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Characterization of physical properties of nanofluids for heat transfer application

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Abstract. Water based nanofluids of SiO_2 and Al_2O_3 were characterized regarding its later use in heat transfer applications. Well dispersed nanofluids were prepared at three different volume fractions (0.5% v., 1% v., 5% v.) and for each one the experimental measurements were carried out at three different temperatures (40ºC, 60ºC, 80ºC). Commercial nanofluids acquired in liquid state were compared with nanofluids prepared by dispersing dry powder in water. The thermal conductivity, the viscosity and the specific heat of all the prepared nanofluids were measured in order to calculate the Mouromtseff number and establish the optimal conditions. Finally, the stability of the nanofluids was also studied through the evolution of the amount of light scattered by the sample during a period of time.

1. Introduction

The concept of adding small solid particles into a base fluid to increase the thermal conductivity of the suspension has been practiced for a long time [1]. However, most of the studies were performed using suspensions of millimeter- or micrometer- sized particles, which led to problems such as poor suspension stability and channel clogging, which limits its practical applicability. In 1995, S.U.S. Choi proposed the use of nanofluids to increase the thermal conductivity of heat transfer fluids [2]. Nanofluids are defined as dilute suspensions with solid particles smaller than 100 nm. Nanofluids present some important advantages over the conventional colloidal suspensions such as high stability, reduced particle clogging and high heat transfer capabilities.

However, the industrial applicability of using nanofluids in heat transfer systems is limited by the increase of the viscosity of the fluid (which increases the pumping power). For particles in the nanometer range the surface forces become predominant and the interactions are controlled by short-range forces like Van der Waals attraction force [3]. These interactions lead to the formation of agglomerates and the increase in the viscosity of the fluids.

Therefore, it is of great importance the combined study of the thermal properties (thermal conductivity and specific heat) and the rheological properties (viscosity). In the last decade, several authors have reported results of thermal conductivity of oxide nanofluids (Al_2O_3, SiO_2 and CuO mainly) [4-11]. Most of them study the evolution of the thermal conductivity with the solid content. However, these studies were carried out at room temperature. The few ones that analyse the evolution of the thermal conductivity with the temperature are limited to temperatures up to 50ºC.

The rheological behaviour of nanofluids has also been previously studied [6, 10, 12-16]. The influence of the solid content, the pH of the nanofluids and the addition of surfactants or electrolytes was analysed. However, the range of temperatures under study were in all cases lower than 60ºC, and in most of them room temperature was chosen.

The use of the nanofluids in heat transfer devices or installations involves working at high temperature conditions. Therefore, it is of great interest to characterize the nanofluids and to analyse the evolution of the physical properties of interest at those working temperatures.

In this work water based nanofluids of SiO_2 and Al_2O_3 were characterized at different volume fractions (0.5% v., 1% v., 5% v.). The evolution of the thermal conductivity, the specific heat and the viscosity of the nanofluids was measured at high temperature conditions (40ºC, 60ºC, 80ºC). Commercial nanofluids acquired in liquid state were compared with nanofluids prepared by dispersing dry powder in water. The thermal conductivity was measured using a commercial device (KD2 pro) based on the hot transient wire technique. A new processing of the data registered by the apparatus had to be developed in order to obtain reliable results for measurements at high temperature conditions and very low viscous fluids.

The main aim of this work was to use the measurements of all the properties analysed to maximize the Mouromtseff number $Mo = k^{0.6} \cdot \rho^{0.8} \cdot c_p^{0.4} / \eta^{0.4}$, since it provides the best thermal performance with the lowest penalty in the pumping power [17].

Finally, the best nanofluid is that one which has not only the best thermal behavior, but also a good stability. Therefore the stability of the nanofluids was also studied through the evolution of the amount of light scattered by the sample during a period of time.

