THERMAL CONDUCTIVITY DIFFERENCE BETWEEN NANOFLUIDS AND MICRO-FLUIDS: Experimental Data and Theoretical Analysis Using Mass Difference Scattering

by

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In this work, an experimental campaign on different nanofluids and micro-fluids, obtained by the dispersion of three different metal oxides (CuO, ZnO, and TiO$_2$) with diathermic oil or deionized water has been carried out, in order to extend phonon theory to liquids, as already done in a previous work on Al$_2$O$_3$. Thermal conductivity of stable samples was evaluated by time. The experimental results on thermal conductivity of stable micrometric and nanometric particles suspensions in oil and water showed a further proof of mass difference scattering phenomenon.

Key words: nanofluid, micro-fluid, mass difference scattering, thermal conductivity

Introduction

Heat transfer fluids, such as water, ethylene glycol and oil, are nowadays widely used in the heat transfer field. Heat transfer phenomena, being heavily dependent on liquid’s thermal conductivity, get improved by adding solid particles in suspension, as Maxwell discovered in 1881 [1]. However solid particles, even if at micrometric scale, usually tend to settle if suspended in liquids and, during circulation, they yield erosion issues, due to their high kinetic energy. Both the previous issues can be fixed using nanofluids (NF), suspensions of nanometric particles (NP) in heat-transfer base fluids (BF), studied for the first time by Choi et al. [2].

Thermal conductivity of NF is a topic widely discussed in literature, due to some physical effects by which the suspensions could be affected such as: layering, ballistic phonon motion, Brownian motions, clustering, Kapitza resistance and mass difference scattering (MDS), the last being the main topic of the present work.

Formation of stratified fluid layers surrounding the NP is known as layering phenomenon. Models able to take into account layering phenomenon come from a study of Yu and Choi [3] and Leong et al. [4], but the thickness and thermal conductivity of the stratified structures were only assumed and not experimentally evaluated. A way to determine them was provided by Lee et al. [5], who obtained an equation for thickness and thermal conductivity of the layer, using colloidal science. Another interesting investigation way is represented by molecular dynamics simulation, by which Li et al. [6] estimated a thickness of the layer around NP equal to 0.5 nm. Furthermore, comparison of molecular dynamics simulation’s results with

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...experimental thermal conductivity values, for metal and metal oxide NP (respectively Cu and CuO) in water, takes to the presence of the stratified structures only in the case of metals [7].

Another physical phenomenon, which could affect NF thermal conductivity, happens when the phonon diffusion mechanism in the particles turns from diffusive (typical of the bulk dimensions) to ballistic, when the dimension of NP is smaller than the mean free path (MFP) of the phonon itself. In literature experimental data explanation [8] and theoretical model formulation of thermal conductivity of NF [9] were carried out using ballistic phonon motion.

Brownian motions of NP suspended in the BF could constitute a further way to explain thermal conductivity enhancement of NF, because they could:

- transport energy by moving NP [10];
- cause micro-convection effects [10];
- create NP collisions, which could yield solid to solid heat transfer [11].

Although widely studied in the past, nowadays prevalent literature assumes Brownian motions negligible. Demonstration comes, for example, from Evans et al. [12] using molecular dynamics simulations, Keblinski et al. [11] by considerations on NP diffusion in Brownian motions and thermal diffusion in liquid, and from Gao et al. [13], who experimentally compared thermal conductivity of Al$_2$O$_3$ NF in liquid and solid states (where no Brownian motions are present).

Clustering phenomenon, consisting in formation of NP aggregates, may also be considered as another physical phenomenon which could affect thermal conductivity of NF. In such regards, the distribution of the clusters is very important: percolated and well dispersed structures, promoting heat transfer along a main direction, could take to an increase in thermal conductivity enhancement, as found by Iacobazzi et al. [14], Gao et al. [13], Prasher et al. [15], and Tahmooressi et al. [16]. Thermal conductivity of NP is another important parameter as well, as highlighted by Wu et al. [17], who experimentally characterized thermal behavior of clustered silica NF. Using a theoretical model of prediction, they concluded that clustering can yield benefits if NP thermal conductivity is higher than 10 W/mK (able to yield negligible interfacial resistance) and if fractal dimension is optimized. Hong et al. [18] and Karthikeyan et al. [19] showed that clustering significantly decreases thermal conductivity, due to reduction of effective thermal interaction area. Increasing in cluster dimension yields sedimentation phenomena as well [15], to be avoided in order to guarantee a good use of the NF.

