Study of Thermallay accelerated aging of Transformer Insulation Paper in Mineral Oil and Gas-to-Liquid Dielectrics using Scanning Electron Microscopy (SEM), Thermogravimetric Analysis (TGA) and X-ray Diffraction (XRD)

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Abstract: This paper reports novel comparative study results on the degradation of kraft paper immersed in mineral oil and gas-to-liquid dielectrics under accelerated aging at 120ºC and 150ºC for duration up to 1344 hours in a controllable oven. Tensile strength and degree of polymerization (DP)v of paper samples before and after aging were measured. Morphological changes were observed using SEM (scanning electron microscope). Degradation of cellulose from kraft paper including its crystallinity change was investigated through EDX (x-ray diffraction) analysis. Thermogravimetric (TG) analysis was used to study the thermal stability of the paper samples aged in the dielectrics. Experimental results showed that DP, and tensile strength of kraft paper decreased with aging. SEM observation indicated that micro globules detected in the aged paper sample and the diameter of micro globules increased with aging time and temperature. The EDX analysis showed that there were increase of crystallinity of aged paper samples. TG analysis showed that higher mass loss was found in aged samples compared to those new one. The initial decomposition temperature slightly reduced with aging time and temperature.

Keywords: solid and liquid dielectrics, kraft paper, mineral oil, gas-to-liquid oil, power transformer, thermal aging, degree of polymerization, tensile strength, SEM, EDX, TGA

1. Introduction

Delivery of a large scale electric energy has become very effective by using high voltage transmission. Power transformers with different voltage levels have made it possible to realize high voltage transmission to transfer a large amount of electric energy from the point of generation to the load over a long distance with minimum power loss [1-2]. A high voltage power transformer mainly consists of an iron core, a copper winding insulated with paper insulation, a liquid insulation medium and a tank that holds everything together. Typical transformer and paper insulation are shown in figure 1.

Unlike oil that can go through the retro-filing or regeneration process, paper cannot be repaired easily. Therefore, the degradation of paper is considered to be one of the main lifetime deciding factors for transformers and has been studied for several decades [3]–[8]. Insulating oil inside of high voltage transformer has function of insulating and cooling agents, which also can influence insulating paper inside. Because of thermal aging, insulation performance of insulating oil and paper may be degraded.

For the least past 100 years, insulating paper mad from cellulose has been applied for insulation of high voltage transformer [9]. Cellulose consist of D-anhydroglucose units which is linked with glycosidic oxygen link [10]. Kraft paper consist mainly of cellulose, which is also have been use for over a decade. Under operating condition, kraft paper insulation degradation process happened because of heating is important as aging factor.

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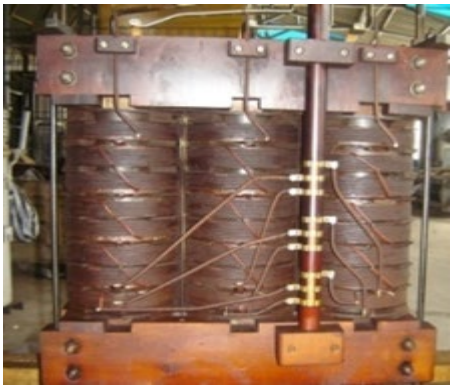
(a)



(b)

Figure 1(a). A transformer at 150 kV substation and
(b) and typical paper insulation inside a power transformer

The lifetime of paper insulation was influenced by several factors such as heating, oxygen and water [11]. There are several aging mechanisms applied to the paper insulation such as pyrolysis, hydrolysis and oxidation [12]. Mineral oil is widely used because a good compatibility with the cellulose insulating paper, good dielectrics , physical and arc quenching properties [13]. While scanning electron microscopy (SEM) is usefull for investigating the physical structure of transformer insulation paper. This technique shows the microstructure of paper surfaces.

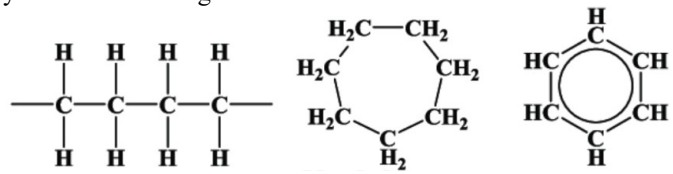
This paper reports the experimental results on the chemical and physical structures of transformer insulation paper using scanning electron microscopy (SEM) and energy dispersive x-ray spectroscopy (EDS). Paper insulation for transformer and copper strip were immersed in mineral oil in a hermetical bottle and subjected to an thermal aging test at temperature 120°C and 150°C for 336, 672 ,1008, and 1334 hours. Dispersive x-ray spectroscopy (EDS) shows the weight percent of the chemical element of transformer insulation paper. Scanning electron microscopy (SEM) shows the microstructure of paper surface which is the micro gap, the micro globule and the roughness of the surface.

