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CHARACTERIZATION OF PHYSICAL PROPERTIES OF Al_2O_3 AND ZrO_2 NANOFLUIDS FOR HEAT TRANSFER APPLICATIONS

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ABSTRACT

Studies demonstrate that nanofluids based on metal oxide nanoparticles have physical properties that characterize them as promising fluids, mainly, in industrial systems in which high heat flux takes place. Water based nanofluids of Al_2O_3 and ZrO_2 were characterized regarding its promising use in heat transfer applications. Three different concentrations of dispersed solutions of cited nanofluids were prepared (0.01% vol., 0.05% vol., and 0.1% vol.) from commercial nanofluids. Experimental measurements were carried out at different temperatures. Thermal conductivity, viscosity and density of the prepared nanofluids were measured.

1. INTRODUCTION

Heat transfer augmentation in engineering processes has been a major challenge for designers of certain strategic sectors like computers microprocessors, electronics, automotive, aviation, among others. A major effort is underway to increase the heat transfer capacity of such systems (macro and micro scales) [1].

Studies are in progress to intensify the heat transfer process between a fluid and a surface for cooling or heating. Among the most studied techniques we can mention the use of structured surfaces, evaporation or condensation of the fluid to reduce the dimensions of the channel characteristics, the mixture of one or more techniques in addition to improve heat transfer capacity [1].

Concerning increase of power density, new materials, cooling fluids, and more efficient heat transfer techniques are in constant development to attend such challenge. Furthermore, due to energy economic aspects, the development of more efficient cooling or heating systems implies on costs-reducing design, manufacture, construction and operation, without losing sight of safety

An example of a critical system in which nanofluids should be efficient for cooling is a nuclear reactor [1]. In this context, nanofluids are a promising technology for application in nuclear reactor systems for high heat flux transport.

Concerning the increasing of efficiency of thermal system, the criterion of thermal quality include pumping power, heat flux removing capacity, and fluid thermal transport capacity, being a commitment between these characteristics. The third criterion, but not less important, is the economic criterion, which regards production, storage and treatment costs.

As demonstrated by recent researches, nanofluids have very interesting physical properties with respect to its ability for heat removing and transport. At present there are research groups in the world conducting investigations on the influence of ionizing radiation on nanofluids and the possibility of its use as cooling fluid of nuclear reactors core in cases of accidents [2-13]. These studies are still focused on more precise knowledge about the thermophysical properties without and with the action of ionizing radiation. In addition, other researchers have investigated the nanofluids heat transfer capacity [14-21].

The nanofluids are as colloidal fluids composed of nanoparticles chemically stable and dispersed in a fluid (water, aqueous solutions, refrigerant, and others) at concentrations ranging from 0.01% to 10% by volume. According to the literature, the more used and researched nanoparticles are the metals (Cu, Ag, Au, Fe, Ti, and others), the oxides (Al_2O_3, CuO, FeO, ZrO_2, SiO_2, MgO, TiO_2, ZnO, and others) and the carbon compounds (graphite, diamond, carbon nanotubes)[14-21].

In nanofluids application for thermal processes, transport properties are very important to classify them a usable fluid. Accordingly to literature, there are many known variables influencing on nanofluids heat transfer capacity like thermal conductivity, particle concentration, Ph, particle size, viscosity, and other unknown variables. Next section describes the main results obtained by researches about those physical properties giving a perspective of nanofluids applications.

1.1 Physical Properties

Nanofluids have slightly higher viscosity than their base fluids and potentially require greater pumping power to have the same thermal performance. They have flow properties similar to the base liquid and have little or modest increase in the turbulent pressure loss. The increase in thermal conductivity can be compensated by an increase in viscosity, decrease in effective specific heat or change in wettability [13,14]. This flow behavior is attractive for the applications in engineering. To obtain good results in practical applications processes, heat transfer fluids should be designed to increase the heat transfer coefficient without penalizing the pressure loss. This requires an accurate selection of particle shape, size, materials and concentrations. In the case of applications in reactor core, as it has been postulated, nanofluids must have low activation characteristic to avoid that high radiation doses occur.