2. Characterization of materials and preparation of nanofluids

Tests were carried out with silica and alumina water-based nanofluids. Well dispersed suspensions were used. In order to achieve the best dispersion state, nanofluids were electrostatically stabilized and therefore, the pH value was modified to be far from the isoelectric point determined for each material. The dispersion was performed using an ultrasonic probe (UP400s from Hielscher Ultrasonics Gmbh, Germany) which has been confirmed as the most effective dispersion system [18].

Two different sorts of nanofluids were prepared:

- Commercial nanofluids were acquired in liquid form and diluted to achieve the desired volume fraction. In this case, after dilution with distilled water, the pH value was adjusted. Finally, the nanofluid was submitted to sonication treatment for 2 minutes.

- Commercial nanoparticles were purchased in dry powder and dispersed in distilled water. This method is known as the two-step method. In this case, initially, the mixture of nanoparticles and the water was submitted to a sonication treatment for 3 minute to disperse all the material and to break the agglomerates. After that, the *pH* of the nanofluid was modified by adding HCl or NaOH solutions. Finally, to ensure correct dispersion of all the components, the nanofluids were submitted to a second sonication treatment for 2 minutes.

The characteristics of the materials used are described in next subsections.

2.1. Silica (SiO_2)

The commercial silica nanofluid used was Ludox TM-50 provided by Sigma-Aldrich. The primary particles have a diameter of 10 nm and a specific surface of 140 m^2/g according to the manufacturer. The particle size of the real agglomerates present in the suspension was measured by the Dynamic Light Scattering technique (DLS) using a Zetasizer Nano ZS (Malvern Instruments Ltd., UK). A median particle size of 38.9 nm was found. The obtained particle size distribution was narrow and the particles were quite well dispersed

The commercial powder purchased was a fumed silica, Aerosil 200, provided by Degussa. The powder is composed of amorphous hydrophilic silica nanoparticles with primary units of 12 nm and a specific surface of 200 m^2/g according to the manufacturer. The particle size of the real agglomerates

present in the suspension was also measured by DLS and a median particle size of 201 nm was found. The 90% of the particles had a diameter lower than 345 nm.

To determine the isoelectric point of the silica nanofluids, the zeta potential of suspensions prepared under different conditions of *pH* was measured using the Zetasizer Nano ZS. All the measurements were carried out in dilute suspension. The isoelectric point was found at *pH* = 2. Therefore, all the silica nanofluids were prepared at *pH* = 9, to ensure a good dispersion.

2.2. Alumina (Al₂O₃)

2.2. Alumina (Al_2O_3)

The commercial alumina nanofluid used was Aerodisp W925 provided by Degussa. The primary particles have a diameter of 11 nm according to the manufacturer. The particle size of the real agglomerates present in the suspension was measured by DLS. A median particle size of 127 nm was found. The 90% of the particles had a diameter lower than 218 nm.

Alumina nanoparticles (Aeroxide Alu C) were also supplied by Degussa, with an average primary particle size of 13 nm, as reported by the manufacturer. The particle size of the real agglomerates present in the suspension was also measured by DLS and a median particle size of 128 nm was found. The 90% of the particles had a diameter lower than 184 nm. Comparing this value with the Aerodisp nanofluid, it can be seen that there are agglomerates of lower size and also that the dispersion is better.

The zeta potential of the alumina nanoparticles was also measured using the Zetasizer Nano ZS. The isoelectric point was found at *pH* = 8.2. Therefore, all the alumina nanofluids were prepared at *pH* = 4, to ensure a good dispersion.

3. Experimental techniques

3.1. Thermal conductivity, *k*

The thermal conductivity of all nanofluids was measured using a KD2 Pro conductimeter (Decagon Devices Inc.). The KD2 Pro is the commercial device that measures the thermal conductivity with the help of the transient hot wire technique. The sample was introduced in a sealed glass tube (22 ml) where the sensor was inserted vertically. It is very important to ensure the vertical position of the needle to reduce convective heat transfer inside the sample and improve the accuracy of the measurements, especially for low viscosity nanofluids. The heat conductance is inversely related to the characteristic dimension of the probe inserted into the fluid which depends on the direction of fluid flow over the probe. When considering a heated probe inserted into a cooler fluid, the fluid flow near the probe from free convection will be upward, as the warmer, less dense material near the probe is forced upwards by forces of gravity working on the surrounding, denser material. If the needle is inserted into a fluid vertically, the fluid flow will be parallel to the axis of the needle, and the characteristic dimension is the length of the needle, not the diameter, thus decreasing the free convection.

