A COMPARATIVE INVESTIGATION OF Al₂O₃/H₂O, SiO₂/H₂O AND ZrO₂/H₂O NANOFLUID FOR HEAT TRANSFER APPLICATIONS


*Research Scholar, Department of Mechanical Engineering, Anna University, Chennai – 600 025, India
**Department of Mechanical Engineering, AVC College of Engineering, Mayiladuthurai – 609305, India
***Department of Mechanical Engineering, Veltech University, Chennai – 600 062, India

In this paper, a direct comparison of the thermo-physical properties such as thermal conductivity and viscosity which are dominating the convective heat transfer phenomena of different nanofluids with three different volumetric concentration (0.5%, 0.75% and 1%) when calculated quantity of nanoparticles like Al₂O₃ (Alumina), SiO₂ (Silica) and ZrO₂ (Zirconia) were dispersed in deionized (DI) water were investigated. Al₂O₃/H₂O and SiO₂/H₂O nanofluids were prepared without using any surfactant and ZrO₂/H₂O nanofluid prepared with surfactant yttriumoxide. The obtained results showed that the thermal conductivity enhancement of 10.13% for Al₂O₃/H₂O, 6.5% for SiO₂/H₂O and 8.5% for ZrO₂/H₂O at 1% volume concentration. Besides, the results showed that the viscosity increases with increase of particle volume concentration. Finally, the experimental results were compared within their corresponding theoretical data outcomes and the results are found to be in good agreement.

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1. Introduction

The conventional heat transfer fluids such as water, ethylene glycol, propylene glycol are used as heat transfer fluids in all heat transfer applications. To improve the heat transferring ability of the heat transfer fluids, nano-sized (1 to 100 nm) particles having higher thermal conductivity are uniformly dispersed and suspended stably in the heat transfer fluid to obtain nanofluid [1]. These nanofluids improve the effective thermal conductivity of the heat transfer fluid due to their higher dispersion stability, high surface to volume ratio and less particle clogging. The selection of nanofluid for a particular heat transfer application plays an important role in the performance of the thermal system. The ultimate goal of this present study was to prepare and to investigate the thermo-physical properties of three different nanofluids.

The studies on effect of the dispersion of nano-sized particles of Al₂O₃, SiO₂ and TiO₂ in water on the change in thermal conductivity and viscosity of the nanofluids at different temperatures and particles concentration was discussed by Masuda et al [2]. It was observed that the effective thermal conductivity has increased by 30% and 10% in the values of Al₂O₃ and TiO₂ nanofluids respectively at 4% particle concentration however, SiO₂ nanofluid at 1% particle volume concentration showed an improvement of only 1% in thermal conductivity. Xie et al. [3] prepared SiC nanofluids by mixing them in water as base fluid and observed thermal conductivity increment of 15.8% for SiC/water nanofluid at 4.2%. Volume concentration, Lee et al [4] investigated SiC nanofluids and inferred thermal conductivity improvement of 7.2% and viscosity increment of 68% at 3% volume concentration. Shahrul et al. [5] prepared Al₂O₃/Water, SiO₂/Water nanofluids without using any surfactant and Polyvinyl Pyrrolidone.

