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Published by the Faculty of Engineering, Khon Kaen University, Thailand

The improved dielectric properties of natural ester oil by using ZnO and TiO_2 nanoparticles

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Received June 2016
Accepted October 2016

Abstract

A transformer is one of the most important devices in a power system. It is used for changing the voltage levels in an electrical system. Its lifetime depends on the heat exchange in the liquid insulation from the core and copper winding inside the transformer. Mineral oil is a widely used liquid insulation that causes problems when it contaminates the surrounding environment because it decomposes slowly. This research is intended to improve properties of a natural oil to replace mineral oil in power transformers. The currents study provides the results of tests on breakdown strength, flash point, fire point, and viscosity of the natural esters in soybean ester and palm esters mixed with nanoparticles. Titanium dioxide (TiO_2) and Zinc oxide (ZnO) nanoparticles were used in the investigation. Different quantities of nanoparticles were tested ranging from 0.01g/l-0.2 g/l. ASTM D1816, ASTM D93 and ASTM D2196 standard methods were used for testing breakdown strength, flash and fire points, and viscosity, respectively. The results indicated that the breakdown strength increased significantly when both TiO2 and ZnO nanoparticles were added to natural oil. The flash and fire points of two types natural oil had higher values than mineral oil. The addition of nanoparticles into the oil had no effect on its flash and fire points. The viscosity of natural oil had lower values than mineral oil. Dielectric properties can be improved by adding a suitable volume fraction of nanoparticles. However, the other properties must be considered before using a natural ester in the field.

Keyword: Nanofluids, Natural ester oil, Dielectric properties, ASTM D1816, ASTM D93, ASTM D2196

1. Introduction

In the power systems field, transformers connect two separate and different voltage systems. They play an important role in the conversion of voltage levels (LV-HV or HV-LV) [1]. When a transformer is operating, it generates heat in the core and copper winding. This heat can increase and cause it to operate ineffectively. Electrical engineers must use a liquid insulator such as mineral oil to transfer the heat. When transformer is operating, it transfers heat into the oil. This oil helps to reduce the heat of the core and copper winding by convection. As a liquid insulator in the transformer, good oil must have good heat transfer properties to dissipate heat. It must also have appropriate values for its breakdown strength, flash point, fire point and viscosity. A transformer may fail [2] if the liquid insulator cannot transfer heat away from core and copper winding.

In the past decade, there have been many studies focused on the process of cooling the core and copper winding of a transformer. Initially, they promoted heat transfer using microparticles mixed with mineral oil [3]. These oils did increase heat transfer, but they did not succeed since the microparticles tended to settle quickly which reduced the breakdown voltage of the oil. Further development shifted into using nano-sized instead of micro-size particles. The nanofluids did in fact promote heat transfer. Their heat transfer coefficients were higher than for unmodified mineral oil [4]. The addition of an appropriate amount of nanomaterials also improved the breakdown voltage of the oil [5-8]. Although mineral oil can be used as an insulator, it is not biodegradable, causing harm to the environment if it contaminates the soil or water. It is better for the environment if a biodegradable vegetable oil is used instead of mineral oil. However, vegetable oil is a poor liquid insulator. The properties of vegetable oil can be enhanced by conversing it to the form of an ester oil. This is a "natural ester oil" which is more environmentally friendly and has properties suitable as an insulating liquid [9].

This research aims to develop a natural oil for use as a liquid insulator and improve heat exchange by adding nanomaterials. Herein, we report the results of experiments measuring the dielectric breakdown strength, flash point, fire point, and viscosity of natural ester oil, i.e., soybean ester and palm ester to which the nanomaterials, ZnO and TiO_2, were added. The amounts of ZnO and TiO_2 nanoparticles added to two types of natural esters, ranged from 0.01g/l, 0.05g/l, 0.10g/l, 0.15g/l and 0.2 g/l.