To carry out the test at high temperature conditions the tube is submerged in a thermostatic bath where the temperature is controlled. From the Grashof number, it is known that free convection is proportional to the temperature difference between the fluid and the object inserted into it. Hence, free convection can be decreased by limiting the heating of the needle. The KD2 Pro KS-1 sensor is specifically designed for use in fluids, and provides a very small heat pulse. The reading time was set at 1 minute to minimize the temperature difference between the fluid and the needle, being the maximum difference obtained of 0.3 degrees. A period of time of 1 hour was waited for the sample to reach the desired temperature before any test. After that time, 6 measurements were done for each nanofluid sample. Within this time the bath was switched off. It was important to wait for about fifteen minutes between readings to recover the temperature after the heat pulse applied to the nanofluid.

The KD2 Pro uses special algorithms to analyze measurements made during a heating and a cooling interval. The device was specially design to measure the thermal conductivity of solids or liquids at low temperatures where the temperature is not affected by external changes. However, in this case where the sample is introduced in a bath, when the bath is switched off to avoid vibrations, the decrease of the bath temperature during the reading time influences the evolution of the fluid

temperature when the heat pulse is applied. Therefore, the internal equations of the KD2 Pro and the values registered by it could not be used because in the fitting of the data, as those influences and contributions were not taken into account.

Therefore, complete evolution of the temperature of the needle with time was obtained for all tests, and data were fitted to the following equations. For the heating pulse:

$$T = m_0 + m_2 t + m_3 \ln t \tag{1}$$

where m_0 is the ambient temperature during heating, m_2 is the rate of background temperature drift and m_3 is the slope of a line relating temperature rise to logarithm of temperature.

During cooling the model is:

$$T = m_0 + m_2 t + m_3 \ln(\frac{t}{t - t_h}) \tag{2}$$

where t_h is the total cooling time.

The thermal conductivity can be computed from the parameter m_3 in both curves, heating and cooling:

$$k = \frac{q}{4\pi m_3} \tag{3}$$

where k is the thermal conductivity and q is the power of the heat pulse applied.

For nanofluids with high thermal conductivity, the heat generated in the needle when the pulse is applied, is removed faster and the increase of temperature in the needle is lower (lower value for m_3). In this way, it is very important to reduce the free convection because when there exist fluid flow inside the tube, the fluid removes the heat from the needle even faster and the thermal conductivity is increased due to the contribution of the convective heat transfer.

3.2. Specific heat, c_P

The specific heat for each nanofluid was measured in a Differential Scanning Calorimeter (DSC), model DSC1 (Mettler Toledo, USA). The calculation of the specific heat capacity is based in the DIN standard (DIN 51007),

The sequence used in the determination was as follows: isotherm of 5 minutes at 25ºC, dynamic segment from 25ºC to 95ºC at heating rate of 10ºC/min and isotherm of 5 minutes at 95ºC.

As a consequence of the nature of the sample (liquid) the crucible (Aluminium) was sealed in order to avoid loss of material by evaporation.

3.3. Viscosity, η

The viscosity and rheological behaviour of nanofluids were obtained by conducting tests under steady state conditions using a Haake RheoStress 1 rotational rheometer (Thermo Scientific). A cylinders system composed of two concentric cylinders was used. In the gap between the inner cylinder (diameter = 34 mm) and the outer cylinder (diameter = 36.88 mm) the sample was introduced. Before each test, a pre-treatment, in which the samples were submitted to a constant shear stress, was applied to the nanofluids for 30 seconds to ensure similar starting conditions for all the measurements.

