Indonesian Journal of Electrical Engineering and Computer Science
Vol. 33, No. 1, January 2024, pp. 10~19
ISSN: 2502-4752, DOI: 10.11591/ijeecs.v33.i1.pp10-19

Impact of low molecular weight acid and moisture on the thermal ageing properties of palm oil

Muhammad Muzamil Mustam[1,2], Norhafiz Azis[2,3], Jasronita Jasni[2], Rasmina Halis[4], Mohd Aizam Talib[5], Nur Aqilah Mohamad[6], Zaini Yaakub[7]
[1]Faculty of Electrical Engineering, Universiti Teknologi MARA (UiTM) Johor, Pasir Gudang Campus, Pasir Gudang, Malaysia
[2]Advanced Lightning, Power, and Energy Research Centre (ALPER), Faculty of Engineering, Universiti Putra Malaysia, Serdang, Malaysia
[3]Institute of Nanoscience of Nanotechnology (ION2), Universiti Putra Malaysia, Serdang, Malaysia
[4]Department of Natural Resources Industry, Faculty of Forestry and Environment, Universiti Putra Malaysia, Serdang, Malaysia
[5]Advanced Diagnostic Services, TNB Labs Sdn. Bhd., Kajang, Malaysia
[6]Department of Electrical Power Engineering, Faculty of Electrical Engineering, Universiti Teknologi Malaysia, Johor Bahru, Malaysia
[7]Hyrax Oil Sdn. Bhd., Klang, Malaysia

Article Info

Article history:

Received Oct 3, 2023
Revised Nov 2, 2023
Accepted Nov 10, 2023

Keywords:

Insulation paper
Mineral oil
Refined, bleached, and deodorised palm oil
Thermal ageing

ABSTRACT

This study examines the effect of low molecular weight acid (LMA), moisture and oxygen on the thermal ageing characteristics of refined, bleached, and deodorised palm oil (RBDPO). The paper moisture was varied between 0.5% and 3.5%. The oil was initially subjected to 0.2 g of LMA and 20 mbar of oxygen pressure. The thermal ageing experiment was performed at 120 °C and 140 °C for 28 days. Several dielectric and physiochemical parameters were measured which included dielectric dissipation factor, relative permittivity, resistivity, moisture in oil, acidity, and thermogravimetric analysis (TGA). It is found that LMA and moisture in paper do not affect the relative permittivity of RBDPO and mineral oil (MO). The dielectric dissipation factor of RBDPO and MO reveals slight increment trends within the ageing time. The decrements of resistivities occur after 7 days of ageing for both RBDPO and MO while only RBDPO shows decrement trend of moisture in oil. The ageing patterns of relative permittivities, dielectric dissipation factors and resistivities for RBDPO are similar to MO. The increment of acidity for RBDPO is more apparent that MO throughout the ageing time. All RBDPOs are more resistant to ageing than MO based on the TGA.

Corresponding Author:

Norhafiz Azis
Advanced Lightning, Power and Energy Research Centre (ALPER), Faculty of Engineering
Universiti Putra Malaysia
Serdang, Selangor, Malaysia
Email: norhafiz@upm.edu.my

1. INTRODUCTION

Palm oil is widely available in Southeast Asia countries and extensive investigations have been conducted to determine whether it is a viable substitute for dielectric insulating fluid for transformers [1]–[4]. Different types of studies are conducted on palm oil to explore more about its applications [5]–[7]. One of the common types of palm oil known as refined, bleached and deodorised palm oil (RBDPO) is able to meet with the dielectric properties requirement for transformer application [2]. Furthermore, the AC breakdown voltage and flash/fire points of unaged RBDPO are high as compared to mineral oil (MO) [8], [9]. The performance of aged palm oil has been investigated under open and sealed ageing conditions [10]. The relative permittivity of RBDPO, dielectric dissipation factor and AC breakdown voltages are found to be unaffected by ageing under both conditions [2], [10], [11]. However, the resistivity of RBDPO can be affected by ageing under open and sealed conditions [2], [12], [13]. The ageing has no clear impact on the moisture, viscosity and acidity trends of RBDPO under open conditions [10]. One of the main findings of a previous study is the RBDPO can delay the ageing process of the paper whereby it is able to maintain tensile strength retention higher than 50% [10]. The thermogravimetric analysis (TGA) for RBDPO aged under sealed ageing conditions reveal that the onset temperature is higher than MO [1]. The ageing resistance of RBDPO is due to its high oxidative stability, which is contributed by the presence of the hydroxyl group in partial esters [1], [14], [15].

