Experimental Investigation for the Thermo-physical Properties and Stability of CeO_2, ZrO_2, and Al_2O_3 Mixed with Ethylene Glycol and Distilled Water

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In this work, the effects of nanoparticles concentration on the density, thermal conductivity, and viscosity of Al_2O_3, CeO_2 and ZrO_2 suspended in 20% of ethylene glycol (EG) and 80% of distilled water (DW) is experimentally investigated. By using two step method, the nanofluid samples are provided at different concentrations, including 0.5%, 1% and 2 %. Visual observation of the nanofluid samples showed that CeO_2-EG/DW and ZrO_2-EG/DW have higher stability for one week more that Al_2O_3-EG/DW. The results indicate that the density, viscosity and thermal conductivity of the nanofluids increased with increasing the nanoparticles concentration. The highest enhancement of the thermal conductivity was found to be 9.6% for 2% concentration of CeO_2-EG/DW at 25°C. Al_2O_3-EG/DW shows the lowest density and viscosity between all types of the nanofluids.

Keyword: nanofluids, Ethylene glycol-water mixture, thermo-physical properties

In many industrial applications which including cooling processes, power generation, transportation, chemical processes, and so on, the heat transfer pure fluids such as ethylene glycol, water, engine oil, and transformer oil, are commonly used. However, these types of fluids regularly cannot improve the mechanical device performance to the required level since it has low thermal conductivity. The fluid properties may be enhanced by mixed small particulars of metallic, non-metallic or polymeric to form solutions. Recent developments in the nanotechnology have revealed a new set of heat transfer fluids, called nanofluids, such as those studied 1995 by Choi and Eastman [1] at the National Laboratory. Numerous reviewers select the ethylene glycol and pure water as a base fluid to the nanofluid; this may back to their attractive price. Yet, both of them have obvious shortcomings: the thermal conductivity of the pure water at temperature 20 °C is 0.6 W/m K, however, at extreme temperature the water will boil or freeze. Moreover, the thermal conductivity of the ethylene glycol at 20°C is 0.253 W m K and it has extensive temperature range for operation. Thus, mixing the pure water with the ethylene glycol is always commercially recommended as heat transfer fluid in many systems to have a high ability for cooling or heating. Consequently, it is more reasonable that the mixture of ethylene glycol and pure water as a based fluid to the nanofluids has attracted much attention from scientific workers [3-17]. Other studies have focused on the analyses of different properties of nanofluides [21-23]. The viscosity of 40% of pure water and 60% of EG based CuO nanofluid been firstly reported by Namburu et al. [2] in 2007.

Vajjha and Das [3] stated that the thermal conductivity of Al_2O_3 mixed with based fluid EG/DW (60:40) increase with 21% when the concentration increased to 6% and when the concentration increased to 10% the enhancement increased to 69% at temperature 365 K compared with pure water. At temperature 363 K, the thermal conductivity of nanofluid ZnO EG/DW (60:40) increased by 48.5% when the concentration increases to 7%. When the concentration of CuO nanofluid increased from 0% to 6% the thermal conductivity increased by 60% at temperature 363 K. Yiamsawasd et al. [4] reported that the thermal conductivity of nanofluids was higher than the base fluids and it's increased with the rise of temperature. The thermal conductivity of the TiO_2-EG/DW (20:80) increased by 15% at nanoparticles concentration 4%. While the thermal conductivity increased by about 5 to 20% for Al_2O_3-EG/DW (20:80) when the concentration increases from 1 to 4%. Kumaresan and Velraj [5] concluded that the thermal conductivity of MWCNT-EG/DW (70:30) increases and the viscosity decreases when the temperature increases. Also they reported that thermal conductivity at temperature below 25°C has low enhancement. Yu et al. [6] reported that the viscosity of the Al_2O_3-EG/DW (55:45) increases when the concentration increases, while it decreases when the mixture temperature increases. They also indicated that the thermal conductivity, at 10°C, enhance by 3.8%, 7.7% and 11.6% when the concentration of the nanofluid increases to 1%, 2% and 3%, respectively. Reddy and Rao [7] indicated that the thermal conductivity of TiO_2 at 1% concentrations base pure water enhanced by 6% at temperature 30°C. With the same concentration and temperature, the nanofluid thermal conductivity base EG/W (40:60) increased by 4.38%, while it's increased by 10.42% at based fluid EG/W (50:50). Sundar et al. [8,9] reported the thermal conductivity of Al_2O_3 nanofluid enhanced with 17.47% for the base fluid EG/W (20:80) in 1.5% concentrations and temperature 20°C. With the same concentration and base fluid, the viscosity increased by 1.37 times than the base fluid. At temperature 20°C, the thermal conductivity increased by 21.96% at concentration 2% of Fe_3O_4 mixed with base fluid EG/DW (20:80). Esfe et al. [10] reported that, at temperature 30°C, the thermal conductivity of Cu/TiO_2-EG/W (40:60) increased by 18.2% when the concentration increases to 2%. They also investigated the effect of the temperature (20 – 50°C) on thermal conductivity of MgO with different concentrations suspended in EG/DW (40:60). They showed that when the temperature increased from 25°C to 50°C, the enhancement difference were less that 1% [11]. Usri et al. [12] noted that the nanofluid Al_2O_3 mixed with base fluid EG/W (40:60) have the highest thermal conductivity than

