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Thermal Aging Characteristics of Newly Synthesized Triester Insulation Oil

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ABSTRACT High-stability triester insulation oil derived from the trimethylolpropane esters (TME) was synthesized by esterifying saturated fatty acids and trimethylolpropane (TMP). The TME oil shows higher AC breakdown voltage (V_b, 72.6 kV/2.5 mm) than that of FR3 (65 kV/2.5 mm). TME oil is readily biodegradable whose biodegradation is 82.2% after 28 days. Pressurized differential scanning calorimetry (PDSC) was applied to measure oxidation onset temperature (OOT) of TME oil for initial evaluating the oxidation stability. Due to the elimination of the carbon-carbon double bonds and β-H, the OOT of the TME oil without antioxidants is approximately 25°C higher than that of FR3. To evaluate the realistic performance of the TME oil, the effect of transformer materials on the aging behavior was investigated. The effect of transformer materials on the thermal aging of TME oil and mineral oil (MO) is less than that of FR3. Although the moisture content and acid value in TME oil induced by aging are higher than those in MO. These acids and moisture do not cause obvious degradation of breakdown voltage and dielectric loss factor, which may be attributed to the high moisture solubility in it.

INDEX TERMS Triester, physicochemical and electrical properties, insulation oil, thermal aging, breakdown voltage.

I. INTRODUCTION

Mineral oils (MOs) as the major dielectric fluid in the power transformer for over a century [1]. However, MOs can cause serious problems due to their low flash point which have caused several fire accidents. The poor degradation ability of MO causes disposal problems against environmental laws and regulations in many countries [2]. Therefore, natural ester insulation oils (NEOs) as promising substitutes for MO in transformers have been received increasing attention due to the advantages of renewability and biodegradability and less flammability compared with MOs [2]. The sealed tube thermal aging tests of insulation papers in NEOs have been performed at 90° − 180°C [3]–[6]. The results show that, under these conditions, the thermal aging rate of insulation paper immersed in NEOs is lower than that immersed in MOs.

The oxidation stability of NEOs is generally lower than that for MOs because of the carbon-carbon double bonds and β-H in triglycerides (**Fig. 1**) [2], [7], [8]. Under the effect of heat, light and metal, triglycerides with carbon-carbon double bonds and β-H trend to lose hydrogen atoms and generate free radicals. The free radical then reacts with oxygen to form peroxy radicals, which creates a new free radical by attacking the new triglycerides, forming a chain reaction that can be repeated thousands of times until no hydrogen source or chain reaction is interrupted [9]. During the propagation of free radical, the aging products (moisture, acid, and gases) increase, which would significantly decrease

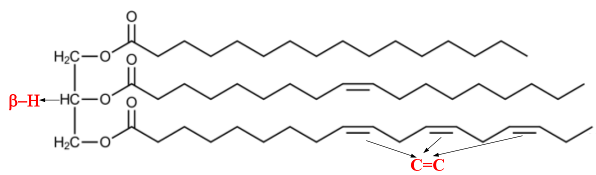

FIGURE 1. Molecular structure of the triglyceride.

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the dielectric breakdown strength, increase dielectric dissipation factor (tan δ) and decrease the safety level [10], [11]. From the above descriptions, the NEOs, such as FR3 and BIOTEMP [12] oils, do not satisfy the request for application in ultrahigh voltage and large power transformers due to the low oxidation stability.

To enhance the oxidation stability of NEOs, the antioxidant was used. Antioxidants include free radical scavengers, oxygen quenchers and inactivators of peroxides and metal ion chelators, which inhibit NEOs from oxidation. Butylated hydroxytoluene (BHT) exhibits improvement in the stability of both MOs and NEOs [2], [13]. The oxidation stability of NEOs with combined antioxidants is better than that with individual dosage at similar concentrations [14], [15]. The addition of antioxidants in NEOs can improve the oxidation stability, but it is still lower than that of MOs [16]. Therefore, it is necessary to synthesize a new ester insulation oil with high oxidation stability.

In this paper, to eliminate the unstable functional group (β−H and C=C), medium-chain saturated fatty acids (C_6-C_{10} acids) and TMP were used to synthesize an insulation oil. The properties and the thermal aging characteristics of TME oil are compared to FR3 and traditional mineral oil.

II. EXPERIMENTAL PROCEDURE

A. MATERIALS

C_6, C_8 and C_{10} acids (purity better than 98%) were purchased from China Agri-Industries Holdings Limited. Commercial MO (25#) and NEO (FR3) was purchased from Sinopec Group and Cargill, respectively. TMP, xylene, $SnCl_2$, were purchased from Aladdin Company Ltd.

