

ANALYSIS OF THE THERMAL CONDUCTIVITY OF THE AQUEOUS-BASED TiO₂ NANOFLUID FOR NUCLEAR APPLICATIONS

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ABSTRACT

This work aims to investigate the thermophysical properties of TiO_2 nanofluids in water base experimentally and also comparing results to the literature. Existing studies indicate that nanofluids presents increase in thermal conductivity compared to the base fluid which in this study will be water, thus, can be classified as promising fluids for heat transport applications. As the proposal is to use it in nuclear applications, the survey of experimental measurements was performed before and after irradiation in the IPEN installations to verify the effect of ionizing radiation on the properties of nanofluids. Thermal conductivity, viscosity and some visualization of nanoparticles in SEM were carried out in order to understand the behavior of radiation influence on nanofluids and it properties.

1. INTRODUCTION

Conventional heat transfer fluids (water, air, oils and refrigerants) are, in specific cases, limited due to low fluid efficiency. In cases where heat transfer fluid must be highly efficient, as in systems with high heat fluxes, such a process needs to be intensified, and this is done by applying more efficient fluids and/or materials. Nanofluids are very promising fluids for such applications.

Nanofluids are solutions composed of a base fluid plus particles on a nanometer scale. These solutions sometimes have very different properties from those of the base fluid. The first experiments that were done used micrometer or millimeter particles to mix solid and liquid. Choi (1995) [1] proposed the suspension of nanoparticles in a base fluid then called nanofluid in which not only the thermal properties but also other physical properties were altered. Since then, many researchers have devoted efforts to the experimental study of nanofluids behavior seeking the characterization of their properties such as Choi and others (2006) [2] and Daungthongsuk and Wongwises (2007) [2,3], as well as a very large number other works.

However, an approach still little explored is the influence of ionizing radiation on nanofluids. For this reason, this work proposes a theoretical and experimental study to evaluate the effect of ionizing radiation (gamma radiation) on the physical properties that are associated with the efficiency in the heat transfer process, such as thermal conductivity, viscosity, density, surface tension and others. In addition to the effects of nanoparticle

concentration on solution and temperature, the effects of ionizing radiation on the mentioned properties are investigated.

The aqueous based nanofluid of TiO_2 was tested. The choice of these type of nanoparticle is based on it stability and thermophysical properties, which are proven to be most interesting from the point of view of heat transfer and also from the amount of data available in the literature ([x,y,z]).

It was carried out a theoretical and experimental study of the thermophysical properties of TiO2 nanofluid samples, and the effects of concentrations and temperature, before and after the action of ionizing radiation. Analysis of theoretical models, influence parameters and experimental results available in specialized literature.

The results from this research project are expected to contribute to the advancement of knowledge of nanofluid applications in thermal systems and specifically under radiation conditions.

1.1 TiO2 Thermophysical Properties

Among the main physical properties of TiO₂ nanofluids viscosity and thermal conductivity are the main properties due to its influence on heat transport capacity.

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 [4,5]. 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 Al₂O₃-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 [6,7]. 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) [8,10], a theoretical and experimental study combining heat conduction and particle agglomeration in nanofluids were carried out. In the experimental part, nanofluids Al₂O₃ 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 [11-13].

The thermal conductivity enhancement ratio can be defined as the ratio of thermal conductivity of the nanofluids (K_n) to the thermal conductivity of the base fluid (K_{bf}) , or (K_n/K_{bf}) [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. [14] observed increasing of 10-25% in thermal conductivity of water based nanofluid with 4.1 % vol. of Al2O3 nanoparticles. Moreover, 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 [15].

Static and dynamic mechanisms were introduced as a good strategy to predict thermal conductivity of nanofluids [16]. 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) [16], 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 [17].

Murshed and Estellé [18] present a comprehensive literature review on the viscosity of various nanofluids. They show that the experimental data from various works yield very scattered and inconsistent results.

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 [19,20].

