Influence of Conductive and Semi-Conductive Nanoparticles on the Dielectric Response of Natural Ester-Based Nanofluid Insulation

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Abstract: Nowadays, studies of alternative liquid insulation in high voltage apparatus have become increasingly important due to higher concerns regarding safety, sustainable resources and environmentally friendly issues. To fulfil this demand, natural ester has been extensively studied and it can become a potential product to replace mineral oil in power transformers. In addition, the incorporation of nanoparticles has been remarkable in producing improved characteristics of insulating oil. Although much extensive research has been carried out, there is no general agreement on the influence on the dielectric response of base oil due to the addition of different amounts and conductivity types of nanoparticle concentrations. Therefore, in this work, a natural ester-based nanofluid was prepared by a two-step method using iron oxide (Fe2O3) and titanium dioxide (TiO2) as the conductive and semi-conductive nanoparticles, respectively. The concentration amount of each nanoparticle types was varied at 0.01, 0.1 and 1.0 g/L. The nanofluid samples were characterised by visual inspection, morphology and the dynamic light scattering (DLS) method before the dielectric response measurement was carried out for frequency-dependent spectroscopy (FDS), current-voltage (I-V), and dielectric breakdown (BD) strength. The results show that the dielectric spectra and I-V curves of nanofluid-based iron oxide increases with the increase of iron oxide nanoparticle loading, while for titanium dioxide, it exhibits a decreasing response. The dielectric BD strength is enhanced for both types of nanoparticles at 0.01 g/L concentration. However, the increasing amount of nanoparticles at 0.1 and 1.0 g/L led to a contrary dielectric BD response. Thus, the results indicate that the augmentation of conductive nanoparticles in the suspension can lead to overlapping mechanisms. Consequently, this reduces the BD strength compared to pristine materials during electron injection in high electric fields.

Keywords: natural ester; dielectric properties; electrical insulation; high voltage engineering

1. Introduction

Mineral-based insulation oil has been widely used in oil-immersed power transformers. The key success of mineral oil is mainly due to its dual functionality as a good dielectric material with cooling abilities and thee low cost associated with its availability. Today, the expansion of urbanisation processes has led to ultra-high power demands and is consequently pushing power networks to their limits. Hence, the failure rate of large power transformers increases, resulting in major environmental
damage and safety concerns. For instance, mineral-oil-based transformer explosions can cause toxicity from the methane gas and oil spill onto the ground [1]. Hence, greater safety concerns have resulted in the search for an alternative insulation oil. One of these is natural ester.

Natural ester (NE) is a new class of fully biodegradable dielectric insulation liquid that is mainly developed from vegetable oils such as canola, rapeseed, soya beans, sunflower and palm oil. Since the 1990s, work on suitability of NE as insulation for power industrial applications in natural conditions, mixed with mineral oil or commercially produced as synthetic forms have been widely reported [2–5]. For instance, NE can be synthesized into several derivatives with different molecular structures and physiochemical properties. For example, increasing the length of the side branches of chemically-modified alkyl ester of palm kernel oil will result in a higher breakdown strength compared to mineral oil [6]. NE also has a higher molecular solubility and thereby can resist pressboard ageing and moisture-rich environments [7,8].

Nowadays, the existence of nanotechnology has been utilized and the addition of nanoparticles in insulating oil has produced a new remarkable material, known as nanofluids. NE-based nanofluids have higher dielectric strength, cooling efficiency, and ability to withstand electrical discharges [9,10]. For instance, the discovered role of nanoparticles when being dispersed into base oil is to energise the potential well. Therefore, it will trap the passing electrons and consequently lead to an enhancement of dielectric strength [11]. In spite of this, it has been reported that the addition of conductive nanoparticles in a certain amount reduces the dielectric strength of base oil [12–14]. When the number of nanoparticles is increased, the collision rate between the particles increases due to Brownian motion, but it appears as bridging between two conductors and could lead to breakdown [15]. The question is why an identical phenomenon is also reported for semi-conductive and dielectric-based nanofluids [16]. It was believed that the enhancement of dielectric strength in nanofluids not only depends on the intrinsic properties and interface regions formed between particles, as in solid phases [17–19]. However, it strongly depends on the potential mobility of charged particles in nanofluid due to electric field orientation. For instance, Ibrahim et al. [20] reported that the increasing number of conductive nanoparticles (Fe$_2$NiO$_4$) in the suspension will decrease the AC dielectric strength, and accumulated particles were observed at the electrode under the DC electric field. However, there is no general agreement regarding the influence of the conductivity and nanoparticle loading in nanofluid insulation. Therefore, this work provides an opportunity to gain a better understanding of this matter in the fluid phase by performing a comparative study using conductive and semi-conductive nanoparticles from low to high amount of concentration using different dielectric measurement techniques.