Being NF composed by BF and NP, which form a large interface area, Kapitza resistance [20] has to be considered, since it can be responsible for thermal conductivity decrease (differently from the previously discussed effects, which cause increment in thermal conductivity). Kapitza resistance, $R_K$, is defined as the ratio between the temperature discontinuity at the interface, $\Delta T$, and the thermal power, $\dot{Q}$, flowing across the boundary of surface $A$:

$$R_K = \frac{A \Delta T}{\dot{Q}} \quad (1)$$

The value of $R_K$ depends on the intensity of the interaction NP-BF. The weaker is the atomic bond, the larger is the thermal resistance, as demonstrated by Xue et al. [21] using molecular dynamics simulations, with Lennard-Jones interatomic potential simulating both liquid and solid systems. Furthermore, thermal conductivity, $\lambda$, of NP seems to have importance, as showed by Wu et al [17], who suggested a value larger than 10 W/m K in order to shadow $R_K$’s effect. In such regards, Gao et al. [13] and Iacobazzi et al. [22] confirm this scenario, concluding that Kapitza resistance has a small weight, both of them using Al$_2$O$_3$ NP ($\lambda \approx 25.1$ W/mK [23]) in their experimental campaigns.
Another phenomenon that, as Kapitza resistance, causes a decrement of thermal conductivity is related to MDS (i.e., the phonon scattering at the interface NP-BF). This phenomenon, already highlighted by Kim et al. [24] and Vines et al. [25] for crystalline semiconductor matrix where NP were embedded, has been extended to NF in a work by Iacobazzi et al. [22]. Extension of such a phenomenon to NF means to consider phonon propagation in liquids, feasible through the new phonon theory of Bolmatov-Brazhin-Trachenko [26]. This model returns theoretical heat capacity of several liquids in a wide range of temperature and pressure, in agreement with experimental data. This innovative theory uses Frenkel’s theoretical prediction, which defines the relaxation time, $\tau_F$, as the average time between two consecutive atomic jumps at one point in the space, and admits the existence, for a liquid, of three types of atomic motion, consisting of one longitudinal mode and two transverse modes (shear modes) with frequency $\omega > \omega_F$, where $\omega_F = 2\pi/\tau_F$ is the Frenkel frequency [27-29]. Experimental validation of Frenkel’s theoretical prediction came, after 70 years, from Brazhkin et al. [30].

In this scenario scattering in the NF, responsible for thermal conductivity enhancement decreasing, can be taken into account by Matthiessen’s rule, which allows to evaluate the combined relaxation time of phonons in NF ($\tau_{NF}$):

$$\frac{1}{\tau_{NF}} = \frac{1}{\tau_{BF}} + \frac{1}{\tau_{MDS}}$$

(2)

where $\tau_{BF}$ is the relaxation time of phonons in BF and $\tau_{MDS}$ is the contribution of relaxation time due to the mass-difference impurity related to the presence of particles (scatters).

Being $\tau_{MDS}$ and $\tau_{BF}$ related, respectively, to the number of scatters and to the properties of BF, as consequence, $\tau_{NF}$ is related to the same parameters. Furthermore, if $\tau_{MDS}$ is fixed, its weight on $\tau_{NF}$ rises with $\tau_{BF}$, taking to a bigger influence of a given number of scatters within BF with high $\tau_{BF}$. Therefore, according to eq. (3) [31]:

$$\lambda = \frac{C_v c^2 \tau_{NF}}{3} = \frac{C_v c MFP}{3}$$

(3)

where $C_v$ is the volumetric heat capacity and $c$ is the average velocity of sound in the medium.

Thermal conductivity, $\lambda$, is more affected by the presence of NP in case of high MFP.