2. Materials and Methods

A.1. Sample

Uninhibited mineral oil (Nynas Nitro Libra) and GTL inhibited oil (Shell Diala S4 ZX- I) were used as liquid dielectrics in this experiment. The hydrocarbon compounds in mineral oil can be divided into paraffin, naphthenic and aromatics. It has been utilized in transformers since more than 100 years ago. The chemical structure of mineral oil can be seen on Figure 2. GTL oil, as new generation of liquid dielectric was developed recently. It is produced from natural

gas (methane) through gas to liquid(GTL) technology based. In the manufacturing process as seen in Figure 3, methane is first reacted with oxygen (O_2) to produce a mixture of carbon monoxide (CO) and hydrogen (H_2). Then the mixture is converted into liquid waxy hydrocarbons through the Fischer-Tropsch process. These predominantly iso-paraffinic hydrocarbon oils have fewer impurities compared to traditional mineral oils and hence are expected to perform better than mineral oil. Commercially available kraft paper commonly used in power transformer was used as paper sample. Paper is composed of cellulose that is obtained from softwood. Cellulose is an unbranched homopolysaccharide consisting of β-D-glucopyranose rings that are connected through 1,4-β-glycosidic bonds. Figure 4 shows the chemical structure of cellulose.



(a). (b). (c)

Figure 2. Hydrocarbon structures in mineral oil (a) paraffin, (b) napthene, and (c) aromatic

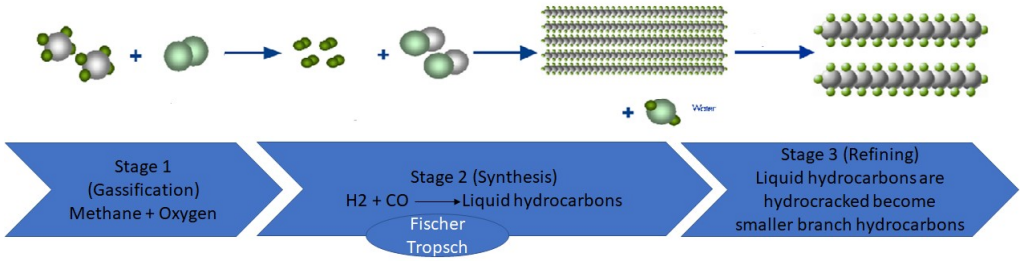


Figure 3. Gas to liquid process using Fischer Tropsch method

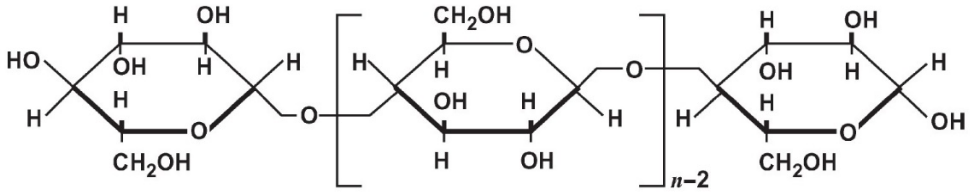


Figure 4. Chemical structures of cellulose

The paper sample is 0.01 mm thick and the density is 0.011 g/cm^2. Each aging test bottle contained 40 g of insulating paper, 850 g mineral or GTL oil and copper metal strips of 32 g. The ratio among paper insulation, liquid dielectric and metal in each bottle is typical ratio for a transformer [14]. Pictures of paper wounded at cooper strips and bottles with samples inside are shown in figure 5. The paper wrapped around the copper metal strip. Mineral oil and GTL oil gets pretreatment process before getting accelerated thermal aging test. Mineral oil and GTL oil samples in the glass bottles without sealed were subjected to heating at 100°C for 24 hours.

(a).

(b).

(c).