3.4. Stability

The stability of the nanofluids was analysed through the evolution of the amount of light backscattered by the nanofluid from an incident laser beam. A Turbiscan Lab Expert (Formulaction SA, France) was used to carry out the tests. Measurements are based on the multiple light scattering theory. This equipment consists of a pulsed near-infrared light source and a detector that measures the light backscattered by the sample. For each nanofluid, the backscattering profiles were obtained along the height cell. To analyse the stability of nanofluids the measurements were carried out at different time intervals up to a total time of 48 hours.

## 4. Results and discussion

4.1. Thermal conductivity, *k*

Figure 1 shows the evolution of enhancement of the thermal conductivity of the nanofluid, k_n, related to the conductivity of the water, k_w. In Figure 1(a) results comparing silica nanofluids can be observed. The enhancement of thermal conductivity increases with the solid content due to the higher number of particles present in the suspension and the higher number of contacts between them [19]. However, the thermal conductivity decreases at 80ºC in almost all cases. Only for the Aerosil suspension at 5% of volume fraction the conductivity remains constant and its value is the highest obtained. This suspension is the one in which the particles are more agglomerated due to the high solid content. The Ludox nanofluid provides worse conductivities because of the better dispersion. In Figure 1 (b) results comparing alumina nanofluids can be observed. The enhancement of thermal conductivity is the same at 40 ºC for both materials. However, for the Aerodisp the enhancement of thermal conductivity remains constant and only increases at high solid content due to the formation of agglomerates. For the Aeroxide nanofluids, the thermal conductivity always decreases. The abnormal behaviour of the thermal conductivity at high temperatures is related to the solubility of the nanoparticles and the procedure of measurement. In the nanometrical size range kinetics of dissolution of particles is enhanced due to the small size according to the Kelvin equation [20, 21]. The dissolution increases with temperature, degree of dispersion and time. So, at high temperature, more solid is dissolved and the conductivity decreases. This decrease is higher for better dispersed nanofluids. Also the procedure of measurement influence, because of the time that the sample remains inside the bath at high temperature. This new variable influencing conductivity must be further studied in detail. However, a long test in which the sample has remained inside the bath more time than the usual was performed and the solubility decreases from 0.693 W/m·K to 0.658 W/m·K. Therefore, the time at which the sample is submitted to high temperature must be taken into account and must be similar to those found in real applications.
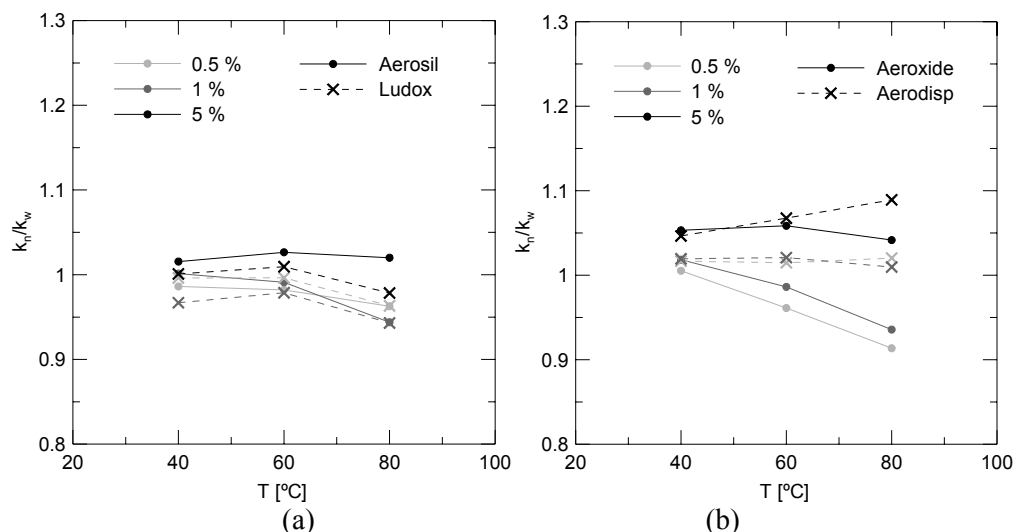
Figure 1. Evolution of thermal conductivity enhancement with solid content and temperature for (a) SiO_2 and (b) Al_2O_3.