Researches carried out in this specific field show that there is linearity correlating data and behavior of Newtonian fluid for nanofluids analyzed. Particle size is a factor that must also be considered. Results show, for example, that Al_2O_3-based nanofluids viscosity not only increases in a non-linear way with concentration, but also with the nanoparticles size in the tube sides. There are findings that show zero viscous shear stresses for CuO/ethylene glycol based nanofluids, and that change abruptly when the volume fraction of particulate becomes greater than 0.2%. Therefore, the volume fraction is regarded as the limit dilution. Substantial improvement in thermal conductivity is achievable only when the concentration of particles is less than the dilution limit. At concentrations above this limit, where both rotation and translational Brownian are restricted, there is no further increase in conductivity predictions beyond the effective medium theory. For some nanofluids the aggregate particles have a strong effect on the viscosity as much on the thermal conductivity of nanofluids.

The nanofluids thermal conductivity is constantly observed to be higher than the base fluid (water, oil or another fluid). The first experimental studies on thermal transport properties of nanofluids were aimed to study the surprisingly changes created by high concentrations of metal oxides nanoparticles in a water based fluid [14,17]. Currently, studies on nanofluids thermal conductivity are focused on fluid behavior due to the increase of that property. There are, for example, studies indicating a nonlinear relationship between the thermal conductivity and the concentration in case of nanofluids containing carbon nanotubes. Furthermore, it is observed that thermal conductivity is strongly temperature dependent and increasing of critical heat flux (CHF) at boiling heat transfer processes. Reports demonstrated that the presence of some nanofluids in the base fluid exhibit 50 % higher thermal conductivity.

According to the article published by Timofeeva et al (2007) [19,20], a theoretical and experimental study combining heat conduction and particle agglomeration in nanofluids were carried out. In the experimental part, nanofluids Al_2O_3 in water and ethylene glycol are characterized by measurements of thermal conductivity, viscosity, dynamic light scattering, and other techniques. Results show that the particle agglomeration state evolves in time, even using surfactants. The data also show that the thermal conductivity is predicted within the range by the effective medium theory. On the theoretical side, a model was developed for heat conduction in a fluid containing nanoparticles and clusters of different geometries. Calculations show that the elongated and dendritic structures are more efficient in increasing the thermal conductivity than the compact spherical structures with the same volume fraction; and surface tension is the major factor resulting in lower thermal conductivity.

Recent studies have sought to explain how nanofluids thermal conductivity widely varies depending on variables such as nanoparticle concentration and temperature. Some effective theories introduced by Mossotti, Clausius, Maxwell and Lorenz in the late 19th century, firmly established with the work of Bruggeman (1935), have been extensively verified and applied in many fields of science and engineering [14-16].

The thermal conductivity enhancement ratio can be defined as the ratio of thermal conductivity of the nanofluids (Kn) to the thermal conductivity of the base fluid (Kbf), or (Kn/Kbf) [2]. Buongiorno (2006) [6] reported a 40% increase in thermal conductivity of ethylene glycol with 0.3 vol% of copper nanoparticles of 10 nm in average diameter. Das et al. [13] observed increasing of 10-25% in thermal conductivity of water based nanofluid with 4.1 % vol. of Al2O3 nanoparticles. Moreove, it seems that the increasing of nanofluids thermal conductivity with temperature is greater than for pure fluids.

The simplest models to explain the effects of increased thermal conductivity composites require that the particles are spherical, where the interface effects are negligible. In other words, at this stage, we do not consider the finite thermal conductance of interface particle/fluid. In the limit of low concentrations of nanoparticles, all versions of the theories presented so far converge to the same solution, but in the limit of high concentrations, there is no consensus among the theories presented yet.

Many authors have studied the effect of temperature on the thermal conductivity enhancement and the data clearly shows that the thermal conductivity is intrinsically associated with increased temperature [2].

Static and dynamic mechanisms were introduced as a good strategy to predict thermal conductivity of nanofluids [27]. In this way, a modified model was proposed to include different materials like metal, metallic oxide and nonmetallic oxide, different volume fractions, or different nanoparticle diameters in a new model.