*Corresponding author: iqbalmech18@gmail.com
(PVP) surfactant was used to stabilize ZnO/Water nanofluid. It was observed experimentally that 35%, 26% and 12% of improvement in overall heat transfer co-efficient for 0.3 vol % of ZnO/Water with PVP surfactant, 0.5 vol.% of Al2O3/Water and SiO2/Water nanofluids respectively compared to water and about 50%, 15% and 9% improvement in heat transfer co-efficient and around 51%, 32% and 26% enhancement in actual heat transfer respectively. Mahdi et al. [6] developed a viscosity prediction through graphical and statistical analyses of water based nanofluids of Al2O3, TiO2, SiO2 and CuO over a wide range of temperature from 283.15K to 345.15K, nanoparticle size from 11 to 100nm, volumetric concentration from 0.03 to 13% and viscosity from 0.412 to 4.864. Anoop et al. [7] dispersed SiO2 nanoparticles in deionized water using an ultrasonic bath for 30 minutes and then subjected to intensified ultrasonication using a probe type sonicator and finally by cyclic ultrasonic pulses for about 15 minutes to achieve maximum de-agglomeration so that the SiO2 nanofluid exhibited good stability. Similarly, Fazeli et al. [8] dispersed SiO2 nanoparticles in distilled water using an ultrasonic bath for at least 90 min and found that silica nanofluids stayed stable for 72 hours. M.A. Ahmed et al. [9] developed numerical and experimental investigations on the heat transfer enhancement in corrugated channels using SiO2/water nanofluid and he found that enhancement of heat transfer and pressure drop increases as the nanoparticle volume fraction increases. Suresh et al. [10] synthesized Al2O3 nanoparticles by using chemical precipitation method and then dispersed them in water using an ultrasonicator for about 6 hours and found that the Al2O3/water nanofluid was very stable for several weeks. Beck et al.[11] subjected a mixture of Al2O3 nanoparticles and ethylene glycol to ultrasonic mixing for several minutes and the resulting solution remained stable for the duration of the entire duration of the experiments because of surface charges of the particles. Chandrasekar et al. [12] synthesized Al2O3 nanoparticles using a microwave assisted chemical precipitation method and then dispersed them in water using ultrasonic vibration for 6 hours and no sedimentation was found.

Bashirnezhad et al. [13] had made the review on viscosity of nanofluids and they justify that increase in viscosity with an increase in nanoparticle volume concentration and decrease in temperature. Park et al.[14] had developed a synthesis of surfactant free SiO2 nanoparticles through emulsion method and concluded that the synthesis of mono-dispersed SiO2 nanoparticles without the use of any surfactant or toxic solvents have potential in drug delivery and as a catalyst.

The surveys of various studies, thermal conductivity [15, 16], viscosity [17, 18] and density [19, 20] hikes accordingly with the increase of volume concentration. More over thermal conductivity increases with increase in temperature [21, 22] however, viscosity and density decreases with the increase of temperature [19, 23, and 24]. Chopkar et al. investigated within 1.5vol% of the ZrO2/EG nanofluid and found it to be stable with no agglomeration [25]. Rathinakumar et al added acacia surfactant of wt 1% to the CNT/water and kept three hours sonication, this nanofluid was stable for one month [26].

From the above literature review, it was found that most of the researchers focused their research on single nanoparticle dispersed fluid. Very few researchers compared different nanoparticles for their thermo-physical properties. The main objective of this present study is to compare the different thermo-physical properties of Al2O3/H2O, SiO2/H2O and ZrO2/H2O nanofluids for different volume concentrations.

2. Experimentation

2.1 Materials

The thermo-physical properties of Aluminum Oxide (Al2O3) purchased from Nano-labs, India. Silicon Dioxide (SiO2) purchased from Sigma Aldrich, USA, Zirconium Dioxide (ZrO2) was purchased from Alfa Aesar, UK is listed in Table 1.
2.2. Preparation of nanofluids
The preparation of nanofluids is a key step in the use of nanoparticles to improve the thermal conductivity of the fluids. Three different volumetric concentrations (0.5%, 0.75%, 1%) were used for the preparation of nanofluids. Al₂O₃ and SiO₂ nanofluids were dispersed into DI water without adding any surfactant whereas for ZrO₂ nanofluid, yttrium oxide as surfactant was added to the DI-water, due to the higher density of ZrO₂ as shown in Table 1. Then, by using magnetic stirrer each sample of volumetric concentration was stirred for about 90 minutes, and then followed by ultra-sonication process for 30 minutes by immersing the nanofluid in bath type ultrasonicator, to achieve maximum possible dispersion of the nanoparticles.