B. PREPARATION OF TME OIL

The setup of the reaction is shown in **Fig. 2**. TMP (0.5 mol), The medium-chain saturated fatty acids (1.6 mol) and stannous chloride (catalyst, 0.8 wt.%) and xylene (300 ml) were stirred at 140°C for 5 hours. The reaction was performed in nitrogen gas. The esterification process was monitored by the acidity and the generated water collected by Dean-Stark cap [17], [18]. The **Fig. 3** shows the reaction between saturated fatty acids and TMP.

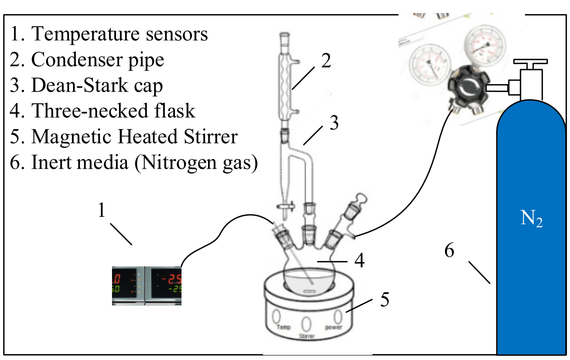

FIGURE 2. Setup of the reaction.

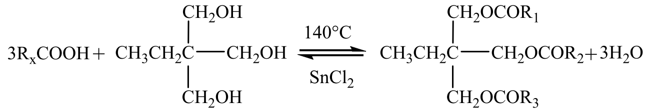

FIGURE 3. Reaction between the saturated fatty acids and TMP.

C. ANALYSIS AND MEASUREMENT

1) PHYSICAL AND CHEMICAL PROPERTIES

The density of insulation oils was measured by automatic density measuring instrument VIDA 40 (ISL, France). The measured viscosity at 20°C was the average of four measured data.

The pour point of oils was measured based on standard GB/T 261-2008 using CPP 5GS (ISL, France). The flash point of oils was measured according to Standard GB/T 261-2008 using PMA-5 (Anton Paar, USA). GB/T 3536-2008 was used to measure the fire point of oils.

The viscosity of oils was measured at 40°C according to ASTM D455. The test instrument is DV-II+PRO viscometer (Brookfield, USA).

The transient hot-wire method is a mature technology In testing the thermal conductivity of liquid [19]. Therefore, this method was used to measure the thermal conductivity of oils at 20°C. The test instrument is TC3010 (XIATECH, China). The specific heat of oils was measured according to ASTM E1269-2011. DSC (Q200, TA) was used to measure the conductivity need to calibrate.

2) DIELECTRIC PROPERTIES

The tan δ, relative permittivity and electrical conductivity of oils at 90°C was measured by DTL C (Baur, Austria) according to the IEC 60247 standard.

The AC breakdown voltage (V_b) is an essential parameter of the insulation oil, which was measured based on the GB/T 507 standard with a rate of 2 kV/s. The test cell contains two identical spherically capped brass electrodes with 2.5 mm of diameter which arranged horizontally. The measured AC V_b was the average of 12 measure data.

3) OOTS OF INSULATION OILS

PDSC test was performed according to ASTM E2009 using NETZSCH DSC 204 HP (Bavarian, Germany) shown in **Fig.4**. The OOTs of insulation oils were obtained according

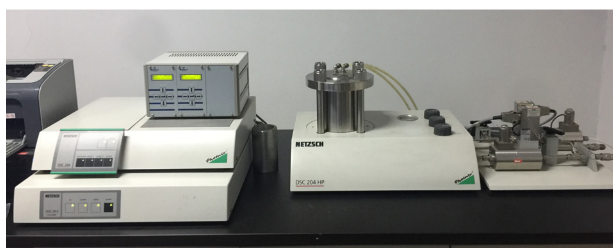

FIGURE 4. Equipment used to measure OOTs of the insulation oils.

