

## Research Article

# Polarization and Conduction Characteristics of Mineral Oil and Natural Ester Mixed with Nanoparticles

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Mineral oil (MO) has been widely used in transformers because of its reasonable price and acceptable properties. Currently, alternative liquid insulations have been proposed with some excellent properties. Therefore, the mineral oil characteristics need to be enhanced for high performance to compete with such alternative liquid insulations. This paper aims to study the power frequency dielectric dissipation factor ( $\tan\delta$ ) along with polarization and depolarization current (PDC) characteristics of the mineral oil-based nanofluid and natural ester (NE)-based nanofluid. Besides, Fourier-transform infrared spectroscopy (FT-IR) measurement of the test specimen was performed. Titanium dioxide ( $\text{TiO}_2$ ) and zinc oxide (ZnO) with 0.01% and also 0.03% volume concentration were used to mix with the unmodified liquid to create liquid insulation-based nanofluid. Then, unmodified mineral oil, unmodified natural ester, mineral oil-based nanofluid, and natural ester-based nanofluid were comparatively investigated.  $\tan\delta$  at power frequency and the PDC characteristics of such liquids were examined. For PDC measurement,  $\text{TiO}_2$  clearly affected the polarization current and conduction loss factor, whereas ZnO evidently impacted the depolarization current and polarization loss factor. In the case of FT-IR test results, there was no significant change when the modified insulating liquid was compared with those of the unmodified insulating liquid.

## 1. Introduction

A power transformer is an important equipment in the power system, which increases or decreases the voltage level for the transmission and distribution system. Most power transformers use mineral oil (MO) as electrical insulation and cooling media. Mineral oil has been developed for a long time to be a crucial part of the transformers that desire more rated power needed as detailed in [1–3]. In general, liquid insulation can be divided into two types, i.e., organic and inorganic liquid insulation. Mineral oil, petroleum oil, plant

oil, etc., are examples of organic liquid insulation. Inorganic liquid insulation, such as liquid nitrogen, deionization water, etc., was produced by chemical synthesis. Mineral oil has been widely used in the transformer industry because of its reasonable price with acceptable properties. Currently, alternative liquid insulation such as natural ester (NE) and synthetic esters has been proposed with some excellent properties. Therefore, the mineral oil characteristics need to be enhanced to compete with such alternative liquid insulations for high performance. Regarding health and environment-friendly, natural ester is preferred. The natural

ester can biodegrade by 90%, but the mineral oil can only biodegrade by 10% when compared after 28 days of the experiment. The natural ester has higher water dissolved because its chemical structure polarizes its molecule and the polar molecule acts like a magnet. Therefore, the molecule of the natural ester can attract water molecules better than nonpolar molecules of mineral oil; this causes high water tolerance characteristics. Moreover, the natural ester has relative permittivity ( $\epsilon_r$ )  $\approx 3.2$ , but the mineral oil has  $\epsilon_r \approx 2.2$ . If the two types of liquid insulation were used in an identical liquid immersed transformer, the natural ester tends to undergo lower electric stress than the mineral oil. However, the natural ester has some disadvantages, i.e., high viscosity, high pour point, and ease of oxidizing. The viscosity affects the cooling rate of the electrical apparatus. The pour point of the natural ester is higher than that of the mineral oil; therefore, a usage under zero degree Celsius seems not very appropriate. The oxidation significantly affects the by-product existence that is not desired for the application of natural ester in the transformers [1–13].

The application of nanomaterials in the high voltage field shows a lot of promise. The authors [14] mixed titanium dioxide ( $\text{TiO}_2$ ) nanoparticles within the polyvinyl chloride (PVC)-based material. Then, the AC breakdown strength, permittivity, and dielectric loss factor of such specimen were examined. The test results show that AC breakdown strength increased and the permittivity, including dielectric loss factor, decreased compared with the unmodified PVC. The surface morphology of the test specimen was captured using the field emission scanning electron microscopy (FE-SEM) technique. The surface energy of  $\text{TiO}_2$  restricts the mobility of the polymeric chain and nanoparticles agglomeration.

On the other hand, it withholds on free space charge to decrease dielectric loss factor and capacitance. Shen et al. [15] have researched zinc oxide (ZnO) nanofluid insulation; the solvothermal approach was used to make ZnO nanofluids insulation. The electrical conductivity of ZnO nanofluids was increased 973 times when the oil insulation was mixed with a 0.75% volume fraction of nanoparticles [15]. The authors observed the relationship of electrical conductivity with nanoparticles volume fraction and temperature, which was linear and nonlinear, respectively. Mansour et al. [16] showed the cutting edge of nanocomposite application in high voltage technology. Breakdown strength, permittivity, conductivity, dielectric loss, space charge accumulation, tracking, erosion, and partial discharge are all fundamentally improved dielectric properties.

This paper proposed an alternative improvement of the electrical properties of mineral oil and natural ester by mixing with nanoparticles, i.e.,  $\text{TiO}_2$  and ZnO. The dielectric loss factor ( $\tan\delta$ ) at power frequency, including the polarization and depolarization current (PDC) measurement, was performed with the mineral oil-based nanofluid and natural ester-based nanofluid compared with those of the unmodified one. Furthermore, Fourier-transform infrared spectroscopy (FT-IR) experiment was conducted with the nanofluids samples.

## 2. Polarization and Depolarization Current

**2.1. Polarization Current.** The polarization process is the response of the insulation under the applied DC voltage by which such voltage does not make the insulation breakdown. Molecules of the insulation are rearranged following the direction of the DC electric field. Polarization current ( $i_{pol}(t)$ ) theoretically consists of capacitive current ( $i_c(t)$ ), conduction current ( $i_{con}(t)$ ), and absorption current ( $i_{ab}(t)$ ). The capacitive current occurs and immediately decreases. The conduction current depends on material conductivity. The absorption current occurs due to polarization phenomena. The components of the polarization current and PDC test circuit diagram are shown in Figure 1.

$$i_{pol}(t) = i_c(t) + i_{ab}(t) + i_{con}(t). \quad (1)$$

Polarization current theoretically can be written as

$$i_{pol}(t) = C_0 V_0 [(\sigma_0/\epsilon_0) + \epsilon_{\infty} \delta(t) + f(t)], \quad (2)$$

where  $C_0$  is the vacuum capacitance,  $V_0$  is a DC voltage applied to the test object,  $\sigma_0$  is the pure DC conductivity,  $\epsilon_0$  is the vacuum permittivity,  $\epsilon_{\infty}$  is the high frequency component of the permittivity,  $\delta(t)$  is the delta function, and  $f(t)$  is the dielectric response function [17–26].