Cinematic viscosity of nanofluids is an important parameter concerning convective heat transfer capacity of nanofluids in hydraulic circuits. According to Motta (2012) [28], in a general way, nanofluids viscosity follow the base pure fluids behavior. For most metal oxide base nanofluids investigated, cinematic viscosity of nanofluids increase with volumetric concentration of nanoparticles; temperature is inversely related to the cinematic viscosity of nanofluids [28].

1.2 Surface contact angle

The surface contact angle of a cooling fluid has been shown as an important variable concerning heat transfer capacity, mainly in boiling conditions. The critical heat transfer (CHF) of a certain fluid is intrinsically related with surface wettability that is closely influenced by surface contact angle of that fluid. The surface contact angle of measured metal oxide nanofluids varies with particle volumetric concentration [28].

1.3 Heat Transfer Capacity

For laminar convection heat transfer process, studies show that the laminar heat transfer coefficient of some nanofluids increases rapidly to values of higher Reynolds number (Re) and may be up to 150%. This coefficient increases with the dimensionless axial distance x/D. Heat transfer coefficient of the laminar nanofluids nearly doubled the upper limit of the Reynolds number range tested, but also decreased with increasing concentration in the range from 1.1 to 4.4 %vol.

For nanofluids turbulent flow heat transfer the Nusselt number (Nu) increases the heat transfer coefficient up to 3-12%. Heat transfer coefficient in turbulent nanofluids based on Cu/water increased by about 40% for a volumetric concentration of 2%. The friction coefficient is not affected by the concentration of nanoparticles for a given for laminar or turbulent flow Reynolds number (Re). Research conducted at the Massachusetts Institute of Technology (MIT) [4-10] showed that, for heat transfer in single-phase convection, heat loss in nanofluids Al_2O_3/H_2O ZrO_2/H_2O present no resemblance to the behavior of pure fluids. In this case, the temperature dependence and thermal load with respect to thermophysical properties were measured and used for definitions of dimensionless numbers Reynolds (Re), Prandtl (Pr), and Nusselt (Nu).

In addition, with respect to particle size influence on convective heat transfer in single phase flow, it was shown that thermal conductivity increases with decrease in nanoparticle size. The nanoparticles in the range of 95-210 nm have a marginal effect on the coefficient of heat transfer. However, there are recent studies showing the opposite trend [25]. This finding is consistent with the recent results for turbulent flow, showing dependence of the heat transfer coefficient during convective flow on the nanoparticles size.

In the case of boiling heat transfer, most of the experiments concerning boiling of nanofluids in a pool show that the nanoparticles deteriorate the heat transfer coefficient. However, for diluted nanofluids ratio with ratio of 0.3 % vol. it was observed that Al_2O_3 nanoparticles can improve the boiling heat transfer coefficient related to the based fluid up to 40%. Interestingly, some studies have also shown that nanofluids heat transfer coefficient increases for small volume fractions by the order of 2%, but decreases for volume fractions larger than this. Therefore, the nanoparticle concentration has been shown to be an important factor in the heat transfer process.

An increase of up to three times of critical heat flux (CHF) for nanofluids Al_2O_3/H_2O relatively to the base fluid (water) at a concentration of 10 ppm has been observed. The critical heat flux increase has been confirmed also for SiO_2/H_2O particles, besides the fact that nanofluid pH increases the CHF up to 350%. In studies on heat transfer after-critical heat flux (post-CHF), a group of MIT [6-11] first demonstrated that nanofluids may improve significantly post-CHF boiling (film boiling). Other studies also have shown that nanofluids have a higher rate of heat transfer in film boiling. These findings are significant because they could pave the way to make improvement of nuclear reactors safety. Furthermore, it was shown that in a mini heat pipe, the heat transfer coefficient and the CHF of nanofluids increases for CuO based nanofluids considerably with decreasing pressure, compared with water.