4.2. Specific heat, c_P

Figure 2 shows the evolution of the specific heat of all nanofluids with the temperature and the solid content. The specific heat describes the amount of heat required to increase the temperature of the sample by a given amount. As expected, the specific heat increases with increasing the temperature and decreases with increasing the solid content, due to the lower specific heat of the solid phase in relation to the liquid phase. In general, nanofluids prepared from the dry powder present a lower specific heat than the commercial nanofluids tested.
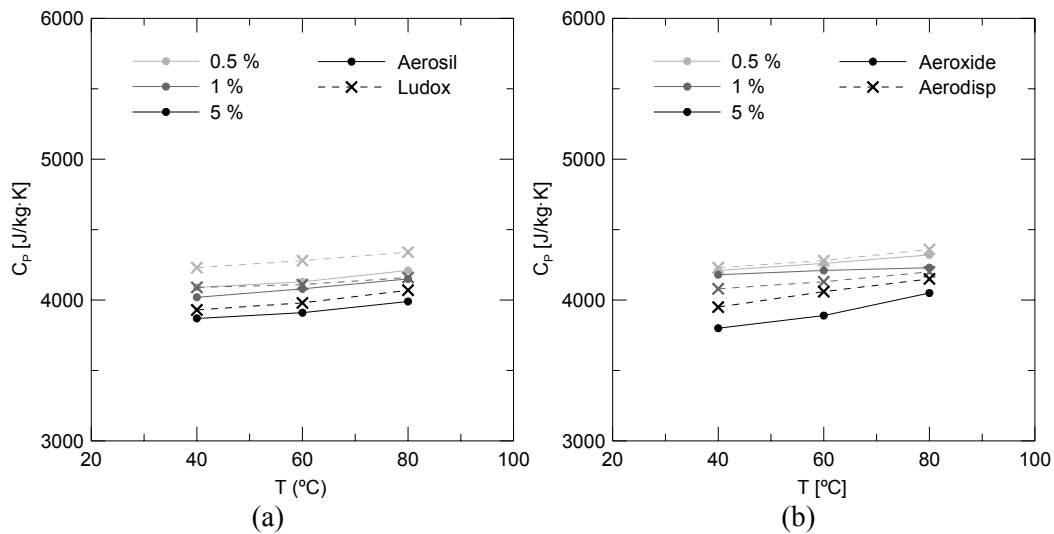
Figure 2. Evolution of specific heat with solid content and temperature for (a) SiO$_2$ and (b) Al$_2$O$_3$.