It is widely known that oxygen, moisture and acid can speed up the ageing process of oil and paper. Several studies are performed to evaluate the impact of these properties on the paper degradation in MO [16]–[18]. The presence 4% of moisture in paper can enhance the rate of paper degradation in MO by 20% [17], [18]. In addition, the oxygen level up to 500 ppm can accelerate paper degradation in MO by a factor of 3 [16], [17]. Low molecular weight acid (LMA) is detrimental for paper aged in MO as compared to high molecular acid (HMA). LMA can cause up to 40% reduction of degree of polymerization for paper ageing in MO in the presence of LMA [19]. Currently, there is limited work on the thermal ageing of natural ester such as RBDPO especially related to the synergy effect by moisture, oxygen, and acids.

This paper discusses the impact of LMA, moisture and oxygen on the ageing performance in term of dielectric and physiochemical properties for RBDPO and paper. The physiochemical properties of RBDPO, which include dielectric dissipation factor, relative permittivity, resistivity, moisture, acidity and TGA are measured and analysed. This study can provide detail insight on the impact of LMA and moisture on the dielectric and physiochemical characteristics of RBDPO under thermal ageing.

2. EXPERIMENTAL DESCRIPTION

2.1. Thermal ageing procedure

The filtration of RBDPO and MO was performed 3 times using a membrane filter that has a pore size of 0.2 m. The properties of the RBDPO and MO used in this study can be found in [14]. These oils were dried for 2 days at 85 °C in a vacuum oven and subsequently purged with nitrogen to reduce the moisture. The final moisture contents of RBDPO and MO are 109 ppm and 12 ppm. The insulation paper was dried at 90 °C or 105 °C in a vacuum oven at different times to produce samples with specific moisture content as seen in Table 1. The low, medium and high moisture contents in paper were defined based on the range from 0.5% to 1.5%, from 1.51% to 2.5% and from 2.51% to 3.5%. The paper was then impregnated in either RBDPO or MO through a vacuum oven for 1 day at 85 °C. In total, 0.2 g of formic acid known as LMA was added into RBDPO and MO and 20 mbar of oxygen gas pressure were applied above the bottle’s oil surface known as a vapour pressure. The procedure to introduce the oxygen into the samples was carried out in accordance to previous studies [16]. The oil-to-paper ratio chosen was 20:1 as per [2], [10], whereby the weight of the oil and paper in this study were 450 g and 22.5 g. Borosilicate glass was used for the ageing experimental work. The borosilicate glass was sealed with caps reinforced with polytetrafluoroethylene tape to minimise environmental interaction. The ageing temperatures used in this study are 120 °C and 140 °C whereby the ageing was performed for 7 days, 14 days, 21 days and 28 days. For each of the ageing times, RBDPO and MO were kept at room temperature for up to 24 hours before the moisture in oil, acidity, resistivity, relative permittivity, dielectric dissipation factor and TGA were determined.

Table 1. Moisture contents in paper for different conditions

Sample	Conditions	Temperature (°C)	Time (h)	Moisture content (%)
MO (A)	Low	105	96	0.55
MO (B)	Medium	90	48	1.66
MO (C)	High	90	24	2.65
RBDPO (A)	Low	105	96	0.55
RBDPO (B)	Medium	90	48	1.66
RBDPO (C)	High	90	24	2.65

3. TEST DESCRIPTION

3.1. Dielectric dissipation factor, relative permittivity, and resistivity

The dielectric properties such as dielectric dissipation factor, relative permittivity and resistivity were all measured through BAUR DTL C oil tester based on IEC 60247 [20]. The measurement was performed at 90 °C and 50 Hz. The oil volume used for the testing was around 40 ml and 1 reading was recorded for each of the parameter as per similar approach in previous studies [2].

3.2. Moisture in oil and paper

Moisture in oil and paper were determined through Metrohm 831 and oven drying method as per ASTM D6304 [21] and ISO 287-2017 [22]. In total, 1 ml of oil was utilized to determine the moisture in oil and 0.5 g of paper was required to determine the moisture in paper. In total, 2 measurements of moisture in oil and paper were taken for the analysis as per similar approaches in previous studies [2], [23].

3.3. Acidity in oil

A Metrohm 877 oil Titrano plus measured acidity of oil based on ASTM D974 [24]. For each of the testing, 10 g of oil was used and 1 reading was taken for the analysis. The number of measurement is similar to the approach in [2]. Furthermore, the measurement of acidity for RBDPO was quite long and due to the time constrain, 1 measurement was taken to standardize with MO.

