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Experimental investigation on stability and thermal conductivity of SiO_2 nanoparticles as green nanofluids for application thermal system

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Abstract

In the last few years, much research has focused on the stability and improvement of the thermo-physical properties of single-component nanofluids. Some studies have not made many improvements to the stability and thermophysical properties of various types of green nanofluids from several variations of nanoparticles. Green nanofluids must be developed to improve heat transfer performance from their stability and thermal conductivity factors. Stability and thermal conductivity of Nano-silicate suspended in a base mixture of water /ethylene glycol with the ratio of 60:40, different volume concentrations were investigated. The experiments carried out were the stability of the green nanofluids investigated for volume concentrations of 0.1~0.3% and temperature conditions from 30 to 70°C for thermal conductivity measurement using TEMPOS Thermal Properties Analyzer. The experimental results showed that the stability analysis of the green nanofluids prepared by the UV-Vis method was stable up to 30 days after preparation with a sonication time of 1 hour with a ratio of 70-80%. The evaluation of the zeta potential for green nanofluids obtained a value of 33.57 mV with a moderate stability classification. The highest thermal conductivity for the green nanofluids was obtained at 0.3%, and the maximum increase was 17% higher than that of the base liquid (W/EG). Green nanofluids with a concentration of 0.1% gave the lowest effective thermal conductivity of 1.09 time at 70°C.

Keywords:
Empty Palm Oil Shell;
Green nanofluids;
Nano-Silicate;
Stability;
Thermal conductivity;

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INTRODUCTION

Indonesia is the largest producer of palm oil in the world. Indonesia in 2020 produced 44.8 million tons of palm oil with a total area of palm oil plantations of 14,457 million hectares [1]. In the palm oil processing process, palm oil is produced as the main product, and waste is also produced as a by-product. Waste is produced in the form of liquid waste and solid waste. Liquid waste occurs in the processing of fresh fruit bunches, while solid waste is the first waste produced from palm oil processing [2].

A material that has high porosity and a large surface area is silica (SiO_2). There are many uses for it in pharmaceuticals, fillers, and catalysts [3]. To produce SiO_2 in an industrial environment, sodium silicate is used, where the smelting of quartz sand and sodium carbonate requires a large amount of energy [4]. Extensive studies have attempted to extract silica from various agricultural waste products such as rice husk, corn and coconut cob ash and palm oil waste [2][5]. As far as we know, studies on OPEFB (empty palm oil bunches) as a source of silica still need to be explored.

Recently, nano-silica has been used as a fluorophore carrier [5][6], for the controlled release of drug molecules, as an antiseptic agent [7][8], and as a biosensor [9]. Different silanol groups in nanosilica can change the crystalline behavior of the polymer matrix, increase the effect of hydrogen bonding, and change the fluorescence properties. The most important property of polymers is their heat resistance; this affects the spectral stability, mechanical properties, lifetime, and other material properties [4, 10, 11, 12, 13, 14, 15].

Nanofluid is a liquid suspension containing metal or non-metal nanoparticles with a special size (1-100 nm) that is dispersed into the base fluid. In 1995, the concept of nanofluids was first introduced by Choi, et al. [16]. This new method is proven to increase heat transfer by increasing the thermo-physical properties of nanofluids. Nanofluids are known for applications in heating and cooling processes. The main cooling process is an important part of industrial applications such as power plants, chemical processes, microelectronics, transportation, and automotive cooling systems [17, 18, 19, 20, 21]. The influence of solid particles can change the thermal properties of the working fluid. Several researchers have conducted research on thermal conductivity, dynamic viscosity and stability of the produced nanofluid [22, 23, 24, 25, 26, 27, 28]. Green nanofluids or bio-nanofluids, or environmentally friendly nanofluids derived from several sources of natural materials [29, 30, 31]. Highly colloidal stable green nanofluids as alternative working fluids are essential for their effective use in different thermal systems [32][33]. Based on previous research, there is very little research on the development of green nanofluids in the world.