Iacobazzi et al. [22] studied thermal conductivity enhancement of Al$_2$O$_3$ suspensions made by micrometric particles (MP) and NP, in different BF (oil and water). Comparing experimental campaign data, the authors found out a variation of thermal conductivity going from micrometric to nanometric scale, $\Delta\lambda_{MF\rightarrow NF}$ (where $MF$ stands for micro-fluid), greater than 0 and larger going from the BF with smaller MFP (oil) to the one at higher MFP (ice). These data, combined with MDS theory previously illustrated, efficiently explain Al$_2$O$_3$ NF thermal conductivity behavior. Purpose of the present work is to extend results obtained using Al$_2$O$_3$ to other materials. In order to do so, CuO, ZnO, and TiO$_2$ (most investigated NP in literature) were considered, even though only stable and well dispersed suspensions were studied by the thermal conductivity point of view (back scattering technique was used to evaluate sedimentation phenomena), how it can be evaluated in the Parameters measured column in tab. 1.

**Experimental set-up**

With the objective of evaluating MDS phenomenon in CuO, ZnO and TiO$_2$ suspensions, all the nanoparticles were dispersed in diathermic oil and deionized water ($pH = 5.1$). The Al$_2$O$_3$, already studied in [22], was dispersed only in oil, in order to confirm MDS at $T = 283$ K.
Working with MP (some of them even characterized by high density, such as ZnO and CuO), stability of the suspension has to be taken into account, being thermal conductivity measurement dependent on sedimentation phenomena [14]. Sedimentation has been evaluated applying the backscattered light spot (BS) technique [32], by means of a Turbiscan LAB – Formulation. The thermal conductivity was instead measured by the hot wire (HW) method, by means of a Flucon LAMBDAsystem (accuracy 1%). The MF considered not stable were not analyzed under the thermal conductivity point of view, and the respective NF was not considered at all, neither for BS nor for thermal conductivity measurements. Table 1 summarizes the samples features, together with the measured parameters (last column). Volume fraction, \( X_v \), of stable samples (\( \text{Al}_2\text{O}_3 \) in oil, \( \text{ZnO} \) in oil and \( \text{TiO}_2 \) in water) was varied in the range 0.5% < \( X_v \) < 4%. The \( \text{TiO}_2 \) in oil, also stable, was studied in the range 1% < \( X_v \) < 2%, due to the impossibility of adding more particles in the NF (getting hardly viscous). The BS of unstable samples, \( \text{CuO} \) MP both in oil and water and \( \text{ZnO} \) MP in water, were measured at \( X_v = 4\% \).

During thermal conductivity measurements, temperature of the sample was kept at a constant value by means of a thermal bath. In particular, samples with water as BF were measured at 293 K, while the ones with diathermic oil were measured at 283 K, in order to get a high value of viscosity and to reduce sedimentation phenomena.

Table 1. List of experimental tests; the Particle mean diameter values are expressed in different ways, being reported as found in the manufacturer datasheets.