3.4. Thermogravimetric analysis

The TGA measurement of oil was performed under non-isothermal conditions based on the ASTM E1131-08 [25] using Mettler Toledo, TGA-DSC HT 3. The oil used in the measurement had a weight of 5 mg. The system was first purged with nitrogen gas at a 50 ml/min rate for 20 minutes at 25 °C to release the trapped gases in the furnace. Next, the sample was heated to 600 °C and held for 10 minutes whereby the rate of increment was set to 10 °C/min.

4. RESULT AND DISCUSSION

4.1. Dielectric dissipation factor

The dielectric dissipation factors for RBDPO and MO slightly increase as the ageing temperature increases from 120 °C to 140 °C as shown in Figure 1. At both ageing temperatures, the dielectric dissipation factors of RBDPO are higher than MO even after 28 days of ageing. At the ageing temperature of 120 °C, the range of dielectric dissipation factors of the RBDPO is from 0.023 to 0.601, but for all MO, the range is from 0.00095 to 0.00481 as shown in Figure 1(a). The same pattern is found at the ageing temperature of 140 °C whereby the range of dielectric dissipation factors for all RBDPO is from 0.021 to 0.558 while the range for MO is from 0.00091 to 0.00416 as seen in Figure 1(b).

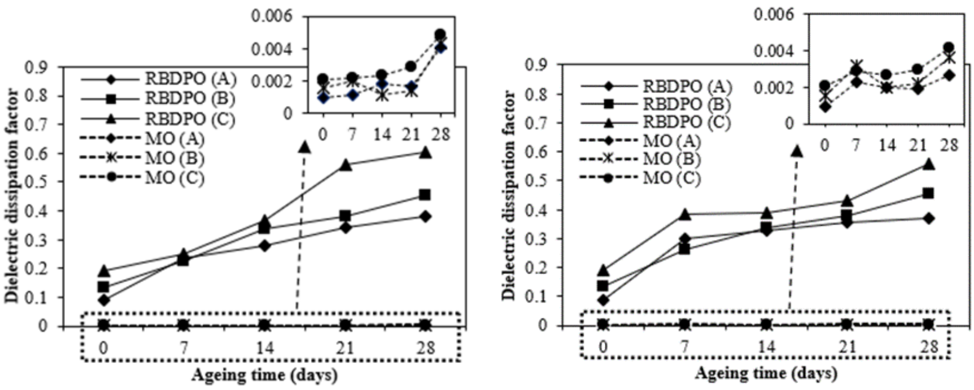


Figure 1. Dielectric dissipation factor for RBDPO and MO in the presence of LMA, moisture and oxygen at (a) 120 °C and (b) 140 °C

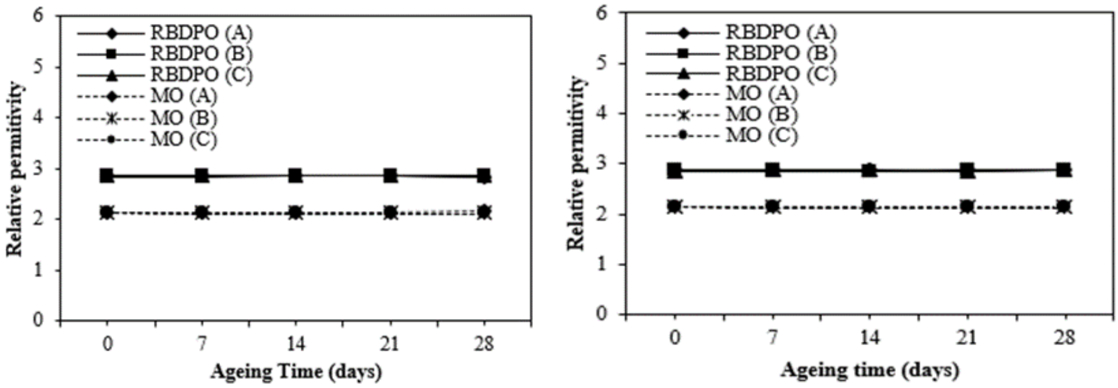
LMA and high moisture in paper have an apparent impact on the dielectric dissipation factor increments for RBDPO and MO at both ageing temperatures in this study. The highest percentages of increments of dielectric dissipation factors at the ageing temperature of 120 °C for RBDPO and MO are 34.4% and 68.72% which occur at the ageing times of 21 days and 28 days as shown in in Figure 1(a). A similar pattern is found at the ageing temperature of 140 °C in Figure 1(b) whereby the dielectric dissipation factors for RBDPO and MO increase as LMA and high moisture in paper are present. RBDPO and MO exhibit up to 50.87% and 28.44% percentages of increments for dielectric dissipation factors after 7 days of ageing.