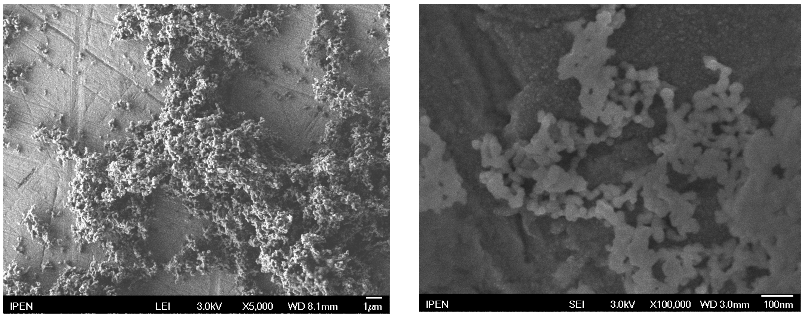
2. EXPERIMENTS

2.1 Sample Preparation and Characterization

Commercial nanofluids containing Al_2O_3 and ZrO_2 nanoparticles dissolved in deionized water of 10% and 5% volume fraction, respectively, were supplied by Sigma-Aldrich and studied in this work. The Al_2O_3 nanoparticles were declared to have distribution into a range of 30-60 nm, a mass fraction purity of 0.99 and a declared value of density $\rho = 1.06$ gcm^{-3} at 25 ^{o}C. The ZrO_2 nanoparticles were declared to have a distribution range < 100 nm. Nanofluids were dispersed into a predetermined water volume to obtain the desired volume concentrations of 0.01%.

Samples were initially prepared using an ultrasonic disrupter to make a homogeneous solution. This is an important step on samples analyses concerning the homogeneity influence on thermal conductivity measurements, as described by Maheshwary and Nemade (2015) [29].

Nanofluids samples were prepared and visually analyzed in a scanning electron microscope (SEM-FEG). The samples were diluted and dispersed in deionized water in an ultrasonic bath for 10 minutes before being placed directly on the sample holder (brass or aluminum) and analyzed by scanning electron microscope JEOL, model JSM 6701F, operating at 5 kV at a distance work of 3.0 mm, as shown in Fig 1.



(a) (b)

Figure 1. Images of nanoparticles obtained in a scanning electron microscope (SEM-FEG): a) Al_2O_3 nanoparticle, b) ZrO_2 nanoparticles.

2.2 Experimental Tests

Preliminary tests for determining the thermophysical properties of nanofluids based on Al_2O_3 and ZrO_2 without the effect of ionizing radiation were performed. These tests were necessary and arose from initial studies to establish a preliminary analysis of the compatibility of nanofluids concentrations into the nuclear reactor environment in which they would be tested. Preliminary tests for determining the thermophysical properties of nanofluids were:

a) Preliminary tests to measure the density of nanofluids: this step consists on measuring densities of the nanofluids samples of Al_2O_3 and ZrO_2 in the volume concentration of 0.01%. The densities were measured with the aid of precision scales by the volumetric flask method [26]. The results are shown in Table 1;

b) Temperature effect: preliminary tests for measurement of the thermal conductivity of nanofluids: this step consists on measuring the thermal conductivities of nanofluids of Al_2O_3 and ZrO_2 in a concentration of 0.01 % by volume at 25°C and 35°C. The thermal conductivities were measured with the hot wire method by KD2-PRO conductivity meter. The results are shown in Table 2;

3. RESULTS

It was possible to observe the agglomeration of nanoparticles of Al_2O_3 forming flat large particles. It was observed that the dispersion of nanoparticles was not sufficient with ultrasonic bath. New tests were carried out with dispersion of nanoparticles into an ultrasonic disruptor, obtaining better results. The same visual analysis was carried out for the ZrO_2 nanofluid. Visualization results of ZrO_2 nanofluids samples in volume concentration of 5% in a scanning electron microscope (SEM-FEG) shows a good dispersion in which small spherical nanoparticles was observed. Both results are important for the qualitative analysis.

Size distribution analysis of nanoparticles showed higher concentration of Al_2O_3 nanoparticles between 10 and 30 nm. Likewise, there is a higher concentration of ZrO_2 nanoparticles between 10 and 25 nm.

Table 1. Nanofluids density measures with volumetric concentration of 0.01% at 20°C; average of 3 measurements).

Fluid	Volume (ml)	Density (kg/m^3)	Uncertainty (%)
H_2O	100	99.9107	±1.0
Al_2O_3/H_2O	100	100.02	±1.0
ZrO_2/H_2O	100	100.49	±1.0

Table 2. Physical properties measures of nanofluids (average of 3 measurements).