2. EXPERIMENTS

2.1 Sample Preparation and Characterization

The TiO2 nanofluids in water base solutions were prepared for this study using the ultrasonic dispersion technique for three distinct volume concentrations: 0.1%, 0.01% and 0.001%.

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 [21].

With all samples prepared, some steps were followed to ensure the dispersion of nanoparticles and thus obtaining the results are more accurate, so the following steps were used for each sample to be studied to obtain the properties:

- 1) Solution homogenization using the ECO-SONICS QR 500 ultrasonic mixer for 30 minutes:
- 2) A sample vial was dedicated to each sample to eliminate cross-contamination between nanofluid types;
- 3) For the thermal conductivity measurements it is necessary that the entire solution completely fills the vial to avoid measurement errors, as the KD2 PRO thermal conductivity measuring equipment averages the value of the air conductivity contained in the vial and the fluid itself.

Following the above steps, samples were prepared to be irradiated. They were numbered so that there was no mixing between the fluids since ID strips would not withstand irradiation and would degrade. A support was used as shown in Figure 1 and the nanofluids were taken to the IPEN Multipurpose Irradiator.

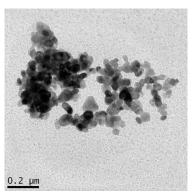
Figure 8 – Nanofluid samples prepared for irradiation.



Source: Authors

Nanofluids samples were visualized in a scanning electron microscope (SEM) JEOL, model JSM 6701F at IPEN. Figure 2 shows the TiO2 nanoparticles image observed.

Figure 2. SEM image of TiO2 nanoparticles obtained.



Source: Authors.

Preliminary tests for determining the thermophysical properties of nanofluids were: density, thermal conductivity and viscosity.

- a) Preliminary tests to measure the density of nanofluids: this step consists on measuring densities of the TiO₂ nanofluid samples in the volume concentration of 0.01%. The densities were measured with the aid of precision scales by the volumetric flask method.
- b) Temperature effect: preliminary tests for measurement of the thermal conductivity of nanofluids: this step consists on measuring the thermal conductivities and viscosities of nanofluids for all concentrations (0.001%, 0.01% and 0.1% vol.) at 25°C and 35°C.

2.1.1 The viscosity measurements

The TiO_2 nanofluid samples (non-irradiated and irradiated) were submitted to ultrasonic dispersion during 30 minutes for homogenization and after, put into the Brookfield DV-I Prime viscometer. It provides continuous detection and measurement display throughout the test. For this work the rotation used was 100 rpm, thus using around 22% of

the torque. The measurement range (in centipoise or milliPascal-seconds). Figure 3 shows the viscometer used.

Figura 3 –Brookfield DV-I Prime viscometer used.



Source: Authors.

2.1.2 The thermal conductivity measurements

The linear probe method based on the transient hot wire technique. This is the method adopted for the present study in determining the thermal conductivity of nanofluids and is thus described in detail below.

The mathematical formulation of this method considers the probe to be an ideal source of heat with zero mass and, therefore, also zero thermal capacity, infinitely long, null diameter, surrounded by an infinite medium whose thermal conductivity is to be determined.

ASTM D5334-08 (2008) describes the standard procedure for determining thermophysical properties and is based on the classical Linear Probe Method also known as the Transient Hot Wire Method.

The thermal properties meter using the hot wire technique (KD2 PRO) used in this work (Fig. 4) calculates the resistivity, thermal conductivity and diffusivity values by monitoring the heat dissipation of a linear heat source given a known voltage.

Figure 4 – Thermal conductivity meter KD2PRO.



Source: Authors.

3. RESULTS

It was possible to observe the agglomeration of nanoparticles of TiO₃ 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. Visualization results of TiO₂ 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 TiO₂ nanoparticles between 40 and 70 nm.

The data collection for density calculations has as its principle to measure the mass of the beaker when it is full and empty, so the difference of the masses of the two divided by the volume it occupies will be the final measure of the density of the fluid.

Figure 5- Thermal conductivity of TiO₂ nanofluids, 0.1%, before irradiation for different temperatures.