Previous studies show that the natural ester such as RBDPO has a higher dielectric dissipation factor than MO [2], [26]. Palm oil has 50% of saturated fatty acids in its molecular structure which leads to the increment of its polar property and subsequently increases the dielectric dissipation factor [2], [27]. At both ageing temperatures and in the existence of LMA and high moisture in paper, as seen in Figure 1, RBDPO exceed the IEC 62770 [28] limit of 0.5 after 21 and 28 days of ageing. The dielectric dissipation factor of MO still maintains within the IEC 60296 [29] limit of 0.005 even after being subjected to the ageing temperature of 140 °C as seen in Figure 1(b).

4.2. Relative permitivity

At both ageing temperatures, LMA and moisture in paper have no impact on the relative permittivities for RBDPO and MO as seen in Figures 2. The relative permittivities for RBDPO maintain between 2.84 and 2.87 while for MO, the range is from 2.14 to 2.16 at ageing temperature of 120 °C according to Figure 2(a). With the temperature set for ageing at 140 °C, the range of permittivities for RBDPO is from 2.86 to 2.88 while for MO, it is between 2.14 and 2.15 as seen in Figure 2(b).

It is expected that the relative permittivities for RBDPO and MO maintain unchanged even in the presence of LMA, moisture and oxygen. The presence of triglycerides structure in the natural ester leads to high permittivity of the RBDPO as compared to the MO [12], [30]. The connection of an ester group to alkyl chain can increase spontaneous polarisation and lead to the increment of relative permittivity [31]. It is also shown the presence of dipole polarisation in the natural ester leads to the high relative permittivity [32].



(a) (b)

Figure 2. Relative permittivity for RBDPO and MO in the presence of LMA, moisture and oxygen at (a) 120 °C and (b) 140 °C

4.3. Resistivity

The pattern for resistivity RBDPO versus ageing time is different as compared to MO at both ageing temperatures are shown in Figure 3. The resistivity of RBDPO initially increases at both ageing temperatures and starts to experience decrement pattern after 7 days of ageing. Similar patterns are observed for MO at the ageing temperature of 120 °C except that resistivity decreases after 14 days of ageing as depicted in Figure 3(a). However, the resistivity of MO is unaffected even the ageing temperature is at 140 °C as shown in Figure 3(b).

LMA and high moisture in paper can affect the resistivity for RBDPO, whereby the lowest values at ageing temperatures of 120 °C and 140 °C are 0.00018 TΩm and 0.00016 TΩm respectively as shown in Figure 3. In total, 52.63% and 57.89% decrements of resistivities are recorded for RBDPO at ageing temperatures of 120 °C and 140 °C after 28 days of ageing. The impact of LMA and high moisture in paper on the resistivity of MO at both ageing temperatures is not apparent as compared to RBDPO as seen in Figure 3. At the ageing temperature of 120 °C, the resistivity of MO decreases up to 80.36% as LMA and high moisture in paper are present as seen in Figure 3(a). In the presence of LMA and medium moisture in paper, the resistivity of MO decreases by 69.41%.

Previous studies have shown that the resistivities of MO and vegetable based oils can be affected by moisture and acidity [2], [10], [12], [33]. The resistivity of MO remains higher than RBDPO at both ageing temperatures and for the entire ageing time, even when there is high moisture content in the paper and LMA is present. MO has a higher resistivity than natural ester as a result of its low electrostatic charge and ion mobility that is attributed to its low water saturation level [34]. In addition, the molecular weight and viscosity MO can affect the electron mobility which in turn affect the resistivity [35]. The high hygroscopicity nature of natural ester due to the hydrogen bonding of the water within the molecular structure linkage also tends to reduce its resistivity [36].