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doi: 10.14456/easr.2017.22

2. Materials and methods

2.1 Procedures to do ester oil and nanofluids

Soybean and palm oils were chosen for experimentation. The procedure of preparation involved reacting fatty acids in the oil by methyl transesterification [10]. Methyl alcohol was used in this reaction and the catalyst was sodium hydroxide. First, 10 grams of sodium hydroxide were added to 250 ml of methyl alcohol. The mixture was stirred using magnetic stirrer for 10 minutes at 300 rpm. Then, the temperature of the oil was increased to 60 °C to speed up the reaction. This solution was then added to the oil at a 1:4 solution:oil ratio and stirred for 10 minutes. It was left to stand for four hours for the reaction as shown in the Figure 1 to take place. When the transesterification reaction was completed, glyceride and methyl ester were separated into two layers. Ester oil was obtained by removing the methyl ester layer and washing it five times with water. Finally, it was heated to 100 °C to evaporate the water from the oil.

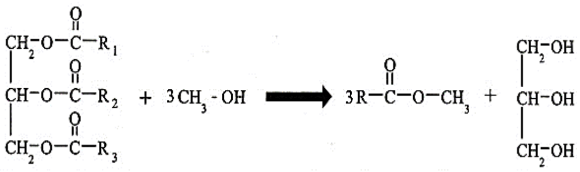

Figure 1 Reaction of oil with methyl ester

After obtaining ester oil, the two types of nanoparticles, ZnO and TiO_2, were added to the ester oil in concentrations ranging from of 0.01-0.2g/l using sonication. Cetyl trimethyl ammonium bromide (CTAB) of 3g/l was used as a surfactant to suspend the nanomaterials. The preparation of nanofluids was carried out by sequentially stirring, ultrasonicating and dehumidifying. as shown in Figure 2. First, 3g/l of surfactant was added to 10 ml of methanol and mixed using a magnetic stirrer for 15 minutes. After that, the magnetic stirrer was used to blend the solution with natural ester oil for 15 minutes. Then, this mixed solution was heated to 100 °C overnight to evaporate the methanol from the natural ester oil (boiling point of methanol, 64.6 °C). Next, the excess surfactant will form double bonds and precipitate. Therefore, this solution must be filtered. Then, the nanoparticles were mixed into the natural ester oil using a magnetic stirrer for 15 minutes. After that, the mixture was subjected to 50 kHz ultrasonic waves for two hours. Finally, moisture in the natural oil was removed in an oven at 60 °C for one day before its properties were determined. Various amounts of nanoparticles were tested in this step using both palm and soybean ester oil.

2.2 Moisture content and AC breakdown voltage tests

Before measurements of dielectric breakdown began, the moisture contents of natural oils were determined for all samples. The ASTM D1816 standard was used for measurement of dielectric breakdown voltage using a Megger OTS100AF electrical breakdown tester. The electrode is a brass cylinder with the gap was 2.0 mm. The frequency was 61.8Hz. The level of voltage was increased to 0.5kV/s during the measurements. Each sample was tested for AC breakdown voltage five times and the average breakdown voltage determined. The measurements of the moisture content and breakdown voltage were done at 26 °C.

2.3 Flash point and fire point test

Flash and fire points were measured according to ASTM D93 using a Koehler K16200 Pensky-Martens closed cup flash tester. The natural oil was placed in a brass test cup and fitted with a lid. The samples were heated and stirred inside the brass test cup. When the heat was increased, an ignition source was directed into the cup. The procedure was repeated until a flame was briefly seen inside the cup. This temperature was the flash point. After the flash point, the fire point was observed at a temperature that was about 5- 30 °C higher. At the fire point, a flame was observed burning continuously for at least 2-3 seconds. An ignition source is directed into the cup when the temperature was increased by 2 °C. These experiments were done in duplicate.

2.4 Viscosity

A nanofluid is a non-Newtonian material. In the current study, nanofluid viscosity was measured using a Brookfield DV-E viscometer according to ASTM D2196 over a range of temperatures from 27°C to 70°C. Spindle number one was selected for testing viscosity. The rotation of the spindle was adjusted to 100 rpm. The temperature of the nanofluid was increased using a hot plate and monitored using a digital thermometer. Viscosity values were recorded once the readings stabilized. All measurements were done in duplicate.

3. Experimental results and discussion

3.1 Natural ester oil and nanofluids

The dispersant could not be dissolved in oil by itself. It required an addition of methanol as a solvent. Methanol will adversely affects the quality of the oil by increasing acidity and decreasing its breakdown voltage When the solvent was removed, it caused the dispersant to precipitate.

