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Effect of temperature and sonication time on nanofluid thermal conductivity measurements by nano-flash method

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Abstract

A detailed procedure is presented for the implementation and evaluation, by Nano-Flash Method, of thermal conductivity of nanofluids with water as base fluid and at different volume concentrations of Al₂O₃. The main advantages of this measurement technique are short measurement times, easy sample preparation and high accuracy. The ultra-sonication technique was employed for preparing the mixture and results showed that the stability and thermal conductivity enhancements of Al₂O₃– H₂O nanofluids are highly dependent on sonication time and the energy supplied to the fluid for its preparation. An estimation of the optimal supplied energy and time for sonication was suggested, while temperature and volumetric concentration effects on thermal conductivity are evaluated by measurements. An enhancement of thermal conductivity with respect to temperature and volumetric concentration is found, as expected. Comparisons with data from literature confirmed the validity and the suitability of the nanoflash technique applied to thermal conductivity evaluation.

Keywords: Nanofluids Properties, Thermal Conductivity Measurements, Temperature and

Sonication Time dependence, Two-Steps Nanofluids Preparation

Introduction

Applications in engineering of nano-technologies, have allowed the production of a new class of fluids for the heat transfer, called nanofluids [1]. The two-phase mixture nanofluids are obtained through the dispersion of solid particles in a base fluid in the liquid phase. The average size of nanoparticles ranges from 5 nm to 100 nm and the most commonly used fluids base are water, mineral oils and ethylene glycol. The nanofluids have heat transfer properties greater than conventional heat transfer fluids, making them particularly suitable for electronic applications, automotive, solar energy conversion systems and in the nuclear field, as reported in [2-10]. The concept of nanofluid was introduced by Choi in 1995 [11], who first investigated the effects of the dispersion of nanoparticles in a fluid, showing their significant thermal capacity. Later, several investigations [12–14] evaluated the thermal properties of nanofluids at low concentration. Choi et al. [15] experimentally showed an increase in the thermal conductivity between 8% and 20% per volume concentrations of Al_2O_3 included between 0.5% and 6%. The numerical and experimental data have shown that TiO₂ particles dispersed in water results in an increase of heat transfer between 3% and 7% for volume concentrations from 0.2% to 2% [12,16].

The identification of nanofluids properties has since been under investigation. Research works have proposed several relations which allow us to calculate the properties such as thermal conductivity, density, viscosity, heat capacity and thermal expansion coefficient. Experimental investigations have shown that thermal conductivity depends on thermal conductivity of both base fluid and nanoparticles, temperature, nanoparticle volume fraction, shape and surface area of nanoparticles. Dependence of temperature on the thermal conductivity of CuO, Al₂O₃, TiO₂, ZnO dispersed nanofluids have been studied by [15–20]. The temperature raise increases the thermal conductivity to one that is generally accepted, does not exist [21–24]. Although there are no theoretical results available

in the literature that predict accurately the thermal conductivity of nanofluids, there are a number of semi-empirical relations that take into account the most significant parameters.

There are several experimental methods for the evaluation of the thermal properties of nanofluids [21,22]. Optical non-intrusive measures have been proposed [25,26], measures with transient hot wire (THW) [27-29], and temperature oscillation method [30], $3-\omega$ method [31]. The transient hot-wire method, is suitable for electrically conducting fluids, however, possible concentration of ions of conducting fluids around the hot wire due to the electrical field may give errors [21]. A problem in the THW method is the impact of natural convection on the measurements, as recently underlined in [32]. A steady state method was employed in [33].

At the same time, in order to improve the stability of nanofluids, the use of surfactant additives and also methods based on the PH control, [34,35] were tested. Systems to obtain a uniform dispersion of the nanoparticles within the base fluid, were analyzed by Lee et al. [36], specializing the investigation of low concentrations of Al₂O₃, by sonication and by also Yang et al. [37] using the same technique. The same technique has been used also for other kinds of nanoparticles, Tajiks et al. [38]. Further analysis allowed evaluation of the stability of properties such as the viscosity and the thermal conductivity over time, in the presence of low concentrations of nanoparticles [27]. Dispersion behaviors and thermal conductivity of Al₂O₃–H₂O nanofluids were experimentally studied under different pH and sodium dodecylbenzenesulfonate (SDBS) concentration values in [39]. The results showed that the stability and thermal conductivity enhancements of nanofluid mixtures are strongly dependent on pH values and dispersant concentration. Ismay et al. [40] studied the thermal conductivity of water-based TiO2 nanofluid over a range of volume fractions, temperatures, pH levels, particle sizes and sonication times. Recently, an experimental investigation was performed on two nanofluid mixtures without surfactant by circulating them in a circuit in [41]. The study was accomplished in order to evaluate thermophysical properties after a long-term exposure to heat. The degradation of the nanoparticle suspensions was also confirmed with a

dynamic light scattering (DLS) measurements and it provided an increase of the particle-size distribution. of the spent experimental fluid samples.