Figure 5. Paper samples are put into inserted into glass bottles inside oven (a) 120°C, (b) 150°C, and (c) paper wrapped around copper metal strip

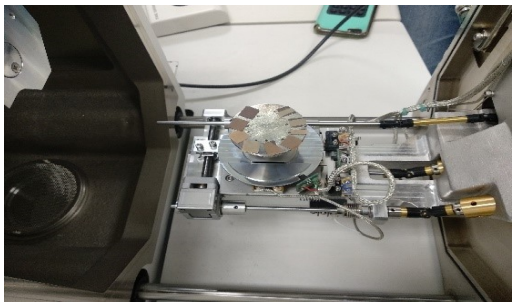
The paper sample and copper strip were then placed in glass bottles and sealed according to the IEEE recommendation loading guide in modern sealed transformers [15-16]. All the glass bottles sealed were put in the different aging oven with controlled temperature of 120°C or 150°C for aging time of 336, 672, 1008 and 1334 hours. Aging temperature of 150°C was selected based on Mc Shane, et al [17]. The 120°C represents IEEE transformer hot spot [18]. Table 1 shows samples and their treatments.

Table 1. Sample used in the experiment

Sample	Aging	Sample	Aging
MO.T0	Initial Condition	GTL.T0	Initial Condition
MO.T2.120	120°C for 336 hours	GTL.T2.120	120°C for 336 hours
MO.T4.120	120°C for 672 hours	GTL.T4.120	120°C for 672 hours
MO.T6.120	120°C for 1008 hours	GTL.T6.120	120°C for 1008 hours
MO.T8.120	120°C for 1334 hours	GTL.T8.120	120°C for 1334 hours
MO.T2.150	150°C for 336 hours	GTL.T2.150	150°C for 336 hours
MO.T4.150	150°C for 672 hours	GTL.T4. 150	150°C for 672 hours
MO.T6.150	150°C for 1008 hours	GTL.T6. 150	150°C for 1008 hours
MO.T8.150	150°C for 1334 hours	GTL.T8.150	150°C for 1334 hours

A.2. Scanning Electron Microscopy (SEM) and Energy Dispersive X-ray Spectroscopy (EDS)

(a).



(b).

(c).

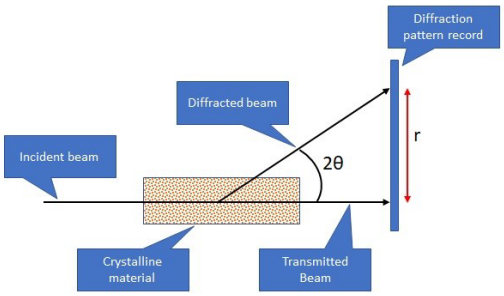
Figure 6. (a) Hitachi SU3500 and EDAX (b) Paper samples are placed in the specimen holder c) coating process using MC1000 Ion Sputter

The SEM-EDX analysis is an analysis that aimed at determining the condition of the sample. This analysis can be divided into 3 aims, the first is topography which is analyze the surface and texture of materials, the second is morphology which analyze the shape and size of the sample, and the third is composition which analyze the composition of the sample quantitatively and qualitatively [19]. To understand changes in microstructure of the transformer insulation paper surface of the transformer insulation paper, scanning electron microscopy measurement (SEM) was performed. The roughness, the micro gaps and the microglobule of paper surfaces can be detected by using SEM. The chemical structure was studied by using energy dispersive x-ray spectroscopy (EDS) analysis. In this research, Hitachi SU 3500 and EDAX were used to conduct SEM and EDS analysis. The equipment along with specimen holder are shown in Figure 6 (a) and (b). The paper samples were cut into 1x1 cm^2 and measured with 200, 500 and 1000 times of magnification at 10-15 kV accelerating voltage. This Hitachi SU 3500 can work with high resolution at low accelerating voltage. The SU 3500 can achieve 7 nm secondary electron image resolution at 3 kV accelerating voltage and 10 nm backscattered electron image resolution at 5 kV accelerating voltage. Prior SEM and EDS measurement , insulation paper sample was laid on an holder to get coating process using MC1000 Ion Sputter as shown in figure 6(c). The

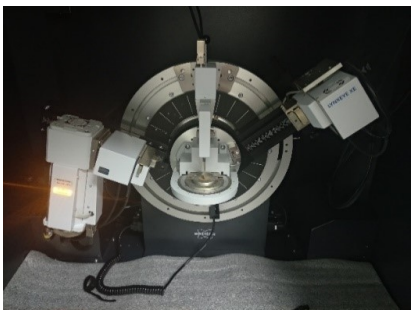
MC1000 is designed to deposit a thin metal coating, such as platinum (Pt), or gold (Au) in order to make the surface of sample electrically conductive and avoid charge build-up during observation in a SEM. The results of EDS analysis are the chemical elements of material and the weight percent of chemical elements as a quantitative data.

