Thermal Conductivity of Al₂O₃ and CeO₂ Nanoparticles and Their Hybrid Based Water Nanofluids: An Experimental Study

Mohammed Saad Kamel¹,2*, Otabeh Al-Oran¹,3, Ferenc Lezsovits¹

¹ Department of Energy Engineering, Faculty of Mechanical Engineering, Budapest University of Technology and Economics, H-1111 Budapest, Műegyetem rkp. 3., Hungary
² Department of Mechanical Techniques, Al-Nasiriya Technical Institute, Southern Technical University, 64001 Thi-Qar, Baghdad street, Al-Nasiriya, Iraq
³ Department of Mechanical Engineering, School of Engineering, The University of Jordan, 11942 Amman, Queen Rania str., Jordan
* Corresponding author, e-mail: kamel@energia.bme.hu; kamel86@stu.edu.iq

Received: 05 December 2019, Accepted: 05 March 2020, Published online: 18 June 2020

Abstract
In many heat exchange systems, there is a demand to improve the thermal conductivity of the working fluids to make those fluids more efficient, and this can be done by dispersing solid nanomaterials into conventional liquids. In the present work, the thermal conductivity of alumina, ceria, and their hybrid with ratio (50:50) by volume-based deionized water nanofluids was experimentally measured. The nanofluids were prepared by two-step method with a range of dilute volume concentration (0.01-0.5 % Vol.), and measured at various temperatures (35, 40, 45, and 50 ºC). The experimental data for basefluid and nanofluids were verified with theoretical and experimental models, and the results have shown good agreement within the accuracy of the thermal conductivity tester. The results demonstrated that the higher thermal conductivity enhancement percentages for Al₂O₃, CeO₂, and their hybrid nanofluids were (5.3 %, 3.3 %, and 8.8 %) at volume concentration (0.5 % Vol.) and temperature (50 ºC) compared to deionized water, respectively. Moreover, a correlation was proposed for the thermal conductivity enhancement ratio of the hybrid nanofluid and showed good accuracy with measured experimental data.

Keywords
thermal conductivity, Al₂O₃ nanoparticles, CeO₂ nanoparticles, hybrid nanofluid, experimental study

1 Introduction
Recently, conventional fluids thermal properties have been modified by dispersing ultrafine solid particles within a range of 1-100 nm, which are consist of metallic or non-metallic nanoparticles as well as carbon nanotubes to produce new thermal fluids so-called nanofluids [1-3]. Nanofluids have been studied from numerous investigators due to their potential impact by heat transfer enhancement in heat exchange applications. Thermal conductivity is an important thermal transport property to which the applicability of using the nanofluids is attributed as it influences the heat transfer performance. However, this property considered as a major key to enhancing the nanofluids heat transfer performance in many heat exchange systems, which are included the boiling process [4-6], cooling of electronic devices [7, 8], solar energy [9], geothermal energy [10], etc. Hence, enhancement of the thermal conductivity of the working fluids could offer a good opportunity to increase the heat transfer rate, which, in turn, improves the thermal efficiency of the heat exchange systems. According to the significant increase in thermal conductivity of nanofluid compared to conventional fluids, great efforts have been paid from many researchers to study thermal conductivity of the nanofluids from several aspects: the influence of the types of the nanoparticles, types of the basefluids, nanoparticles size and shapes [11-14], the effect of volume concentration and temperatures [15, 16] and preparation methods by mean of sonication time and surfactant effects [17, 18]. All the above-mentioned influence parameters that affect the thermal conductivity of nanofluids were summarized and discussed in interesting review studies [19, 20]. However, thermal conductivity enhancement was reported from the literature by using several types of nanomaterials based on different types of basefluids such as Al₂O₃ [21], TiO₂ [22], MgO [23], MWCNT [24], ZnO, CuO, and SiO₂ [25, 26].