From the numerical point of view there are various models for the estimation of the conductivity of a mixture which is stabilized in the assumptions of uniform dispersion of the particles, as the Maxwell model [22,40,41]. In some cases, the experimental data are in agreement with such a model, and in some other cases they disagree, either overestimating or underestimating when compared to the Maxwell model [42–46]. In fact, the nanofluids are not mixtures ready for use, which can lead to some variability in thermal- and fluid- dynamic characteristics depending on the particular preparation process, especially the sonication process, in terms also of its characteristics such as energy and sonication time [34,40,47]. More recently, a comparison between thermal conductivity measurements of nanofluids by nanoflash and hot disk techniques was accomplished in [48] and the agreement was satisfying. The flash method was for the first time employed in the nanofluid thermal conductivity measurements.

The present paper proposes a procedure for the implementation and evaluation of the properties of nanofluids at different volume concentrations of alumina (Al₂O₃) which is valid also for fluids with different particle materials. Specifically for the preparation of the mixture, the sonication technique was employed, whereas for the measurement of diffusivity and thermal conductivity the nano-flash method was employed [48,49]. The present investigation extends and improves significantly the nano-flash method application in the estimation of thermal conductivity of nanofluids mixture. Furthermore, the evaluation of nanofluids thermal conductivity dependence on sonication by means of nano-flash method is being presented in the this paper for the first time.

Experimental Procedure

Preparation

A prerequisite to obtain thermal properties improvement is to provide a stable and durable nanofluid. Several materials might be used to realize nanoparticles for particular applications which

4

can be dispersed into fluids. Nanofluids are achieved as a mixture of nanoparticles of metals, oxides, nitrides, metal carbides, and other nonmetals, with or without surfactant molecules, and water, ethylene glycol or oils [35]. Two techniques are used to produce nanofluids. First is the single-step direct evaporation method, which simultaneously makes and disperses the nanoparticles directly into the base fluids, and the other is the two-step method, which first makes nanoparticles and then disperses them into the base fluids [35]. In either case, a well-mixed and uniformly dispersed nanofluid is needed for successful reproduction of properties and interpretation of experimental data. Nanofluids can lose their potential to transfer heat because they are prone to aggregation.

In the present investigations ultrasonic vibration, by a sonicator, was used for dispersing and disaggregating the particles. Experiments were carried out starting from a mixture of 50% in mass, i.e. 20% in volume, of alumina (Al₂O₃) dispersed in water which was acquired from Alfa Aesar. A surfactant was present at a concentration less than 1%. The nanofluid was diluted using bi-distilled 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. The medium size of particles was spherical with a diameter around 40 nm as certificated by the producer. The dispersion of the particles was obtained by first mixing the required volume of mixture in a calibrated flask with distilled water for obtaining the desiderated concentration and then using ultrasonic vibration, by a sonicator, in order to disperse it. Particle aggregate dimensions after mixture preparation and sonication were about 120 nm for all concentrations as reported in [48]. The sonicator used was the "'Hielscher UP 400S''

(frequence=24kHz).

The standard density of alumina is equal to 3940 kg/m³ and that of water is equal to 998.2 kg/m³. Effective density and specific heat of nanofluids are evaluated by using the following equations:

$$\rho_{nf} = (1 - \phi)\rho_{bf} + \phi\rho_p \tag{1}$$

$$\left(c_{p}\rho\right)_{nf} = (1-\phi)\left(c_{p}\rho\right)_{bf} + \phi\left(c_{p}\rho\right)_{p}$$
⁽²⁾

where ρ_{nf} , ρ_{bf} and ρ_{p} are the density of nanofluid, base fluid and nanoparticles, respectively, ϕ is the volumetric concentration and c_{pnf} , c_{pbf} and c_{pp} are the specific heat of nanofluid, base fluid and nanoparticles, respectively. Eq.(1) is valid for a mixture and it is usable also for nanofluids [50,51]. Eq. (2) is derived by assuming thermal equilibrium between the nanoparticles and the base fluid phase, as shown by Khanafer and Vafai [52]. In Table 1 density and specific heat are reported for different concentrations of alumina in the nanofluids. The properties of alumina are assumed constant in the temperature range considered from 20 °C to 65 °C and the values are as follows. Density is equal to 3940 kg/m³, specific heat is equal to 880 J/kg K and thermal conductivity equal to 35.0 W/m K.

φ	ρ [kg/m ³]	c _p [J/kgK]
0%	998	4182
0.1%	1003	4176
0.5%	1006	4163
1%	1028	4146
2%	1057	4112
3%	1086	4077.8
4%	1116	4043.7
5%	1145	4009.6
20%	1586	3498.6

Table 1. Density and Specific heat of nanofluids for different concentration of Al₂O₃ in the mixture.

Nanoparticles rapidly tend to aggregate with time and the agglomeration can give the decreasing of thermal conductivity of nanofluids. In order to evaluate the stability of nanofluids, the method of sedimentation was used. In Fig.1 photographs of test holder are reported for different concentrations and period of observation. After thirty days the sedimentation is observed to be already clear, especially for low volume concentrations. It is therefore essential to make the measurements in the hours immediately after nanofluid preparation and ultasonication.



Figure 1. Sedimentation test: a) observation period of 30 days; b) observation period of 300 days.