![Fig. 1 XRD, SEM and EDAX characterization test results of a) Al₂O₃, b) SiO₂ and c) ZrO₂ nanoparticles](image)

Table 1. Thermo-physical properties of nanoparticles

<table>
<thead>
<tr>
<th>Nanoparticle</th>
<th>Density (g/cm³)</th>
<th>Mean Diameter (nm)</th>
<th>Thermal Conductivity (W/mK)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al₂O₃</td>
<td>3.95</td>
<td>40</td>
<td>30</td>
</tr>
<tr>
<td>SiO₂</td>
<td>2.65</td>
<td>15</td>
<td>1.4</td>
</tr>
<tr>
<td>ZrO₂</td>
<td>5.68</td>
<td>25</td>
<td>2.2</td>
</tr>
</tbody>
</table>

Fig. 1 XRD, SEM and EDAX characterization test results of
a) Al₂O₃, b) SiO₂ and c) ZrO₂ nanoparticles
2.3. Analysis techniques

The X-ray Diffraction (XRD) analysis is used to determine crystalline size, structure, and purity. The microstructure and surface morphology of the Al$_2$O$_3$, ZrO$_2$, SiO$_2$ nanoparticles have been observed using a SEM (Scanning Electron Microscope) (Model: ZEISS-EIGMA with a Gemini column). The EDAX is used to identify the atomic percentage of the nanoparticles [27]. Three nanoparticles were tested on a Bruker AXS D8 advanced X-ray diffractometer by Cu-k$_1$ radiation in the range of 20-60°. The Scherrer formula was used for obtaining the mean size of the crystalline domain $D=\frac{k\lambda}{\beta \cos \theta}$, $\lambda$ is the X-ray wave length, ‘$K$’ is the shape factor, ‘$\beta$’ is the line broadening at half the maximum intensity and ‘$\theta$’ is the Bragg angle.

2.3.1. Characterization of Al$_2$O$_3$ nanoparticle

The diffraction peaks in the XRD image of Al$_2$O$_3$ nanoparticle is shown in Fig.1A (i). The morphological analysis of Al$_2$O$_3$ nanoparticle was analysed using SEM image as shown in Fig. 1A(ii). It was identified that Al$_2$O$_3$ nanoparticles has a spherical shape. The EDAX analysis confirms the elemental composition of the Al$_2$O$_3$ nanoparticle as shown in Fig.1A (iii), which was evident from the atomic composition of Al and O as the only elementary representations present in the Alumina nanoparticle.

2.3.2. Characterization of SiO$_2$ nanoparticle

The XRD pattern for SiO$_2$ identified as shown in Fig.1B (i). Fig. 1B (ii) shows the SEM image of the SiO$_2$ nanoparticle. It identified that the SiO$_2$ nanoparticle has a spherical shape. EDAX analysis confirms the presence of the composition of crystalline SiO$_2$ nanoparticle in the Fig.1B (iii), which showed that the Si and O are the only elementary species present in the SiO$_2$ nanoparticle.

2.3.3. Characterization of ZrO$_2$ nanoparticle

The XRD pattern for ZrO$_2$ identified as shown in Fig. 1C(i). Fig. 1C (ii) shows that the SEM image of the ZrO$_2$ nanoparticle. It depicts spherical shape of the nanoparticle Fig. 1C (iii) shows an elemental examination of the ZrO$_2$ nanoparticles, in which the peaks of Zr and O are evident. EDAX analysis confirms the presence of the composition of crystalline ZrO$_2$ nanoparticle was done by quantitative analysis, which showed the Zr and O as the only elementary species present in the result signifying the absence of impurities in the tested sample.

2.4. Thermo-physical properties measurement of nanofluids

2.4.1. Thermal conductivity measurement

Thermal conductivity was measured by using KD2-Pro thermal property analyzer (Decagon devices Inc., USA). The transient hot wire method is a transient dynamic technique based on the measurement of the temperature rise of a linear hot wire embedded in testing material. This device consists of a probe with 1.3mm in diameter and 60mm in length, a thermistor and a microprocessor to measure the conduction in the probe. The instrument has an accuracy of ±5%. Before measurement, the calibration of the sensor needle was implemented by measuring the thermal conductivity of the DI water at room temperature of 30°C and found to be 0.6 W/mK.