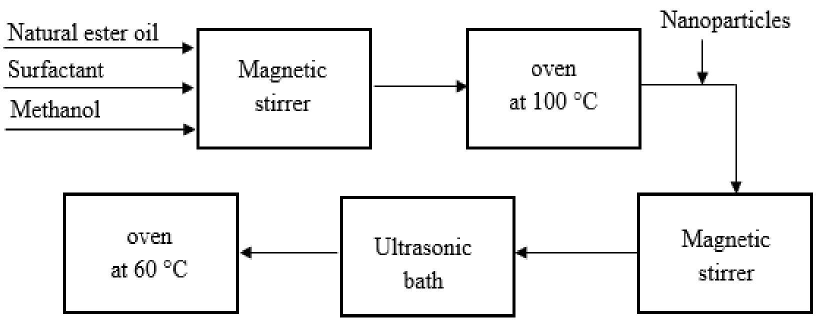

Figure 2 The procedures of preparation nanofluids

The dispersant itself will recombined with an excessive amount of dispersant used called double chain. The volume fraction of dispersant therefore affects the dispersion of nanoparticles. If little dispersant is used, the nanomaterials cannot be suspended in oil. However, an excessive amount of dispersant will form double chains among the dispersant molecules at the surface of the nanoparticles. It is necessary to use the correct amount of dispersant. Otherwise it will modify the nanoparticle surfaces, acting against attractive forces and preventing the formation of double chains [11].

3.2 AC breakdown voltage tests

All samples were tested for dielectric breakdown strength using the ASTM D1816 standard. The results of this testing are shown in Table 1. Humidity has a significant effect on breakdown strength. The oil must have a reduced moisture content. All samples had similar moisture contents.

Table 1 Dielectric breakdown and moisture content for each of the oils.

Types of oil	Breakdown strength (kV)	Moisture content (ppm)
Mineral oil	19.8	22
Soybean ester	20.6	23
Soybean ester mixed TiO_2 0.01 g/l	22.3	23
Soybean ester mixed TiO_2 0.05 g/l	24.1	24
Soybean ester mixed TiO_2 0.10 g/l	25.7	23
Soybean ester mixed TiO_2 0.15 g/l	29.4	23
Soybean ester mixed TiO_2 0.20 g/l	33.6	24
Soybean ester mixed ZnO 0.01 g/l	21.7	23
Soybean ester mixed ZnO 0.05 g/l	22.6	23
Soybean ester mixed ZnO 010 g/l	26.5	23
Soybean ester mixed ZnO 0.15 g/l	29.1	24
Soybean ester mixed ZnO 0.20 g/l	28.7	23
Palm ester	18.9	23
Palm ester mixed TiO_2 0.01 g/l	20.2	23
Palm ester mixed TiO_2 0.05 g/l	22.6	23
Palm ester mixed TiO_2 0.10 g/l	25.2	23
Palm ester mixed TiO_2 0.15 g/l	26.1	23
Palm ester mixed TiO_2 0.20 g/l	29.1	23
Palm ester mixed ZnO 0.01 g/l	20.0	23
Palm ester mixed ZnO 0.05 g/l	21.6	23
Palm ester mixed ZnO 0.10 g/l	26.2	24
Palm ester mixed ZnO 0.15 g/l	26.8	23
Palm ester mixed ZnO 0.20 g/l	27.3	23

The breakdown voltages of TiO_2 nanofluids increased when more nanoparticles were added. The change in breakdown voltage with increasing levels of TiO_2 is shown in Figure 3. The best result was with addition of0.2g/l TiO_2 in both types of oil.

Oils containing ZnO behaved in a similar fashion as those with TiO_2. The addition of ZnO nanoparticles increased the breakdown voltages of both types of oil, as shown in Figure 4. Oils containing 0.2g/l of ZnO nanoparticles had had the highest breakdown voltages, similar to TiO_2.

The effects of nanoparticles on ZnO and TiO_2 were compared in soybean ester and palm ester, as shown in Figure 5 for soybean ester and Figure 6 for palm ester, respectively. The highest breakdown voltage was observed when 2.0 g/l of nanoparticles were added to the oil. TiO_2 increased the breakdown voltage to a greater degree than ZnO. However, both TiO_2 and ZnO nanoparticles can improve the breakdown voltage of oil.