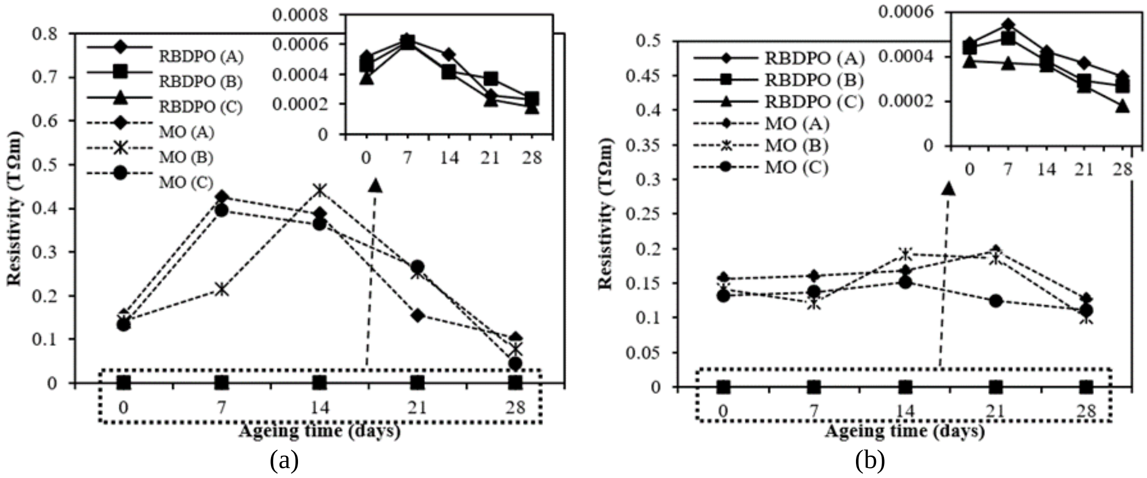


Figure 3. Resistivity for RBDPO and MO in the presence of LMA, moisture and oxygen at (a) 120 °C and (b) 140 °C

4.4. Moisture

The moisture in RBDPO decreases as the ageing time increases for both ageing temperatures as shown in Figure 4. Moisture in RBDPO is within the similar range as MO when the ageing process comes to an end. The moisture in MO remains almost unchanged at the range between 20.2 ppm and 54.4 ppm at both ageing temperatures. Between 7 days and 28 days of ageing, the moisture in the RBDPO is within the limit in IEC 62770 [28], which is less than 200 ppm at both ageing temperatures. Meanwhile, the moisture in MO at both ageing temperatures maintains within the IEC 60296 [29], which is less than 40 ppm. At the ageing temperature of 120 °C, the moisture in RBDPO and MO decrease by 64.97% and 60.32% after 7 days of ageing in the presence of LMA and high moisture in paper as shown in Figure 4(a). The decrement of moisture content in RBDPO is quite significant with 73.07% at the ageing temperature of 140 °C while the decrement of moisture in MO decreases to 21.81% as seen in Figure 4(b).

The decrement of moisture in RBDPO with the increment of ageing time is likely caused by the natural ester capacity to interact with moisture through acid-catalysed hydrolysis [37], [38]. Due to its hygroscopicity and polar nature, natural ester moisture solubility is higher than MO [39]. It is related to the bond numbers around the triglycerides molecule where the occurrence of the free rotations is present [32]. The C=O bonds in natural esters cause a significant imbalance in the charge imbalance due to the different electronegativity of both oxygen and carbon atoms [40]. This distinction generates permanent dipoles that increase the propensity and polarity of natural esters [40]. Dipole moments of both ester and water molecules can interact that results in strong attractive interactions for these molecules [41]. The dipole moment-induced effect can allow the ester to absorb moisture from the surrounding [42].