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Al_2O_3 mixed with EG/W (50:50) or (60:40). Li and Zou [13] showed that the thermal conductivity of SiC at 1% concentration nanofluid increased by 33.8% than base fluid, while the viscosity increase by 22.4% at the same concentration. Hamid et al. [14] stated that the thermal conductivity of TiO_2 based EG/W (40:60) increased by 7% when the concentration of the nanoparticles increased from 0.5 to 1.5%. Cabaleiro et al. [15] reported that the temperature increases has a high effect on the density and viscosity of the acid-functionalized graphene nanoplatelets, but no enhancement was found for the thermal conductivity. Hamid et al. [16] stated that the mixture 20:80 (TiO_2:SiO_2) mixed with EG/DW (40:60) with concentration 1% show higher thermal conductivity than other ratios which increased by 10.1% at temperature 30°C. While the ratio 50:50 (TiO_2:SiO_2) nanofluid shows the highest viscosity.

Consequently, in order to fill the research gap in the review, this work investigate the thermophysical properties of CeO_2 (Cerium oxide), ZrO_2 (Zirconium dioxide) and Al_2O_3 (Aluminium oxide) nanoparticles suspended in 20% ethylene glycol (EG) and 80% distilled water (DW). In many research papers, the properties such as viscosity and thermal conductivity are estimated from the experimental work, while specific heat and density are estimated from the equations of the solid–fluid homogeneous. The viscosity, thermal conductivity, and density were experimentally studied after the nanofluids stable and homogenous with concentration varying from 0 to 2%. We consider this study would cover a lack of information in this scientific field.

Experimental part

Materials and preparation

Three nanoparticles (Al_2O_3, ZrO_2, CeO_2) used in this research were purchased from SkySpring Nanomaterials, Inc (Houston, USA). To measure the nanoparticles diameter, Scanning Electron Microscopy (SEM) was used. As shown in figure1, 2 and 3, the SEM proved that all the nanoparticles have diameter less than 50 nm. Table 1 shows the properties of the nanoparticles. The distilled water made in the laboratory and the ethylene glycol purchased from Sigma-Aldrich (Bucharest, Romania). The base fluid consists of 20% of ethylene glycol and 80% of distilled water. The 20:80 EG/DW base Al_2O_3, ZrO_2 and CeO_2 nanofluids were prepared based on the two-step method [13,14]. The two step method technique was conducted as follow. Firstly, the nanoparticles were mixed into the base fluid to give the necessary volume concentration varying from 0 to 2%. The amount of the concentration was confirmed by using Eq. (1) [17].

$$\varphi\% = \left[\frac{\left(\frac{w}{\rho}\right)_{np}}{\left(\frac{w}{\rho}\right)_{np} + \left(\frac{w}{\rho}\right)_{bf}}\right] \qquad (1)$$

where φ is the volumetric fraction, ρ is the density, and w is the materials weight. The subscripts *np* represent the nanoparticles and *bf* is the base fluid. An electronic balance (AND-EJ610) was used to quantify the nanoparticles weights. In order to diminish the agglomeration and maintain the stability of the nanoparticles in base fluid, two techniques were used such as heating magnetic stirrer and ultrasonic probe mixer. The beaker which contains