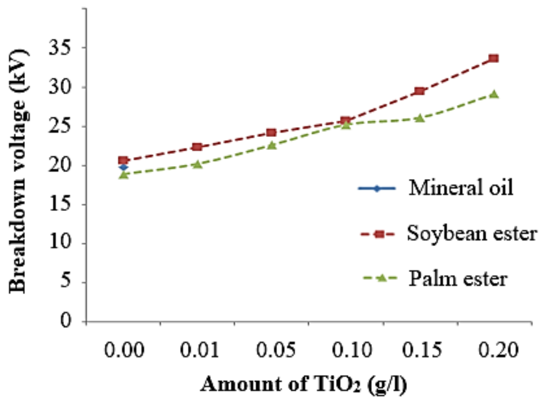

Figure 3 Comparison of breakdown voltage in two types of the oils with on TiO_2 nanoparticles

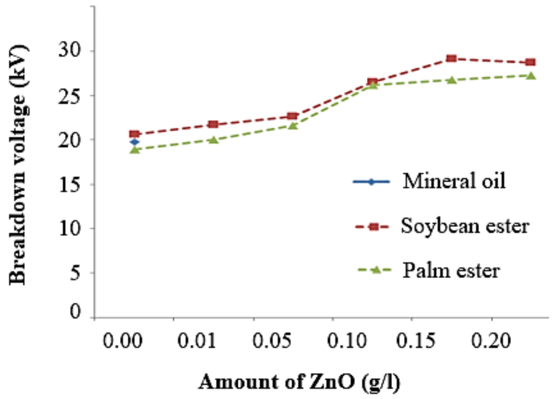

Figure 4 Comparison of breakdown voltage in two types of the oil based on ZnO nanoparticles

3.3 Voltage breakdown strength and relaxation time constant

The breakdown voltage of both types of ester oils increased with the addition of increasing levels of nanoparticles. The results of the experiment involved the theory of a charge relaxation with a time constant [12] and its effect on the dielectric breakdown strength when nanoparticles were mixed with a liquid insulator. The charge relaxation time constant of nanomaterials has a large effect on streamer propagation. It involves a timescale over which streamer development is inhibited. If the timescale of a nanofluid is short, it will quickly capture free electrons that will flow around its surface. In this case, streamer propagation will be slow. Alternatively, if the timescale of a liquid insulator is longer, the movement of free electrons is not as well inhibited since its surfaces are negatively charged and will not attach to free electrons. In this case, streamer propagation will be faster. The equation for the charge relaxation time constant (τ) is:

$$\tau = \frac{2\varepsilon_1+\varepsilon_2}{2\sigma_1+\sigma_2} \quad (1)$$

where ε_1, σ_1 are the conductivity and permittivity of the liquid insulator. ε_2 , σ_2 are the permittivity of nanoparticles. In the literature [13], magnetic nanomaterials have shorter charge relaxation time constants than semiconductor nanomaterials. ZnO nanoparticles are a magnetic material that has low permittivity and high

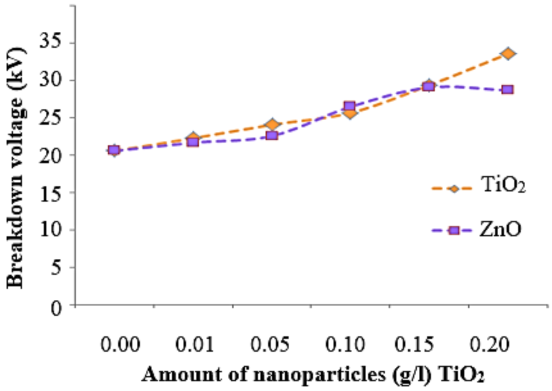

Figure 5 Comparison of breakdown voltage between TiO_2 and ZnO in soybean ester based on nanofluids