Cite this article as: Kamel, M. S., Al-Oran, O., Lezsovits, F. *Thermal Conductivity of Al₂O₃ and CeO₂ Nanoparticles and Their Hybrid Based Water Nanofluids: An Experimental Study*, Periodica Polytechnica Chemical Engineering, 65(1), pp. 50–60, 2021. https://doi.org/10.3311/PPch.15382
The following literature studies [27-39] presents the main works related to thermal conductivity measurements of single and hybrid nanofluids during recent years. The thermal conductivity of alumina oxide nanoparticles with different types of liquids nanofluids was significantly studied in the literature compared to other nanomaterials. Das et al. [27] studied the thermal conductivity enhancement by inserting alumina oxide nanoparticles with a size of 38 nm into the water as a base fluid. The thermal conductivity measurements were done under the temperature range between (21-51 °C), and volume concentration less than 4 %. Their results demonstrated that the best thermal conductivity enhancement was about 24.3 % under higher temperature and volume concentration. In another study, Esfe et al. [28] conducted an experimental investigation using small aluminum oxide nanoparticles with a size of 5 nm based water under temperature ranging between 25-55 °C and volume concentration 0.25-5 % Vol. Their results showed a linear enhancement behavior in the thermal conductivity when the temperature and volume concentration increased. Besides, the best enhancement was demonstrated to be 34 %. Moreover, they presented a new correlation regarding the thermal conductivity ratio under the tested conditions.

Chon et al. [29] examined the effects of different sizes of alumina nanoparticle (i.e., 11, 47, and 150 nm) based water nanofluids on thermal conductivity of nanofluids with various temperatures and volume fractions. They reported that the Brownian motion of nanoparticles plays a key role in the thermal conductivity enhancement with increasing temperature and decreasing nanoparticle sizes. Mostafizur et al. [30] measured the thermal conductivity using alumina oxide nanoparticle-based methanol nanofluid with various volume concentrations and temperatures. The authors have shown that the thermal conductivity using nanofluid was improved compared to other types of tested nanoparticles, as well as the methanol as basefluid. Besides, they proposed a correlation for the thermal conductivity ratio as a function of volume concentration.

Sundar et al. [31] conducted an experimental study to measure the thermal conductivity of Al₂O₃ nanoparticle-based mixture base fluid. The base fluids were a mixture of ethylene glycol EG and water (i.e., 20:80, 40:60, and 60:40 by weight). Different volume concentrations and temperatures were used to see the effects of those parameters on the thermal conductivity of the nanofluids. Results showed that the higher thermal conductivity was about 32.26 % for the nanofluid with 20:80 EG: water at volume concentration 1.5 % and temperature 60 °C compared to basefluid.

According to reported works in literature, there are only a few studies related to cerium oxide nanoparticles based liquids on enhancing the thermal properties. Beck et al. [32] compared the thermal conductivity results by using two sizes of cerium oxide nanoparticles based water. The obtained results were done at room temperature (25 °C), and volume concentrations 2, 3, 4 %. Their results showed that the higher thermal conductivity was reported using large nanoparticle and high concentration. Elis et al. [33] examined the thermal conductivity of CeO₂ nanoparticles with diameter size ranging (30-50 nm) dispersed in EG as a base fluid without adding surfactant. Major results evaluated under volume concentration ranging from (0-1 % Vol.). Their results have shown that the thermal conductivity enhancement percentage for concentration (1 % Vol.), and temperatures (10 and 30 °C) equal 17% and 10.7 % respectively. Keyvani et al. [34] experimentally investigated the thermal conductivity by using cerium oxide CeO₂ nanoparticles based ethylene glycol under volume concentration ranging from 0.25 % to 2.5 % Vol., and the particle diameter (10-30 nm). The thermal conductivity of various samples was measured using the transient hotwire method under the temperature range from (25-50 °C). Results demonstrated that the higher enhancement reached 22 % for the sample has concentration and temperature equal to 2.5 % Vol., and 50 °C, respectively. Besides, a new correlation was proposed using curve fitting their obtained experimental data to present the thermal conductivity ratio under the mentioned conditions.

Growing attention to enhance thermal transport properties of the working fluids that synchronize with a demand for reducing the cost and improve the efficiency led to producing a modified new nanofluid called hybrid nanofluid. Hybrid nanofluids consist of two or more nanomaterials that dispersed into a base fluid to enhance the thermal properties, especially the thermal conductivity. Recently, many researchers have studied the thermo-physical properties of hybrid nanofluids. Asadi et al. [35] investigated the thermal conductivity, the dynamic viscosity of hybrid nanofluid contains alumina oxide, and Multi-walled carbon nanotubes MWCNT dispersed in thermal oil under volume concentrations and temperatures ranging from 0.125-1.5 % Vol. and 25-50 °C, respectively. Their obtained results showed that the higher enhancement reached 45 % at the higher volume concentrations and temperatures.