Table 1
THE PROPERTIES OF Al_2O_3, ZrO_2, AND CeO_2 NANOPARTICLES

Nanoparticle	Color	Purity	Diameter	Density	Shape
Al_2O_3	White	99.9 %	>50 nm	3.97 g/cm³	Nearly spherical
ZrO_2	White	99.9 %	>50 nm	5.6 g/cm³	Nearly spherical
CeO_2	Off white	99.9 %	>50 nm	7.22 g/cm³	spherical

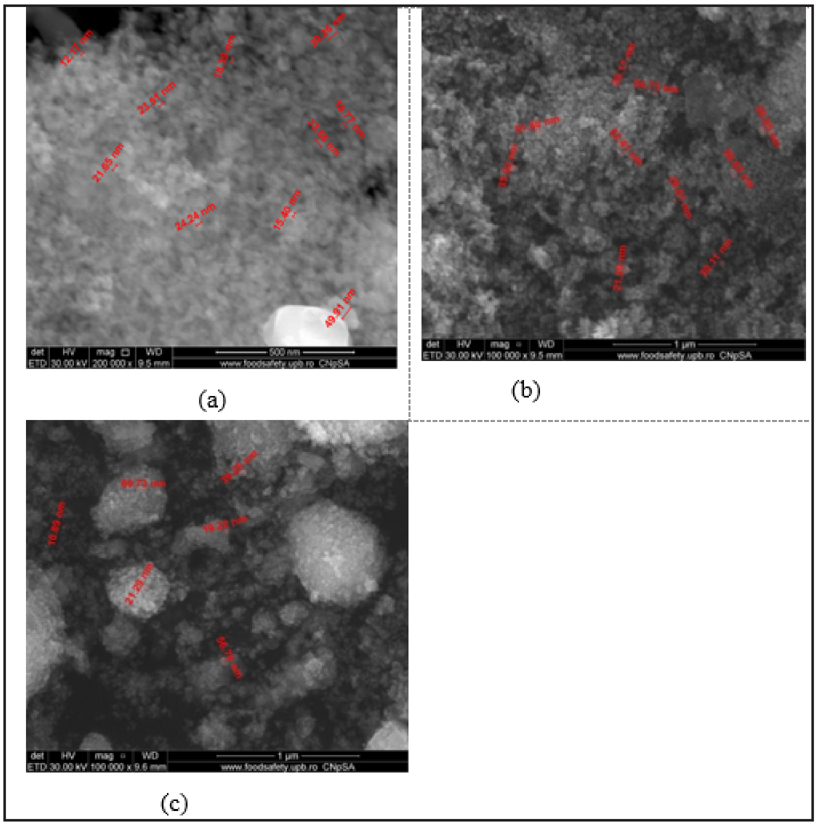


Fig. 1. Scanning Electron Microscopy (SEM) for (a) Al_2O_3, (b) ZrO_2, (C) CeO_2

100 ml of nanofluid is placed on the magnetic stirrer and stirs it for 60 minutes with 1100 rpm and heating 55°C, followed by ultrasonic vibration probe for 1 hour to ensure high stability. This ultrasonic probe (Sonics and materials, USA) produce pulses with the maximum power of 500 Watts and 20 kHz frequency. The process of ultrasonic probe generates temperature varying between 35 and 45°C. The surfactants may reduce the thermal properties of the nanofluids thus no surfactants added. To evaluate the stability of the nanofluids with different concentrations, observation method was used.

Measurement of thermal conductivity

To measure the thermal conductivity of the CeO_2, ZrO_2 and Al_2O_3 nanofluids base EG/DW (20:80), KD2 Pro thermal analyzer (Decagon Devices Inc., USA) was used (see Figure 2). The transient hot-wire method is an operation mechanism applied to find out the thermal conductivity of the nanofluids. The sensor KS-1 single needle with diameter 1.3 mm and length 6 cm was used to operate as a line heat source and to measure the thermal conductivity. The sensor was placed into 40 ml vessel filled with nanofluid. To obtain an accurate reading, before each measurement, the vessel was maintained at ambient temperature for 20 min in order to reach an equilibration. The measurement of the thermal conductivity for all nanofluids was taken with room temperature (25°C). The measurements were started with base fluid EG/DW (20:80) for data verification. After that the results were compared with DOW guide (Dow Chemical Company) [18] to validate the accuracy of the device and measurement procedure. The data were taken three times, and the averages of these readings were used for analysis. In this paper, the thermal conductivity of Al_2O_3, ZrO_2, and CeO_2 nanofluids with different concentration base EG/DW (20:80) was measured with 25°C.