Ambient and nanofluid temperatures were measured by means of two 0.50 mm OD ironconstantan (type J) thermocouples which had an accuracy of $\pm 0.1^{\circ}$ C.

Several samples were taken at regular intervals, by ascertaining the variation of thermal conductivity as a function of the time of sonication. Samples were taken at different locations of the sonicated bath. It was found that during the sonication, thermal conductivity values are initially highly variable, while measured conductivity tends to a constant value. When that happens, further steps of sonication do not lead to significant changes in the conductivity of the nanofluid, making it possible to suspend the process considering uniformity of the property. The process depends on the particular nanofluid. In fact, the type of nanoparticle, the concentration, the nature of the base fluid, the temperature of the mixture and the environment as well as the amount of material to sonicate can condition the measurement. The first critical point in the sonication process involves the transformation of mechanical energy into thermal one. The immediate consequence of this phenomenon is the heat generation which, in the vicinity of the vibrating element, causes a rapid temperature increase. Water evaporation gives rise to variations in the concentrations which could invalidate the measurement. This gives accumulations of alumina which, due to its higher

temperature than the rest of the mixture, can emerge on the surface. To avoid this risk, a 0.50 mm OD iron-constantan (type J) thermocouple was located close to the vibrating element, which is the hottest zone, and another thermocouple external to the bath, in order to control the ambient temperature. The supplied power at the sonicator was assigned in order to ensure a temperature increase of nanofluid in the bath lower than 4.5 K.

Furthermore, in order to ensure an effective dispersion of the nanoparticles inside the mixture, it is required that the right amount of energy is supplied to the mixture by extending the sonication until the mixture properties do not become constant. Therefore, the sonication time should be established as a function of the particular mixture. In the present work, this procedure was first employed for the nanofluid with 20% of nanoparticles volume concentration. The sonication time is the highest for the highest particles concentration because this mixture has the highest mass of solid particles and the highest aggregated mass. Therefore, specific mechanical energy for the sonication of 20% volume concentration is assumed as parameter to estimate the time at different concentrations. This energy was obtained by multiplying the mechanical power delivered by the sonicator with time sonication for the mixture at 20% volumetric concentration. The procedure was stopped when the difference between two successive measurement of the considered properties, in this case the thermal diffusivity, was lower than 3%.

$$E_{m,s} = \frac{E_{m,tot}^{20\%}}{m_{20\%}} = \frac{1}{m_{20\%}} \left(\sum_{i=1}^{n} P_i t_i \right)$$
(3)

where $E_{m,s}$ is the supplied mechanical energy to the sonicator per unit of mixture mass, $E_{m,tot}^{20\%}$ is the supplied mechanical energy to the sonicator in a bath with a nanofluid mixture at 20% volumetric concentration, $m_{20\%}$ is the mass of the mixture at 20% volumetric concentration, P_i is the *i*-th supplied mechanical power to the sonicator for a time t_i .

By assuming that the supplied mechanical energy for mass unit equal to 1.0 kg, at the highest considered volume concentration (20%), is at least sufficient to uniformly disperse the nanoparticles even at lower concentrations, the sonication time for different concentrations was estimated as:

$$t_{s\%} = \frac{E_{m,s}\rho_{nf\%}V_s}{P_w} = \frac{E_{m,s}m_{\%}}{P_w}$$
(4)

where $t_{s\%}$ is the sonication time at assigned mixture volumetric concentration, V_s is the volume of the mixture, ρ_{nf} is the density of the mixture, P_w is the supplied power and m% is the mass of the assigned mixture.

Thermal Diffusivity Measurement

The thermal diffusivity measurements were carried out by means of the "NanoFlash NETZSCH LFA 447" device. The "Nano-flash" technique [49] was used in the experimental tests. In this method the front side of a plane-parallel sample is heated by a short light pulse as shown in Fig.2. In Fig. 3 drawing and sizes of the sample holder are reported. The resulting temperature rise on the rear surface is measured using an infrared detector. By analysis of the resulting temperature-versustime curve, the thermal diffusivity can be determined.

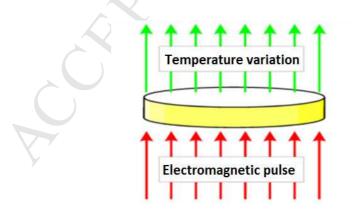


Figure 2. Sketch of the working principle of nanoflash system.

The nano-flash method is based on the evaluation of thermal diffusivity by means of the following equation:

$$t_{1/2} = \frac{1.38l^2}{\pi^2 \alpha}$$
(5)

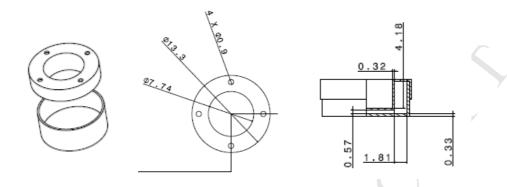


Figure 3. Sample drawing and sizes (in mm).

where *l* is the thickness of the sample and $t_{1/2}$ is the time at which the half maximum temperature value rise at the rear surface of the sample is attained. Eq. (5) is obtained assuming the following hypothesis as also indicated in [53]:

- a) Fourier law is validity and the heat conduction is parabolic.
- b) The initial temperature is uniform.
- c) Thermo-physical properties are assumed constant with temperature.
- d) Heat losses toward the external ambient are negligible.
- e) The hot spot on the surface is considered uniform.