**2.2. Depolarization Current.** When stopped applying the DC voltage and then short-circuiting the test object, the energized molecules from the polarization process are rearranged to a normal state. The discharging current was measured, which is called the depolarization current ( $i_{dep}(t)$ ) as expressed in equations (3) and (4). Depolarization current consists of the capacitive discharge current ( $i_{c(\text{discharge})}(t)$ ) and the absorption discharge current ( $i_{ab(\text{discharge})}(t)$ ). The capacitive discharge and the absorption discharge current from this state have reverse polarity from the charging current. The depolarization current does not consist of the conduction current because the applied voltage is terminated.

$$i_{dep}(t) = i_{c(\text{discharge})}(t) + i_{ab(\text{discharge})}(t), \quad (3)$$

$$i_{dep}(t) = -C_0 V_0 [f(t) - f(t + T_c)], \quad (4)$$

where  $T_c$  is the duration of applying voltage.

When considering (1) and (3), the conduction current is obtained from the subtraction of the depolarization current from the polarization current [17–23, 26–29].

$$i_{con}(t) = i_{pol}(t) - i_{dep}(t) = (i_c(t) + i_{ab}(t) + i_{con}(t)) - (i_{c(\text{discharge})}(t) + i_{ab(\text{discharge})}(t)). \quad (5)$$

## 3. Experimental Procedure

**3.1. Nanofluids Preparation.** For experiment preparation, mineral oil was heated at 80 degrees Celsius in a vacuum oven for 12 hours. Decreasing dissolved water content in liquid insulation is the purpose of this process.

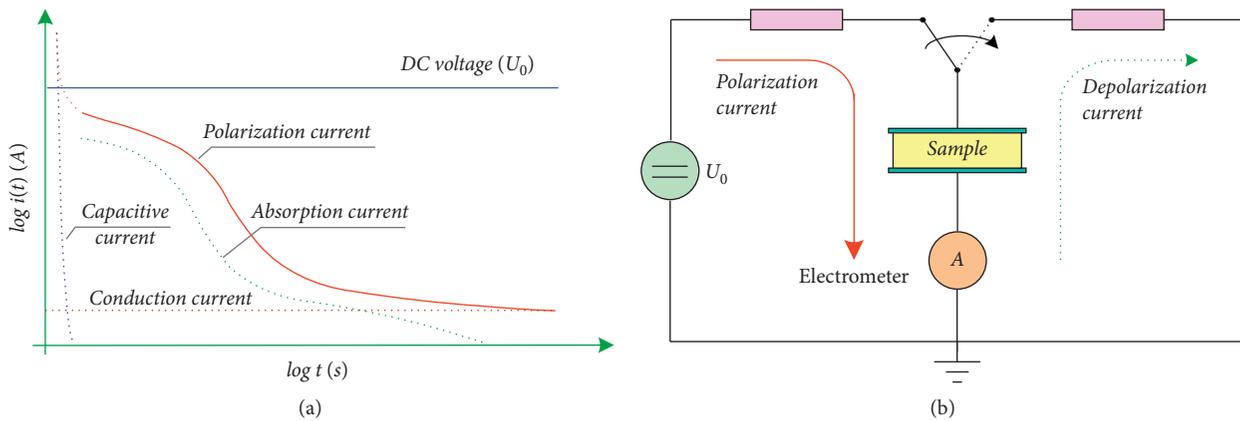


FIGURE 1: (a) Component of polarization current and (b) PDC test circuit diagram [17–20].

Nanoparticles were also heated for the same reason. Then, they were naturally cooled down at room temperature; two types of nanoparticles, i.e.,  $\text{TiO}_2$  and  $\text{ZnO}$ , were weighed and mixed with mineral oil at 0.01% and 0.03% of volume fraction. The mass of nanoparticles was calculated. After filling nanoparticles into the mineral oil, a magnetic stirrer was used for 30 minutes; this process distributed nanoparticles thoroughly in the mineral oil. An ultrasonic device was also utilized for an hour to guarantee the homogeneity between the mineral oil and nanoparticles. Finally, the mineral oil-based nanofluids were ready to test. The nanofluids preparation devices are shown in Figure 2. Additionally, the natural ester-based nanofluids were also prepared as the same as preparing the mineral oil-based nanofluid by which the natural ester was used instead of mineral oil. The nanofluids preparation process is illustrated in Figure 3. From the moisture content measurement of the sample preparation, the prepared unmodified mineral oil and the mineral oil-based nanofluid had water content less than 20 ppm, whereas the prepared unmodified natural ester and the natural ester-based nanofluid had water content less than 200 ppm. The case studies in this experiment are summarized in Table 1 [5–7, 30–36].

**3.2. Power Frequency  $\text{Tan}\delta$  Measurement.** The test sample was filled in a liquid electrode of about 50 ml. Before filling, the liquid electrode was cleansed and then kept dry. The C and  $\text{tan}\delta$  meter used in this experiment and the test circuit setup according to IEC 60247 are shown in Figure 4 [37].

**3.3. PDC Measurement.** To perform PDC measurement of the liquid samples, the PDC test circuit setup is shown in Figure 5. PDC measurement procedure was divided into three steps as follows:

- (i) In the first step, the remaining current of the test sample was measured.
- (ii) In the second step, a 100 DC volt was applied to the test object for 4 seconds for polarization current measurement. The current should not exceed  $\pm 1$  mA because of the limitation of the measuring

instrument. Then, short-circuiting the test sample, the discharging current was measured, so-called depolarization current. This process was performed as long as the depolarization current approached the remaining current [38]. The polarization and depolarization current obtained from this step would be used as a guideline for PDC measurement in the third step.

- (iii) In the third step, a 200 DC volt was applied to the test sample for 1500 seconds for the polarization current measurement. Then, the depolarization current was measured by short-circuiting the test sample for 1500 seconds.

**3.4. FT-IR Measurement.** To perform FT-IR measurement of the liquid samples, the FT-IR spectroscope was set up in the absorbance mode with wave numbers of  $650\text{--}4000\text{ cm}^{-1}$ . The sensor area was cleaned and dried; then, the liquid sample was dropped into FT-IR testing sensor for the measurement. The FT-IR spectroscope measurement system is depicted in Figure 6. Intermolecular connections vibrated according to their physically normal frequencies and modes of vibrations when exposed to infrared radiation. Following that, the vibration modes and dependent frequencies were used to determine the chemical functional groups of the investigated material.

## 4. Experimental Results and Discussion

**4.1. Power Frequency  $\text{Tan}\delta$  Test Results.** The power frequency  $\text{tan}\delta$  test results of the unmodified mineral oil, the unmodified natural ester, the mineral oil-based nanofluid, and the natural ester-based nanofluid are shown in Figure 7.

From Figure 7(a), there is no effect on the relative permittivity for the unmodified mineral oil and mineral nanofluid. For the natural ester-based nanofluid, the permittivity increases a bit compared with the unmodified natural ester. From Figure 7(b), the unmodified liquids mixed with  $\text{ZnO}$  nanoparticles have higher  $\text{tan}\delta$  for most cases compared with the unmodified liquids mixed with  $\text{TiO}_2$ .