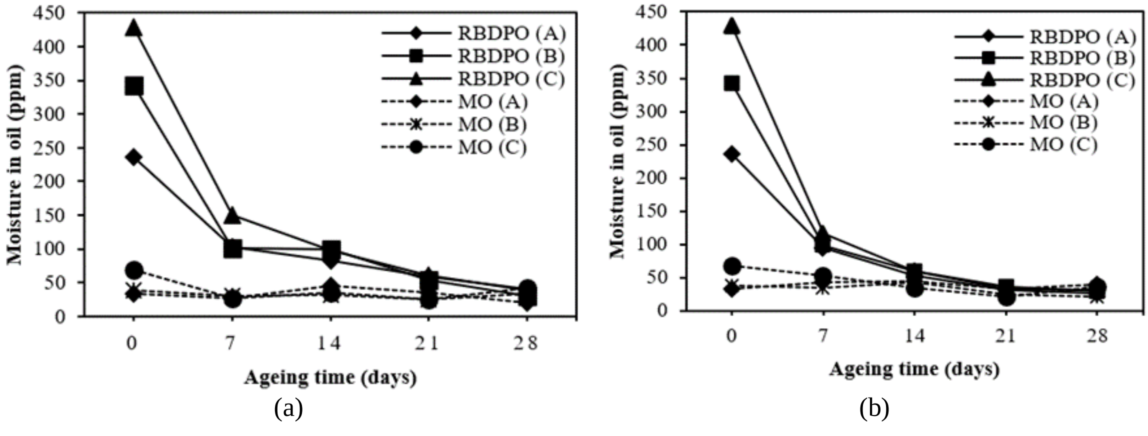


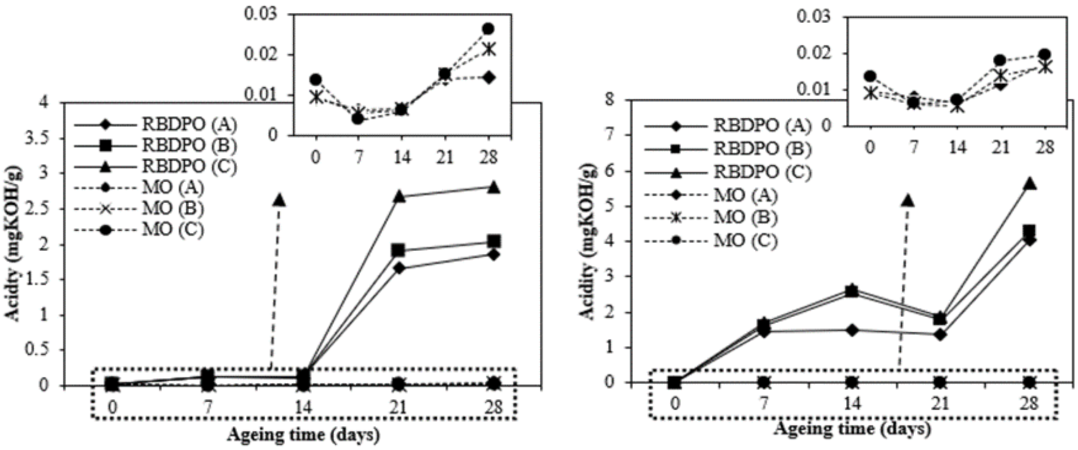
Figure 4. Moisture in RBDPO and MO in the presence of LMA, moisture and oxygen at (a) 120 °C and (b) 140 °C

4.5. Acidity

Generally, the acidities for RBDPO and MO in Figure 5 increase as the ageing temperature increases. At both ageing temperatures, the RBDPO has higher acidity than MO. The highest acidity of RBDPO at the ageing temperature of 120 °C is 2.8114 mg KOH/g while for MO, it is 0.0261 mg KOH/g as seen in Figure 5(a). Similar pattern is found at the ageing temperature of 140 °C whereby the highest acidity for RBDPO is 5.6531 mg KOH/g whereby for MO, it is 0.0194 mg KOH/g as seen in Figure 5(b). The acidity of RBDPO from 7 days to 28 days of ageing exceeds the IEC 62770 [28] limit of 0.06 mgKOH/g while the acidity of MO from 14 days to 28 days of ageing exceeds the IEC 60296 [29] limit of 0.01 mgKOH/g.

It is apparent that LMA and high moisture in paper can affect the acidity increments for RBDPO and MO at both ageing temperatures as shown in Figure 5. The highest increments of acidities for RBDPO and MO at the ageing temperature of 120 °C can be up to 96.22% and 60.66% after 21 days of ageing as seen in Figure 5(a). Similar pattern is found at the ageing temperature of 140 °C whereby the acidities for RBDPO and MO can exhibit up to 66.71% and 60.89% percentages of increments after 28 days and 21 days of ageing as shown in Figure 5(b).

The apparent increment of acidity for RBDPO can be due to the existence of LMA and moisture. Furthermore, the fatty acid composition can also affect the acidity level [27], [37]. The decrement of the acidity of MO during the early stages of ageing are related to the carboxylic acid consumption [35]. As the ageing time increases, both HMA and LMA continue to increase in MO and further affect the total acid number [43]–[45].