Nanofluid preparation methods are important to minimize the agglomeration of nanoparticles so as to increase stability. The most common processes used in the manufacture of nanofluids are one-step and two-step methods. The one-step method is the process of synthesizing nanoparticles and simultaneously dispersing them in an alkaline liquid. Another method of nanofluid preparation is known as the two-step method. There are two processes in this method, namely (i) synthesis of nanoparticles in powder form (ii) dispersion of nanoparticles into the base liquid to form a stable and homogeneous solution [34][35].

The stability of the nanofluid and the size of the nanoclusters affect parameters in thermal conductivity [25, 27, 28, 36, 39, 40]. Nanofluid stability is defined as the resistance of nanoparticles to aggregation. Factors such as Van der Waals attraction cause aggregation, which results in the formation of nanoclusters contained in nanofluids [41, 42, 43, 44]. The formation of nanoclusters depends on their size, which causes the sedimentation of particles in the nanofluid. The stability of nanofluids has a significant effect. There are two testing methods, quantitative and qualitative. UV-Vis spectrophotometric measurements have been used to quantitatively characterize the stability of nanoparticles from dispersions [25][37]. Can be applied to all base fluids, while zeta potential analysis has the limitation of base fluid viscosity [45]. In other literature studies, different techniques are used to stabilize nanoparticles in liquids such as ultra-sonication, additional surfactants, surface modifiers and pH adjustment [44, 46, 47]. For qualitative testing by visually seeing the sedimentation from the nanofluid made. The thermal conductivity of nanofluids is highly dependent on the stability of nanoparticles dispersed in the base fluid [39, 48, 49, 50].

Thus many researchers carry out important thermal conductivity investigations in understanding the behavior of nanofluids for further application in heat transfer applications. Thermal conductivity is an important factor affecting heat transfer enhancement [21, 51, 52, 53, 54, 55]. There are several factors that affect thermal conductivity such as: concentration, temperature, particle size, surface-to-volume ratio of nanoparticles, and nanofluid stability [56, 57, 58, 59]. Turgut et al. [58], proved that the thermal conductivity increased by 7.4% with the volume fraction of the particles above the alkaline liquid. Investigation of Al_2O_3-Cu composite nanofluids with water as the base fluid was carried out by Suresh, et al. [60], they reported an increase of up to 12% with increasing volume concentration.

The stability and thermo-physical properties of various types of green nanofluids are very important to study. This aims to understand the stability and behavior of nanofluids as well as factors that affect properties that can improve heat transfer performance. Based on the information obtained by the author, the study of the influence of nanoparticles on green nanofluids is limited to the literature. Based on these problems, this research was carried out by emphasizing the influence of nanosilicate from empty palm shell (EPS) on the stability and thermal conductivity of green nanofluids.

METHOD

Preparation of green nanofluids

The production of green nanofluids involves mono nanofluids, namely SiO_2 nanoparticles or Nano-Silicates that are mixed and dispersed in a base fluid mixed with water/EG (60:40). Nano-Silicate is obtained from Empty Palm Oil Shell (EPS) made using the Ultra-Sonication Method. Nano-Silicate nanoparticle size is 51 nm. Nano-Silicate characteristics are given in Table 1. Table 2 presented information properties of Ethylene Glycol.

A two-step method is used for the preparation of green nanofluids. Green nanofluid is prepared by mixing Nano-silicate with water/EG mixture, and sonication. The production of green nanofluid begins with the calculation of the required volume according to its concentration. In this research, green nanofluids were made with volume concentrations of 0.1, 0.2, and 0.3%. The nanofluid was first prepared at the highest concentration, 0.3% and then diluted to a lower concentration. Arrangement of green nanofluids based on Figure 1.

SiO_2 nanoparticles or Nano-Silicates are available in powder form with a weight concentration of 25% for SiO_2.