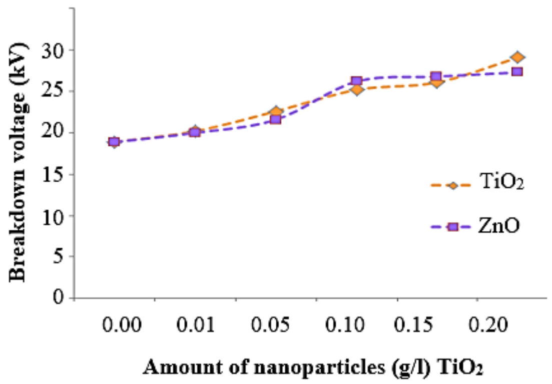

Figure 6 Comparison of breakdown voltage between TiO_2 and ZnO in palm ester based on nanofluids

conductivity. So, its charge relaxation time constant is shorter than for TiO_2 nanoparticles, a semiconductor. The dielectric breakdown strengths of all samples containing nanoparticles were higher than for either ester oil or transformer oil with no nanoparticles. This is because the surfaces of nanoparticles in oil attracted free electrons. So, both ZnO and TiO_2 can attract and trap electrons when an electrical field is applied. This is supported by the data in Table 1 showing that the breakdown voltages of all nanofluids increased with higher levels of nanoparticles because the spacing between particles becomes relatively small. Then, electrons were trapped in the gaps between the nanoparticles. This result indicated that an appropriate volume fraction of nanoparticles can be used to develop the desired dielectric breakdown strength in a liquid insulator [14].

The action of nanoparticles can be explained using Figure 7. This is a model of the electrodynamics within an electrically stressed natural ester oil based on a nanofluid. The condition assumed perfectly conducting nanoparticles with infinite permittivity. Electrons were injected into the system. Electrons travel along the electric field lines and approach the nanoparticles where the radial electric field is positive. The electric field lines will terminate at the bottom and create a negative surface charge that emanates from the top side with a positive surface charge on the particle, as shown Figure 7(a) and 7(b). The electrons in the natural ester oil near nanoparticles will move in the opposite direction to the field lines and deposit on the nanoparticles, where the surface charge is positive, as shown in Figure 7(c). The nanoparticles can capture electrons rapidly, depending on its charge relaxation time constant. Nanoparticles with a short relaxation time constant can quickly capture free electrons.

The relaxation time constant of ZnO was shorter than for TiO_2. As such, the breakdown voltage of ZnO nanoparticles should be higher than for TiO_2. However, in this experiment, it was observed that TiO_2 had a little effect on the breakdown strength than ZnO nanoparticles. This may be due to the different grades of nanoparticles used. The TiO_2 nanoparticles were laboratory grade with a 21 nm size. However, the ZnO nanoparticles were industrial grade with a size of less than 100 nm. The ZnO nanoparticles were much larger surface area when compare with the TiO_2. So, an equal mass of TiO_2 has a higher number of particles. As such, TiO_2 can capture more free electrons than ZnO. Therefore, the value of breakdown voltage when adding TiO_2 was higher than ZnO.

3.4 Flash point and fire point test

The flash and fire points all of samples in this experiment were tested to ensure for usability according to the ASTM D93 standard. Table 2 shows the flash and fire points of all samples.

It can be observed that flash and fire points the samples were generally higher for than mineral oil with flash and fire points of 158 °C and 180 °C, respectively. Natural ester oil has higher flash and fire point values than mineral oil because mineral oil is composed of more volatile petroleum derived substances that burn faster at lower temperatures. Higher flash and fire points are very desirable because this condition reduces the risk of explosion and fire when transformers operate at high temperatures. Addition of nanoparticles to a natural ester oil has no effect on flash and fire point values.

Table 2 Measured flash and fire points of all samples

Types of oil	flash point (°C)	fire point (°C)
Mineral oil	158	180
Soybean ester	178	206
Soybean ester mixed TiO_2 0.01 g/l	182	210
Soybean ester mixed TiO_2 0.05 g/l	180	210
Soybean ester mixed TiO_2 0.10 g/l	178	204
Soybean ester mixed TiO_2 0.15 g/l	178	210
Soybean ester mixed TiO_2 0.20 g/l	178	208
Soybean ester mixed ZnO 0.01 g/l	180	208
Soybean ester mixed ZnO 0.05 g/l	178	204
Soybean ester mixed TiO_2 0.10 g/l	180	206
Soybean ester mixed ZnO 0.15 g/l	180	210
Soybean ester mixed ZnO 0.20 g/l	178	202
Palm ester	177	208
Palm ester mixed TiO_2 0.01 g/l	178	218
Palm ester mixed TiO_2 0.05 g/l	172	206
Palm ester mixed TiO_2 0.10 g/l	174	202
Palm ester mixed TiO_2 0.15 g/l	178	202
Palm ester mixed TiO_2 0.20 g/l	178	200
Palm ester mixed ZnO 0.01 g/l	178	208
Palm ester mixed ZnO 0.05 g/l	180	210
Palm ester mixed ZnO 0.10 g/l	176	212
Palm ester mixed ZnO 0.15 g/l	176	200
Palm ester mixed ZnO 0.20 g/l	178	190