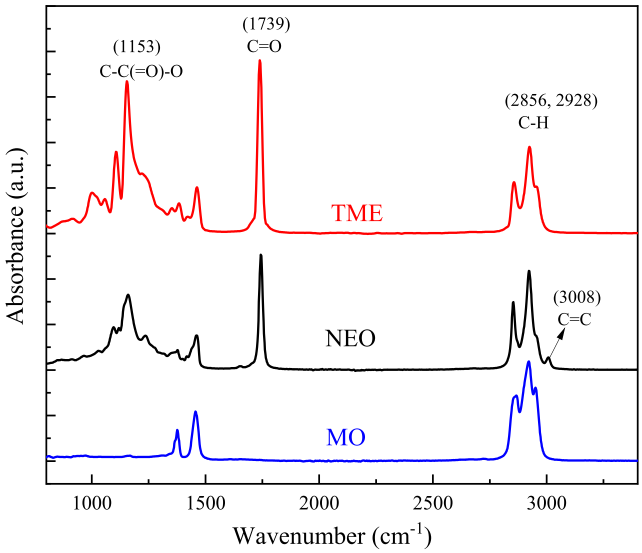

FIGURE 5. FTIR spectrum of TME oil.

to ASTM E2009 using PDSC shown in Figure 10. The Higher OOT means that the oils are more stable. The heating rate is 10 °C/min. The oxygen flow was 100 ml/min and the pressure of oxygen is 3.5 MPa.

III. RESULTS AND DISCUSSION

A. ANALYSIS OF PRODUCTS

Fig. 5 shows the FTIR spectra of TME oil. Within 1800-1700 cm^{-1}, the peak at 1739 cm^{-1} is caused by the stretching of C=O, which is a typical signature from esters [20]. The carbon-oxygen (C(=O)-O-C) stretching peak shows at 1151.9 cm^{-1}. The peaks at 2856.0-2923.8 cm^{-1} and 1463.9-1151.9 have attributed to the stretching and bending of C-H. These results reveal the successful synthesis of TME oil. To eliminate C=C, saturated chain fatty acids are used as raw materials to replace unsaturated free fatty acids in natural ester insulation oils. Therefore, the peak at 3008 cm^{-1} which is attributed to the C=C is not detected in TME oil.

Table 1 shows the comparison of the properties of FR3 and TME oil. The standards used to measure the main properties are shown in the fourth column of **Table 1.**

The convection heat transfer is the most important method in cooling transformer. In particular, the low viscosity is very critical for transferring the insulation oil into narrow passages between windings to restrict the local overheating. Therefore, the viscosity of the insulation oil is a crucial factor in the heat transfer performance of a transformer. The relatively high viscosity of NEO limits the lifetime and load capacity of power transformers.

The low viscosity of the insulation oil will accelerate the initial impregnation of the insulation paper. Therefore, the low viscosity of the insulation oil is important for the transformer. The TME oil has lower kinematic viscosity (23.3 cSt) compared with NEO FR3 (34 cSt). The pour point of FR3 oil is 21°C below zero, which is notably higher than that of TME oil (−45°C). The TME oil exhibits excellent breakdown strength (72.6 kV/2.5mm) compared with FR3 (65 kV/2.5mm). Flash point of TME is 248°C which is higher than that MOs (140-170°C).

TABLE 1. The main properties of TME and FR3 oil.

Property (Unit)	FR3	TME	Test methods
Appearance	clear	clear	GB/T 5525-2008
Viscosity (cSt)	34	23.3	40°C ASTM D455
Density (g/cm)	0.92	0.934	20°C ASTM D-4052
Thermal conductivity (W/m·K)	0.16	0.15	20°C Transient hot wire
Specific heat (J/kg·°C)	1939	2294	ASTM E1269-2011
Expansion coefficient	0.00075	0.00086	DL/T 1204-2013
Pour point (ºC)	-21	-45	GB/T 3535-2006
Flash point (ºC)	316	248	GB/T 261-2008
Fire point (ºC)	358	298	GB/T 3536-2008
Acid value (mgKOH/g)	0.04	0.02	GB/T 264-1983
Permittivity	2.8	3.2	90ºC IEC 60247
Resistivity (10^{10} Ω·cm)	4.5	1.1	90ºC IEC 60247
Tanδ (%)	0.58	2	90ºC IEC 60247
AC V_b / (kV)	65	72.6	2.5 mm GB/T 507

B. BIODEGRADABILITY OF PRODUCTS

The biodegradability test of TME oil was conducted using standard OECD 301B, which is the more common test used to evaluate oil biodegradability of oils [21]. TME oil was added into a specific inorganic nutrient solution inoculated with micro-organisms, which was prepared according to the guideline of OECD 301B.

Sodium acetate was used as the reference material to valid the biodegradability test. If the biodegradation of reference material more than 60% within 14 days, the test is valid based on OECD 301B.

According to the standard OECD 301B, there are three parts included in the setup of the biodegradation test (**Fig. 6**). (a) An empty flask to ensure the stability of the gas. (b) a flask use to conduct the degradation. (c) The $Ba(OH)_2$ solution was used to absorb CO_2 produced during the degradation process. The amount of CO_2 produced is determined by acid-base titration, and the biodegradation rate can is calculated using equation 1.