Under these hypothesis the thermal conductive model which describes the experiments is onedimensional and the conductive problem is:

$$\frac{\partial^2 T(x,t)}{\partial x^2} = \frac{1}{\alpha} \frac{\partial T}{\partial t} \qquad 0 \le x \le 1$$
(6a)

$$T(x,0) = T_o \qquad 0 \le x \le 1 \tag{6b}$$

$$\frac{\partial T(0,t)}{\partial x} = \begin{cases} q & \text{for } 0 \le t \le \tau \\ 0 & \text{for } t > \tau \end{cases}$$
(6c)

$$\frac{\partial T(l,t)}{\partial x} = 0 \qquad t > 0 \tag{6d}$$

As indicated in [53, 54], problem (6) can be solved analytically to yield the dimensionless temperature at the rear surface of the sample: $\theta = T(x,1)/T_{\text{max}}$ in terms of the dimensionless "Fourier" time: $\tau = \frac{\pi^2 \alpha t}{l^2}$. The result is plotted in Fig. 4 in terms of time vs temperature variation. From this

curve relationship (5) can be easily inferred.

In the nano-flash technique the previous hypothesis, from a) to e) are satisfied or assumed because:

- a) The pulse width is at least $100 \ \mu$ s, greater than the nanoseconds range which allows the electrons and lattice to reach thermal equilibrium [55].
- b) The sample is at uniform temperature.
- c) The maximum increase in temperature is less than 2 K.
- d) Negligibility of heat losses depends on temperature of measurement and for temperature close to the ambient level, is verified. For higher temperature the software related to the nano-flash system [56] allows to select other one-dimensional models that take into account the heat losses from the sample [38, 56, 57].

Moreover, in case of measurements on liquids, such as water, some other important concerns due to the sample holder presence should be noted. The sample holder is made of aluminum and therefore the measurements are on a composed medium. However, in terms of thermal resistance ratio between the holder and the liquid sample, with reference to the thicknesses given in Fig. 3, the order of magnitude of holder thermal resistance is 10⁻² times the one of water or of low concentration nanofluid water based thermal resistance. Furthermore, the penetration time ratio, between the holder (aluminum) and liquid sample (water or low concentration nanofluids water based), is of the same order of magnitude of the ratio between the water and aluminum thermal diffusivity, especially when their thickness is of the same order of magnitude and it is equal to 10⁻³. This indicates that thermal disturbance penetrates very quickly in the aluminum thickness and therefore it does not affect significantly the thermal diffusivity measurement of the liquid.

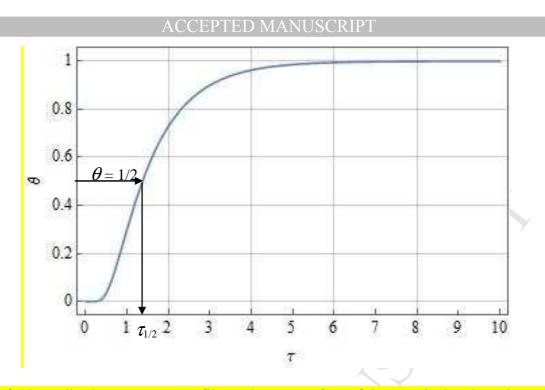


Figure 4. Normalised temperature profile on the rear surface of the sample due the pulse energy.

The higher the thermal diffusivity of the sample, the faster the energy reaches the other side. For each measurement 15 tests were run.

Thermal diffusivity measurement depends on liquid volume in the sample. The optimal volume of liquid in the sample holder was obtained by calibrating the device. In Fig. 5, the overfilled sample holder and the optimal volume of liquid in the sample holder are schematically shown. Tests for different volumes are reported in Fig. 6 for a temperature equal to 25°C. The measured values are compared with respect to the thermal diffusivity given in [58] at same temperature value. In this way an optimal value of the liquid sample volume was found equal to 0.055 ml.

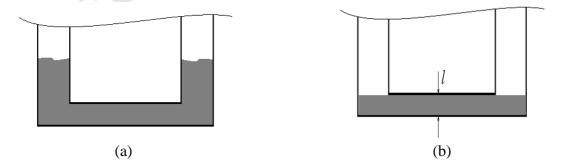


Figure 5. Sketch of sample holder: (a) Overfilled sample holder, (b) optimal volume of liquid in the sample holder (c).