<table>
<thead>
<tr>
<th>Item</th>
<th>Material</th>
<th>Particle mean diameter (datasheet) [nm]</th>
<th>Manufacturer</th>
<th>Concentration [vol.%]</th>
<th>Temperature [K]</th>
<th>BF</th>
<th>Measured parameters</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>( \text{Al}_2\text{O}_3 )</td>
<td>45</td>
<td>A.Aesar</td>
<td>0.5</td>
<td>283</td>
<td>Oil</td>
<td>BS, ( \lambda )</td>
</tr>
<tr>
<td>2</td>
<td>( \text{Al}_2\text{O}_3 )</td>
<td>( d_{50} = 500, d_{90} = 2000 )</td>
<td>Almatis</td>
<td>0.5</td>
<td>283</td>
<td>Oil</td>
<td>BS, ( \lambda )</td>
</tr>
<tr>
<td>3</td>
<td>( \text{Al}_2\text{O}_3 )</td>
<td>45</td>
<td>A.Aesar</td>
<td>2</td>
<td>283</td>
<td>Oil</td>
<td>BS, ( \lambda )</td>
</tr>
<tr>
<td>4</td>
<td>( \text{Al}_2\text{O}_3 )</td>
<td>( d_{50} = 500, d_{90} = 2000 )</td>
<td>Almatis</td>
<td>2</td>
<td>283</td>
<td>Oil</td>
<td>BS, ( \lambda )</td>
</tr>
<tr>
<td>5</td>
<td>( \text{Al}_2\text{O}_3 )</td>
<td>45</td>
<td>A.Aesar</td>
<td>4</td>
<td>283</td>
<td>Oil</td>
<td>BS, ( \lambda )</td>
</tr>
<tr>
<td>6</td>
<td>( \text{Al}_2\text{O}_3 )</td>
<td>( d_{50} = 500, d_{90} = 2000 )</td>
<td>Almatis</td>
<td>4</td>
<td>283</td>
<td>Oil</td>
<td>BS, ( \lambda )</td>
</tr>
<tr>
<td>7</td>
<td>( \text{ZnO} ) ≤ 50</td>
<td>S.Aldrich</td>
<td>0.5</td>
<td>283</td>
<td>Oil</td>
<td>BS, ( \lambda )</td>
<td></td>
</tr>
<tr>
<td>8</td>
<td>( \text{ZnO} ) ≤ 76000</td>
<td>S.Aldrich</td>
<td>0.5</td>
<td>283</td>
<td>Oil</td>
<td>BS, ( \lambda )</td>
<td></td>
</tr>
<tr>
<td>9</td>
<td>( \text{ZnO} ) ≤ 50</td>
<td>S.Aldrich</td>
<td>2</td>
<td>283</td>
<td>Oil</td>
<td>BS, ( \lambda )</td>
<td></td>
</tr>
<tr>
<td>10</td>
<td>( \text{ZnO} ) ≤ 76000</td>
<td>S.Aldrich</td>
<td>2</td>
<td>283</td>
<td>Oil</td>
<td>BS, ( \lambda )</td>
<td></td>
</tr>
<tr>
<td>11</td>
<td>( \text{ZnO} ) ≤ 50</td>
<td>S.Aldrich</td>
<td>4</td>
<td>283</td>
<td>Oil</td>
<td>BS, ( \lambda )</td>
<td></td>
</tr>
<tr>
<td>12</td>
<td>( \text{ZnO} ) ≤ 76000</td>
<td>S.Aldrich</td>
<td>4</td>
<td>283</td>
<td>Oil</td>
<td>BS, ( \lambda )</td>
<td></td>
</tr>
<tr>
<td>13</td>
<td>( \text{TiO}_2 ) ≤ 25</td>
<td>S.Aldrich</td>
<td>1</td>
<td>283</td>
<td>Oil</td>
<td>BS, ( \lambda )</td>
<td></td>
</tr>
<tr>
<td>14</td>
<td>( \text{TiO}_2 ) ≤ 45000</td>
<td>S.Aldrich</td>
<td>1</td>
<td>283</td>
<td>Oil</td>
<td>BS, ( \lambda )</td>
<td></td>
</tr>
<tr>
<td>15</td>
<td>( \text{TiO}_2 ) ≤ 25</td>
<td>S.Aldrich</td>
<td>2</td>
<td>283</td>
<td>Oil</td>
<td>BS, ( \lambda )</td>
<td></td>
</tr>
<tr>
<td>16</td>
<td>( \text{TiO}_2 ) ≤ 45000</td>
<td>S.Aldrich</td>
<td>2</td>
<td>283</td>
<td>Oil</td>
<td>BS, ( \lambda )</td>
<td></td>
</tr>
<tr>
<td>17</td>
<td>( \text{TiO}_2 ) ≤ 25</td>
<td>S.Aldrich</td>
<td>1</td>
<td>293</td>
<td>Water</td>
<td>BS, ( \lambda )</td>
<td></td>
</tr>
<tr>
<td>18</td>
<td>( \text{TiO}_2 ) ≤ 45000</td>
<td>S.Aldrich</td>
<td>1</td>
<td>293</td>
<td>Water</td>
<td>BS, ( \lambda )</td>
<td></td>
</tr>
<tr>
<td>19</td>
<td>( \text{TiO}_2 ) ≤ 25</td>
<td>S.Aldrich</td>
<td>2</td>
<td>293</td>
<td>Water</td>
<td>BS, ( \lambda )</td>
<td></td>
</tr>
<tr>
<td>20</td>
<td>( \text{TiO}_2 ) ≤ 45000</td>
<td>S.Aldrich</td>
<td>2</td>
<td>293</td>
<td>Water</td>
<td>BS, ( \lambda )</td>
<td></td>
</tr>
<tr>
<td>21</td>
<td>( \text{TiO}_2 ) ≤ 25</td>
<td>S.Aldrich</td>
<td>4</td>
<td>293</td>
<td>Water</td>
<td>BS, ( \lambda )</td>
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</tr>
<tr>
<td>22</td>
<td>( \text{TiO}_2 ) ≤ 45000</td>
<td>S.Aldrich</td>
<td>4</td>
<td>293</td>
<td>Water</td>
<td>BS, ( \lambda )</td>
<td></td>
</tr>
<tr>
<td>23</td>
<td>( \text{CuO} ) ≤ 45000</td>
<td>S.Aldrich</td>
<td>4</td>
<td>293</td>
<td>Water</td>
<td>BS, ( \lambda )</td>
<td></td>
</tr>
<tr>
<td>24</td>
<td>( \text{CuO} ) ≤ 45000</td>
<td>S.Aldrich</td>
<td>4</td>
<td>283</td>
<td>Oil</td>
<td>BS, ( \lambda )</td>
<td></td>
</tr>
<tr>
<td>25</td>
<td>( \text{ZnO} ) ≤ 76000</td>
<td>S.Aldrich</td>
<td>4</td>
<td>293</td>
<td>Water</td>
<td>BS, ( \lambda )</td>
<td></td>
</tr>
</tbody>
</table>
The steps to prepare and analyze the samples were the following:
– weighing of BF, particles and dispersant (only in water-based NF) with a precision scale (accuracy 0.01 g);
– mixing of suspensions with a magnetic stirrer for 10 min;
– sonication by ultrasonic probe for 20 min at 20 kHz and 70 W;
– thermal conditioning of the NF using thermal bath (according to tab. 1);
– BS and thermal conductivity analysis (only stable samples), the last step was repeated 10 times for reducing the instrumental errors.