A comparison of nanoparticle conductivity with different concentrations used to synthesise a novel, natural, ester-based nanofluid intended as a high voltage insulation material is presented in this work. The study compares the influence of the surface conductivity of nanoparticles and the amount of concentration in relation to the dielectric responses of natural ester-based nanofluids in frequency-dependent, current–voltage relation and breakdown strengths in high electric fields. Iron oxide (Fe$_2$O$_3$) and titanium dioxide (TiO$_2$) were used in this work instead of other metal oxide nanoparticles, e.g., magnesium oxide (MgO) and zinc oxide (ZnO) due to the latter particles showing dissolution in the fluids with different pH values [21–23]. The results show that titanium dioxide is more promising as a nanoparticle in natural ester for insulation purposes. The ability of TiO$_2$ nanoparticles to remain as electron scavengers at varying concentrations is also discussed. A stable suspension and particle size distribution after preparation is also described in this investigation.

2. Experimental

2.1. Materials

In this experiment, the natural ester used as the base oil was highly-refined, bleached, deodorised palm oil (RBDPO), which was commercially obtained from Lam Soon Edible Oils Sdn. Bhd. (Shah Alam, Malaysia). Iron oxide, Fe$_2$O$_3$ ≥ 97% and titanium dioxide, TiO$_2$ ≥ 99.5% were both obtained
from Sigma Aldrich (St. Louis, MO, USA) in nanopowder form. They were used as conductive and semi-conductive nanoparticles, respectively.

2.2. Sample Preparation

First, all materials were dried at a temperature of 80 °C (for base oil) and 300 °C (for nanoparticles) in a thermal vacuum (90 kPa) chamber for one to two days. After drying and cooling-down processes, nanoparticles were carefully added into 500 mL of natural ester (palm oil) with certain ratios as shown in Table 1. In order to achieve a stable suspension, the sonication treatment was performed for 30 min at room temperature using an ultrasonic bath (output power = 120 W, frequency = 40 kHz).

| Sample Abbreviation ** | Amount of Nanoparticles [g/L] |
|------------------------|------------------------------|
|                        | Iron Oxide       | Titanium Dioxide |
| NE                     | -               | -               |
| NE/Fe-a                | 0.01            | -               |
| NE/Fe-b                | 0.1             | -               |
| NE/Fe-c                | 1.0             | -               |
| NE/Ti-a                | -               | 0.01            |
| NE/Ti-b                | -               | 0.1             |
| NE/Ti-c                | -               | 1.0             |

** NE = Natural ester, Fe = Iron oxide, Ti = Titanium dioxide, a = 0.01 g/L, b = 0.1 g/L, c = 1.0 g/L.

2.3. Sample Characterisation Techniques

The nanofluid samples were characterized through three techniques: visual inspection, morphology analysis and dynamic light scattering (DLS). Visual inspection is a technique for evaluating the tendency of nanofluids to aggregate and eventually boost the sedimentation process with time. After the natural ester-based nanofluid samples have been prepared, each sample was poured into a cylindrical glass tube. The samples were left for 24 h without shaking to observe any changes of the condition due to natural ambience before measurements were performed. The morphology analysis was conducted using a scanning electron microscope (SEM) (Zeiss EVO® MA10, Oberkochen, Germany). The nanofluid sample was deposited on the sticky tape of a SEM specimen holder and was dried until solid particles were obtained. The dynamic light scattering (DLS) method was applied to compare the average aggregate size of the nanoparticles at 0.01 g/L. Measurements were performed at 25 °C using a Malvern Zetasizer (Malvern, UK).

2.4. Dielectric Response Measurements

Frequency dielectric spectroscopy (FDS) response and current-voltage (I-V) characteristic measurements for the liquid under test were used in this work. The setup consists of two cylindrical copper plates with a diameter of 25 mm and 100 µm gap distance. Both electrodes were fully immersed in each nanofluid sample. For FDS measurement, a sinusoidal AC voltage of 0.5 Vrms was applied to the test cell at room temperature with various frequencies, ranging between 0.1 Hz and 0.1 MHz. The impedance results from the measurement were plotted in terms of dielectric constant, \( \varepsilon' \) and dielectric loss, \( \varepsilon'' \). For I-V characteristic measurements, the parameters were adjusted using linear sweep voltammetry (LSV) with a scan rate of 0.05 V/s and a step potential of 0.20 V. The applied DC voltage was within a range of 1 to 10 V.