Table 1. Properties of SiO_2 nanoparticles from Empty Palm Oil Shell [61]

Properties	SiO_2
Molecular mass, g mol^{-1}	60.08
Average particle diameter, nm	51
Density, kg m^{-3}	2220
Thermal conductivity, W m^{-1} K^{-1}	1.4
Specific heat, J kg^{-1} K^{-1}	745

Table 2. Properties of Ethylene Glycol (EG) [62]

Properties	EG
Vapour pressure, mmHg at 20 °C	0.08
Boiling point, °C	195–198
Melting point, °C	-13
Density, g ml^{-1} at 25 °C	1.113

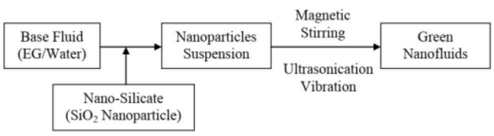

Figure 1. The preparation of green nanofluids

Equation (1) [43][63] is used to convert from weight concentration to volume concentration. Dilution from a higher volume concentration to a lower volume concentration using (2) [64] by adding the base fluid (*ΔV*).

$$\phi = \frac{\omega \rho_w}{\frac{\omega}{100}\rho_w + \left(1-\frac{\omega}{100}\right)\rho_p} \qquad (1)$$

$$\Delta V = (V_2 - V_1) = V_1\left(\frac{\phi_1}{\phi_2} - 1\right) \qquad (2)$$

Nano-Silica is mixed together with a mixture of Water/EG in a ratio of 60:40 to form a green nanofluid. A total volume of 100 mL was prepared for the concentration of green nano liquid. The green nanofluid solution was mixed together using a magnetic stirrer for 15 minutes. The solution then undergoes a sonication process for 1 hour using ultrasonic immersion to increase stability.

Stability of green nanofluids

Investigating the stability of green nanofluid in this research was done through UV-Vis spectrophotometer measurement, visual observation and Zeta potential. UV-Vis was performed for 30 days (720 hours) with varying sonication times. The wavelength of the UV-Vis spectrophotometer is set at 850 nm following the research of Hamid, et al. [65]. UV-Vis measures the absorption and light scattering intensity of the nanofluid by comparing the intensity level with the base fluid. The absorbance ratio of the sonication time is different during the sedimentation time at a constant wavelength (*λ*) of 850 nm. Stability evaluation with UV-Vis was also used by previous research [40]. Sedimentation with visual observation is done for up to 30 days. Green Nanofluid will be considered stable when its concentration is constant. Previously, the same method to observe the visual sedimentation of nanofluids was prepared by Habibzadeh, et al. [34]. Measurements with zeta potential were performed using a Zetasizer Nano ZS (Malvern Instruments Ltd., GB) [40].

Thermal conductivity measurement of green nanofluids

The thermal conductivity measurement method follows the ASTM D5334 and IEEE 442-1981 standards, using the TEMPOS Thermal Property Analyzer shown in Figure 2.

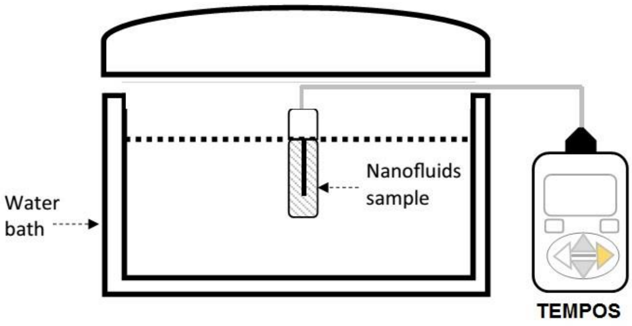

Figure 2. TEMPOS Thermal Properties Analyzer to the measurement of thermal conductivity

Thermal sample conductivity test section, KS-1 sensor to read k [W/m K], measuring bottle to enter the sample to be tested, and the TEMPOS controller is an important part of the thermal conductivity measuring device. The TEMPOS Thermal Property Analyzer instrument uses a transient line heat source to measure thermal properties. Thermal conductivity measurements were performed for temperatures varying from 30 to 70 °C. A water bath is used to keep the sample temperature constant. Previously, the thermal conductivity value from the thermal conductivity sensor was validated using liquid glycerine standards supplied by Meter Group. The measured K is 0.286 W/m K with an accuracy of ±0.35%. The measurement of thermal conductivity was done several times and the average was taken, the measurement time was 15 minutes for each set of data at different temperatures. This is important to minimize the occurrence of errors in measurements with free convection due to temperature variations along the sensor that is in direct contact with the liquid sample.