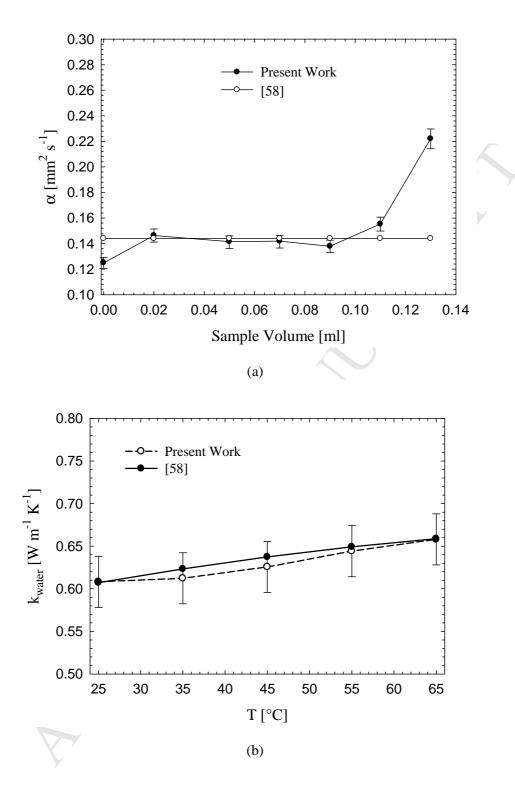


Figure 6. Thermal diffusivity and conductivity measurements: a) thermal diffusivity as a function of the volume of fluid in the sample holder and comparison with the data in [58] for temperature equal to 25°C, b) Comparison between measured thermal conductivity estimated from eq. (7) and data given in [58] in temperature range from 25 °C to 65 °C.

Uncertainty. The uncertainty of the "Nanoflash LFA 447" system for thermal diffusivity measurements is higher than the uncertainty of the volume inside the sample holder system. In fact, the uncertainty of the apparatus, as indicated by the datasheet, is $\pm 3\%$ whereas the uncertainty on the volume inside to the sample holder is $\pm 2.5\%$ for the minimum considered volume, equal to 0.02 ml, and $\pm 0.5\%$ for the maximum volume, equal to 0.10 ml. The maximum volume which should be employed in the measurements is evaluated by Fig. 6a and its value is 0.10 ml. Fig. 6a shows that for volumes higher than 0.10 ml the measurement is very different from the thermal diffusivity value of the distilled water, as given in [58]. The volumetric quantity used in the present thermal diffusivity measurements is therefore equal to 0.055 ml. The uncertainty for this value is $\pm 0.91\%$ which is lower than the uncertainty of the Nanoflash system. The tests were repeated on three different samples and temperature ranged between 25 °C and 65 °C for each sample. At assigned temperature and for each sample, the Nanoflash system carried out ten shoots in order to estimate the thermal diffusivity measurement as the mean value. The corresponding variance resulted less than $\pm 3\%$. The same procedure was employed to evaluate the uncertainty for each volume concentration of nanofluid mixture,

Thermal Conductivity Evaluation

If thermal diffusivity of nanofluids, α_{nf} , is known, thermal conductivity of nanofluid mixture, k_{nf} , is calculated by means of the relation:

$$k_{nf}(T) = \alpha_{nf}(T)c_{nf}(T)\rho_{nf}(T)$$
(7)

with ρ_{nf} , density, and c_{nf} , specific heat, of the nanofluid mixture evaluated by Eqs.(1) and (2), respectively; k_{nf} , thermal conductivity and α_{nf} thermal diffusivity of nanofluid.

A validation test was accomplished by comparing the thermal conductivity estimated by Eq. (7) for pure distillated water with the data reported in[58]. Thermal diffusivity employed in Eq. (7) was measured by the Nanoflash system in temperature range from 25 °C to 65 °C. In Fig. 6b the

estimated data from the measurements and the data from [58] are reported and both were found to agree significantly.

Uncertainty

The main contributing factors to the uncertainty are the temperature, concentration, thermal diffusivity and sample volume.

The uncertainty in the calculated quantities is determined according to the standard single sample analysis recommended in [59]. The uncertainty of a dependent variable R, as a function of the uncertainties in the independent variables X_{i} , is given by the relation

$$\delta R = \left[\left(\frac{\partial R}{\partial X_1} \ \delta X_1 \right)^2 + \left(\frac{\partial R}{\partial X_2} \ \delta X_2 \right)^2 + \dots + \left(\frac{\partial R}{\partial X_n} \ \delta X_n \right)^2 \right]^{1/2} \tag{8}$$

On the basis of Eqs. (1,2,7) and of the maximum percent uncertainties in the values of the independent variables, the maximum uncertainty of thermal conductivity are reported in Table 2 as a function of volume concentration.

		Uncertainty on measured		Uncertainty of instruments	
	_	qu	antities		
ϕ	Error	$\frac{\delta\phi}{\phi}$	$\frac{\delta k}{k}$	Balance: Weight [g]	Netzsche LFA 447: diffusivity α [mm ² /s]
0.1%	Absolute	± 0.0006	± 0.0175	± 0.2 g	$\pm 4.2 \times 10^{-3}$
0.170	Relative	$\pm 0.5\%$	± 2.9%(max)	Not indicated	± 3%
5.0%	Absolute	negligible	± 0.0186	± 0.2 g	$\pm 4.8 \times 10^{-3}$
5.070	Relative	negligible	± 3.11%(max)	Not indicated	± 3%

Table 2. Maximum percent uncertainties in the measurements.