![Fig.2. KD2-Pro thermal analyzer](image1)

![Fig.3. Brookfield Viscometer](image2)
The nanofluid sample of 45ml was taken in a glass vial of 30mm diameter whose cap is equipped with a septum through which the sensor needle was inserted fully into the fluid and oriented vertically with the vial without touching the inner circumferential walls of the vial. The glass vial is then turned upside-down to reduce errors from free convection as shown in Fig.2. Thus, the thermal conductivity for each volume concentration of the nanofluid was measured at an interval of 15 minutes for the period of about 3 hours after sonication. The measurement cycle consists of 90sec, in the first 30sec the instrument will equilibrate which is then followed by heating and cooling of sensor needle for 30sec each.

2.4.2 Viscosity measurement
Viscosity is one of the most dominant thermo-physical properties in nanofluid usage especially in chemical industries. Viscosity is another important property like thermal conductivity that has a crucial impact on heat transfer. Pumping power, pressured drop in laminar flow and convective heat transfer directly depend on the viscosity of fluids. The viscosity of the nanofluids of various volume concentration of nanoparticles were measured by Brookfield Viscometer (Model: DV-I PRIME) shown in Fig. 3, and it has a cone and plate arrangement with small gap provided for allowing the fluid to measure viscosity in the range of 0.3 mPa-sec to 1000 mPa-sec. The viscosity of the distilled water measured at room temperature of 30°C and found to be 0.776 mPa-sec.

![Fig. 4 Comparison of experimental and theoretical thermal conductivity values of a) Al₂O₃/H₂O, b) SiO₂/H₂O, c) ZrO₂/H₂O nanofluids](image-url)
3. Results and discussion

3.1. Thermal conductivity

The base fluid (water) thermal conductivity is 0.6 W/mK. The thermal conductivity of nanofluid depends upon many factors like size of nanoparticle, temperature of the medium, type of the base fluid and preparation process. It is known that increase of the thermal conductivity of nanofluid increases with the volume concentration and temperature [28]. The thermal conductivity of different volumetric concentration of nanofluids such as Al$_2$O$_3$/H$_2$O, SiO$_2$/H$_2$O and ZrO$_2$/H$_2$O at temperature 30°C was tested and compared. Several theoretical models for thermal conductivity have been proposed by Maxwell [29], Hamilton Crosser [30], and Timofeeva et al. [31].

The Maxwell model [29] is given by the following equation:

$$k_{nf} = \frac{k_p + 2k_{bf} + 2(k_p - k_{bf})\varphi}{k_p + 2k_{bf} - (k_p - k_{bf})\varphi}$$

Where $k$ is the thermal conductivity, $\varphi$ is the volume fraction also the subscripts nf, bf and p signifies nanofluid, base fluid and nanoparticle respectively.

The Hamilton crosser model [30] is applied to predict the thermal conductivity of homogenous mixtures containing spherical particles as shown in the formula given below:

$$k_{nf} = \frac{\alpha + (n - 1) - (n - 1)(1-\alpha)\varphi}{\alpha + (n - 1) + (1-\alpha)\varphi} k_{bf}$$

Where, $\alpha = k_p/k_{bf}$, $n=3$ for spherical shape.

Timofeeva et al., [31] used the effective medium theory to calculate the thermal conductivity of nanofluids as given below:
The thermal conductivity calculated using the theoretical models for the three different nanoparticles of different volume concentration was compared with the experimental thermal conductivity of nanofluid.

3.1.1. Effect of volume concentration on the thermal conductivity
Solid nanoparticle having higher thermal conductivity is dispersed to water to improve the effective thermal conductivity of the nanofluid. In this study, it is observed that with increase in volume concentration of the nanofluid, the thermal conductivity of the nanofluid increases due to the higher interactions of the nanoparticles with the basefluid. The enhancement of thermal conductivity is believed due to particle shape, particle interaction, particle clusters, liquid layering, Brownian motion of the nanoparticles and also pH value [32] of the nanofluid.