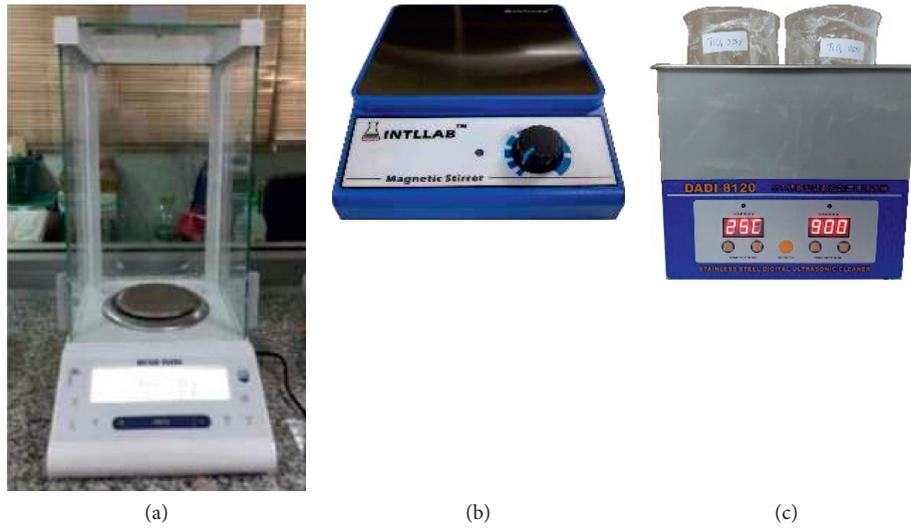


FIGURE 2: Nanofluid preparation devices: (a) balance, (b) magnetic stirrer, and (c) ultrasonic device.

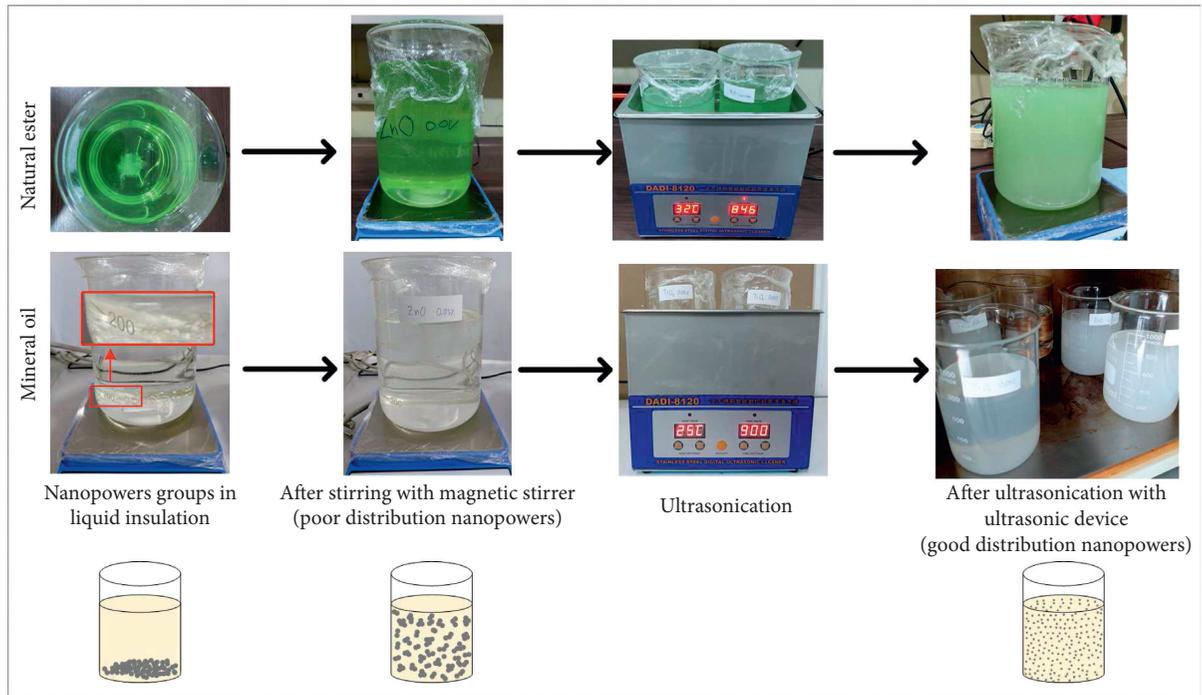


FIGURE 3: Nanofluids preparation process.

TABLE 1: Test specimens in this experiment.

Case	Insulating liquids	Volume fraction of nanoparticle mixed	Nanofluid samples
1	Mineral oil	—	Unmodified mineral oil
2	Mineral oil	TiO <sub>2</sub> 0.01%	Mineral oil + TiO <sub>2</sub> 0.01%
3	Mineral oil	TiO <sub>2</sub> 0.03%	Mineral oil + TiO <sub>2</sub> 0.03%
4	Mineral oil	ZnO 0.01%	Mineral oil + ZnO 0.01%
5	Mineral oil	ZnO 0.03%	Mineral oil + ZnO 0.03%
6	Natural ester	—	Unmodified natural ester
7	Natural ester	TiO <sub>2</sub> 0.01%	Natural ester + TiO <sub>2</sub> 0.01%
8	Natural ester	TiO <sub>2</sub> 0.03%	Natural ester + TiO <sub>2</sub> 0.03%
9	Natural ester	ZnO 0.01%	Natural ester + ZnO 0.01%
10	Natural ester	ZnO 0.03%	Natural ester + ZnO 0.03%

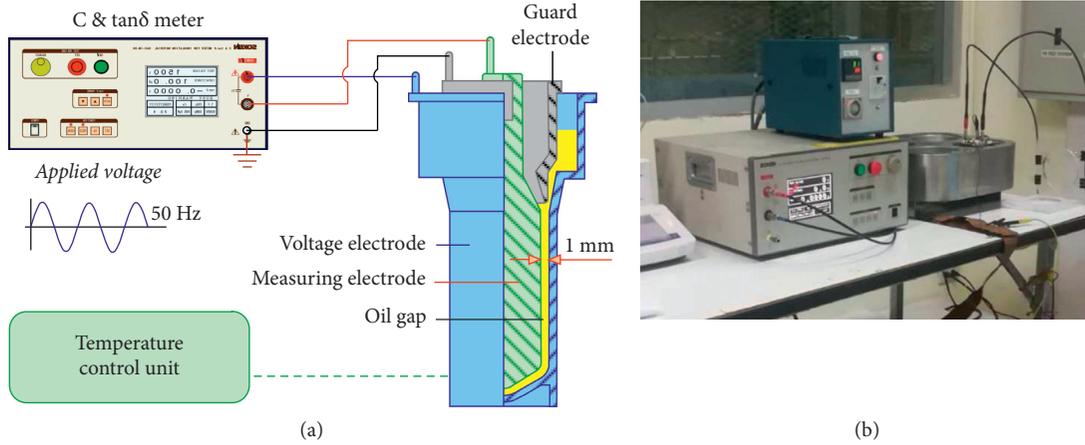


FIGURE 4: (a) Tanδ circuit diagram and (b) tanδ test circuit setup [33].

