the instrument is the sensor, shown in figure 4a, made of a double spiral of thin Nickel, immersed in the nanofluid, contained in a proper box. The sensor works as a continuous plane heat source and, at the same time, as temperature sensor. Actually, through the sensor, a power input is supplied to the sample and the increase of temperature is measured by the electrical resistance of the sensor itself. According to the Fourier’s law of heat conduction, the increase in temperature can be calculated as

$$\Delta T(\tau) = \frac{Q}{\pi^{1.5} r \lambda} D(\tau) \tag{3}$$

where
Figure 3. Signal carried out from the LFA 447: (a) theoretical response curve; (b) response curve and time to evaluate the thermal diffusivity, (c) and (d) effective response curve at (c) 25°C and (d) 65°C.

\[ \tau = \sqrt{\frac{L\alpha}{r^2}} \]  

and \( r \) is the probe radius, \( Q \) is the heat power, supplied as electric power, and \( D \) is a dimensionless time function of \( \tau \). Without natural convection, fitting the line given by equation (3) to the experimental data, its slope gives \( 1/\lambda \).

Figure 4b shows the Hot Disk model TPS 2500 S set up. This instrument can measure a wide range of materials, after suitable sample preparation and choosing the proper sensor diameter. The thermal
conductivity which can be detected ranges from 0.005 W/m·K to 500 W/m·K, over a wide temperature range. The declared instrument uncertainty is 5%. However, before measuring nanofluids, thermal conductivity of a well known fluid, as bidistilled water, was measured at ambient pressure in the temperature range between 10°C and 70°C to test the sensor of the instrument and to evaluate the instrument accuracy. All the measured data were compared literature data [24], obtaining an absolute average deviation less than 1%, well within the 5% accuracy declared by the constructor.

3. Results

3.1. Stability
Stability of nanofluid was studied considering two important parameters: the nanoparticles volume concentration and the sonication time. The figure 6 shows the particle size distribution, according to the intensity detected by the Zetasizer, for the water-Al$_2$O$_3$ nanofluids at the volume concentration of 2% and 4%. In the figure 7, the nanoparticles size is represented as a function of the sonication time for nanofluid at several concentrations. Al$_2$O$_3$ diameter decreases after 30 min sonication, for all the compositions, and slightly continues to decrease until 120 min of sonication. However, further sonication seems to have no effects on nanoparticle size. At a sonication time of 120, 150 and 180 min, nanoparticles diameter remains constant, indicating that there is not an optimal sonication time, but a value beyond which the further sonication is unnecessary. This value is the same for all the concentrations. Nanofluids at a sonication time 180 min were chosen as samples to measure thermal conductivity.

![Figure 6. Particle size distribution for the water Al$_2$O$_3$ nanofluids at different sonication times for volume concentration of (a) 2% and (b) 4.](image)
3.2. Thermal conductivity comparison

Thermal conductivity measurements were performed in a temperature range between 25°C and 65°C, with step of 10°C, at ambient pressure. Tests on pure water were initially made in order to verify the accuracy of the instrument, using a referential fluid and in order to compare base-fluid and nanofluid properties. Results indicate good agreement with literature data [24], all the deviations being within 2%, in the entire temperature range and for both the used instruments.

In table 1, thermal conductivity data are reported, for both the laser flash and the hot disk methods. Nanofluid was studied in a volume concentration between 0.1 and 4 vol.%. Deviations between the two methods are below or similar to the experimental uncertainty, the maximum deviation being 5.4% at the higher volume fraction. It confirms the goodness of the measurements and indicates that these different techniques are able to properly characterize the nanofluid. Thermal conductivity increases with nanoparticles concentration and with temperature.

In figure 8, the enhancement in thermal conductivity with respect to that of the base-fluid is reported as a function of the temperature, from 25°C to 65°C. Thermal conductivity is very similar to that of the base-fluid for nanofluid at temperature up to 45°C and low concentrations, while at higher temperatures, nanofluid thermal conductivity is always higher than water thermal conductivity. As expected, the maximum enhancement was found for the nanofluid at 4 vol.%, resulting 13.3% and 10.2% with nano flash and hot disk methods, respectively.

4. Conclusions

Stability of water-based nanofluid containing Al₂O₃ nanoparticles was studied considering the nanoparticles volume concentration at 1.0%, 2.0%, 3.0% and 4.0% and different sonication time, between 0 and 180 min. Increasing the sonication time, Al₂O₃ diameter decreases until 120 min of sonication, for all the compositions. However, further sonication seems to have no effects on nanoparticle size.

Thermal conductivity measurements were performed on nanofluid at 0.1, 0.5, 2, 3 and 4 vol.%, in a temperature range between 25°C and 65°C, using laser flash and hot disk methods and a comparison between the two sets of data was done. Good agreement between the two methods was found. Thermal conductivity increases with nanoparticles concentration and with temperature. The maximum
enhancement, i.e. 13.3% and 10.2% for laser flash and hot disk method, respectively, was found for the nanofluid at 4 vol.%.

Table 1. Thermal conductivity data.

\[
\Delta\% = \frac{\lambda_{\text{hot disk}} - \lambda_{\text{nanoflash}}}{\lambda_{\text{nanoflash}}} \cdot 100
\]

| \( T \) [°C] | \( \text{Thermal Conductivity} \) [W/mK] | \( \Delta\% \) |
|---------------|--------------------------------|-----------------|
|               | \( \text{Nano flash} \) | \( \text{Hot disk} \) |          |
| 25.0          | 0.612                         | 0.610           | -0.4     |
| 35.0          | 0.631                         | 0.635           | 0.7      |
| 45.0          | 0.649                         | 0.657           | 1.1      |
| 55.0          | 0.677                         | 0.680           | 0.3      |
| 65.0          | 0.712                         | 0.695           | -2.4     |
| 0.5%          |                               |                 |          |
| 25.0          | 0.614                         | 0.614           | -0.1     |
| 35.0          | 0.635                         | 0.636           | 0.1      |
| 45.0          | 0.653                         | 0.658           | 0.7      |
| 55.0          | 0.687                         | 0.682           | -0.8     |
| 65.0          | 0.721                         | 0.692           | -4.0     |
| 2%            |                               |                 |          |
| 25.0          | 0.619                         | 0.619           | 0.0      |
| 35.0          | 0.641                         | 0.643           | 0.3      |
| 45.0          | 0.662                         | 0.667           | 0.5      |
| 55.0          | 0.699                         | 0.683           | -2.4     |
| 65.0          | 0.725                         | 0.699           | -3.7     |
| 3%            |                               |                 |          |
| 25.0          | 0.626                         | 0.636           | 1.6      |
| 35.0          | 0.649                         | 0.656           | 1.0      |
| 45.0          | 0.678                         | 0.684           | 0.9      |
| 55.0          | 0.711                         | 0.702           | -1.4     |
| 65.0          | 0.732                         | 0.693           | -5.4     |
| 4%            |                               |                 |          |
| 25.0          | 0.636                         | 0.642           | 0.9      |
| 35.0          | 0.653                         | 0.666           | 2.1      |
| 45.0          | 0.691                         | 0.696           | 0.7      |
| 55.0          | 0.721                         | 0.707           | -2.0     |
| 65.0          | 0.747                         | 0.726           | -2.7     |
Figure 8. Thermal conductivity enhancement at 0.1(a), 0.5(b), 2(c), 3(d) and 4 vol%(e). Full symbol for nano flash method, empty symbols for hot disk method.
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