RESULTS AND DISCUSSION

Stability of green nanofluids

Observations of the absorbance for volume concentrations of 0.1, 0.2 and 0.3% are shown in Figure 3. The absorbance of green nanofluid increases linearly with increasing volume concentration. This trend is in accordance with the Beer-Lambert Law, namely the absorbance value is equal to the concentration. The stability of the green nanofluid was further confirmed by the absorbance ratio, A_r and presented in Figure 4.

The absorbance ratio at 0.1% volume concentration of green nanofluid for 4 different sonication variations. The ideal absorbance ratio is one (100%) indicating ideal fluid stability. Condition variations with 3 hours of sonication began to decrease after 50 hours by 70% and continued to decrease until it stabilized at 30 days (720 hours) by 50%. While the sonication time is 2 hours, it remains at a good concentration ratio value of about 70~80% up to 30 days later. The condition of sonication time for 1 hour, reduced the absorbance ratio by 60%. For variations of sonication 0 hours or after preparation there was a decrease of 40%.

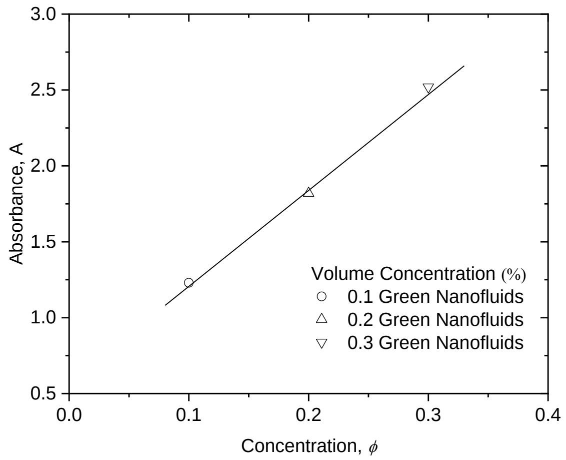

Figure 3. UV-Vis spectrophotometer linear relation graph between absorbance and green nanofluids concentration

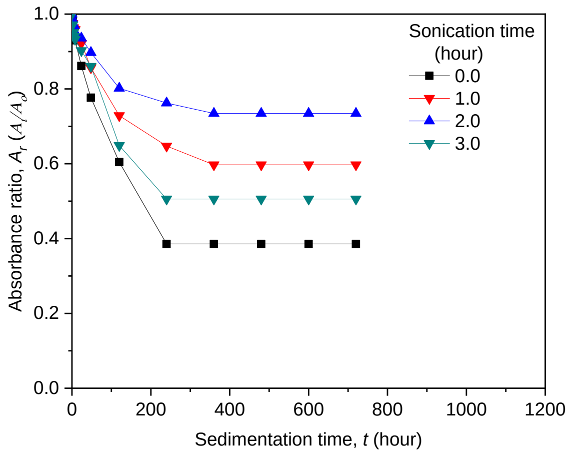

Figure 4. The absorbance ratio of green nanofluids for different sonication time within 30 day

From the figure it can be seen that the sonication time of 2 hours shows the best absorbance ratio compared to other sonication times. Thus, the manufacture of green nanofluid in this study used 2 hours for the sonication process. The absorbance ratio finding was obtained 70-80% for 30 days. The absorbance ratio of the green nanofluid in this study was compared with Hwang, et al. [66] and shown in Figure 5.