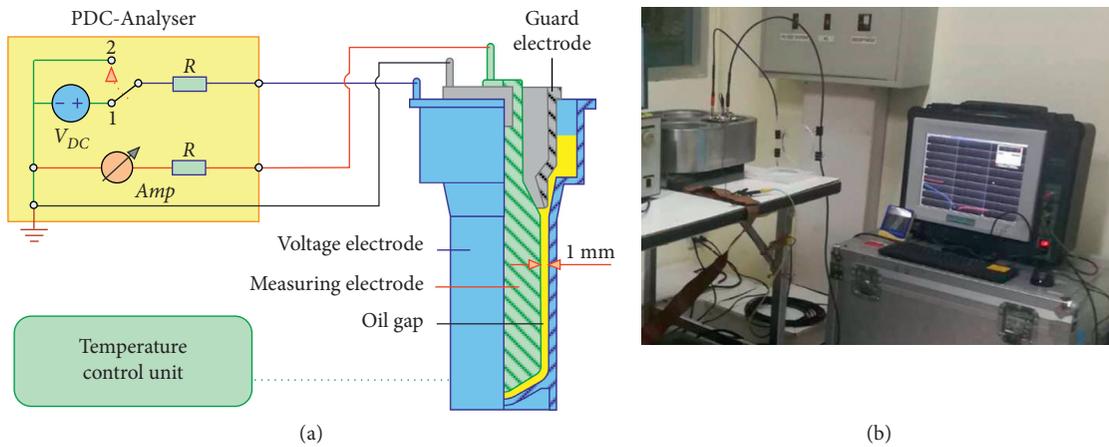


FIGURE 5: (a) PDC circuit diagram, (b) PDC test circuit setup [33].

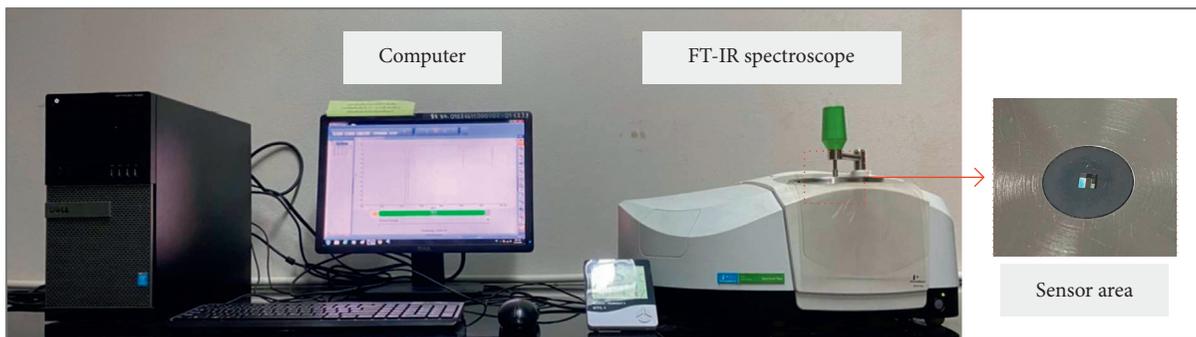


FIGURE 6: FT-IR spectroscopy measurement system.

4.2. PDC Test Results. The PDC test results of the mineral oil mixed with nanoparticles compared with these of unmodified mineral oil are shown in Figure 8. The PDC test results of the natural ester mixed with nanoparticles compared with these of unmodified natural ester are presented in Figure 9.

From Figure 8(a), mineral oil mixed with  $\text{TiO}_2$  with 0.01% volume fraction, at the initial state, shows a higher polarization current than the unmodified mineral oil. Then, the polarization current of the mineral oil mixed with  $\text{TiO}_2$

with 0.01% volume fraction decreases and lowers than that of the unmodified mineral oil after 100 seconds of measurement. In the case of mixing with  $\text{TiO}_2$  with 0.03% volume fraction, the polarization current of the mineral oil nanofluid is clearly lower than that of the unmodified mineral oil. The depolarization currents of such liquids are invariable. From Figure 8(b), the polarization current of the mineral oil mixed with the  $\text{ZnO}$  is clearly lower than that of the unmodified mineral oil. However, the mixed nanoparticles cause a bit

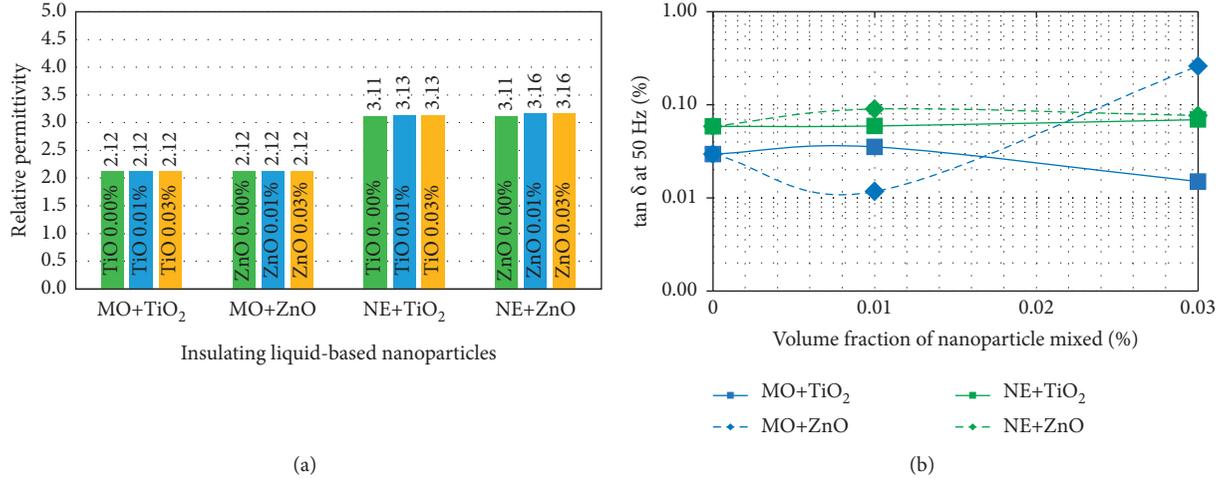


FIGURE 7: Comparison of relative permittivity and loss factors for mineral oil and natural ester-based nanoparticles. (a) Relative permittivity and (b) loss factors at 50 Hz.