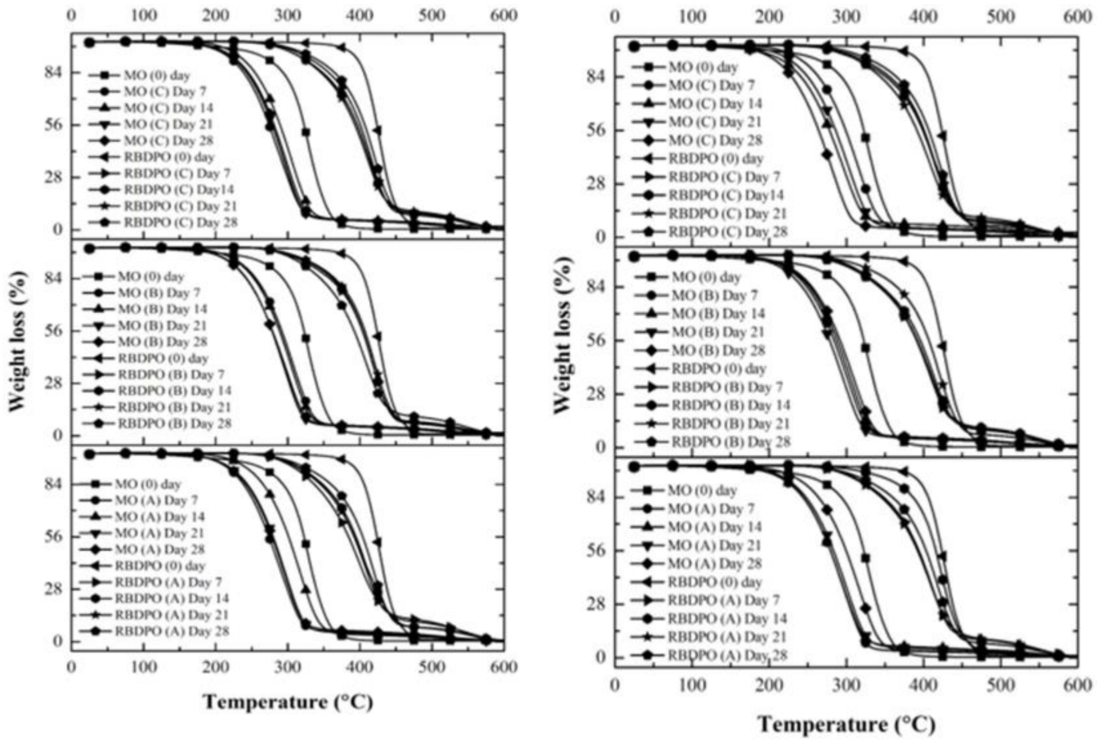
(a) (b)

Figure 5. Acidity for RBDPO and MO in the presence of LMA, moisture and oxygen at (a) 120 °C and (b) 140 °C

4.6. Thermogravimetric analysis

TGA records the weight loss of oil as it is heated where it reflects the evolution of volatile components and degradation products. Figure 6 show the thermogravimetric curves of weight losses as a function of temperature for RBDPO and MO at both ageing temperatures. For both ageing temperatures, the RBDPO show significant weight loss at a temperature higher than 300 °C while MO shows the similar phenomenon at temperature higher than 200 °C. At the ageing temperature of 120 °C, the onset temperatures of RBDPO and MO decrease up to 42.48% and 52.22% after 28 days of ageing with LMA and high moisture in paper as seen in Figure 6(a). The decrement of onset temperatures for RBDPO and MO slightly decrease to 42.18% and 48.60% at the ageing temperature of 140 °C as seen in Figure 6(b).

The finding demonstrates that RBDPO has lower weight loss than MO, which indicates high resistance during ageing. This is because the triglycerides in RBDPO have a lower susceptibility to oxidation and other degradation reactions than hydrocarbons in MO. Due to its structure low molecular weight components, MO has high volatility, which indicate low oxidative stability. Another factor that can contribute to high thermal stability of RBDPO is the presence of natural vitamin E that act as antioxidant, which retard the oxidation process [11], [46]. The thermal performance of RBDPO is better that MO attributable to the high content of mono-unsaturated fatty acids where it can increase the oxidative stability of the oil [47], [48].



(a) (b)

Figure 6. Thermogravimetric curves of weight loss for RBDPO and MO in the presence of LMA, moisture and oxygen at (a) 120 °C and (b) 140 °C

5. CONCLUSION

This study investigates the influence of LMA, different moisture contents and oxygen on the dielectric and physiochemical properties of aged RBDPO and MO. Under current thermal ageing study, both moisture in paper and LMA do not affect the relative permittivities of RBDPO and MO. Dielectric dissipation factors of RBDPO and MO show slight increment trends in the presence of LMA and moisture in paper. RBDPO and MO experience decrements of resistivities after 7 days of ageing while only RBDPO shows decrement trend of moisture in oil in the presence of LMA and moisture in paper. Acidity of RBDPO experiences clear increment trend as compared to MO throughout the ageing time. According to the TGA analysis, RBDPO is more age-resistant than MO in the presence of LMA and moisture in paper.

ACKNOWLEDGEMENT

The authors would like to express sincere gratitude for The Matching Grant University of Putra Malaysia provided the funding for this study. Kyushu Institute of Technology (UPM.RMC/800/2/2/4/Matching UPM–KYUTECH/2021/9300481), Ministry of Higher Education under the FRGS scheme of FRGS/1/2019/TK07/UPM/02/3 (03-01-19-2071FR), Universiti Teknologi MARA (UiTM) and Majlis Amanah Raya (MARA). For the technical assistance, we would especially like to thank Hyrax Oil Sdn. Bhd. and Malaysia Transformer Manufacturing Sdn. Bhd.