Investigations by Hwang, et al. [66], used multi-wall carbon nanotubes (MWCNT) in an aqueous base fluid. As shown in the graph, MWCNT nanofluid has poor stability. In this study, Nano-Silicate was used in the EG/Water mixed base liquid and used a sonication time of 2 hours. The absorbance ratio finding was obtained 70-80% for 30 days. This condition is stable.

Zeta potential measurement is one of the most critical tests to validate the stability quality of nanofluids through electrophoretic behavior studies [45][67]. Zeta potential evaluation is the standard quantitative method for stability evaluation [25][42]. Usually, the accepted zeta potential values are summarized on Table 3.

Generally, suspensions with a measured zeta potential above 30 mV (absolute value) are considered to have good stability [67]. The absolute value of the zeta potential was used to show the agglomeration of SiO_2 nanoparticles in EG/W. The higher the absolute value, the better the dispersion of the particles, was resulting in better stability.

Measurement of zeta potential for nanofluid green recorded up to 33.57 mV. The results were then compared with the stability classification of nanofluids based on zeta potential with moderate stability values compiled by Ghadimi, et al. [67] as shown in Figure 6.

Table 3. Zeta potential and classification stability [67]

Zeta potential (mV)	Stability
0	Little or no stability
15	Some stability
30	Moderate stability
45	Good stability
60	Very good stability

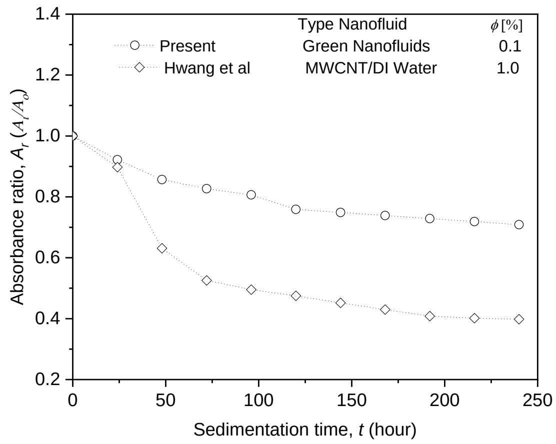

Figure 5. Comparison of absorbance ratio with sedimentation ratio from literature

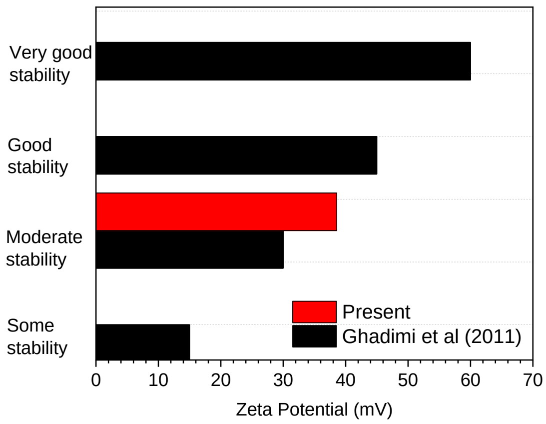

Figure 6. Zeta potential of green nanofluids

An absolute value above 30 mV is desired for good stability of the green nanofluid. It is proven that the green nanofluid is beyond the stable limit of 30 mV. Therefore, the evaluation of zeta potential has confirmed the stability of green nanofluid.

Figure 7 shows an image of the green-W/EG nanofluid for a volume concentration of 0.1. 0.2, and 0.3%. Green Nanofluids-W/EG images were taken only after preparation and after 30 days.

Based on Figure 7a–c, no particle sedimentation was observed after the nanofluid was prepared after 21 days. Agglomeration appears in the 30-day sample in Figure 7 d.

Validation of thermal conductivity with ASHRAE

Thermal conductivity measurements using the TEMPOS Thermal Properties Analyzer are presented in Figure 8. Thermal conductivity measurement data needs to be validated by comparing with ASHRAE [68] for EG/Water (60:40). The deviation for the measured data is less than 1.0% compared to ASHRAE [45]. Reddy et al. [69] conducted a validation test for deviation from the base fluid of up to 2.5% compared to ASHRAE [70]. Therefore, further measurements and investigations for the thermal conductivity of the green nanofluids were carried out.