Five parallel experiments were performed to test the biodegradation: Two parallel experiments were carried out to test the TME oil in duplicate. Two parallel experiments served as the blank control group. One parallel experiment using to test the biodegradation of reference material (sodium acetate).

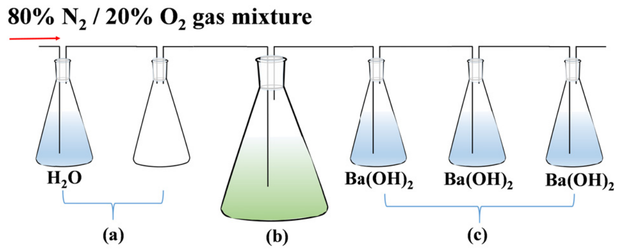

FIGURE 6. The setup of the biodegradation test.

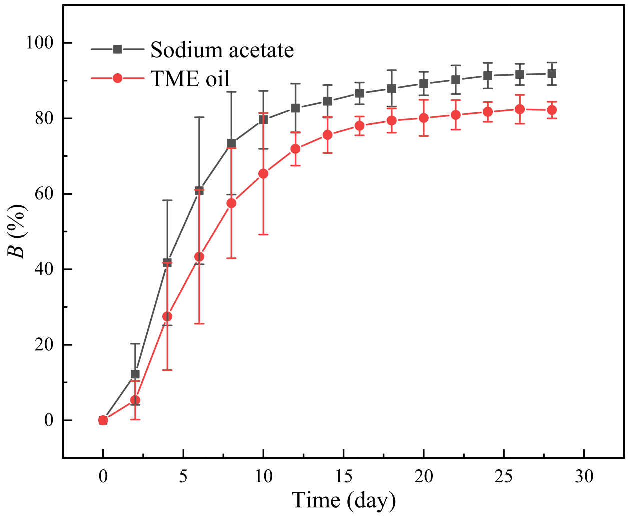

FIGURE 7. Biodegradation of sodium acetate and TME oil.

The biodegradation of the material (B_T) is calculated by equation 1:

$$B_T(\%) = \frac{(CO_2)_T - (CO_2)_b}{m_T \times Ca \times (44/12)} \times 100 \quad (1)$$

where $(CO_2)_T$ (mg) and $(CO_2)_b$ (mg) represent the mass of CO_2 emitted from the material and the blank, respectively. m_T (mg) is the mass of material used to test and Ca (%) is the mass percentage of carbon in this material. The CO_2 amount of five parallel experiments was measured every 2 days during the 28 days period.

As shown in **Fig. 7**, biodegradation of the reference material (sodium acetate) is about 84% at 14th day, which proves that the test is valid according to OECD 301B. The biodegradation of TME oil is 82.2% after 28 days. The biodegradation of NEOs is more than 85% [22]. FR3 oil is produced from soybean oil. The biodegradation of FR3 oil is about 98% in the bulletin [23], but the detail of the test method to evaluate the biodegradation did not clarify. Though the biodegradation of TME oil is lower than that of NEOs, the biodegradation of TME oil reaches 65.3% after 10 days in this test, which demonstrates that it is readily biodegradable (OECD 301B). TME oil is a synthetic ester, and the inherent biodegradability of ester molecules cause it is relatively easy to degrade [24].

C. OOT OF PRODUCTS

Rotary bomb oxidation test (RBOT), oven oxidation stability test (OOST), Rancimat and PDSC [25]–[27] are the common methods to evaluate the oxidation stability of oils. PDSC and rotary bomb oxidation test (RBOT) can well assessment the oxidation of NEOs [25]. PDSC test is faster more convenient compared with RBOT, OOST and Rancimat methods. Furthermore, the use of PDSC can reduce the evaporation from the oil [26]. Therefore, PDSC was used to measure OOT to evaluate the oxidation stability of FR3, MO and TME oils. The higher the OOT of the oil, the better the thermal oxidation stability. TME oil has good oxidation stability because there are no unstable functional groups (β-H and C=C) in it. Therefore, the thermal oxidation stability of TME is comparable with that of commercial MO, whose OOT is only 6°C higher than that of TME oil without antioxidants (205.3°C), as shown in **Fig. 8**. The OOT of TME oil is about 25°C higer than that of commercial FR3.