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BIOGRAPHIES OF AUTHORS

Ts. Muhammad Muzamil Mustam obtained his B.Tech., (Edu) in Electrical Engineering with Education and M.Eng. degrees in Electrical Engineering (Power) from Universiti Teknologi Malaysia (UTM) in 2010 and 2013, respectively. He is currently pursuing Ph.D., degree in Electrical Power Engineering at Universiti Putra Malaysia. He is currently Senior Lecturer at the Department of Electrical Engineering, Universiti Teknologi MARA (UiTM) Johor, Pasir Gudang Campus. His research interest includes the high voltage material for liquid and solid insulation and thermal ageing of vegetable oil as alternative liquid for transformers. He can be contacted at email: dmuzam87@gmail.com.

Assoc. Prof. Ir. Dr. Norhafiz Azis received the B.Eng., degree in electrical and electronic engineering from Universiti Putra Malaysia, Serdang, Malaysia, in 2007, and the Ph.D., degree in electrical power engineering from The University of Manchester, Manchester, United Kingdom, He is currently an Associate Professor with the Department of Electrical and Electronic Engineering, Universiti Putra Malaysia. His research interests are in-service aging of transformer insulation, condition monitoring, asset management, and alternative insulation materials for transformers. He can be contacted at email: norhafiz@upm.edu.my.

Assoc. Prof. Dr Jasronita Jasni received the B.Eng., degree in Electrical Engineering and the M.Eng., degree in Electrical Engineering from Universiti Teknologi Malaysia, Johor Bahru, Malaysia, in 1998 and 2001, respectively, and the Ph.D., degree in Electrical Power Engineering from Universiti Putra Malaysia, Serdang, Malaysia, in 2010. She is currently an Associate Professor with the Department of Electrical and Electronic Engineering, Universiti Putra Malaysia. Her research interests include power system analysis for static and dynamics, load flow analysis, embedded generation, and renewable energy. She can be contacted at email: jas@upm.edu.my.

Assoc. Prof. Ts. Dr. Rasmina Halis obtained bachelor's degree at University of Manchester and Master Degree from Universiti Sains Malaysia. She completed his Ph.D., study from The University Putra Malaysia and currently is an Associate Professor under the Department of Natural Resources Industry, Faculty of Forestry and Environment Universiti Putra Malaysia (UPM). Her main research area are paper science (hydrogen bond failure, conservation ageing paper) and biotechnology in Pulp and Paper (Kenaf Bio Kraft Pulping). She can be contacted at email: rasmina@upm.edu.my.

Ir. Dr. Mohd Aizam Talib received his B.Eng., in Electrical Engineering from the University of Portsmouth, United Kingdom in 1997, M.Eng., in Electrical Engineering from Universiti Tenaga Nasional (UNITEN), Malaysia in 2001 and Ph.D., degree at Universiti Teknologi Malaysia in 2016. He has over 24 years of experience in dealing with transformer research, testing, diagnostic and life assessment of in-service power transformers. He has held several positions previously with TNB Research Sdn. Bhd. as Senior Research Engineer, Manager for High Voltage Testing Laboratory and Senior Technical Expert (Transformer). Presently, he is Head of Advanced Diagnostic Services at TNB Labs Sdn. Bhd. His research interests are in transformer condition monitoring, insulation diagnostic and dielectric measurements. He can be contacted at email: aizam.talib@tnb.com.my.

Dr. Nur Aqilah Mohamad She received the B.Eng., degree in Electrical and Electronic Engineering (Power) from Universiti Pertahanan Nasional Malaysia, Kuala Lumpur, Malaysia. In 2021, she received Ph.D., degree in Electrical Power Engineering from Universiti Putra Malaysia, Selangor. She is currently a Senior Lecturer with the Department of Electrical Power Engineering Universiti Teknologi Malaysia. Her research interests are in-servic transformer insulation, condition monitoring and alternative materials for transformers. She can be contacted at email nuraqilah.m@utm.my.

Zaini Yaakub was awarded the degree of Applied Chemistry with Honours from Sheffield Hallam University, United Kingdom in 1993. He joined Caleb Brett Malaysia in the same year as a Chemist before joining Hyrax Oil in 1994. Currently, he is the General Manager at Hyrax Oil after having had a working experience for more than 20 years on various responsibilities and roles. His research interests are in the field of electrical insulating oils. He can be contacted at email: zaini@hyraxoil.com.