Esfe et al. [36] studied the thermal conductivity of the hybrid nanofluids containing single-walled carbon nanotubes and magnesium oxide nanoparticles based EG as a basefluid. They tested the hybrid nanofluids under volume
concentrations 0.015 %-0.55 % Vol. and temperatures range 25-50 °C. Their results demonstrated that the thermal conductivity enhanced at the volume concentration of 0.55 % Vol. and temperature of 50 °C, and the higher enhancement percentage was 35 % compared to basefluid. Besides, their results showed that when using MgO nanoparticle, the cost can decrease to producing a new nanofluid.

Moldoveanu et al. [37] investigated experimentally the thermal conductivity enhancement resulted by using unitary nanofluids of Al₂O₃, TiO₂ and their hybrid combination with water as basefluid. The experimental results were presented for different cases in a correlation to cover mentioned nanofluid under volume concentration range from 1 % to 3 % Vol.

In addition to a large number of experimental studies that have been reported in the literature regarding the thermal conductivity of single and hybrid nanofluids, there is a demand to predict this important property to reduce the time-consuming and to avoid using the expensive instruments to measure the thermal conductivity [38]. Some studies have reported in the literature to predict the thermal conductivity of liquids and nanofluids using different statistical and artificial approaches, for example, surface response methodology RSM and artificial neural networks ANNs, and analysis of variance ANOVA [1, 16, 26, 38, 39].

According to our best knowledge and from all reported studies in literature related to the thermal conductivity of nanofluids, there is no study related to the thermal conductivity of (alumina and ceria 50:50 by volume) based on deionized water hybrid nanofluid with dilute volume concentration. Therefore, this study aims to measure the thermal conductivity of alumina, ceria, and their hybrid based deionized water nanofluids at dilute volumetric fractions within a range of (0.01 %- 0.5 % Vol.) and temperatures ranging from (35-50 °C). In addition, a correlation was introduced for the thermal conductivity ratio of the hybrid nanofluid as a function of volume concentration and the temperature. The importance of the obtained experimental results and the proposed model could be involved to improve the efficiency of the heat exchange systems by using this type of nanofluids (hybrid nanofluids) as a working new fluid with high thermal conductivity which, in turn, saving the energy and enhance the economic aspects for many thermal systems.

2 Experimental methods
2.1 Preparation of nanofluids
The formation of nanofluid is a crucial step when we talk about the thermos-physical properties of these fluids. Great effort should be put on the preparation of the nanofluid to ensure a homogenous suspension to avoid the sedimentation and aggregation that might happen during the dispersing of nanoparticles inside the conventional fluids. In this work, all types of nanofluids were prepared at various dilute concentrations ranging from (0.01- 0.5 % Vol.) by dispersing the Al₂O₃, CeO₂ and their hybrid (50:50) by weight into deionized water. A two-step method was adopted to formation those three types of nanofluids. The nanoparticles used in this study purchased from (US Research nanomaterials Inc., USA). The properties of those nanomaterials from the supplier are shown in Table 1.

![Fig. 1 The preparation stages of nanofluids.](image)

Table 1 Nanoparticles specifications from the supplier.

<table>
<thead>
<tr>
<th>Specifications</th>
<th>Alumina Oxide (Al₂O₃)</th>
<th>Ceria Oxide (CeO₂)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Purity</td>
<td>99 %</td>
<td>99.97 %</td>
</tr>
<tr>
<td>APS nm</td>
<td>20</td>
<td>50</td>
</tr>
<tr>
<td>SSA m²/g</td>
<td>138</td>
<td>30-35</td>
</tr>
<tr>
<td>Morphology</td>
<td>nearly spherical</td>
<td>-</td>
</tr>
<tr>
<td>Color</td>
<td>white</td>
<td>light yellow</td>
</tr>
<tr>
<td>Specific heat capacity J(kg⁻¹K)</td>
<td>880</td>
<td>-</td>
</tr>
<tr>
<td>Density kg/m³</td>
<td>3890</td>
<td>7132</td>
</tr>
</tbody>
</table>

Fig. 1 presents the steps of nanofluid preparation during this study. The first step is that the nanoparticles were scaled with an electronic balance with accuracy (0.001 gram), then the desired weight added to the desired quantity of water according to the volume concentration, which was suggested in this work by using the formulation used by [40], as shown in Eq. (1):

$$\varphi, \% = \left( \frac{W_{p1} + W_{p2}}{\rho_{p1} + \rho_{p2}} \right) \times 100.$$

(1)
The second step was to mixing those dry particles into the water by using physical techniques such as stirrer and sonication process. Afterward, the mixture stirred for 1 hour for each type of nanofluid, and next, the ultra-sonication probe (Type: Bandelin, SONOPULS HD 2200, Germany) was inserted into the suspension for 45 min to disperse the nanoparticles inside water.