Fig. 2. KD2 Pro thermal analyzer

Measurement of viscosity

In this reasrch paper, Brookfield viscometer (DV-I prime) was used to measure the viscosity of nanofluids with different concentrations (0-2%) base EG/DW (20:80). For calibration, base fluid was used to measure the viscosity then nanofluids were used. All the measurements have been taken directly from the device screen and under steady-state conditions with fixed temperature 25°C. RTD Temperature Probe DVP-94Y which fixed with the Brookfield viscometer was used to read the temperature for all nanofluids.

Measurement of density

The density of nanofluid is directly proportional to the volume ratio of nanoparticles (solid) and base fluid (liquid) in a system. For the nanofluid density, the base fluid considers a significant role. The size and the shape of nanoparticles can be neglected since they have no effect on the density of the nanofluid. Pycnometer used to find out the density of nanofluids as it can be seen in figure 3. The data were taken three times, and the average of these readings was used for analysis. The density of (Al_2O_3, ZrO_2 and CeO_2) nanofluids at volume concentrations 0.5, 1, and 2 % was measured at temperatures at 25°C. Based on the Eq. (1), the density for all types of nanofluids been calculated [19].

$$\rho_{nf} = \frac{m_{nf} - m_0}{m_{bf} - m_0} \qquad (2)$$

where m_{nf} is the pycnometer weight contain nanofluid (gram), m_{bf} is the pycnometer weight contain base fluid (gram) and m_0 is empty pycnometer (gram). The expermintal resultes of the nanofluids density were compared with Pak and Cho [20] model equation;

$$\rho_{nf} = (1-\varphi)\rho_{bf} + \varphi\rho_{np} \qquad (3)$$

Fig. 3. Pycnometer

Results and discussions

Stability

There are many methods to evaluate the stability of the nanofluids such as observation, zeta potential, centrifugation method, TEM (Transmission Electron Microscopy), etc. In this work, the observation method been used which been confirmed in previous works [11,12,14]. The CeO_2, Al_2O_3, and ZrO_2 nanofluids with concentrations 0.5%, 1%, and 2% were examined in terms of particles sedimentation for one week. Figure 4 show the samples freshly prepared and after one week, respectively. It was noted that Al_2O_3 nanofluid with different concentration show a slight sedimentation after one week, while CeO_2 and ZrO_2 nanofluids appeared high stability.