The dielectric breakdown voltage (BDV) of each sample was measured using an automated test kit with a maximum applied voltage of 60 kV. The setup and procedure for each breakdown of samples were in accordance with the ASTM D1816 standard. The gap space of the hemisphere electrodes was set to 1.0 mm. The measured breakdown voltage of each sample was compared with
the average breakdown voltage and Weibull distribution. The cumulative distribution function of Weibull distribution for any random variable $x$ is defined as

$$f(x) = 1 - \exp(-x/a)\beta$$  \hspace{1cm} (1)

where $a$ is the scale parameter that corresponds to a probability of 63.2% and $\beta$ is the shape parameter that is related to the scattering of the data and failure rate. To estimate the probability of $x$ as a function of breakdown voltage, the Weibull cumulative distribution function can be transformed into a straight line form as given by

$$\ln\left[\ln\left(\frac{1}{1-f(x)}\right)\right] = \beta\ln(x) - \ln(a)$$  \hspace{1cm} (2)

Hence, $\beta$ is directly represented by the slope of the line and $a$ is evaluated from the $y$ data that intercepts with it.

3. Results
3.1. Sample Characterisation Results

Figure 1 shows the natural ester samples with different concentration levels of iron oxide and titanium dioxide nanoparticles. When the nanoparticle concentration was increased, the nanofluid samples with iron oxide took on a dark colour while the nanofluid samples with titanium dioxide became milky. Visual inspection was made at the base of the glass tube to observe any sedimentation occurrence. If sedimentation occurred, the sample was rejected and a new sample was prepared. In this work, no sedimentation occurred for 24 h, indicating that the dispersion of nanoparticles within the sample is uniform and the sample was accepted for further experiments.

![Figure 1](image1.png)

**Figure 1.** Samples of natural ester (NE)-based nanofluids; with: (a) Iron oxide; (b) Titanium dioxide.

Figure 2a,b compares the distribution of the particles cross-sectional diameter and the volume of the diversely-sized particle constituted. It can be seen that the iron oxide particles occupied a higher distribution compared to titanium dioxide. Therefore, it has a marked impact on the dielectric response of the suspension. Figure 2b shows the morphology of the formed particles from the titanium dioxide when a higher amount 1.0 g/L was deposited from the suspension. It was presumed that the nanoparticles were dispersed homogeneously due to only a small agglomeration size being observed. A Van Der Waals force from ultrasonication created a repulsive electrostatic potential between the electric double layers (EDL) surrounding the particles [24]. Consequently, the fluid stability was enhanced.
3.2. Frequency Dielectric Response (FDS) Results

Figure 3a,b shows the measured dielectric spectroscopy at different frequencies of natural ester-based nanofluid samples. It can be seen clearly that the dielectric constant, $\varepsilon'$ of NE/Fe a, b, and c exhibits slightly stronger frequency-dependency with an increase of iron oxide concentration at 0.01, 0.1 and 1.0 g/L, respectively. Surprisingly, the dielectric constant, $\varepsilon'$ of the NE/Ti shows quite different trends, especially for the concentration amount of titanium dioxide at 0.1 and 1.0 g/L. Although the presences of the hydroxyl group on the surface of particles or the impurities, such as polar species, can affect the polarization, it can be minimized by pre-heat treatment during sample preparation [25,26]. Therefore, the apparent polarization in this study is the summation of the polarization of the base oil, inner polarization of nanoparticles and orientation polarization of charged nanoparticles as polar molecules due to Miao et al. [27]. Since the electrical properties of iron oxide are more conductive than titanium dioxide, it can be more easily charged and polarized than titanium dioxide when being dispersed in natural ester. The mobility of charged particles may also influence the polarization process at low frequencies.

Figure 4a,b shows the measured dielectric loss $\varepsilon''$ as a function of frequency for natural ester-based nanofluid samples with conductive and semi-conductive nanoparticles. At very low frequencies, natural ester-based nanofluid samples with conductive, NE/Fe a, b and c exhibit higher dielectric loss $\varepsilon''$ than the samples with semi-conductive nanoparticles, NE/Ti a, b, and c. At higher frequencies, relaxation spectra are dominant and clearly visible for all samples. This is associated with a descend
conduction mechanism due to the low mobility of charge carriers, which is likely to be suppressed by a high frequency field. However, at lower frequencies (0.1 Hz to 10 Hz), when nanoparticles were dispersed in a base oil, free charges in the natural ester are trapped by nanoparticles. This hinders charge carriers to move between two electrodes and consequently, the dielectric loss decreases. However, as the concentration of nanoparticles is higher in the sample, the probability of the electric double layer (EDL) region overlapping is higher, producing a higher conductive region along the natural ester ions. The effect on the conductive path of the overlapping EDL is more sensitive in highly-conductive nanoparticles, which can be seen from the response of NE/Fe-c in Figure 4a.