The theoretical correlations produce similar results, while the experimental values were higher. The addition of nanoparticles has increased the effective thermal conductivity of the Al₂O₃/H₂O nanofluid by 5%, 7.3% and 10.13% for 0.5% 0.75% and 1% volume concentration compared to the basefluid.

The theoretical correlations developed by Maxwell and Hamilton Crosser to measure the thermal conductivity of SiO₂/H₂O nanofluid produced similar results. However, the correlation developed by T’mofeva produced different results compared to the other correlations considered. Yet, experimental values were higher than the theoretical results produced by the correlations. The addition of SiO₂ nanoparticles has increased the effective thermal conductivity of the nanofluid by 2.66%, 4% and 6.5% for 0.5% 0.75% and 1% volume concentration compared to the basefluid.

The addition of ZrO₂ nanoparticles have increased the thermal conductivity of the ZrO₂/H₂O nanofluid by 3.6%, 5.34% and 8.5% for 0.5% 0.75% and 1% volume concentration compared to the basefluid.

Fig. 4 depicts the comparison of measured thermal conductivity values and predicted values using different existing models. It was noticed that the obtained experimental thermal conductivity are higher than the predicted values.

3.2. Viscosity
In this study, the existing correlation of viscosity of water based nanofluids of Al₂O₃, SiO₂ and ZrO₂ were compared with the experimental results obtained. The Einstein equation [33] for particle volume concentration is given by the equation.

\[ \mu_{nf} = \mu_{bf} (1 + 2.5\varphi) \]

where \( \mu \) is the viscosity.

The Brinkman model[34] gives the viscosity prediction as,

\[ \mu_{nf} = \mu_{bf} / (1 - \varphi)^{2.5} \]

The Wang et al model[35] for the prediction of viscosity is given as,

\[ \mu_{nf} = \mu_{bf} (1 + 7.5\varphi + 123\varphi^2) \]

Thus, viscosity changes with volumetric concentration for each nanofluid. In fact, no model is able to predict the exact value of viscosity of nanofluids. A broad range of variations occurs on comparing the experimental results and theoretical calculations.

3.2.1. Effect of volume concentration on the viscosity
The viscosity, which is defined as the fluid’s resistance to flow, was measured using a Brookfield Viscometer for the different nanofluids. The viscosity of the nanofluid increases with
increment of volume concentration. This effect is due to the inter-molecular attraction among the nanoparticles. Thus with the rise in volume concentration of the nanofluid, the increase in viscosity turns out to be detrimental to the heat transfer phenomena.

The viscosity values obtain using the correlations developed by Einstein, Brickman and Wang et al were similar. The viscosity value obtained from Wang et al correlation was different from the theoretical values produced by other correlations, but the experimental results are higher than predicted model for all the nanofluids. It is also inferred that higher density Zirconia nanoparticle yielded more viscosity in base fluid than alumina and silica nanoparticles.

Fig. 5 depicts the comparison of measured viscosity values and predicted values using different traditional models. It was observed from that the obtained experimental viscosity values are not in good agreement with the predicted values from theoretical models. The difference may be due to the effect of Brownian motion, assumptions made while deriving the theoretical models, dispersion methods, particle size distribution, particle aggregation, surfactant, etc.

4. Conclusions

The comparative study on the thermal conductivity and viscosity of three different nanofluids such as Al$_2$O$_3$/H$_2$O, SiO$_2$/H$_2$O and ZrO$_2$/H$_2$O were performed. Thermal conductivity of nanofluid increases with increment of volume concentration, these three nanofluids exhibited enhanced thermal conductivity than base fluid DI water. Among the three nanofluids Al$_2$O$_3$/H$_2$O showed 10.16% highest thermal conductivity then ZrO$_2$/H$_2$O showed 8.5 %, SiO$_2$/H$_2$O showed 6.5% at 1% volume concentration.

The augmentation of thermal conductivity of nanofluid is negated by the increase in viscosity of the nanofluids, which is detrimental to the enhancement in heat transfer and causes lower thermal performance due to the increase in pumping power and pressure drop. An optimum volume concentration which to justify the increase in thermal conductivity and viscosity is required for an effective nanofluid as heat transfer fluid.

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