4.3. Viscosity, η

Figure 3 shows the evolution of the viscosity of all the nanofluids prepared. As a general rule the viscosity of the nanofluid increases with solid content and decreases with temperature. In Figure 3 (a) it can be observed that the Ludox suspensions are very low viscous due to the high degree of dispersion. The Aerosil suspensions present viscosities higher than the Ludox, being the Aerosil at 5% of volume fraction the most viscous. In Figure 3 (b) it can be observed that both sorts of alumina nanofluids have similar viscosities, being the Aerodisp nanofluid slightly more viscous than the Aeroxide one.
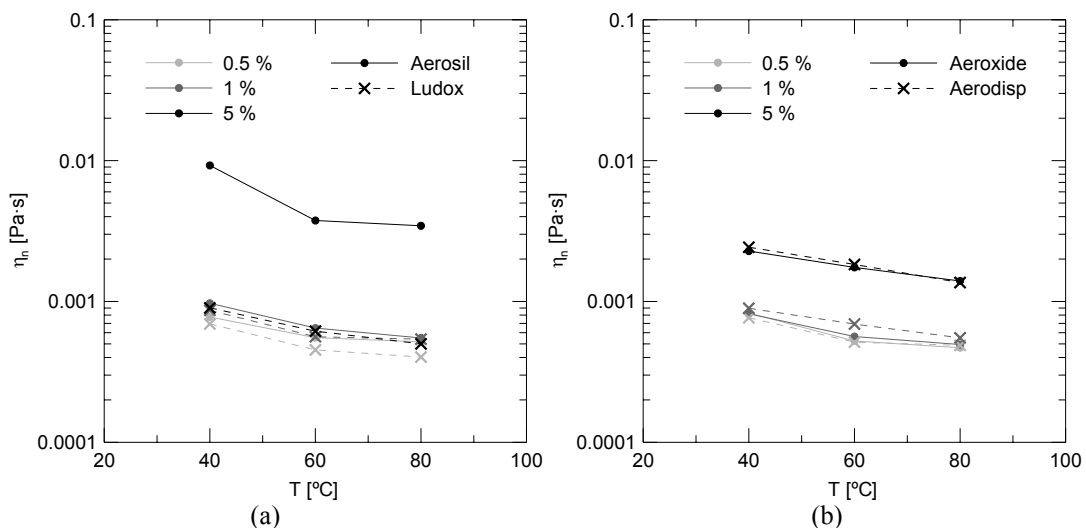

Figure 3. Evolution of viscosity with solid content and temperature for (a) SiO$_2$ and (b) Al$_2$O$_3$.

4.4. Stability

Figure 4 shows the evolution of the amount of light backscattered from the sample (*BS*) with time, at the centre of the sample. This property is proportional to the turbidity of the sample and its change with time means that particles agglomerate. As a general rule, the backscattering increases with the solid content and it can be observed that the influence of the temperature is negligible. Values are similar for silica nanofluids. However, for alumina the turbidity of the Aeroxide is always higher than

for the Aerodisp. The most important conclusion is that all nanofluids were stable and their state of agglomeration did not change with time.
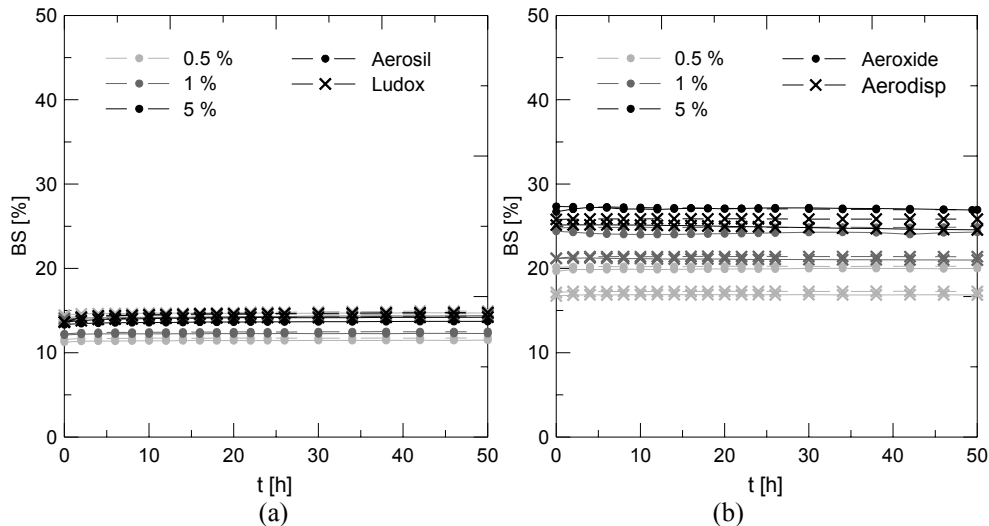
Figure 4. Evolution of the backscattering with time and solid content for (a) SiO$_2$ and (b) Al$_2$O$_3$.
(dashed line $T = 40$ºC, solid line $T = 60$ºC)