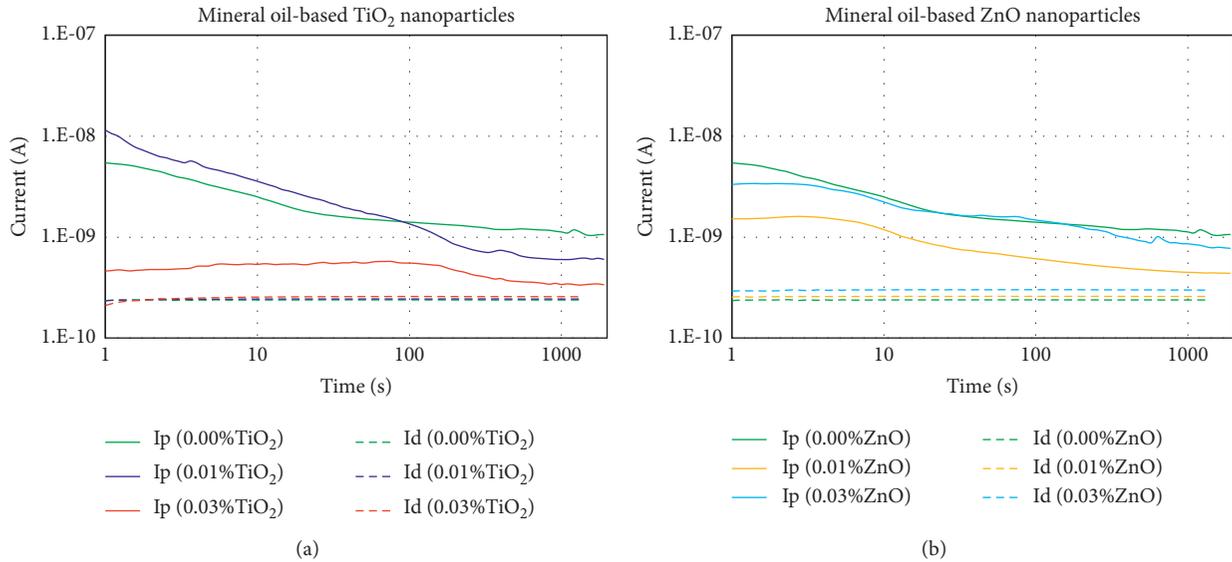


FIGURE 8: The PDC of the mineral oil mixed with nanoparticles. (a) Mineral oil mixed with TiO<sub>2</sub> and (b) mineral oil mixed with ZnO, where  $I_p$  is the polarization current and  $I_d$  is the depolarization current.

higher depolarization current than that of the unmodified mineral oil.

From Figure 9, nanoparticles used in the experiment seem not to have an effect on the polarization current, but it is evident that the employed nanoparticles cause a higher depolarization current than that of the unmodified natural ester. It is obviously seen that the nanoparticles used have a different effect on the nonpolar (mineral oil) and polar liquid (natural ester). The volume fraction of the TiO<sub>2</sub> and ZnO does not have the same impact on the unmodified liquids as well.

4.3. Analysis of Conductivity in Insulating Liquid-Based Nanoparticles. Further analysis for PDC test results can be performed with the analysis of apparent conductivity and

$\tan \delta$  of the insulating liquid [2]. Generally, the measured polarization and depolarization current is directly proportional to the time-dependent conductivity by which the polarization current can be neglected. The conductivity of the examined insulating liquid ( $\sigma_{oil}$ ) can be approximately calculated with the equation as follows [23,24,26,29]:

$$\sigma_{oil}(t) \approx \frac{\epsilon_0}{V_0 \cdot C_0} [i_{pol}(t) - i_{dep}(t)]. \quad (6)$$

The conduction currents of the unmodified liquids compared with that of the nano liquids are demonstrated in Figure 10.

From Figure 10, most types and volumes of applied nanoparticles lead to reduce the conductivity compared to the conductivity of unmodified mineral oil, whereas the nanoparticles lead a bit increase in the conductivity at the

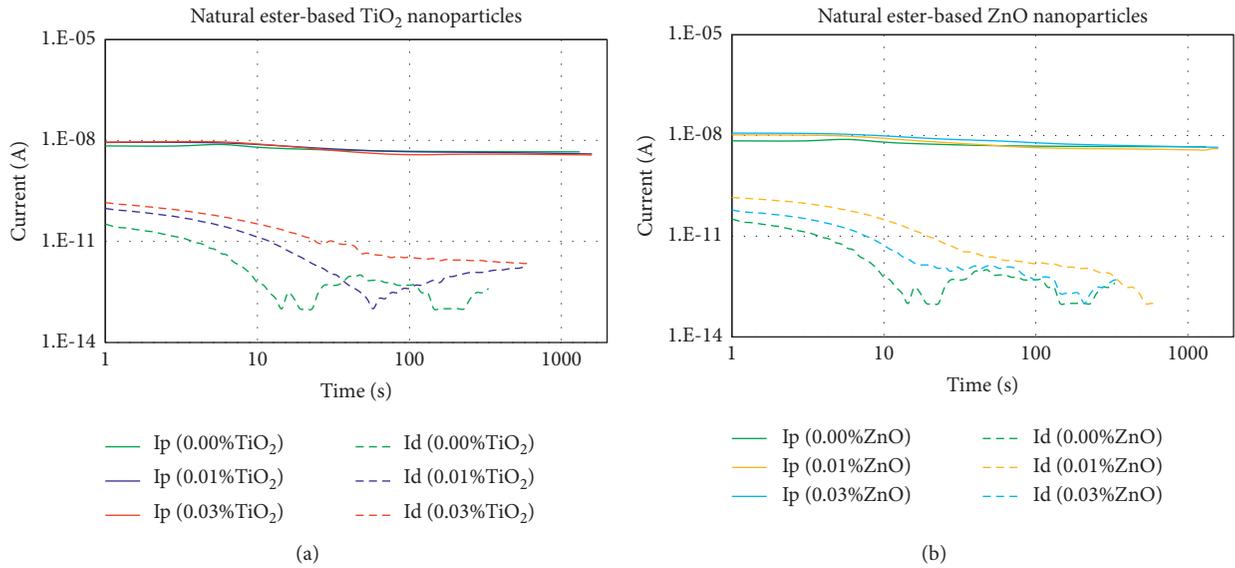


FIGURE 9: The PDC of the natural ester mixed with nanoparticles. (a) Natural ester mixed with TiO<sub>2</sub> and (b) natural ester mixed with ZnO.