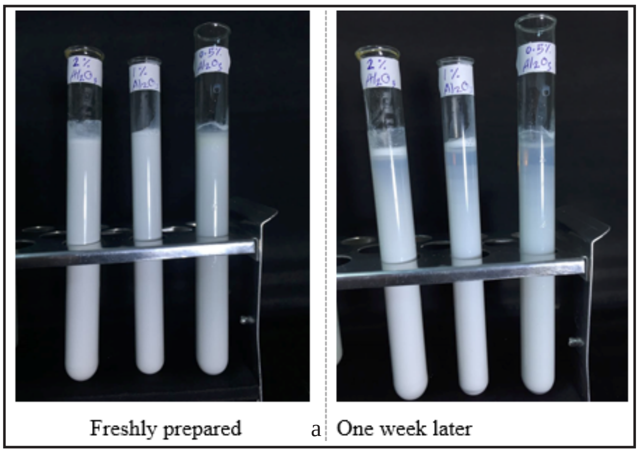
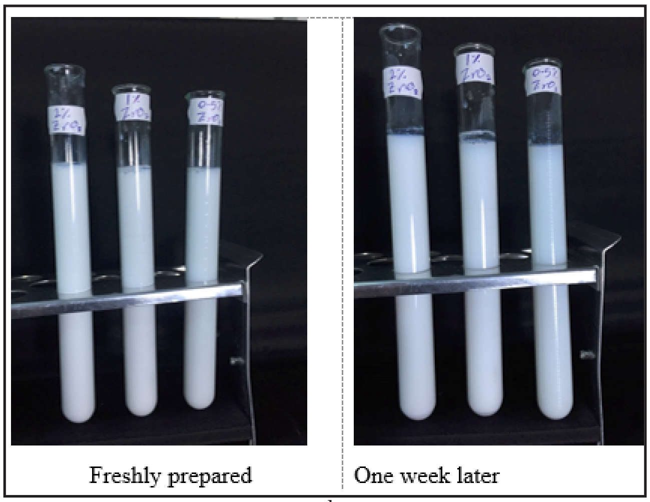
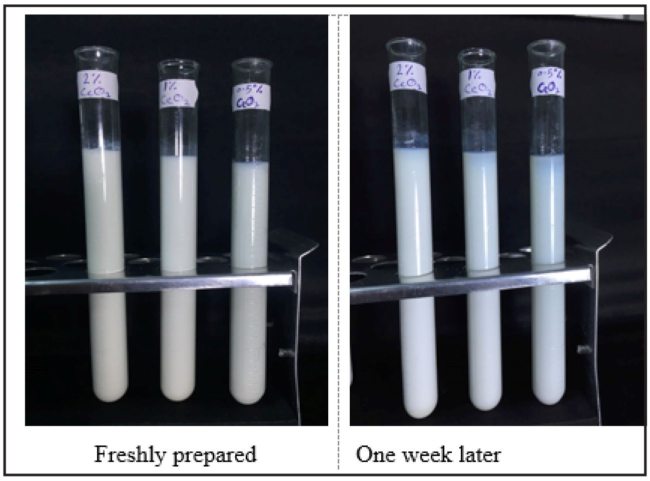


Fig. 4. Sedimentation of the nanoparticles at concentration 0.5, 1, and 2 % mixed with EG/DW (20:80) for (a) Al_2O_3-EG/WD



b

Fig. 4. Sedimentation of the nanoparticles at concentration 0.5, 1, and 2 % mixed with EG/DW (20:80) for (b) ZrO_2-EG/WD



c

Fig. 4. Sedimentation of the nanoparticles at concentration 0.5, 1, and 2 % mixed with EG/DW (20:80) for (c) CeO_2-EG/WD

Thermal conductivity

The thermal conductivity of the nanoparticles CeO_2, Al_2O_3 and ZrO_2 based EG/DW (20:80) with different concentrations is shown in figure 5. All the results been recorded at temperature 25°C. It's very clear from the figure that the thermal conductivity increases when the nanoparticles concentration increases. This phoneme may happen back to the reason that increasing the concentration leads to stick the nanoparticles more to each other, thus the heat transfer faster through these particulars which in turn caused increases the thermal conductivity. The thermal conductivity of the base fluid is 0.498 W/m k which is very close to the results reported by Sundar et al. [8]. However, at temperature 25°C and 2% concentration, ZrO_2-EG/DW shows 4% enhancement, while Al_2O_3-EG/DW show 9.4% enhancement. At the same temperature and concentration, CeO_2-EG/DW shows 9.6% enhancement. Yiamsawasd et al. [4] reported that Al_2O_3 at 2% concentration suspended in EG/DW (20:80) show 9% enhancement. Such deviations are reasonable since the nanoparticles purity and ethylene glycol been supplied from different sources. Finally, CeO_2 mixed with EG/DW with concentration ranging from 0.5 and 2% has the highest thermal conductivity, following Al_2O_3 and ZrO_2 nanofluids.

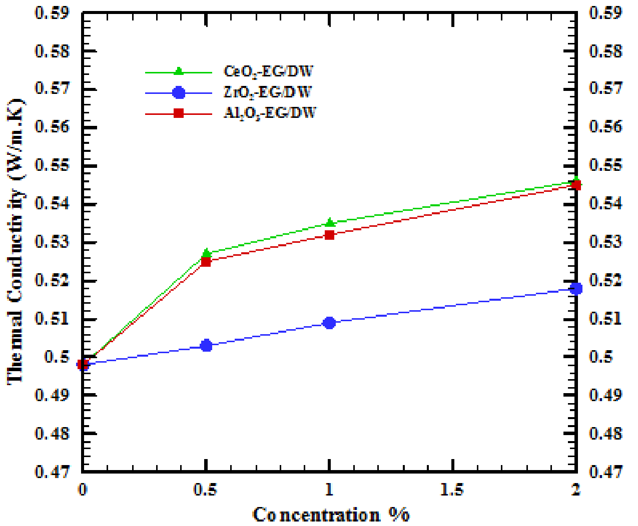


Fig. 5. Thermal conductivity of CeO_2, ZrO_2 and Al_2O_3 suspended in EG/DW (20/80) nanofluid as a function of different concentrations