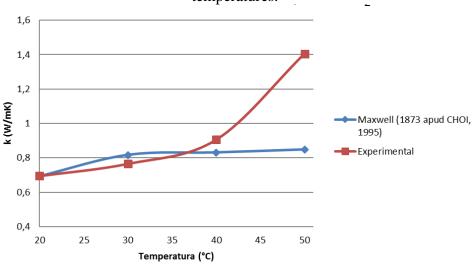
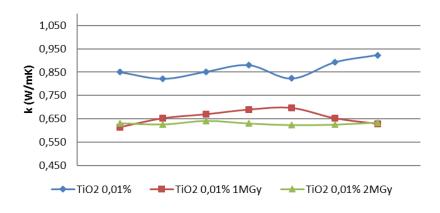


Figure 6- Thermal conductivity of TiO2 nanofluids, 0.01%, after irradiation (1MGy and 2MGy) at 25 $^{\circ}$ C.

Source: Authors.



Source: Authors.

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Figure 7- Thermal conductivity of TiO2 nanofluids, 0.001%, after irradiation (1MGy and 2MGy) at 25 $^{\circ}$ C.

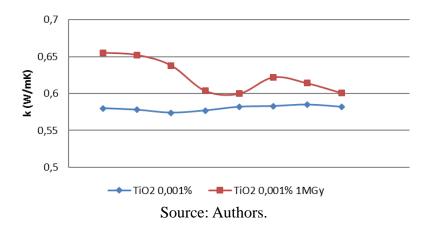
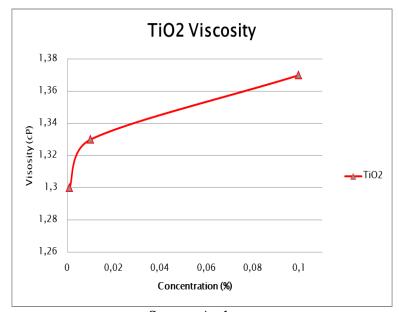


Figure 8- Viscosity of TiO2 nanofluids for 26 °C and all concentrations, before irradiation.



Source: Authors.

Preliminary tests show small nanofluids density variation compared to the water (20 $^{\circ}$ C). Densities of nanofluids and water revealed to be much closed for the concentrations and temperature measurements.

From the results presented on the density, it can be noted that the density increases with the concentration and the nanofluids after irradiation to enter if there was alteration in the properties.

4. CONCLUSIONS

This work present important conclusions concerning the thermophysical properties of TiO₂ nanofluids and its application in heat transport systems, including those with presence of radiation. As the first conclusion we can highlight 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.

Concerning viscosity it can be soncluded that results agree with Batchelor (1977) model at high concentrations, with Maiga et al (2004) model at intermediate concentrations and with low concentrations, and presents the lowest standard deviation at all concentrations for Corcione (2011) model. It can be seen that increasing nanoparticle concentration is directly related to increased viscosity.

Thermal conductivity experiments shower that it increases with nanoparticle concentration and with temperature. Comparison with Maxwell's mathematical model at temperatures of 20 to 40 °C were very close to the experimental results. Experimental measurements above 40 °C start to become very unstable and there is no total convergence of results after irradiation showing any major difference.