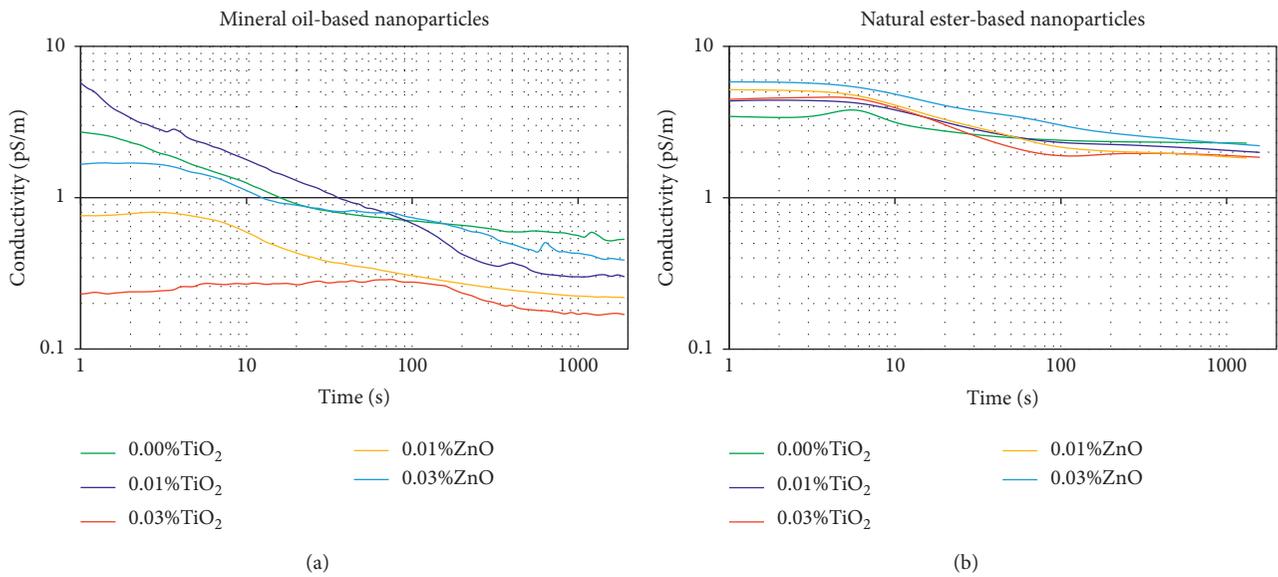


FIGURE 10: Apparent conductivities of the insulating liquid-based nanoparticles. (a) Mineral oil mixed with nanoparticles and (b) natural ester mixed with nanoparticles.

beginning stage of natural ester nano liquids compared with the conductivity of unmodified natural ester. An increase in the conductivity seems to have a negative effect on the insulation integrity; however, the moving electrons or charges may be trapped created by the nanoparticles in the insulating liquid. From the previous experiment by authors, the breakdown voltage of the mineral oil-based nanofluids was improved reported in [39, 40]. The higher breakdown voltage of mineral oil-based nanofluids should be governed by the trapping mechanism created by the mixed nanomaterial related to the lower conductivity of the mineral oil nanofluids. In the case of natural ester nanofluid, the breakdown voltage of the natural ester nanofluid was also

improved and reported in [41, 42], even though the nanomaterials increased the conductivity of the natural ester nanofluids.

**4.4. Analysis of Conductivity and Polarization Loss Factors in Insulating Liquid-Based Nanofluids.** The polarization and depolarization current was measured, and then the curve fitting was performed to determine the individual element  $R_i$  and  $C_i$  of a linear dielectric model [1, 5]. The frequency-dependent  $\tan\delta$  can be calculated from the individual element  $R_i$ ,  $C_i$  of the linear dielectric model by applying the following equations:

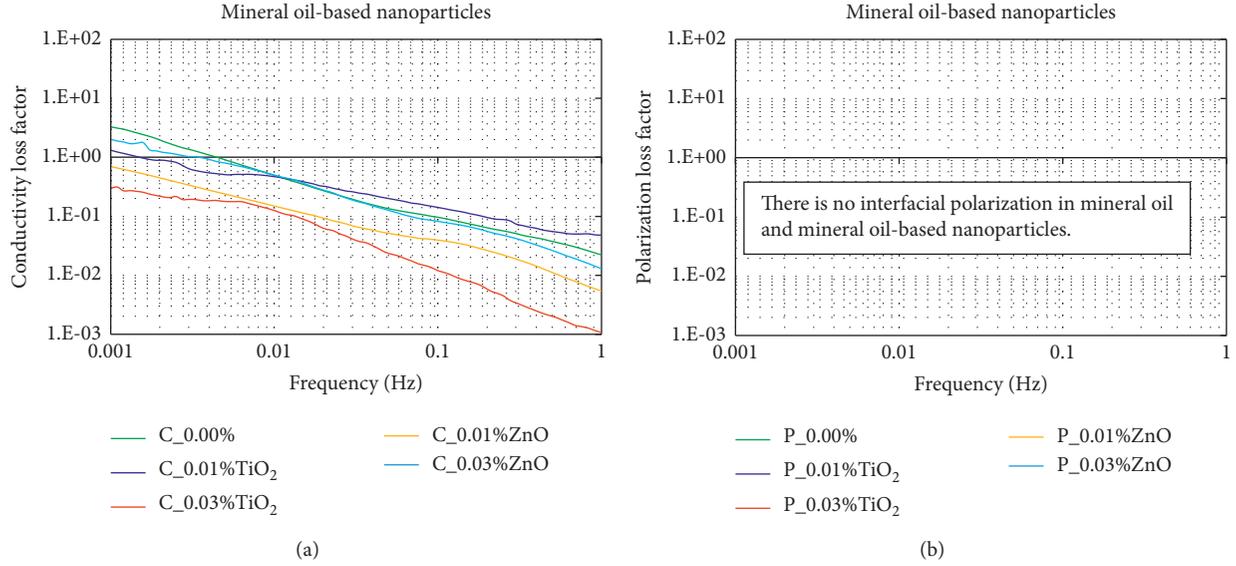


FIGURE 11: Frequency-dependent conductivity and polarization loss factors of the mineral oil-based nanoparticles; (a) conductivity loss factor and (b) polarization loss factor.