A.3. X-ray Diffraction (XRD)

Figure 7 shows the principles of XRD analyser (a), Bruker D8 Advance. Bruker D8 Advance (b) and sample holder (c). The Bruker D8 Advance goniometer is equipped with stepper motors controlled by optical encoders and the smallest addressable increment is 0,0001°. The X-ray diffraction was invented by W. H. Bragg in 1913 [20]. If X-ray is targeted to a material then part of the tray will be diffracted and transmitted. The diffraction will be useful for identifying the crystallographic structure.



(a).



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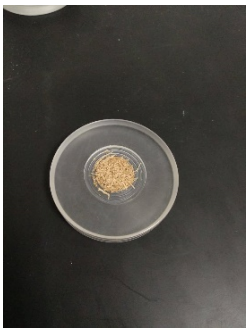


(c).

Figure 7(a). Schematic XRD Characterization, (b) Bruker D8 Advance and (c) paper sample are placed in the specimen holder

A.3. Thermogravimetric and Derivative Thermogravimetric (TG/DTG) analysis

Thermogravimetric (TG) and derivative thermogravimetric (DTG) analysis are used to determine the quantity and the frequency of the weight variation of the samples against temperature and time in a controlled atmosphere (eg, nitrogen gas) [23-25]. TG and DTG can be used primarily to investigate the thermal stability (the strength of the material at a given temperature), oxidative stabilities (the oxygen absorption rate on the material), as well as the compositional properties (eg, fillers, polymer resin, solvents) of the samples. Besides, the weight gain/loss of the samples corresponds to different factors. Generally, weight gain is attributed to the adsorption or oxidation, whereas weight loss is attributed to decomposition, desorption, dehydration, desolvation, or volatilization [26-28]. The thermal degradation characteristics of cellulose was examined by using thermogravimetric analyzer as shown in Figure 8. A computer was used to monitor the temperature and regulate the heat flow. Dry paper of 5 mg, after oil separation, was put into TG analyzer. The sample is then heated inside high temperature furnace with heating temperature in the range of 30-550°C and the heating rate of 10°C/min. The measurement took place under nitrogen gas atmosphere.

Figure 8. Thermogravimetric Analyzer

A.4. Degree of Polymerization, and Tensile Strength

Kraft paper consist mainly of cellulose fibers composed of a polymeric chain of cellulose, which is formed from d-glucose monomers. The reduction in mechanical strength is due to splitting of the cellulose polymer by the action of oxygen, water, temperature, and likely acids. The number of d-glucose monomers in the cellulose fiber is termed the degree of polymerization (DP). New oil-impregnated cellulose typically has 1,000 to 1,100 individual monomers in the cellulose polymer [32]. There is a correlation between the mechanical strength and degree of polymerization, the degradation of degree of polymerization are mainly influenced by moisture, aging duration and aging temperature [29]. Besides that, some experimental studies have established that acids and oxygen are also the major factors effecting the aging of cellulose or in this experiment kraft paper, in high voltage transformers. The tensile strength measurement was conduct with the Dongguan Sinowon universal testing machine as shown in Figure 9.

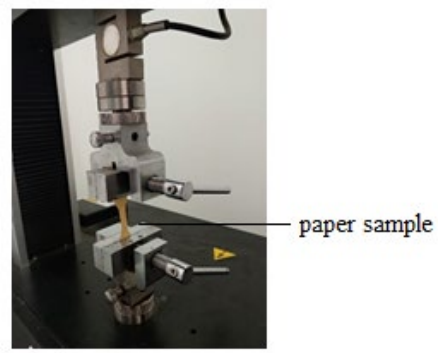


Figure 9. Dongguan Sinowon Universal Testing Machine for tensile strength measurement

The machine is connected with PC and interface board for data collection, processing, and printing test result. The specimen held on the grips, rubber was added to hold the specimen in place. Tensile speed that we used in this experiment is 5 mm/min. All tensile strength values will be recorded on the moment of the breaking momentary. Number of test specimens was 3 specimens for accuracy of data.