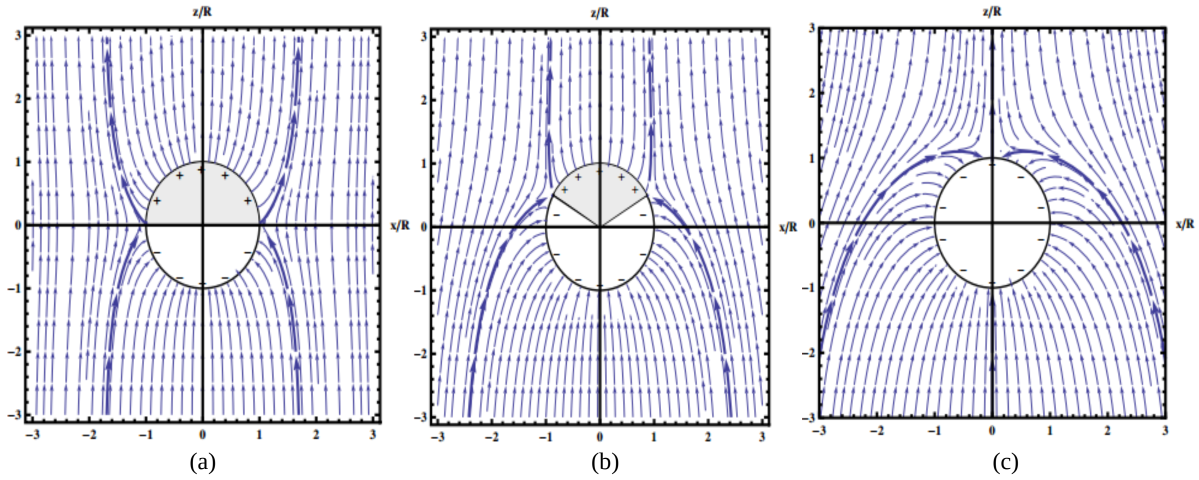

Figure 7 Model of the electrodynamics of a nanoparticles within an electrically stressed natural ester oil based on a nanofluid

Table 3 Viscosity of all samples measured between 27°C and 70°C

Types of oil	Viscosity (cP or mPa s)					
	27°C	30°C	40°C	50°C	60°C	70°C
mineral oil	1.913	1.802	1.640	1.163	1.059	0.919
Soybean ester	1.067	1.059	0.907	0.776	0.685	0.675
Soybean ester mixed TiO_2 0.01 g/l	1.069	1.050	0.881	0.812	0.714	0.666
Soybean ester mixed TiO_2 0.05 g/l	1.100	1.082	0.912	0.817	0.726	0.675
Soybean ester mixed TiO_2 0.10 g/l	1.112	1.079	0.923	0.810	0.713	0.675
Soybean ester mixed TiO_2 0.15 g/l	1.122	1.086	0.941	0.828	0.761	0.672
Soybean ester mixed TiO_2 0.20 g/l	1.156	1.106	0.936	0.844	0.759	0.680
Soybean ester mixed ZnO 0.01 g/l	1.071	1.061	0.909	0.802	0.716	0.666
Soybean ester mixed ZnO 0.05 g/l	1.085	1.085	0.919	0.830	0.721	0.647
Soybean ester mixed ZnO 0.10 g/l	1.128	1.109	0.919	0.815	0.757	0.675
Soybean ester mixed ZnO 0.15 g/l	1.142	1.114	0.921	0.828	0.732	0.670
Soybean ester mixed ZnO 0.20 g/l	1.164	1.112	0.934	0.834	0.766	0.687
Palm ester	1.106	1.078	0.919	0.816	0.715	0.666
Palm ester mixed TiO_2 0.01 g/l	1.122	1.080	0.900	0.820	0.729	0.669
Palm ester mixed TiO_2 0.05 g/l	1.148	1.088	0.919	0.817	0.729	0.672
Palm ester mixed TiO_2 0.10 g/l	1.153	1.109	0.912	0.824	0.750	0.669
Palm ester mixed TiO_2 0.15 g/l	1.159	1.105	0.927	0.826	0.747	0.672
Palm ester mixed TiO_2 0.20 g/l	1.172	1.144	0.925	0.837	0.778	0.677
Palm ester mixed ZnO 0.01 g/l	1.114	1.086	0.909	0.816	0.728	0.668
Palm ester mixed ZnO 0.05 g/l	1.126	1.097	0.919	0.823	0.745	0.666
Palm ester mixed ZnO 0.10 g/l	1.159	1.100	0.929	0.834	0.757	0.670
Palm ester mixed ZnO 0.15 g/l	1.162	1.117	0.922	0.836	0.757	0.668
Palm ester mixed ZnO 0.20 g/l	1.183	1.146	0.934	0.855	0.782	0.675