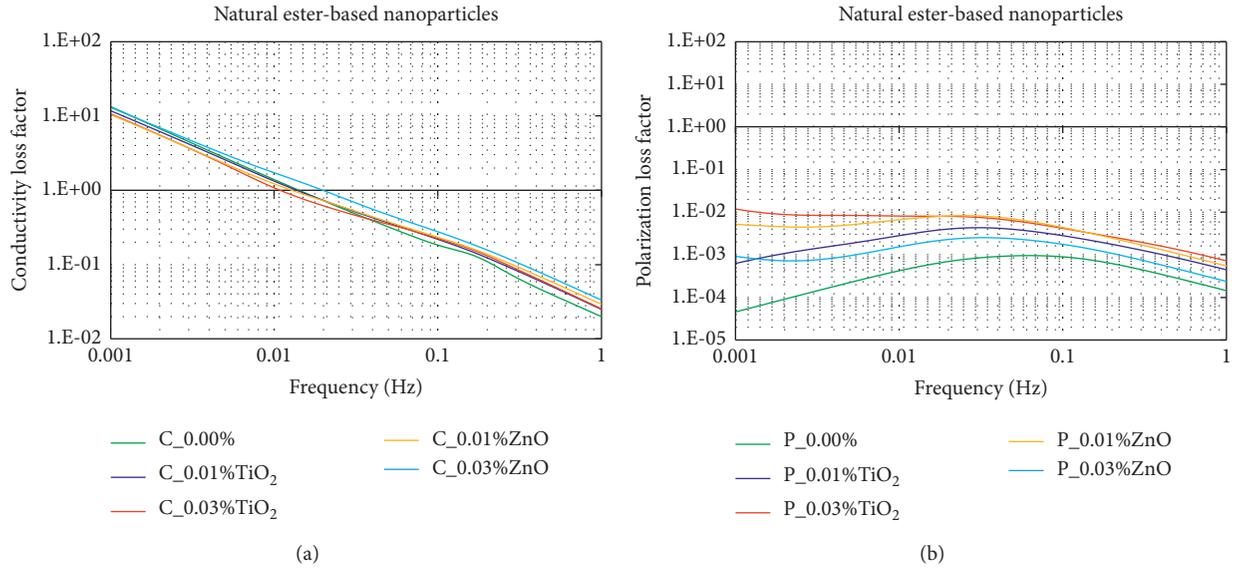


FIGURE 12: Frequency-dependent conductivity and polarization loss factors of the natural ester-based nanoparticles; (a) conductivity loss factor and (b) polarization loss factor.

$$\tan\delta_{\sigma}(\omega) = \frac{1}{\omega R_{oil}(\omega) \left\{ C_{50Hz} + \sum_{i=1} C_i / (1 + (\omega R_i C_i)^2) \right\}}, \quad (7)$$

$$\tan\delta_{pol}(\omega) = \frac{\sum_{i=1} \omega R_i C_i^2 / (1 + (\omega R_i C_i)^2)}{C_{50Hz} + \sum_{i=1} C_i / (1 + (\omega R_i C_i)^2)}. \quad (8)$$

where  $\tan\delta_{\sigma}(\omega)$  is the frequency-dependent conductivity loss factor,  $\tan\delta_{pol}(\omega)$  is the frequency-dependent polarization loss factor,  $\omega$  is the angular frequency,  $R_{oil}$  is the oil resistance,  $C_{50Hz}$  is the capacitance at 50 Hz,  $C_i$  is the individual capacitance, and  $R_i$  is the individual resistance [19, 26, 29].

The frequency-dependent conductivity and polarization loss factors of the unmodified liquids compared with

these of the nano liquids are demonstrated in Figures 11 and 12.

From Figure 11, mineral oil-based nanofluid shows a lower conductivity loss factor compared with that of the unmodified mineral oil, whereas there is no polarization loss factor in the unmodified mineral oil and mineral oil nanofluid [2]. From Figure 12, the nanoparticles cause a higher polarization loss factor compared with that of the unmodified natural ester.

**4.5. FT-IR Test Result.** The FT-IR experiment was conducted to preliminarily investigate the nanofluid sample's physicochemical characteristics. FT-IR spectra were measured,

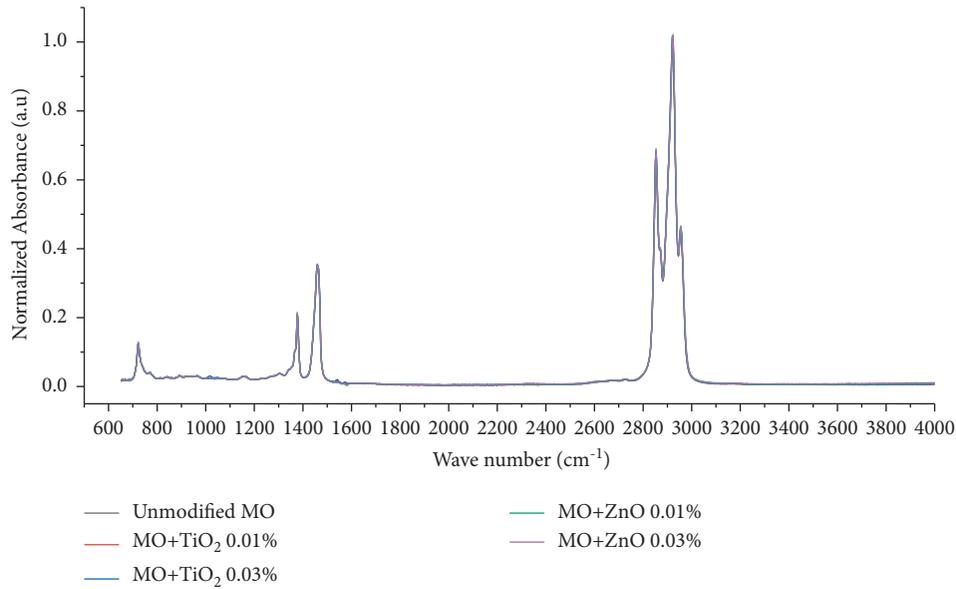


FIGURE 13: FT-IR test results of the mineral oil-based nanoparticles compared with that of the unmodified insulating liquid.

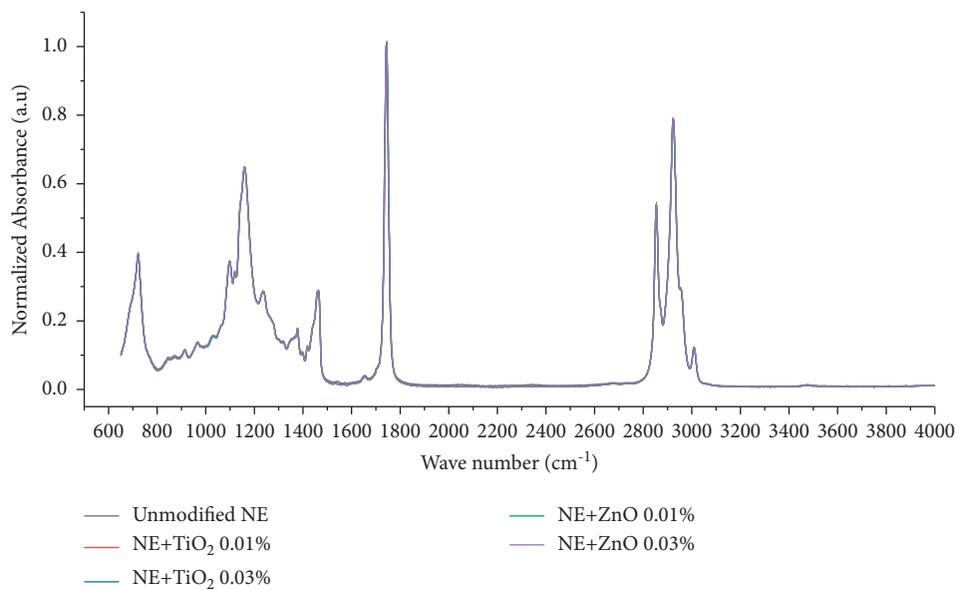


FIGURE 14: FT-IR test results of the natural ester-based nanoparticles compared with that of the unmodified insulating liquid.

and then the data was normalized to be compared. Test results are presented in Figures 13–15.