Density

Since there is no available review studying the density of the CeO_2 and ZrO_2 mixed with 20% ethylene glycol and 80% of distilled water, the densities are measured and listed in the figure 6 to fill the experimental records. All the experimental data been recorded with the ambient temperature 25°C. Many research papers used Pak and Cho [20] model to evaluate the density experimental data. Figure 5 present the measured density compare to Eq. (3). As can be seen, the theoretical equation and experimental results are in a good agreement for all types of nanofluids with different concentrations. Also, it was noted that the density of the nanofluid increases when the nanoparticles concentration increases. Al_2O_3-EG/DW has the lowest density, following that ZrO_2-EG/DW and CeO_2-EG/DW.

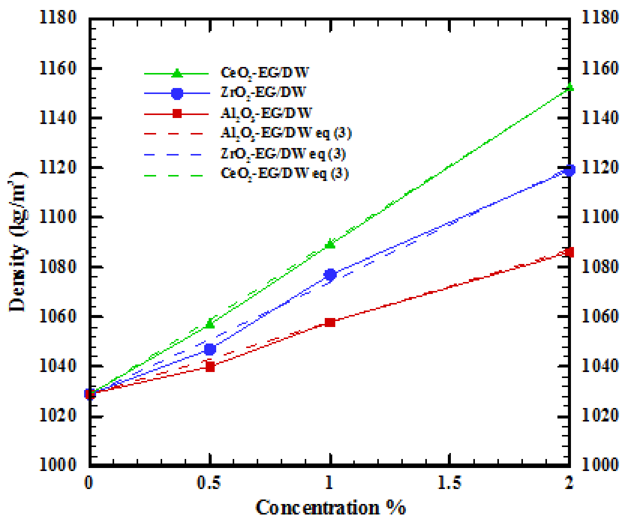


Fig. 6. Density of CeO_2, ZrO_2 and Al_2O_3 suspended in EG/DW (20:80) nanofluid as a function of different concentrations

Viscosity

The viscosity of the nanofluids has an important effect on the pumping power and pressure drop in many energy application and industrials. In this research paper, the viscosity of CeO_2, ZrO_2, Al_2O_3 nanoparticles suspended in EG/DW (20:80) with concentration ranging between 0.5 and 2% were experimentally investigated. The results are showed in figure 7. It was concluded that increasing the nanoparticles in the base fluid lead to increase the viscosity

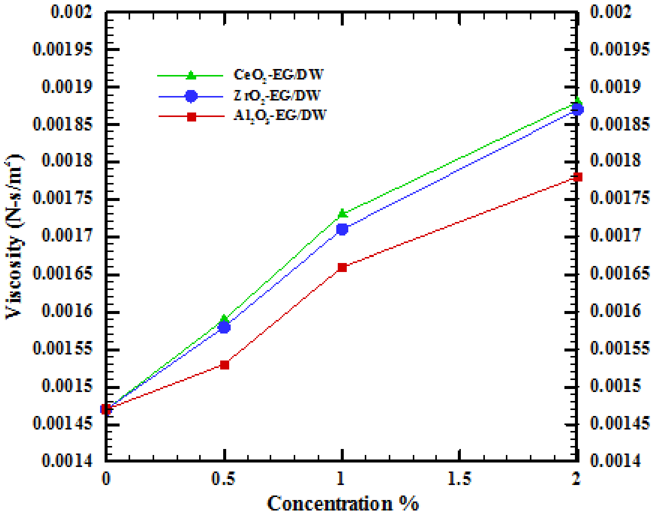


Fig. 7. Viscosity of CeO_2, ZrO_2 and Al_2O_3 suspended in EG/DW (20/80) nanofluid as a function of different concentrations

of the nanofluids. However, CeO_2-EG/DW with concentration ranging between 0.5 to 2% shows the highest viscosity between all the nanofluids, this is due to the fact that CeO_2-EG/DW has high density. Al_2O_3-EG/DW shows the lowest viscosity, while ZrO_2-EG/DW placed between two nanofluids.