4.5. Optimization

In order to chose the nanofluids providing the best thermal behaviour, the Mouromtseff number (Mo=k$^{0.6}$·ρ$^{0.8}$·C$_p^{0.4}$/η$^{0.4}$) was calculated. In Figure 5 it can be observed that, despite the decrease of the thermal conductivity with the temperature, the Mouromtseff number increases when increasing the temperature. That means that the thermal conductivity is not a key factor if the specific heat and the viscosity have the suitable behaviour. Comparing commercial nanofluids with those prepared from the dry powder, it can be observed that for the silica, the Aerosil at 5% of volume fraction provides the best behaviour, while for the alumina the Aerodisp at 5% of volume fraction is the one which provides the best behaviour. This last mentioned nanofluid is the best of all the nanofluids tested.
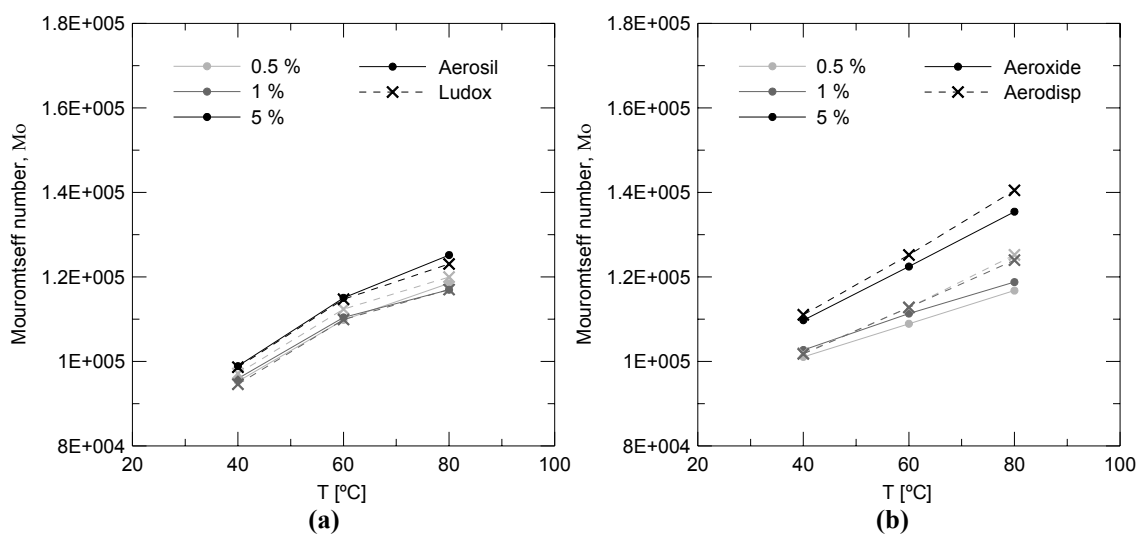
Figure 5. Evolution of Mouromtseff number with solid content and temperature for (a) SiO$_2$ and (b) Al$_2$O$_3$.

5. Conclusions

Silica and alumina nanofluids acquired in liquid and prepared from the dry powder were characterized in terms of thermal conductivity, specific heat, viscosity and stability.

The evolution of the thermal conductivity with the temperature presents an abnormal behaviour at high temperature (80ºC). At this temperature the solubility of the nanoparticles can be the cause of the decrease of conductivity. The time at which the nanofluid remains at high temperature influences the conductivity and hence must be considered and set at a value similar to those in real application.

The specific heat has the expected trend and increases with temperature. Also the viscosity decreases with temperature as expected. That caused an increment in the Mouromtseff number, despite the abnormal behaviour of the thermal conductivity.

The optimal silica nanofluid providing the best thermal behaviour was found to be the Aerosil at 5% of volume fraction, while the optimal alumina nanofluid was the Aerodisp at 5% of volume fraction. The last one presented the best behaviour among all the tested nanofluids.

The stability measurements conclude that all nanofluids were stables for at least 48 h and no agglomeration was observed within this time.

Acknowledgments

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