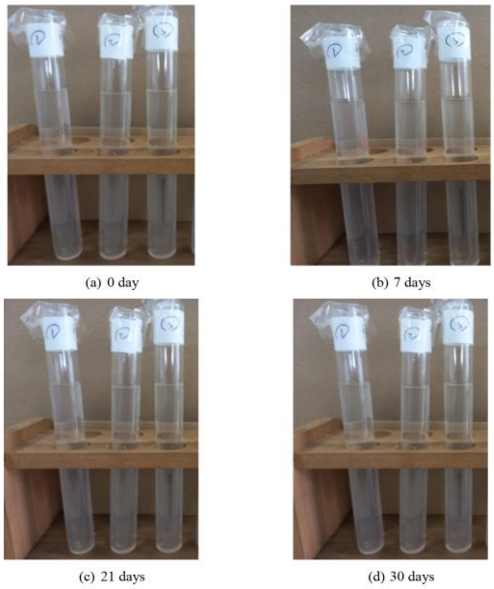

Figure 7. Sedimentation observation of green nanofluids: (a) 0 day, (b) 7 days, (c) 21 days, and (d) 30 days

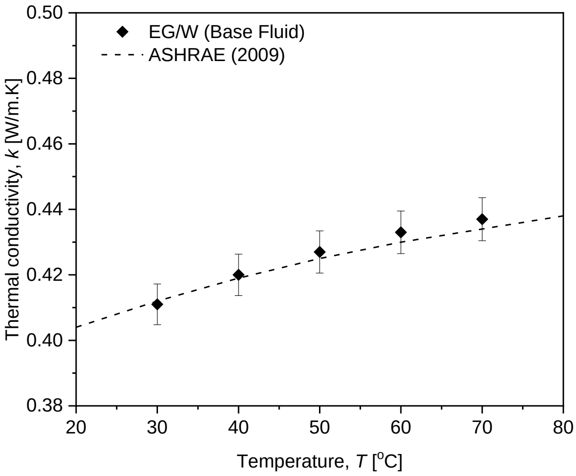

Figure 8. Validation of EG/Water (40:60) with ASHRAE for thermal conductivity

The thermal conductivity of green nanofluids

The relationship between the thermal conductivity of green nanofluid and temperature for volume concentrations of 0.1~0.3% is shown in Figure 9. The thermal conductivity of green nanofluid for variations in volume concentration can increase in relation to temperature and is higher than the base liquid. Furthermore, the highest thermal conductivity was obtained for a volume concentration of 0.3%. While the volume concentration of 0.1% gives the lowest thermal conductivity among the temperatures studied. In this study, the effect of the SiO_2 nanoparticle size of 50 nm on the increase in thermal conductivity. So it has a strong bond in filling the empty space. To increase the contact area for intermolecular conduction, resulting in a higher rate of heat transfer during collisions with Brownian motion [68]. The effective thermal conductivity of the green nanofluid is presented in Figure 10.

The results show that the effective thermal conductivity increases with a volume concentration of 0.3% in the green nanofluid, except for 0.1% volume where the effective thermal conductivity of the nanofluid is lowest. The effective thermal conductivity in green nanofluids can significantly influence the relationship between volume concentration and each temperature series.

Comparison of thermal conductivity with literature

Figure 11 shows a comparison of the effective thermal conductivity from this study with that of Murshed, et al. [71]. In this study, the effective thermal conductivity of green nanofluid increased 1.08-1.17 times compared to base fluid for a concentration of 0.1%. Murshed, et al. [71], using Al_2O_3 nanoparticles with a volume concentration of 0.5% of the basic fluid used was a mixture of EG-Water.