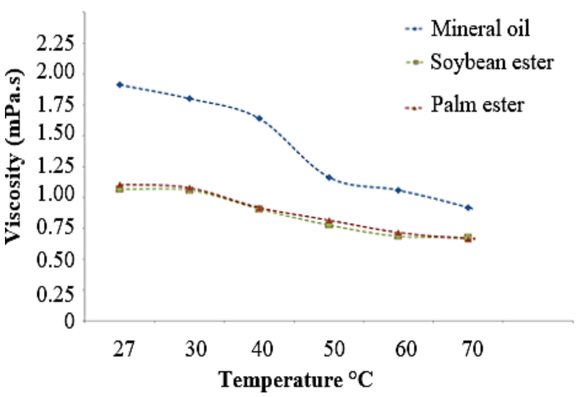

Figure 8 Comparison of the viscosity of soybean ester oil, palm ester oil and mineral oil

3.5 Viscosity

Viscosity is an important property of transformer oil. It influences both its electrical properties and heat transfer. Viscosity is an important indicator of the effect of nanoparticles in oil. The viscosity of oil samples was measured at 27 °C to 70 °C, as shown in Table 3.

From Table 3, it can be seen that viscosity of mineral oil was the highest of any of the materials examined. Viscosity affects the cooling of a transformer because heat transfer is mainly by convection. It is important to have a low viscosity for better convection. Furthermore, solid nanoparticles can also conduct heat faster. When nanoparticles are added, it has little impact on the viscosity of an oil. The viscosities of all samples decreased with increasing temperature. The change of viscosity due to the addition of nanoparticles was negligible, especially above 40 °C.

Both types of natural ester oils had lower viscosities than mineral oil. There was little different between the viscosities of palm ester oil and soybean ester oil. The viscosity of soybean ester oil was the highest at 1.067 mPa.s at 27°C and the lowest value was 0.675 mPa.s at 70°C. The viscosity of palm ester was the highest at 1.106 mPa.s at 27°C and the lowest at 0. 0.666 mPa.s at 70°C, as shown in Figure 8. The viscosity of palm ester oil at 27°C was slightly higher than that of soybean. This may have been due to a five-fold higher ratio of unsaturated/saturated fatty acids in palm oil than in soybean. Even so, the esterification process was done under the same conditions. Perhaps, triglycerides affected viscosity in the oil and they could not be removed from the oil. Therefore, palm ester oil had a slightly higher viscosity than soybean ester oil.

4. Conclusions

The results indicated that the properties of natural ester oil including the dielectric breakdown strength, flash and fire points as well as viscosity were better than for mineral oil. The addition of increasing amounts of nanoparticles can improve the dielectric breakdown properties of a natural ester oil. Increasing amounts of nanoparticles had negligible effect on the flash point, fire point and viscosity of an oil. However, the natural ester oil mixed with nanoparticles may cause sedimentation inside of transformers as it is a solid particle when there are no oil circulation inside transformer tank. So, improvements must be made to ensure that these replacement oils can adequately replace a conventional mineral oil.

5. Acknowledgment

This research was supported by the Centers for Alternative Energy Research and Development (AERD), Khon Kaen University, Mittapab Road, Khon Kaen 40002, Thailand

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