Conclusions

In this work, the thermo-physical properties and stability of the Al_2O_3, CeO_2, and ZrO_2 suspended in EG/DW (20:80) were experimentally investigated. The following conclusions are obtained based on the experimental results.

The thermo-physical properties such as thermal conductivity, density, and viscosity are increases when the concentration increases. CeO_2 and ZrO_2 nanofluids show higher stability for one week than Al_2O_3 nanofluid. CeO_2-EG/DW shows the highest enhancement of thermal conductivity with 9.6% at 2% volume concentration, following Al_2O_3-EG/DW with 9.4% enhancement and 4% enhancement of ZrO_2-EG/DW. Al_2O_3-EG/DW with concentration ranging between 0.5 to 2% shows lower viscosity and density than other nanofluids, while CeO_2-EG/DW shows the highest.

References

1.CHOI, S. U. S.; EASTMAN, J. Enhancing Thermal Conductivity of Fluids with Nanoparticles, Argonne National Lab, Argonne, Illinois, USA, 1995

2.NAMBURU, P. K. et al., Exp. Therm.Fluid Sci., **32**, p. 67, 2007

3.VAJJHA, R. S.; DAS, D. K., International Journal of Heat and Mass Transfer, **52**, p. 4675, 2009

4.YIAMSAWASDA, T.; DALKILIC, A. S.; WONGWISESA, S., Thermochimica Acta, **545**, p. 48, 2012

5.KUMARESAN, V.; VELRAJ, R., Thermochimica Acta, **545**, p. 180, 2012

6.YU, W. et al., Powder Technology, **230**, p. 14, 2012

7.REDDY, M. C. S.; RAO, V. V., International Communications in Heat and Mass Transfer, **46**, p. 31, 2013

8.SUNDAR, L. S. et al., International Communications in Heat and Mass Transfer, **56**, p. 86, 2014

9.SUNDAR, L. S.; SINGH, M. K.; SOUSA, A. C. M., International Communications in Heat and Mass Transfer, **49**, p. 17, 2013

10.ESFE, M. H. et al., International Communications in Heat and Mass Transfer, **66**, p. 100, 2015

11.ESFE, M. H. et al., International Communications in Heat and Mass Transfer, **67**, p. 173, 2015

12.USRI, N. A. et al., Energy Procedia, **79**, p. 397, 2015

13.LI, X.; ZOU, C., International Journal of Heat and Mass Transfer, **101**, p. 412, 2016

14.HAMID, K. A. et al., International Communications in Heat and Mass Transfer, **73**, p. 16, 2016

15.CABALEIRO, D. et al., Heat Transfer Capability of (Ethylene Glycol + Water)-Based Nanofluids Containing Graphene Nanoplatelets: Design and Thermophysical Profile, Nanoscale Research Letters, 2017

16.HAMID, K. A. et al., International Journal of Heat and Mass Transfer, **118**, p. 617, 2018

17.KEYVANI, M. et al., Journal of Molecular Liquids, **266**, p. 211, 2018

18.*** Engineering and Operating Guide, for DOWTHERM SR-1 and DOWTHERM 4000 Inhibited Ethylene Glycol-based Heat Transfer Fluids. [S.l.]. 2008

19.SEKHARA, T. et al., Materials Today: Proceedings, **5**, p. 6176, 2018

20.PAK, B. C.; CHO, Y. I., Experimental Heat Transfer, **11**, p. 151, 1998

21.ALFARYJATA, A. A. et al., Thermal Science and Engineering Progress, **5**, p. 252, 2018

22.GIRBU, M., AGOP, M., BEJINARIU, C., HARABAGIU, A., POPA, C., Rev. Chim. (Bucharest)., **59**, no. 2, 2008, p.195

23.ABACIOAIE, D., PAUN, M.A, FORNA, N., BEJINARIU, C., AGOP, M., Rev. Chim. (Bucharest)., **59,** no. 8, 2008, p.930

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