The stability of the nanofluids in this work was checked by necked eye observation and zeta potential method. First, the sedimentation of dispersed nanoparticles for hybrid nanofluid with time was presented in Fig. 2. This method was applied in previous works [28, 34]; hence, the sedimentation for the prepared hybrid nanofluids with two volume concentrations (0.01 % and 0.5 %) was observed by naked eyes for different periods as shown in Fig. 2. Therefore, the stability of the suspension is significantly stable without any settlement for 1-day (at less during the measurements of nanofluid thermal conductivity). Second, by using the zeta potential device (PALS Zeta potential analyzer from Brookhaven Instruments USA), the stability of nanofluid was checked after the preparation process. The mean value of zeta potential for 0.5 % Vol. was about (-31.53 mV), which considered acceptable physical stability.

2.2 Thermal conductivity measurements

In the present work, the thermal conductivity of three types of nanoparticles Al₂O₃, CeO₂, and their hybrid (Al₂O₃+CeO₂) 50:50 by volume-based deionized water, were measured. Transient Plane Source Method (TPS) is a new technology to measure the thermal conductivity of materials, which was developed based on the hot wire method. Transient Plane Heat Source sensor (type: SKZ1061C) from (SKZ Industrial Co., Ltd) was used as a thermal conductivity tester for both deionized water and nanofluids in this study. The test time was 5 seconds and the accuracy of the sensor within a range of ±5 %.

Fig. 2 Stability checking of hybrid nanofluid with two different concentrations (A) 0.5 % Vol. (B) 0.01 % Vol. at a different period.

All the measured data repeated three times for each test, and the average value was taken. Thermometer utilized to measure the temperatures with hot water insulated vessels after heating the samples for the desired temperature and the accuracy of this thermometer ±1 % of reading temperature. The sensor was calibrated using deionized water; by comparison, thermal conductivity measured data for deionized water with those obtained from NIST under various temperatures [41], and the validation of the obtained results after calibration shows high accuracy behavior with NIST thermal conductivity data as presented in Fig. 3.

3 Results and discussion

3.1 Thermal conductivity of Al₂O₃ based deionized water nanofluid

Thermal conductivity of alumina nanoparticles based deionized water nanofluids was measured at various temperatures (35-50 °C), and different dilute volume concentration (0.01 % - 0.5 %). The working fluid was heated up for the desired temperature and isolated with a thermal container to minimize the temperature changes during the experimental measurements. The thermal conductivity ratio obtained by dividing the thermal conductivity of the nanofluid to the thermal conductivity of deionized water was used to get an indication of thermal conductivity enhancement in this study as represented in Eq. (2) and used by [37]. Besides, this indication compared with two of the most common theoretical models found in literature: Hamilton and Crosser H-C [42], and Yu and Choi [43] as represented in Eqs. (3) and (4), respectively. Fig. 4 shows the thermal conductivity enhancement ratio of alumina nanofluid at a constant temperature 35 °C compared to previous theoretical models when the experimental thermal conductivity of water at this temperature equals 0.6475 (W/m · K).

Fig. 3 Validation of thermal conductivity of deionized water with NIST data [41].
The thermal conductivity enhancement ratio showed a reasonable agreement trend with previous models. According to the obtained results, the thermal conductivity ratio was increased with an increasing volume concentration of nanofluid. In addition, the obtained results show a slight increase compared with previous theoretical models with a maximum deviation equal to 1.9 % at a volume concentration of 0.1 % Vol. This deviation referred to the difference between the temperature of the experimental results, which was tested at 35 °C and the models that were taken at room temperature, and this was stated in previous work [28]. Moreover, the experimental results were compared to Esfe et al. [28] under a volume concentration of 0.5 % and different temperatures. Fig. 5 presents the comparison results that show high accuracy with the empirical model of [28] within a maximum deviation of 1 % at a higher temperature of 50 °C. This small variation can be referred to as nanoparticle size that used in both studies, the preparation methods of nanofluid, and the experimental conditions.