Discussion of results

In this experimental campaign, the same hypothesis of negligibility of layering, Brownian motions, clustering and Kapitza resistance phenomena used by Iacobazzi et al. [22] have been assumed, except for the ballistic phonon motion one, for which some explanations are required. Negligibility of the aforementioned phenomenon was assumed in [22] basing on the comparison between Al$_2$O$_3$ NP dimension (45 nm) and phonon MFP ($\approx$ 35 nm), and MFP value was evaluated by Debye theory [33]:

$$MFP = \frac{10aT_m}{\gamma T}$$

where $a$ is the lattice constant, $\gamma$ – the Gruneisen parameter, $T_m$ – the melting point, and $T$ – the temperature. In the same way, negligibility of ballistic phonon transport can be assumed in this campaign, simply comparing dimension of the NP, $d$, to the phonon MFP, both reported in tab. 2 for all materials considered stable, based on BS analysis. In calculating MFP by eq. (4), $T$ and $\gamma$ were assumed respectively 300 K and 1. By comparison, MFP results always smaller than $d$, taking to ballistic phonon motion phenomenon negligibility.

Unlike Al$_2$O$_3$, the other materials involved in the present experimental campaign are characterized by a high bulk density (particularly ZnO and CuO), which can be responsible for particle sedimentation and convective phenomenon around the platinum wire of the HW system, affecting thermal conductivity measurements [14]. In order to exclude samples with such a problem, thus not being measurable by HW, sedimentation was studied by means of BS light spot technique. In particular, $\Delta BS$ (backscattered signal) with respect to the height of the cuvette, $h$, was evaluated. Referring to fig. 1 the region of interest for BS measurements (zone highlighted with green in Turbiscan cuvette) was set in order to be analogous to the HW one, delimited by $H_1$ and $H_2$. So, $h_1$ and $h_2$ values were carried out simply using proportion calculation, as follows:
– $h_1 = H_1 \times d/D = 15$ mm
– $h_2 = H_2 \times d/D = 40$ mm

Furthermore, $\Delta BS$ was also correlated to the time elapsed from sonication, measuring up to

<table>
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<tbody>
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<td>45</td>
<td>37.4</td>
<td>0.478</td>
<td>2345</td>
</tr>
<tr>
<td>ZnO</td>
<td>50</td>
<td>24.4</td>
<td>0.325</td>
<td>2248</td>
</tr>
<tr>
<td>TiO$_2$</td>
<td>32</td>
<td>26.1</td>
<td>0.37</td>
<td>2116</td>
</tr>
</tbody>
</table>