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REFERENCES

- 1. Collier, J.G., Thome, J.R., Convective Boiling and Condensation, Oxford Engineering Science Series, 3rd Edition.
- 2. Taylor R., Coulombe, S., Otanicar, T., Phelan, P., Gunawan, A., Lv, W., Rosengarten, G., Prasher, R., Tyagi, H., Small particles, big impacts: A review of the diverse applications of nanofluids, Journal of Applied Physics, 113, pp. 011301-1-19, (2013).
- 3. Kakaç, S., Pramuanjaroenkij, A., Review of convective heat transfer enhancement with nanofluids, International Journal of Heat and Mass Transfer, 52, pp. 3187–3196 (2009)
- 4. Kleinstreuer, C., Feng, Y., Experimental and theoretical studies of nanofluid thermal conductivity enhancement: a review, Nanoscale Research Letters, 6:229, 13 p., (2011).
- 5. M-S. Kang, C. Jee, S. Park, I.C. Bang, G. Heo, "Design process of the nanofluid injection mechanism in nuclear power plants", Nanoscale Research Letters, 6, pp. 3636-372 (2003).
- 6. J. Buongiorno, "Convective transport in nanofluids", Transactions of ASME, 128, pp. 240-250 (2006).
- 7. J. Buongiorno, L-W Hu, S.J. Kim, R. Hannink, B. Truong, E. Forrest, "Nanofluids for enhanced economics and safety of nuclear reactors: an evaluation of the potential features, issues, and research gaps", Nuclear Technology, 162, pp. 81-91 (2007).
- 8. J. Buongiorno, L-W. Hu, "Nanofluid heat transfer enhancement for nuclear reactor applications", ASME 2009 2nd International Conference on Micro/Nanoscale Heat and Mass Transfer, 3, pp. 517-522 (2009).
- 9. S.J. Kim, I.C. Bang, J. Buongiorno, L-W. Hu, "Study of pool boiling and critical heat flux enhancement in nanofluidos", Bull. Pol. Ac.: Tech, 55, pp. 211-216 (2007).
- 10. S.J. Kim, I.C. Bang, J. Buongiorno, L-W. Hu, Surface wettability change during pool boiling of nanofluids and its effect on critical heat flux, International Journal of Heat and Mass Transfer, 50, 4105-4116 (2007).
- 11. H. Kim, J. Kim, M. Kim, Experimental study on CHF characteristics os water-TiO2 nanofluids, Nuclear Engineering and Technology, 38, pp. 61-68 (2006).

- 12. J. Buongiorno, L-W. Hu, Nanofluid Coolants for Advanced Nuclear Power Plants, Proceedings of ICAPP '05, Seoul, May, pp. 15-19 (2005).
- 13. A. Chupin, L-W. Hu, J. Buongiorno, Applications of nanofluids to enhance LWR accidents management in in-vessel retention and emergency core cooling systems, Proceedings of International Congress on Advances in Nuclear Power Plants 2008, Anaheim, California, 20 p. (2008).
- 14. I.C. Bang, G. Heo, Y.H. Jeong, S. Heo, An axiomatic design approach of nanofluid engineered nuclear safety features for generation III+ reactors, Nuclear Engineering and Technology, 41, pp. 1157-1170 (2009).
- 15. E. Zafiri, G. Jahanfarnia, F. Veysi, Thermal-hydraulic modeling of nanofluids as the coolant in VVER-1000 reactor core by the porous media approach", Annals of Nuclear Energy, 51, pp (2013) 203-212.
- 16. Motta, F. C, Caracterização da condutividade térmica, viscosidade, dinâmica e ângulo de contato de nanofluidos baseados em partículas de alumina-gama em água, Escola de engenharia da São Carlos da Universidade de São Paulo, 2012.
- 17. P. Keblinski, J.A. Eastman, D.G. Cahill, Nanofluids for thermal transport, Materials Today, June, pp. 36-44 (2005).
- 18. S.K. Das, S.U.S. Choi, H.E. Patel, Heat transfer in nanofluids: a review, Heat Transfer Engineering, 27, pp.3-19 (2006).
- 19. W. Daungthongsuk, S. Wongwises, A critical review of convective heat transfer of nanofluids, Renewable and Sustainable Energy Reviews, 11, pp.797-817 (2007).
- 20. PAULA, M. C.; MANNAA, I.; DAS, P.K. Techniques for measuring the thermal conductivity of nanofluids: A reviewG., march 2010 revista Renewable and Sustainable Energy Reviews (2010).
- 21. Ramadhan. A.I.; Diniardi. E., RASMA. Effect Analysis of Volume Fraction of Nanofluid Al2O3-Water on Natural Convection HeatTransfer Coefficient in Small Modular