Results

First, tests on thermal conductivity nanofluid mixtures as a function of sonication time were performed in order to investigate its effect, as previously mentioned. The thermal conductivity values were nearly constant over a certain sonication time for all concentrations, as reported in

Fig.7. Moreover, in Table 3 sonication times, ts%, evaluated from eq. (3) are reported. They are the minimum time values which guarantee the independence of thermal conductivity of the sonication time and also that the temperature of the bath is lower than 4.5 K of room temperature. In all measurements of the thermal diffusivity, the sample holder was immediately filled after the sonication.

$Vol.Al_2O_3$	V _s [1]	m%[kg]	$P_i[W]$	E _{ms} [kJ/kg]	t _{s%} [min]
20	4.0	6.35	100	110	110
0.1	4.5	4.51	100	110	82
0.5	4.5	4.53	100	110	83
1.0	4.5	4.63	100	110	85
2.0	4.5	4.76	100	110	87
3.0	4.5	4.90	100	110	89
4.0	4.5	5.02	100	110	92
5.0	4.5	5.16	100	110	94

Table 3. Sonication time in minutes for the considered alumina-water nanofluid mixtures.

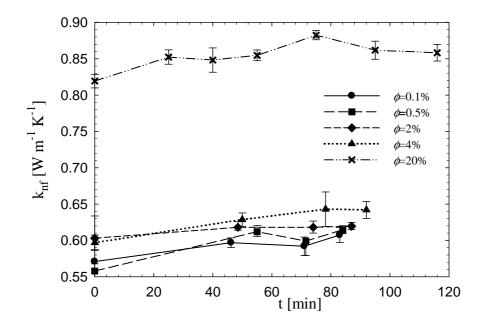


Figure 7. Effect of sonication time on thermal conductivity for different Al₂O₃-water nanofluid mixtures.

Measurements were carried out at room temperature of 25° C for nanofluids mixture at various particle volume concentrations of Al₂O₃. The ratio between the thermal conductivity of nanofluid mixtures and the one related to the pure water are reported in Fig. 8. As expected, the values are greater than 1.0 for all volume concentrations considered and at ϕ =4.0% the increase in thermal conductivity is about 5%. In Fig. 8, a comparison with data from literature are reported for a temperature of nanofluid equal to 25 °C and size of the nanoparticles of 40 nm. The results presented in figure show an excellent agreement with the measurements of Das et al. [30] who utilized temperature oscillation technique for the measurement of thermal diffusivity and thermal conductivity whereas data from Chandrasekar et al. [60], who employed the theoretical Maxwell model, do not match very well. Also the comparison with the data given by Bayat and Nikseresht [61] presents a good agreement between the data. Moreover, Fig. 7 shows that eq. (3) given in [18] is in very good agreement with the present data.

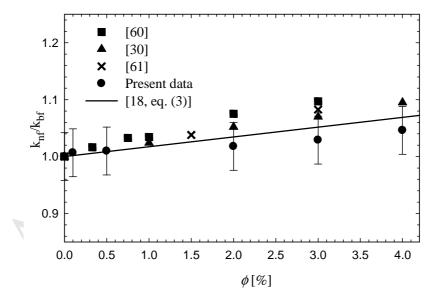


Figure 8. Enhancement of thermal conductivity with respect to the thermal conductivity of water at room temperature of 25°C and comparison with experimental data from refs. [30,60,61] and correlation given in [18].

Thermal conductivity measurements and their relative values in reference to water, as functions of temperature, are reported in Fig. 9, for different volume concentrations. Figures show the thermal conductivity enhancement, with respect to the base fluid, and of nanofluids with temperature for different volume concentrations. The enhancement of nanofluid thermal conductivity with respect to pure water is greater as the temperature increases, as shown in Fig. 9b. With 0,5% particles concentration at room temperature of 25°C the increase is only about 0.57%, but at 65°C this value increases to about 8%. For volume concentration of 4% the enhancement goes from 7.6% to 14.4% with temperature rising from 25°C to 65°C. In this case, the average rate of enhancement increase is much higher compared to that of 0.5% nanofluids. Thus, it can be said that the enhancement of thermal conductivity shows a marked increase with temperature and the rate of this increase depends on the concentration of nanoparticles. The dependence on volumetric concentration of thermal conductivity enhancement for different temperature values is shown in Fig. 10. The effect of volume concentrations on k_{nf} and the ratio k_{nf}/k_{bf} is weaker than the temperature effect. However, the increase is more significant from pure water to a mixture with ϕ =0.1 %. This increment is greater with increasing temperature.