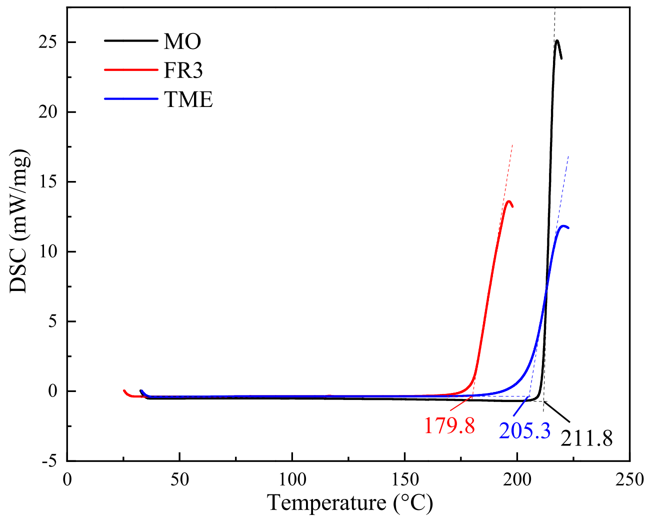

FIGURE 8. Comparison of OOTs of different insulation oils.

D. THERMAL AGING OF PRODUCTS

Thermal aging tests with and without transformer materials were performed to investigate the thermal aging characteristic of the TME oil. The effect of transformer materials on thermal aging was also investigated. The transformer materials (Cu, Fe and insulation paper) can accelerate thermal aging of insulation oils [11], [28], [29]. For use in power transformers, the aging characteristics (acid value, dielectric dissipation factor and V_b) of insulation oil is of vital importance [30]. To evaluate the realistic performance of newly synthesized TME oil, the influence of transformer materials on the aging behavior needs to carry out.

The oven was used to perform the thermal aging test. As shown in **Fig. 9**, each of the tanks was filled with 400 ml of insulation oil. Every two of 6 stainless steel tanks was filled with the same insulation oil, one of which contains transformer materials (Cu, Fe and insulation paper) and the

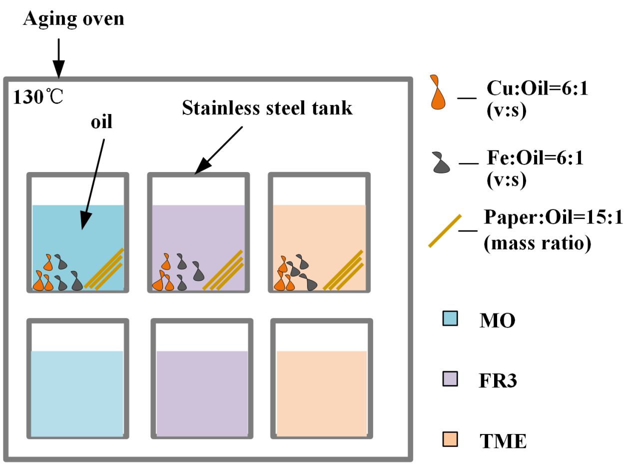

FIGURE 9. Test setup for the thermal aging of insulation oils (v:s = insulation oil volume:surface area).

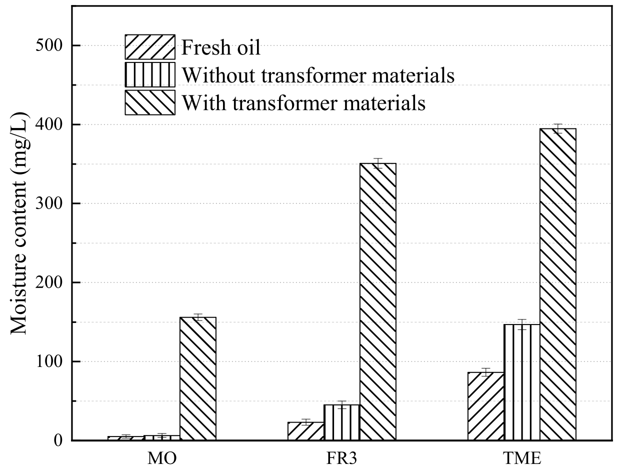

FIGURE 10. Comparison of moisture of insulation oils at various thermal aging conditions.