Table 2. Dimension of the NP, $d$, phonon MFP in the considered materials and values used for evaluation, using Debye theory

Figure 1. Geometric comparison of HW system and Turbiscan cuvette, useful to evaluate region of interest of BS analysis
1200 seconds, duration of thermal conductivity measurement. Measurements of samples considered stable and suitable for thermal conductivity evaluation, are showed in the following figures and are related to micrometric and nanometric Al\textsubscript{2}O\textsubscript{3} particles in oil, fig. 2, ZnO particles in oil, fig. 3, and TiO\textsubscript{2} particles, both in oil, fig. 4, and water, fig. 5. All previous ΔBS trends do not exhibit variation, with respect to the elapsed time, larger than 0.7%, in the measur-

Figure 2. The ΔBS with respect to the height of the cuvette, h, and the elapsed time since the sonication for; (a) micrometric and (b) nanometric Al\textsubscript{2}O\textsubscript{3} in oil, X\textsubscript{v} = 4\%, T = 283 K
(for color image see journal web site)

Figure 3. The ΔBS with respect to the height of the cuvette, h, and the elapsed time since the sonication for; (a) micrometric and (b) nanometric ZnO in oil, X\textsubscript{v} = 4\%, T = 283 K
(for color image see journal web site)

Figure 4. The ΔBS with respect to the height of the cuvette, h, and the elapsed time since the sonication; (a) micrometric and (b) nanometric TiO\textsubscript{2} in oil, X\textsubscript{v} = 2\%, T = 283 K
(for color image see journal web site)
ing range $0 < t < 1200$ seconds. Among them, nanometric TiO$_2$ either in oil or water exhibited more instability with respect to the other materials, although not having an influence on thermal conductivity, as shown in fig. 7, where no convective trends are appreciable.

As previously said, a further indirect criterion to check the discussed samples’ stability comes from the measured thermal conductivity enhancement with respect to the elapsed time since the end of sonication ($0 < t < 1200$ seconds), by which convection around the HW can be highlighted. In fact, convective motions around the HW, due to sedimentation, can be responsible for generating unstable and overestimated measurements [14]. Using previously criteria, a validation of stability is verified for:

- Al$_2$O$_3$ in oil at $X_v = 4\%$ and $T = 283 K$ (fig. 6);
- ZnO in oil at $X_v = 4\%$ and $T = 283 K$ (fig. 6);
- TiO$_2$ in oil at $X_v = 2\%$ and $T = 283 K$ (fig. 7);
- TiO$_2$ in water at $X_v = 4\%$ and $T = 293 K$ (fig. 7);

appearing thermal conductivity enhancement with respect to elapsed time trends almost smooth in all cases.

On the other hand, ΔBS measurements of micrometric CuO both in water and oil, showed, respectively, in figs. 8(a) and 8(b), take to ΔBS variation with respect to the time. Furthermore, a strong instability can be appreciated in micrometric ZnO-water case too, where a Type C sedimentation profile [34] can be observed, characterized by constant rate of particle sedimentation, particles’ rearrangement and compression phenomena (see sedimentation front

Figure 5. The ΔBS with respect to the height of the cuvette, $h$, and the elapsed time since the sonication; (a) micrometric and (b) nanometric TiO$_2$ in water, $X_v = 4\%$, $T = 293 K$

(for color image see journal web site)

Figure 6. Thermal conductivity enhancement with respect to the elapsed time since the end of sonication of micrometric (micro) and nanometric (nano) Al$_2$O$_3$ and ZnO in oil, $T = 283 K$, $X_v = 4\%$
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propagation in fig. 8(c). Therefore, micrometric CuO suspensions, both in oil and water, and micrometric Zinc oxide suspension in water can be considered not suitable for HW measuring.