Fig. 6 shows the thermal conductivity enhancement ratio of the alumina nanofluids against the temperatures for different volume concentrations. The results were increased by increasing temperature and volume concentration. The enhancement ratio at low temperature has a small variation between maximum and minimum concentration equal 1.5 %, while it’s equal to 2.7 % at high temperature, which means the effect of increased concentration has a clear impact in enhancing thermal conductivity at a higher temperature. This was attributed to the collision between the nanoparticles at high concentration, which is responsible for increasing the internal energy of the suspended particles, the led to increasing the thermal conductivity of the nanofluids compared to deionized water. Fig. 7 presents the thermal conductivity enhancement percentage for alumina nanofluid against the temperature for different volume concentrations. The results showed that the thermal conductivity enhancement of alumina oxide increase with temperature and volume concentration, and get a maximum improvement of about 5.34 % at volume concentration 0.5 % Vol. and temperature 50 °C. Particularly, the enhanced percentage value at higher volume concentration compared with deionized water ranged from 2.9 % to 5.3 % for the temperatures 35 °C and 50 °C, respectively. These results can be justified as follow;
the increase in volume concentration of alumina nanofluid reduces the space between the moving particles, which increases the collisions for the solid particles and then led to increasing in the particle’s movements (kinetic energy). Hence, these factors were the main reasons to increase in the thermal conductivity of the suspension [37].

3.2 Thermal conductivity of CeO$_2$ based deionized water nanofluid

The thermal conductivity measurements for the ceria based deionized water nanofluids at various temperatures and volume concentrations were investigated in this study. Fig. 8 presents the thermal conductivity enhancement ratio of cerium oxide nanoparticles based deionized water nanofluid compared to deionized water as a baseline case against volume concentrations and different temperatures. Besides, the comparison between the obtained results and the H-C model was introduced in the same diagram. The results showed a high accuracy trend with H-C model [42], especially at low temperature where the maximum deviation was less than 0.5 % at low temperature. The results proved the validity of using this model at low temperatures, as discussed before. The measured data showed a high thermal conductivity enhancement ratio at a higher temperature; this enhancement varied from 1.37 % up to 3.2 % for the volume concentration of 0.01 % and 0.5 %, respectively. While at the lower temperature, the thermal conductivity enhancement ratio varied from 0.4 % to 0.8 % under the same concentration. The results can be attributed to increasing the number of nanoparticles when used high concentration, and this could increase their kinetic energy (collision rate) with the presence of the high temperature, which resulted in the higher thermal conductivity of the nanofluids. Fig. 9 illustrates the thermal conductivity enhancement ratio against temperatures for different volume concentrations. It can be seen that the thermal conductivity enhancement ratio increased with temperature and volume concentration, but the enhancement of the thermal conductivity ratio was significantly increased at higher concentration to be 1.032 compared to basefluid; this referred to the high conductive between the ceria nanoparticles at this level of loading due to the high collision rate of the nanoparticles. While Fig. 10 explains the percentage of thermal conductivity enhancement versus temperature, the results have shown that the maximum enhancement reached 3.3 % at temperature and volume concentration equal to 50 °C, and 0.5 %, respectively.

3.3 Thermal conductivity of hybrid nanofluid

Nowadays, as noticed from the literature, hybrid nanofluid consider as a modified method used to enhance thermal performance in different applications, where study thermal properties of the different modified fluid showed a variation on various attitudes for a different combination. Therefore,
in the present study, hybrid nanofluid, which consists of ceria and alumina under various concentrations and temperature, was examined experimentally, as shown in Figs. 11 and 12. Whereas Fig. 11 presents the thermal conductivity enhancement ratio against temperature under different volume concentrations for hybrid nanofluids. The mean obtained results have shown that the higher thermal conductivity reached 1.088 at the volume concentration 0.5 % Vol., and the temperature equal to 50 °C. In addition, the variation of the volume concentration (the difference between the high and low concentration) has a considerable effect on the thermal conductivity ratio at the higher temperature, which equals 6.1 % compared with the volume concentration effect at low temperature, which equals 2.1 %. Fig. 12 describes the enhancement percentage of thermal conductivity for the hybrid nanofluid versus temperature for different volume concentrations. It can be clearly seen that at higher volume concentration, the hybrid nanofluid varying from 3.07 % up to 8.8 % for the temperature range from 35-50 °C. Fig. 13 presents the thermal conductivity enhancement ratio for three types of nanofluids (mono nanofluids and their hybrid one nanofluids) against volume concentrations at a constant temperature equal to 50 °C. It was found that the hybrid nanofluid has the best thermal conductivity enhancement ratio compared with the other two mono nanofluids at high volume concentration. This could be attributed to the mixing of different nanoparticles size. In detail, mixing alumina oxide particle that has a small diameter 20 nm with ceria oxide nanoparticle that has diameter 50 nm led to increase the collision rate of solid nanoparticles and liquid molecules and then the Brownian motion of nanoparticles due to the high kinetic energy at high temperature which, in turn, improved the thermal conductivity of hybrid nanofluids compared to other mono nanofluids [44].