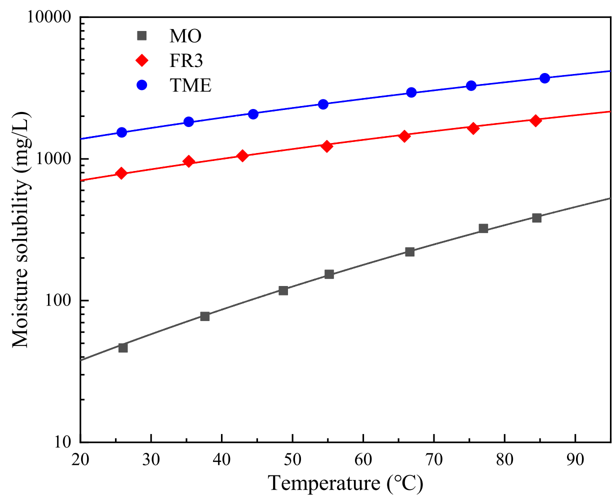

FIGURE 11. Moisture solubility in insulation oils.

other contains no transformer materials. **Fig. 9** shows the test setup to age the insulation oil in the oven. For this test setup, the effect of the transformer materials on oil aging was investigated. Following [31], the ratio of insulation oil volume to copper and iron surface area is 6:1 (v:s). The 0.05 cm thick copper strips are 16.21 cm long and 2 cm wide. The 0.02 cm thick iron strips are 16.30 cm long and 2 cm wide. The mass ratio of insulation oil and insulation paper is 15:1. The aging behavior of natural ester oils, synthetic and mineral oil was investigated in paper [11]. It is necessary to age at 130°C by not less than 164 hours to distinguish the aging behavior among the insulation oils. This work performed the aging tests at 130°C for 10 days (240 hours). Before aging, the TME, FR3 and MO oils were dried in vacuum for 48 hours (0.2-0.6 kPa, 98°C). The insulation paper strips were dried in vacuum for 10 hours (0.2-0.6 kPa, 80°C).

The excess moisture in the insulation oil can increase the $\tan\delta$, the hydrolysis and thermal aging rate of the oil and insulation paper. Measuring the moisture content in the insulation oils is needed. The oxidation products in insulation oil occur during the thermal aging, which increases the moisture content. High moisture content can degrade the dielectric strength and lead to transformer failure.

Fig. 10 shows the variation of moisture of insulation oils before and after thermal aging. Different insulation oils have similar variation trends. In the process of thermal aging, the moisture content is increasing. The transformer materials accelerate the thermal aging of the oil, which causes a strong increase in the moisture content of insulation oil. MO has a significantly smaller moisture content than the FR3 and TME oil, and a small increase in moisture content occurs after thermal aging without transformer materials. For comparison, the moisture content of TME oil is higher than those of FR3 and MO under the same aging conditions.

The moisture solubility of the insulation oils was measured according to the paper [32].

$$W_s = \frac{W_{abs}}{\%R.H} \times 100 \quad (2)$$

In equation (2), $\%R.H$ is the relative humidity of the oils, which measured by relative humidity and temperature transmitter MMT162 (Viasla, Finland). W_{abs} is the absolute moisture in the oil, which was measured using Karl-Fischer Moisture Titrator JF-5.

The moisture solubility of TME oil, FR3 and MO are shown in **Fig. 11**. It is visible TME oil has considerably higher moisture solubility from 20-100°C. Free moisture and dissolved moisture are the two formations exist in the insulation oil. Hydrogen bonds are easy to form between polar and water molecules [33]. Therefore, the weak polarity insulation oil (FR3 and TME) can dissolve more moisture than that of non-polar MO. The permittivity of TME is 3.2, which is higher than that of FR3 (2.8), which means the moisture solubility of TME is higher than that of NE insulation oil. Therefore, the moisture in the aged TME oil is higher compared with the moisture contents of FR3 and MO.

Fig. 12 shows the change in the acid value of insulation oils before and after thermal aging. The acid value has a

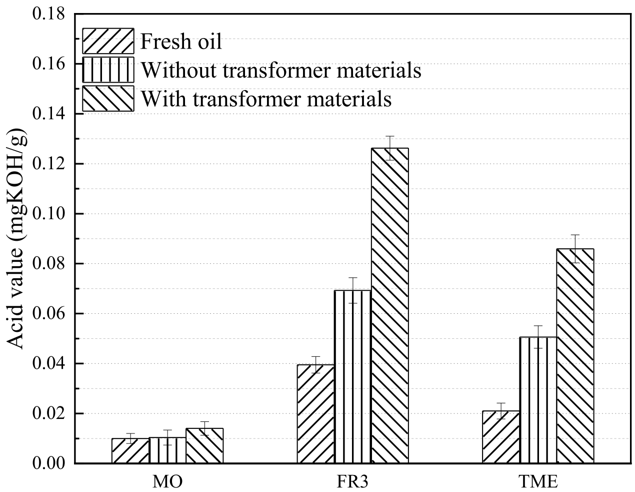

FIGURE 12. **Comparison of acid value of insulation oils at various thermal aging conditions.**

similar variation trend to that of the moisture content. For MO, the transformer materials have a weaker on the acid value than that of FR3 and TME oil. The acid value of thermal aging MO without transformer materials remains nearly unchanged with a comparison to fresh MO. The effect of transformer materials on the thermal aging of FR3 results in an obvious increase of acid value. Although the TME oil has no unstable functional groups (C=C and β-H), there is an apparent increase of the acid value of TME oil after thermal aging; the increase may be attributed to the hydrolysis, which is the inherent properties of ester oil [34]. The transformer materials, such as Cu and Fe, can catalyze the hydrolysis and oxidation of ester oil. Therefore, the acid value of aged TME oil with transformer materials is higher.