<table>
<tr><th rowspan="3">Physical Property</th><th rowspan="3">Temp. (°C)</th><th colspan="6">Nanofluid</th><th rowspan="3">Meas. Uncert. (%)</th></tr>
<tr><th colspan="3">Al_2O_3/H_2O</th><th colspan="3">ZrO_2/H_2O</th></tr>
<tr><th>0.01%</th><th>0.05%</th><th>0.1%</th><th>0.01%</th><th>0.05%</th><th>0.1%</th></tr>
<tr><td rowspan="2">Thermal Conductivity, K (W/m.K)</td><td>25</td><td>2.58</td><td>2.45</td><td>2.25</td><td>1.57</td><td>2.15</td><td>2.37</td><td rowspan="2">5.0</td></tr>
<tr><td>35</td><td>1.91</td><td>1.86</td><td>1.71</td><td>1.47</td><td>2.37</td><td>3.81</td></tr>
<tr><td rowspan="2">Specific Thermal Resistivity, R_{th} (°C.cm/W)</td><td>25</td><td>38.81</td><td>40.91</td><td>44.5</td><td>63.56</td><td>46.6</td><td>42.2</td><td rowspan="2">5.0</td></tr>
<tr><td>35</td><td>52.45</td><td>54.17</td><td>58.4</td><td>68.16</td><td>43.32</td><td>26.24</td></tr>
<tr><td rowspan="2">Electrical Conductivity, μ (μS)</td><td>25</td><td>18.12</td><td>29.7</td><td>45.25</td><td>56.5</td><td>10.25</td><td>15.62</td><td rowspan="2">2.0</td></tr>
<tr><td>35</td><td>31.75</td><td>83.5</td><td>79.25</td><td>91.5</td><td>23.5</td><td>29.5</td></tr>
</table>

Preliminary tests show (Table 1) small nanofluids density variation compared to the water (20 °C). Densities of nanofluids and water revealed to be much closed for the concentrations and temperature measurements.

In Table 2 it is shown the result of preliminary tests for the physical properties of nanofluids for concentrations of 0.01%vol., 0.05%vol. and 0.1%vol., and also two different temperatures (25°C and 35°C). The physical properties analyzed were thermal conductivity, specific resistivity, and thermal conductivity.

It was observed that, for nanofluid of Al_2O_3, the thermal conductivity decreases by about 30% with increasing temperature, for the three concentrations investigated. For ZrO_2 nanofluids the average increasing was more sensitive to the concentration, varying from 10% (0.01%vol.) to 60% (0.1%vol.).

Specific thermal resistivity of Al_2O_3 nanofluid increases with temperature about 32%, independently of concentrations. The same applies to the ZrO_2 nanofluid that increased 62%, and deceased with concentration.

Concerning electrical conductivity of nanofluids, one can note a sensible increasing with temperature. For Al_2O_3 nanofluids, an increasing of more than 75% was observed. For AlO_2 nanofluids, it was noted that temperature has a strong influence over that variable; however, it was very sensitive to the concentration, varying about 60% from 0.01 to 0.05%vol.

4. CONCLUSIONS

An extended literature review on nanofluids properties and applications, mainly for new high efficiency thermal processes was carried out aiming to give an overview on the actual status of such research line worldwide, and to be a conduction line for the new research area in development at IPEN/CNEN-SP in the Nuclear Engineering Center (CEN) group.

Concerning density, thermal conductivity and thermal resistivity of Al_2O_3 and ZrO_2 nanofluids were investigated, and it was possible to conclude that volumetric concentration, particle size/shape and temperature are important variables. More and accurate investigations must be performed to characterize the effects of other variables on nanofluids and its application aiming to prove its ability as a high efficiency fluid in highly efficient cooling systems.

Research proposal concerning the study of nanofluids physical properties and cooling capacity for applications in special systems should be encouraged. The improvement of the efficiency and security of future high efficiency heat transfer processes can be achieved through the following aspects: understanding of: the changing in the nanofluids physical properties compared to their base fluid, especially those related to the high cooling capacity; the relationship between the thermal conductivity and other physical properties against the nanoparticles concentrations. These are some of desired results for the new research line, which has been showing to be one of the most promising on thermal engineering and material application.

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