From Figure 13, the FT-IR spectra of the unmodified mineral oil and the mineral oil nanofluid are very similar. The C–H rocking reaches a high of 1380 and 721  $\text{cm}^{-1}$  (alkanes). The C–H stretching (in-ring or aromatics) can be seen at wave numbers approximately 1460  $\text{cm}^{-1}$  which means a subcomponent of mineral oil. Peaks near 2800–3000  $\text{cm}^{-1}$  wave numbers for C–H stretching indicate the presence of the main component of mineral oil (alkanes).

From Figure 14, the FT-IR spectra of the unmodified natural ester and the natural ester nanofluid are very similar. The C–H rocking reaches a high of 721  $\text{cm}^{-1}$  (alkanes). The  $\text{CH}_2$  bending can be seen in oil samples at wave numbers

approximately 1460  $\text{cm}^{-1}$ . C–O stretching peaks at 1000–1260  $\text{cm}^{-1}$  indicate the presence of alcohol (carboxylic acid, esters, and ethers). Peaks near 1740  $\text{cm}^{-1}$  for C=O stretching suggest the presence of esters. Peaks near 2800–3030  $\text{cm}^{-1}$  wave numbers for C–H stretching are also found.

From Figure 15, it is clearly different between the FT-IR spectra of mineral oil nanofluid and natural ester nanofluid. Peak at 2950  $\text{cm}^{-1}$  is only in unmodified mineral oil and mineral oil nanofluid, whereas peaks at 800–1300  $\text{cm}^{-1}$ , 1740  $\text{cm}^{-1}$ , and 3010  $\text{cm}^{-1}$  are only in unmodified natural ester and natural ester nanofluid. FT-IR results showed no difference between the unmodified mineral oil and mineral oil nanofluids. Besides, there is no difference in FT-IR results between the unmodified ester and ester nanofluids.

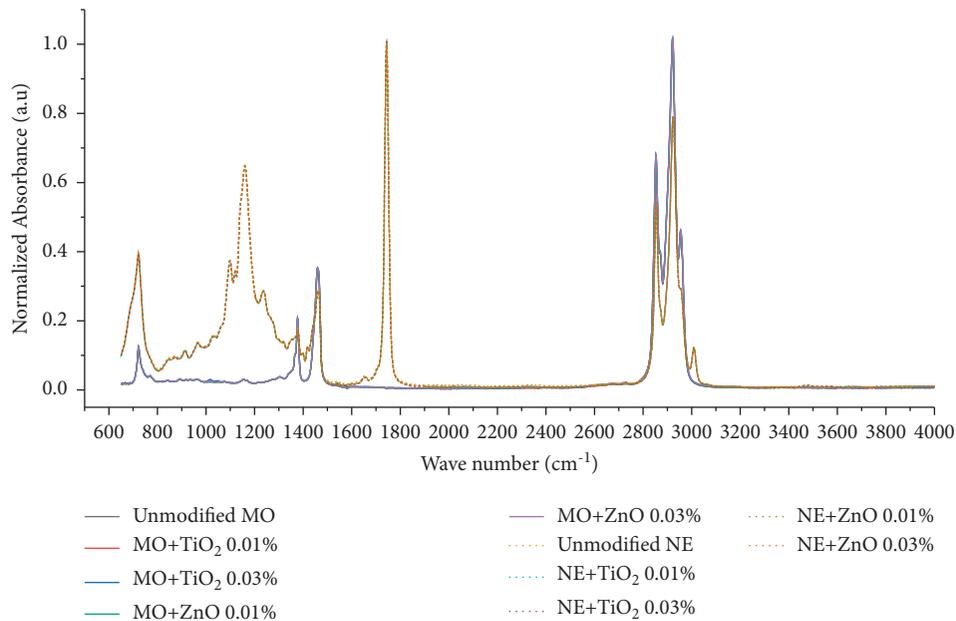


FIGURE 15: FT-IR test results of all test samples.

## 5. Conclusion

The characteristics of mineral oil and natural ester mixed with two types of nanoparticles, i.e.,  $\text{TiO}_2$  and  $\text{ZnO}$ , with the 0.01% and 0.03% of volume fraction were investigated. At power frequency, the applied nanomaterial hardly affected the relative permittivity. Nanomaterial used clearly affected the  $\tan\delta$  value in the case of mineral oil mixed with  $\text{ZnO}$  with 0.03% of volume fraction. The breakdown voltage of the mineral oil nanofluids and natural ester nanofluid was improved according to the former publications of the authors. For PDC measurement, the studied nanomaterials clearly affected the polarization current of the mineral oil nanofluids, whereas such nanomaterial impacted evidently the depolarization current of the natural ester nanofluids. For PDC measurement, mineral oil mixed with  $\text{TiO}_2$  with 0.01% of volume fraction increased the polarization current around 111% at the initial state. On the other hand, other case studies for mineral oil-based nanofluids showed the reduction of the polarization current of 38.8% to 91.5% compared with that of unmodified mineral oil, whereas natural ester mixed with  $\text{ZnO}$  had clearly an effect on increasing the depolarization current about 82.7% to 337.1% at the initial state, compared with that of the unmodified natural ester.

The applied nanoparticles lead to reduce the conductivity compared to the conductivity of unmodified mineral oil, whereas the nanoparticles lead a bit increase in the conductivity at the beginning stage of natural ester nano liquids compared with the conductivity of unmodified natural ester. An increase in the conductivity seems to have a negative effect on the insulation integrity; however, the moving electrons or charges may be trapped created by the nanoparticles in the insulating liquid.

FT-IR results showed no difference between the unmodified mineral oil and mineral oil nanofluids. Besides, there is no difference in FT-IR results between the

unmodified ester and ester nanofluids. The challenge of applying nanofluid in the insulation liquid is the accumulation of nanomaterial at a trap inside high voltage equipment by which the trap may be cellulose, papers, or pressboards in the transformers. The trap may happen at the edge or barrier of the component installed inside the high voltage apparatus.

## Data Availability

The data used to support the findings of this study are included in the article.

## Conflicts of Interest

The authors declare that they have no conflicts of interest regarding the publication of this paper.

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