Frequency Domain Spectroscopy (FDS) was used to measure the dissipation factor and conductivity to indicate the thermal aging status of insulation oil [35]. The dielectric dissipation factor is a parameter that represents the conductivity and polarity components in an insulation oil and is an important parameter of the quality. The aging products, such as moisture, acid and gases, increase the conductivity and relaxation polarization of the oil. The frequency dependence of tan δ measured for all samples in the range of 0.1 Hz-10000 Hz is shown in **Fig. 13**

The tan δ of insulation oil decreases with the increase in frequency (**Fig. 13**). The aging products cause an increase in tan δ. For FR3 (**Fig. 13(b)**), the difference in tan δ is very small between the new and the aged oil without transformer materials at the range of 1-10000 Hz, but it becomes apparent below 1 Hz. The addition of copper, iron and paper greatly increases the tan δ at the range of below 10 Hz. Therefore, the addition greatly accelerates the thermal aging process. A similar trend is shown in **Fig. 13(a)** and **Fig. 13(c)**. For TME oil, the difference in tan δ at various condition is apparent below 10 Hz. It is evident that the effect of the transformer materials on the thermal aging of TME oil is less than that of FR3, which may be attributed to the absence of carbon-carbon double bonds and β-H in TME oil molecules. The effect of transformer materials on the thermal aging of TME oil and MO is lower compared with that of FR3 (*cf.* **Figs. 13(a)** and **13(c)**). The tan δ of MO is much smaller than