3.4 Proposed correlations for hybrid and mono nanofluids
According to the reported studies from the literature related to the thermal conductivity of mono and hybrid nanofluids, there is still no model for predicting the ceria and alumina-based deionized water hybrid nanofluids with a dilute
volume concentration. Hence, in this study, correlations were proposed based on experimental data for the thermal conductivity enhancement ratio for both the mono and hybrid nanofluids as a two-variable functions of dilute volume concentration and temperature. Fig. 14 ((A), (B), and (C)) illustrates the influence of volume concentrations and temperatures on thermal conductivity ratio of hybrid, alumina and ceria nanofluids in three-dimensional 3D surfaces using Matlab curve-fitting tool, respectively. A quadratic polynomial functions were proposed for thermal conductivity ratio of all nanofluids with best fit that obtained by above-mentioned experimental conditions for hybrid and mono nanofluids. The results demonstrated that the temperature has more effect than the volume concentration on thermal conductivity enhancement ratio, and this could be attributed to the increase in the Brownian motion of the nanoparticles during the higher temperature [1, 44]. The correlations for nanofluids introduced as follows (Eqs. (5) to (7)):

**Hybrid nanofluids**

\[
k_{\text{ratio}} = 1.21 - 0.009581T - 0.223\phi + 0.0001223T^2 + 0.006598T \times \phi,
\]

**Alumina nanofluid**

\[
k_{\text{ratio}} = 0.9845 - 0.0008274T - 0.01939\phi + 0.0000293T^2 + 0.00123 \times T \times \phi,
\]

**Ceria nanofluid**

\[
k_{\text{ratio}} = 1.027 - 0.00153T - 0.0577\phi + 0.0000261T^2 + 0.0018 \times T \times \phi,
\]

where: \( T, \phi \) are the temperature and volume concentration of nanofluids, respectively. In order to check the accuracy of the proposed correlations, the following parameter, referred to a Margin of Deviation, is defined in Eq. (8):

\[
The \text{margin of deviation} (\%) = \frac{k_{\text{exp}} - k_{\text{corr}}}{k_{\text{exp}}} \times 100,
\]

where: \( k_{\text{exp}} \) referred to thermal conductivity results obtained from measured data, while \( k_{\text{corr}} \) referred to thermal conductivity obtained from our proposed correlations. The margin of deviation was adopted to check the accuracy of the predicting models, and it was found that the accuracy not exceed \( \pm 4.2 \% \), as shown in Fig. 15, which means reasonable accuracy between the predicting models and experimental data of all nanofluids used in this study. Moreover, for better comparison between the experimental results and the data that obtained from the models, it can be seen there is a reasonable agreement between the predicted models and the measurements data for thermal conductivity enhancement ratio for used nanofluid as shown in Fig. 16.
Conclusion

In the present study, the thermal conductivity of nanofluids so-called (alumina nanofluids, ceria nanofluids, and their hybrid nanofluid 50:50 by volume) was measured at various dilute volume concentrations and different temperatures. The results demonstrated that the thermal conductivity enhancement ratio could improve for all nanofluids with increasing the volume concentration and temperature compared to deionized water as a baseline case. In addition, the results of the thermal conductivity enhancement ratio of hybrid nanofluid shown a significant increase compared to those of mono nanofluids as well as deionized water. The following points summarized the obtained results from this study:

- The thermal conductivity enhancement ratio of all nanofluids showed a considerable enhancement by increasing the temperature and volume concentrations.
- The higher thermal conductivity enhancement ratio for Al₂O₃, CeO₂, and their hybrid nanofluids were (1.053, 1.033, and 1.088), respectively, at higher volume concentration (0.5 % Vol.) and higher temperature (50 °C) compared to deionized water case.
- A correlations were introduced for the thermal conductivity enhancement ratio for hybrid, alumina, and ceria nanofluids using Matlab curve fitting tool. The obtained results from the proposed models have shown a good accuracy with experimental data for all nanofluids with maximum Margin of Deviation of 4.2 %.

Acknowledgment

The authors would like to thanks the Hungary Government for their financial support as the Stipendium Hungaricum Scholarship. In addition, the authors would like to thank the Tempus Public Foundation (TPF) in Hungary for their continued administrative support since the application stage until graduation.

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