![Figure 3](image-url)  
**Figure 3.** Dielectric constant $\varepsilon'$ as a function of frequency for natural ester-based nanofluids with: (a) Conductive nanoparticles; (b) Semi-conductive nanoparticles.

![Figure 4](image-url)  
**Figure 4.** Dielectric loss $\varepsilon''$ as a function of frequency for natural ester-based nanofluids with: (a) Conductive nanoparticles; (b) Semi-conductive nanoparticles.

### 3.3. Current-Voltage (I-V) Results

Figure 5a,b represent the I-V curves of natural ester-based nanofluids with conductive and semi-conductive nanoparticles. It was found that the I-V results show that the conduction current increases gradually with the voltage and verified that the samples have a resistance mode that is similar to the behaviour of nanofluids [28]. The resistivity value for pure natural ester is high, at 3.08 GΩ·m. Under the applied DC electric field, the number of electrons as charge carriers increases in the channel above the threshold voltage. Thus, the conduction current increases in symmetrical non-linear fashion with the voltage. At a low loading of nanoparticles, both the NE/Fe and NE/Ti samples show a reduction of charge mobility in the DC electric field. At this critical interface distance, it shows a trapping charge ability through a polarization and charge induction mechanism, consequently enhancing the insulation properties [29]. However, when the particle loading increases in the suspension, a higher volume of particles in conjunction with electric field orientation may deteriorate the critical interface distance between inter-particles. Therefore, it leads to a new condition...
where the intrinsic properties (particle surface conductivity) are more dominant. It can be seen from the I-V result that the natural ester-based nanofluid containing semi-conductive nanoparticles is reduced especially for NE/Ti-b and NE/Ti-c. This is due to the larger amount of titanium dioxide in natural ester, which could improve the charge trapping and increase the surface resistance. However, for natural ester-based nanofluids with iron oxide, the incorporation of conductive nanoparticles in larger amounts has modified the tube–tube contact resistance, which improved the conduction mechanism of the nanofluid. This proposed mechanism also can be attributed as the percolation threshold in nanofluid cases.

3.4. Dielectric Breakdown Voltage (BDV) Results

The measured dielectric breakdown voltage (BDV) for each sample is presented in Figure 6. The breakdown voltage of natural ester without nanoparticles, as indicated by NE, is around 31 kV. The BDV of NE/Fe-a and NE/Ti-a with 0.01 g/L amount of conductive, semi-conductive nanoparticles respectively is higher than the pure sample, NE. However, the BDV of NE-based nanofluids with conductive nanoparticles shows a higher BDV than the NE pure sample at lower amounts only. The dielectric breakdown voltage of NE/Fe nanofluids decreases significantly with the increasing amount of conductive nanoparticles.

![Figure 5. I-V curves of natural ester-based nanofluids with: (a) Conductive nanoparticles; (b) Semi-conductive nanoparticles.](image)

![Figure 6. Dielectric breakdown voltage of natural ester-based nanofluids.](image)
The average breakdown voltage does not represent the probability that the insulation will breakdown when being stressed with high electric fields. Thus, the Weibull distribution was applied as a statistical tool to provide the degree of withstanding capability of the samples. Since the lowest voltage at which breakdown will occur is the main interest for insulation applications, it is important to observe the lower tail of the Weibull distribution slopes. Referring to Figure 7a,b, it is observed that the range of the breakdown probability of the natural ester-based nanofluid with iron oxide is relatively lower for the NE/Fe-b and NE/Fe-c samples compared to the pure natural ester. The range of breakdown probability of the natural ester-based nanofluid with titanium dioxide is more pronounced than the pure natural ester, especially for the NE/Ti-b and NE/Ti-c samples.

![Figure 7. Weibull distribution of dielectric breakdown of natural ester-based nanofluids with: (a) Conductive nanoparticles; (b) Semi-conductive nanoparticles.](image)

4. Discussion

Electrical breakdown in a liquid dielectric is initiated by the streamer formation between two electrodes when the electric field intensity exceeds their threshold level. At this level, the molecules of the liquid are ionized and more ions and electrons are created, forming a conductive path known as a streamer. After a streamer is formed, it tends to elongate and transverse in the dielectric liquid from the point of initiation towards the lowest potential in the system, which is usually the grounded electrode. When the streamer reaches the grounded electrode, it forms a bridge or short circuit between the two electrodes. Eventually, a current or arc flows through it, which causes a dielectric breakdown [30].