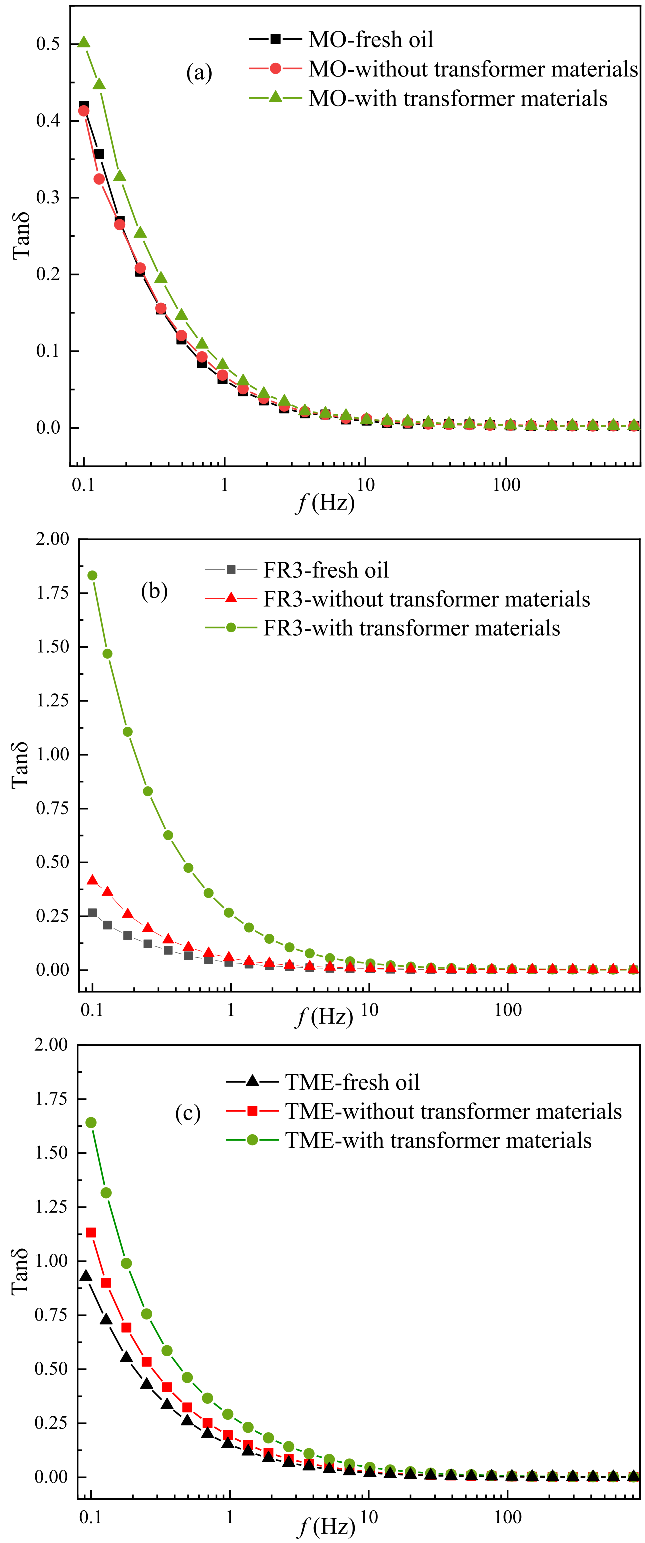

FIGURE 13. **Comparison of tan δ of insulation oils at various thermal aging conditions: (a) MO, (b) FR3, (c) TME oil.**

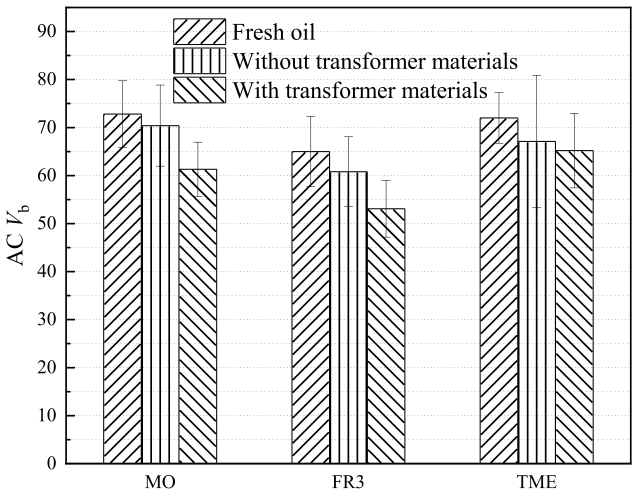

FIGURE 14. Comparison of AC breakdown voltages of insulation oils at various thermal aging conditions.

that of FR3 and TME oil. The tan δ of aged MO without transformer materials is almost identical to fresh MO due to the high oxidative stability of MO aged only ten days without transformer materials.

Fig. 14 presents the comparison of AC V_b of insulation oils before and after the thermal aging process. As shown in **Fig. 14**, the ACV_b of fresh MO is 72.8 kV. The ACV_b of MO decreases to 70.4 after thermal aging without transformer materials. However, a relatively large decrease in ACV_b (61.3 kV) of MO after thermal aging with transformer materials, which is attributed to the increased moisture content in MO (*cf*. **Fig. 10**). It is apparent that the MO is highly sensitive to the moisture (*cf.* **Fig. 11**). A similar variation trend of ACV_b for FR3 is observed in **Fig. 14**. The ACV_b of TME oil with transformer materials is slightly lower than that of TME oil without transformer materials, which is attributed to the high moisture solubility in it (*cf.* **Fig. 11**).

IV. CONCLUSION

In this paper, to obtain ester insulation oil with high oxidation stability, saturated fatty acids (C6, C8 and C10 acids) and trimethylolpropane were used. The TME oil has lower kinematic viscosity (23.3 cSt) compared with NEO FR3 (34 cSt). The pour point of FR3 oil is 21°C below zero, which is notably higher than that of TME oil (−45°C). The TME oil exhibits excellent breakdown strength (72.6 kV/2.5mm) compared with FR3 (65 kV/2.5mm). TME oil is readily biodegradable, whose biodegradation is 82.2% after 28 days. Due to the elimination of the carbon-carbon double bonds and β-H, The TME without antioxidation exhibits high OOT as 205.3°C, which is around 26°C higher than that (179.8°C) of the commercial FR3.

The distinct thermal aging behavior with transformer materials of the TME oil is explicit compared to that of FR3 and MO. The effect of transformer materials on the thermal aging of TME oil and MO is less than that of FR3. Although the moisture content and acid value in TME oil induced by aging are higher than those in MO. These acids and moisture do not cause obvious degradation of the AC V_b and tan δ, which may be attributed to the high moisture solubility in it.

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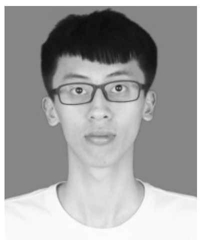

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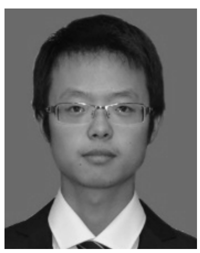

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