Finally, thermal conductivity enhancements of stable suspensions, micrometric and nanometric Al$_2$O$_3$ in oil, ZnO in oil and TiO$_2$ both in oil and water, have been evaluated. The Al$_2$O$_3$ in oil was measured at $T = 283$ K, fig. 9. From the results it is evident that $\Delta\lambda_{\text{MF-NF}} = 4.3\%$ at $X_v = 4\%$, confirming data of previous work [22]. Analysis of ZnO in oil at $T = 283$ K, fig. 9, confirms MDS phenomenon presence and takes to $\Delta\lambda_{\text{MF-NF}} = 5.8\%$ at $X_v = 4\%$, even higher than the case of Al$_2$O$_3$ in oil. Furthermore, ZnO allowed to reach highest thermal conductivity value (both for NP and MP) if compared with the other materials investigated. Differently from Al$_2$O$_3$ and ZnO, TiO$_2$ does not allow to overcome $X_v = 2\%$, due to impossibility to disperse NP in oil. For this reason $X_v$ was varied up to 2%, taking to graph of fig. 10, that confirms MDS phenomenon presence and takes to $\Delta\lambda_{\text{MF-NF}} = 1\%$ at $X_v = 2\%$ and at $T = 283$ K. Finally, MDS phenomenon was once more confirmed in TiO$_2$-water case, studied up to $X_v = 4\%$, at $T = 293$ K. By results, showed in fig. 10, $\Delta\lambda_{\text{MF-NF}} = 2.6\%$ can be appreciated at $X_v = 4\%$, that is less than $\Delta\lambda_{\text{MF-NF}} = 6.9\%$ of Al$_2$O$_3$ in water, obtained in [22].
Conclusions

This experimental campaign has been conceived to extend the MDS theory to suspensions of CuO, ZnO, and TiO$_2$ nanometric and micrometric particles in diathermic oil and water. The obtained suspensions have been selected, before considering the experimental thermal conductivity measurements, under the stability point of view and only the stable suspensions have been analyzed for this campaign, i.e. ZnO in oil and TiO$_2$ both in oil and water. In a previous work, which demonstrate MDS for Al$_2$O$_3$, measurements in liquid state were carried out at 293 K. In the present work, to limit sedimentation phenomenon, oil-based suspensions were analyzed at 283 K, temperature at which oil viscosity is higher with respect to the one at 293 K (dynamic viscosity increases from 123.47 mPa·s to 344.26 mPa·s), thus Al$_2$O$_3$ in oil was once more studied, to confirm MDS at 283 K. The results showed clearly that MDS occurs in these cases and that the increasing in thermal conductivity for all the considered metal oxides is higher for the suspensions with micrometric particles than nanometric ones at the same conditions.

Nomenclature

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
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</thead>
<tbody>
<tr>
<td>$A$</td>
<td>surface, [m$^2$]</td>
</tr>
<tr>
<td>$d$</td>
<td>particles mean diameter, [m]</td>
</tr>
<tr>
<td>$h$</td>
<td>height of the cuvette in the backscattering analysis, [mm]</td>
</tr>
<tr>
<td>$R_K$</td>
<td>Kapitza resistance, [m$^2$K·W$^{-1}$]</td>
</tr>
<tr>
<td>$Q$</td>
<td>thermal power, [W]</td>
</tr>
<tr>
<td>$P$</td>
<td>temperature, [K]</td>
</tr>
<tr>
<td>$X_v$</td>
<td>volumetric fraction of the suspension</td>
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</table>

Greek symbols

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\lambda$</td>
<td>thermal conductivity, [W·m$^{-1}$K$^{-1}$]</td>
</tr>
<tr>
<td>$\tau_{BF}$</td>
<td>relaxation time of phonons in base fluid, [s]</td>
</tr>
<tr>
<td>$\tau_F$</td>
<td>Frenkel relaxation time, [s]</td>
</tr>
</tbody>
</table>
\[ t_{\text{MDS}} \] \text{– contribution of relaxation time due to the mass-difference impurity, [s]}
\[ t_{\text{NF}} \] \text{– relaxation time of phonons in nanofluid, [s]}
\[ \omega \] \text{– frequency, [s\(^{-1}\)]}
\[ \omega_F \] \text{– Frenkel frequency, [s\(^{-1}\)]}

**Acronyms**

- BF – base fluid
- BS – Backscattered light spot
- HW – hot wire
- MDS – mass difference scattering
- MF – micro-fluid
- MFP – mean free path
- MP – microparticle
- NF – nanoparticle
- NP – nanoparticle

**References**