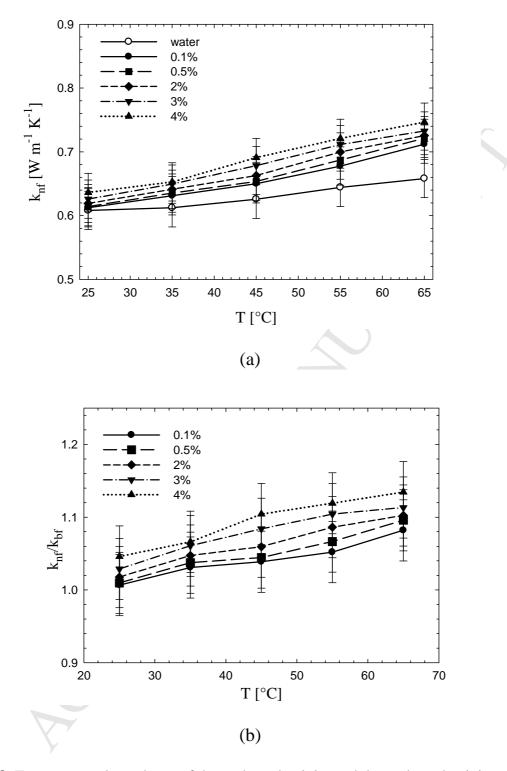


Figure 9. Temperature dependence of thermal conductivity and thermal conductivity ratio for different volume concentration: a) thermal conductivity, b) thermal conductivity ratio.

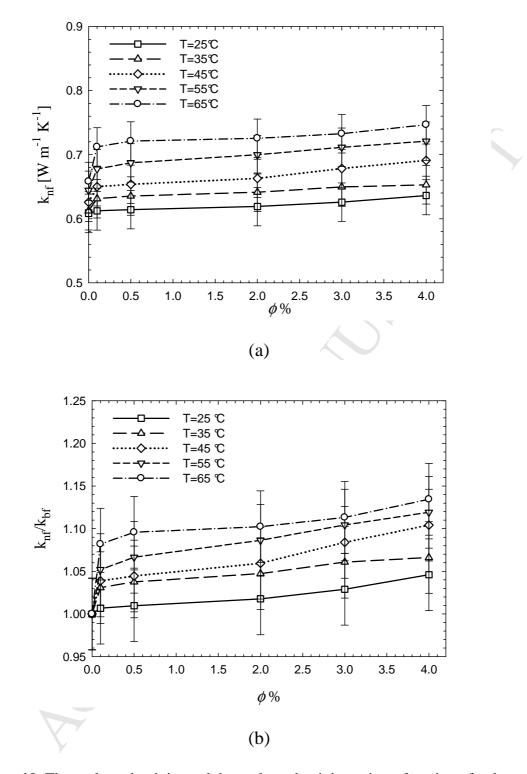


Figure 10. Thermal conductivity and thermal conductivity ratio as function of volumetric concentration for different temperature values: a) thermal conductivity, b) thermal conductivity

ratio.

It is interesting to evaluate the upper and lower limits of thermal conductivity as proposed by Hashin and Shtrikman (H-S) and reported in [47]. The thermal conductivity ratio limits can be written as

$$\frac{k_{ll}}{k_{bf}} = 1 + \frac{3\phi(k_{np} - k_{bf})}{3k_{bf} + (1 - \phi)(k_{np} - k_{bf})}$$
(7)

$$\frac{k_{ul}}{k_{bf}} = \frac{k_{np}}{k_{bf}} \left[1 - \frac{3\phi \left(k_{np} - k_{bf}\right)}{3k_{bf} - \phi \left(k_{np} - k_{bf}\right)} \right]$$
(8)

where k_{ll} and k_{ul} are the lower and upper limits for nanofluid thermal conductivity, respectively; k_{bf} and k_{np} are the thermal conductivity of base fluid (water, in this case) and nanoparticles (Al₂O₃, in the present investigation), respectively; ϕ is the volumetric concentration. As indicated in [47] k_{ll} is evaluated considering a system of plates stacked perpendicular to direction of heat flux whereas k_{ul} corresponds to a system of plates stacked parallel to direction of heat flux. A comparison among thermal conductivity given in [18,30,52,60-62] for two nanoparticle diameters, 40 nm and 120 nm, at mixture temperatures from 21 °C to 65 °C, is reported in Fig. 11. The comparison shows that all data from experimental and theoretical investigations are close to the lower limit, as in Fig. 10a. Present data are in good agreement with correlations proposed in [52] and [62] for a particle diameter equal to 120 nm as reported in Fig. 11b. This diameter value was estimated in previous measurements for the same mixtures reported in [48]. The direct comparisons among present data (pd) and the ones from literature (ld) in terms of measured thermal conductivity, given in Fig.12, with two error ranges $\pm 10\%$ and $\pm 20\%$, show that the greater part of data are in the $\pm 10\%$ error range. The comparisons confirm the value of 120 nm for the nanoparticles diameter. In fact, the correlations proposed in [52] and [62] employed for a nanoparticles diameter equal to 120 nm compared to the present data fall all into the $\pm 10\%$ error range.