It is known that refined, bleached, deodorised palm oil (RBDPO) is a low melting liquid fraction of natural ester, which consists of oleic acid (C18:1) at 48% due to refining and good bleaching processes [31]. Oleic acid acts as a natural surfactant when metal oxide nanoparticles are dispersed into a base oil during the sample preparation process. A mechanically-stable natural ester-based nanofluid sample is obtained when oleic acid is chemisorbed as carboxylate on the nanoparticle surface. Hence, the interfacial zone of the nanofluid as a solid–liquid suspension is enhanced [32]. The role of the interfacial zone in a nanofluid is to create intermolecular forces, which keeps the molecules from each other according to the Van Der Waals theorem. As a result, the nanoparticle consists of two layers, the affected and aligned layers, which are difficult to polarize, trapping charges and slowing down the electron mobility, as suggested by Mansour et al. [33]. For low volumes (0.01 g/L) of nanoparticles in natural ester, the interfaces have a large space to form an efficient distance of double layers. This is a possible mechanism explaining the reduction of the total amount of polarization for both types of nanoparticles, resulting in a decrease of dielectric constant. The aligned layers also create a rigid
intermolecular structure that traps electron and ion mobility between interparticle zones, resulting in a decrease in the conduction current and consequently an increase in the dielectric breakdown voltage.

On the other hand, for higher amounts of nanoparticles (0.1 and 1.0 g/L), the interparticle distance decreases, reducing the efficient width of double layers, and overlapping will potentially occur. In conjunction with the applied electric field, particles can be easily charged and move to bridging the flow of electrons [20]. For highly-conductive nanoparticles, such as iron oxide, when the amount in natural ester is high, a smaller interparticle distance affects the electric field enhancement. This is due to the effect of nanoparticle surface conductivity being more dominant. For semi-conductive and dielectric nanofluids, reduction and overlapping of the interfacial zone space does not affect the dielectric response much because they can attract and trap electrons in shallow traps, as well as having very low conductivity values [20,34]. Nevertheless, if impurities exist on the particle surface, they play an important role in the mechanism, increasing the electrical conductivity of the base material [25].

Figure 8 illustrates the electric displacement field and current density distributions in the nanofluid with the influence of nanoparticle properties using finite element analysis (FEA) between two plane electrodes. When the electrical properties of a nanoparticle are different from the base oil, the electric field intensity surrounding the nanoparticle will be distorted. For instance as described in [20], a larger distortion of the electric field results in a higher magnitude of the electric field on the nanoparticle interface, subsequently exerting a force that attracts free electrons. Also, a high current density at the nanoparticle surface shows that streamer penetration can occur more easily with the presence of highly conductive nanoparticle in natural ester. As can be seen from Figure 8a,b, natural ester-based nanofluids with iron oxide have higher magnitudes of current density compared to titanium dioxide, which can easily initiate a breakdown in high electric fields.

![Figure 8. The magnitude of surface current density on a nanoparticle: (a) Conductive; (b) Semi-conductive.](image)

5. Conclusions

In this work, investigation of the influence of nanoparticles, iron oxide and titanium dioxide on the dielectric response of natural ester-based nanofluid insulation was successfully performed. Some meaningful findings are summarized as follows:

1. Natural ester, such as palm oil, which consists of natural oleic acid as a substantial content, produces a stable nanofluid suspension when dispersed with metal oxide nanoparticles. Oleic acid plays an important role as an artificial surfactant, which enhances the suspension stability and avoids agglomeration.

2. At 0.01 g/L concentration of nanoparticles within natural ester, the effect of the interfacial zone was dominant and a large space for rigid intermolecular forces was created, requiring more
energy to break up. This behaviour was shown through the reduction of dielectric loss and the enhancement of dielectric strength for both types of nanoparticles.

3. When the concentration of nanoparticles within natural ester was increased to 0.1 and 1.0 g/L, respectively, the dielectric response of nanofluids became more dependent on the nanoparticle properties (in this case, surface conductivity). Since iron oxide is highly conductive compared to titanium dioxide, the enhancement of the polarization and conduction current was observed with increasing concentration of conductive nanoparticles. This has also led to the reduction of dielectric breakdown voltage.

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