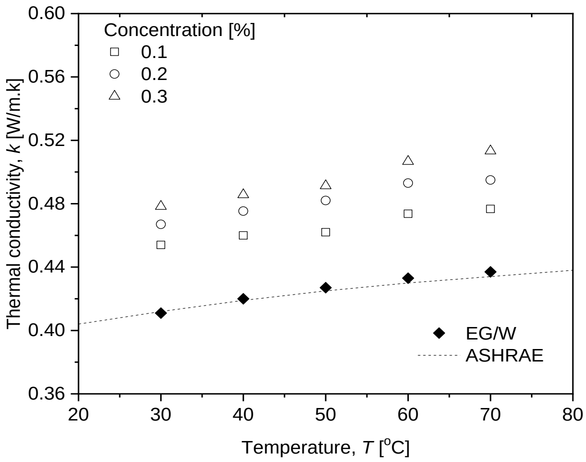

Figure 9. The experimental thermal conductivity of green nanofluids

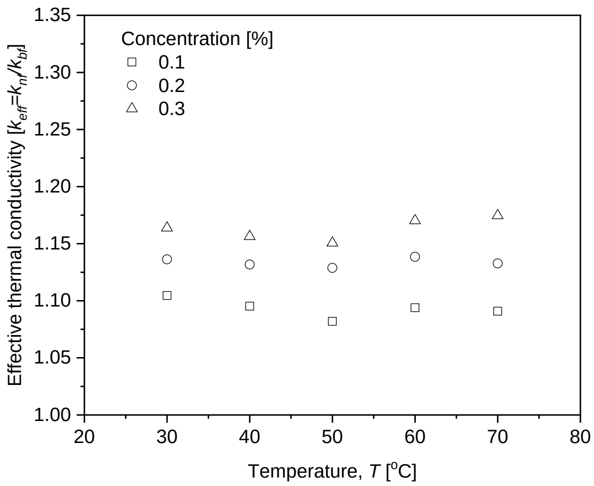

Figure 10. The effective thermal conductivity of green nanofluids

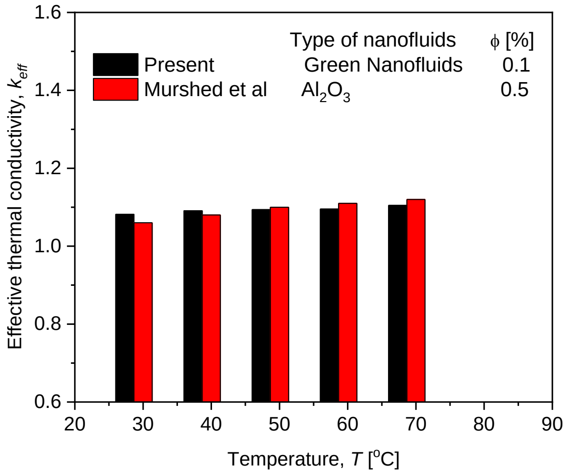

Figure 11. Comparison of green nanofluids of thermal conductivity with the data from literature

They proved that the experimental values of different thermal conductivities of nanofluids increased significantly with liquid temperature. This is due to the high fluid temperature increasing the movement of the Brownian nanoparticles. The effect of intense Brownian motion increases the contribution of microconnectivity to heat transfer, so that the thermal conductivity of the nanofluid increases.

CONCLUSION

In this research, the stability of green nanofluid was investigated for a volume concentration of 0.1~0.3%, and temperature conditions from 30 to 70°C for thermal conductivity measurement. The experimental results show that the stability analysis of the green nanofluid made with the UV-Vis method is stable up to 30 days after preparation with a sonication time of 1 hour. A comparison of data on the ratio of concentration to sedimentation for green nanofluid shows that it remains stable with a value of 70-80%. The zeta potential evaluation performed for green nanofluid obtained a value of 33.57 mV with a medium stability classification. Sedimentation from this visual observation is influenced by the gravitational force of the movement of particles that fall in the tube after 30 days. All methods prove that green nanofluids contain SiO2 nanoparticles or nano-silicates. The concentration volume of 0.3% obtained the best according to the effective thermal conductivity compared to 0.1~0.2%.

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