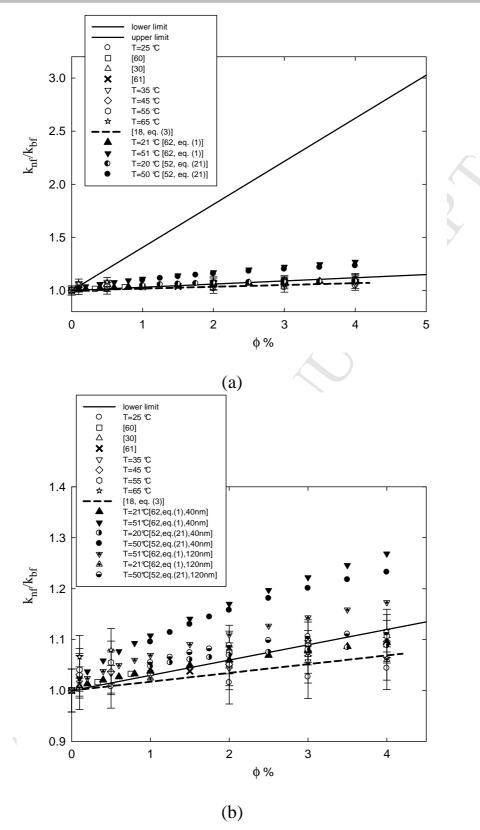


Figure 11. Comparison among thermal conductivity from literature and present data with: (a) upper and lower limits of thermal conductivity by Hashin and Shtrikman (H-S) reported in [47] and (b)

close to the (H-S) lower limit.

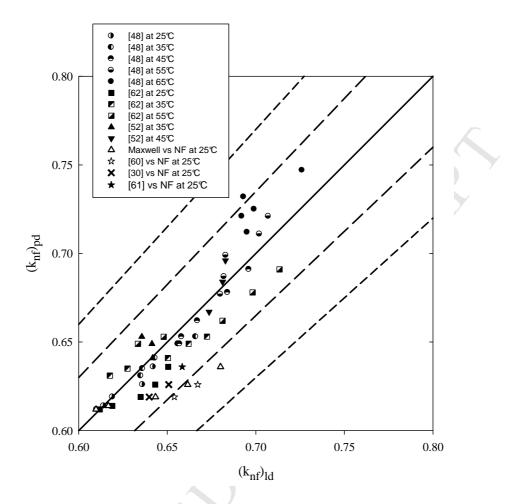


Figure 12. Comparison of the present experimental data and from literature. Error bands are

 $\pm 10\%$ and $-----\pm 20\%$.

Conclusions

Experimental measurements on thermal conductivity of Al₂O₃/water nanofluid mixtures were accomplished by means of the Nanoflash technique for the first time also taking into account the effects of sonication time, temperature and volumetric concentration. Moreover, a simple procedure to obtain the evaluation of minimum sonication time to realize a stable nanofluid mixtures was given. The effect of sonication time on thermal conductivity was investigated experimentally by means of the thermal conductivity measurement which allowed to verify the minimum sonication time estimated by the simple proposed procedure. In fact, it was showed that the agreement between theoretical estimated values and measured ones was very good.

Comparisons among the present measurements with many data reported and correlations, proposed in literature, were performed to validate the nanoflash technique applied to the measurements of thermal conductivity of nanofluids. The comparisons showed a very good agreement between the measured values by nanoflash methods and the existing data from literature. This confirmed the efficiency of nanoflash technique to evaluate the thermal conductivity of nanofluid mixtures. The method presented the following main advantages with respect to the other techniques employed to evaluate the thermal conductivity of nanofluid mixtures: (a) it is a simple method; (b) natural convection is not present inside the liquid sample; (c) the sample has a very small volume and this could be very useful in local analysis of nanofluid mixtures; (d) low uncertainty (<3%) and repeatability of the measurements.

Further, the measurements confirmed the dependence of thermal conductivity on temperature and volumetric concentration, as it was found out that thermal conductivity increased with both increasing temperature and volumetric concentration, as expected. The main indications from the measure values confirm: (a) the enhancement of thermal conductivity due to the temperature increase is more significant than the increase due to volumetric concentration; (b) the greater increase with respect to the volumetric concentration was obtained for lower concentration values, less than 0.2%, whereas the one with respect to the temperature was obtained passing from 55 °C to 65 °C. This suggest that in terms of thermophysical properties, the nanofluid mixtures can be better employed, with greater advantage, at low volumetric concentrations (<0.2%) and high temperatures (>50 °C).

Nomenclature

- c specific heat, $Jkg^{-1}K^{-1}$
- k thermal conductivity, $Wm^{-1}K^{-1}$
- *E* energy, J

- *l* thickness, m
- *m* mass, kg
- *P* mechanical power, W
- *R* depended variable
- t time, s
- *T* temperature, K
- V_s volume, m³

Greek symbol

- α thermal diffusivity, m²s⁻¹
- θ dimensionless temperature, T/T_{max}
- ρ density, kg m⁻³
- τ Fourier number, $\frac{\pi^2 \alpha t}{l^2}$
- ϕ volumetric concentration

Subscript

- *bf* base fluid
- *ll* lower limit
- *ul* upper limit
- m mechanical
- nf nanofluid
- *p* nanoparticle
- s supplied
- tot total

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Research Highlights

- Evaluation of thermal conductivity of nanofluids by Nano-Flash Method is presented;
- Nano-flash technique is a fast and accurate method for measuring thermal conductivity of nanofluids;
- An estimation of the optimal supplied energy and time for sonication is suggested;
- Stability and thermal conductivity enhancements of nanofluids depend on sonication time and energy supplied for fluid preparation;
- Thermal conductivity improves for low volumetric concentrations (<0.2%) and high temperatures (>50 °C);

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