3. Results and Analysis

A.1. Visual appearance, Polymerization Degree (DP) and Tensile Strength(TS)

Table 2 shows the visual appearance and thensile strenhth of new paper sample and paper samples aged in mineral and GTL oils at temperature of 120ºC at different aging time. The color of paper slightly became darker in both oils as the aging time became longer. In mineral oil, the tensile strength of the new sample was 47,46 N. This value decreased to 42,2 N after aging time of 336 h and 31,54 N after aging time of 1008 h. The tensile strength reduced drastically to 5,32 after aging time of 1344 h. Similar pattern was observed for sampled aged in GTL oil. The tensile strength reduced to 78% after 336 h, then 51 % after aging time of 1008 hors and then drastucally dropped to 9 % after 1344 hours. The results clearly indicated that thermal aging strongly affected the mechanical properties of the transformer insulating paper sampels. This is why the operating temperature of a power transformer is very important since it will affect the lifetime of a transformer due to the decrease of the tensile strength.

Table 2. Visual appearances and tensile strengths of new and aged paper samples in mineral and GTL oils at temperature of 120ºC at different aging time

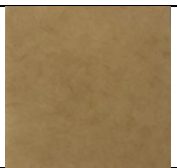
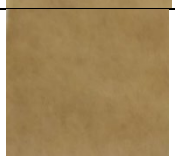
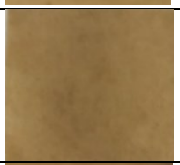
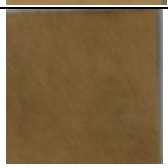
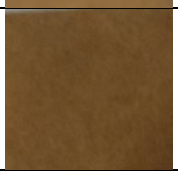
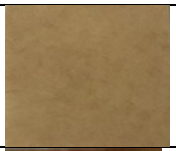
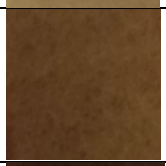
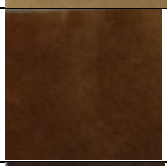
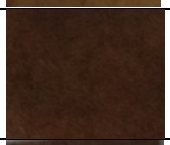
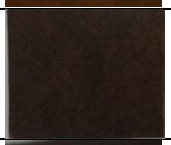
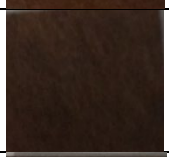
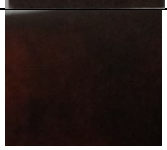
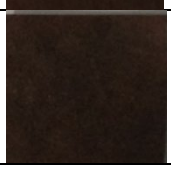
Sample	Paper Appearance	Tensile Strength (N)	Sample	Paper Appearance	Tensile Strength (N)
MO.T0 new		47,46 100	GTL.T0 new		47,46 100
MO.T2.120 336 h		43,2 91	GTL.T2.120 336 h		37,1 78
MO.T4.120 672 h		40,65 86	GTL.T4.120 672 h		35,6 75
MO.T6.120 1008 h		31,54 66	GTL.T6.120 1008 h		24,08 51
MO.T8.120 1344 h		5,32 12	GTL.T8.120 1344 h		4,12 9

Table 3 shows the visual appearance and thensile strength of new paper sample and paper samples aged in mineral and GTL oils at temperature of 150°C at different aging time. From the table it is seen that the visual appearance of paper samples are much darker than those from paper samples aged at 150°C in both mineral and GTL oils. The color became almost black after aging time of 1008 hours.The tensile strength of the samples droped drastically since the aging time of 336 hours from 100 % to less that 2 %. These results showed that aging temperature is a very important factor in the aging process and in the reduction of tensile strength. The change of the tensile strength is stongly correlate with the degree of polymerization of the paper samples.

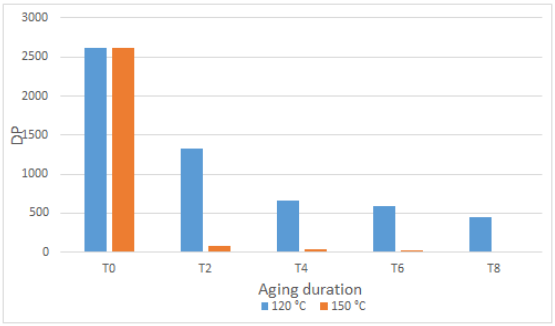
Table 3. Visual appearances and tensile strengths of new and aged paper samples in mineral and GTL oils at temperature of 150°C at different aging time

Sample	Paper Appearance	Tensile Strength (N)	Sample	Paper Appearance	Tensile Strength (N)
MO.T0 new		47,46	GTL.T0 new		47,46
MO.T2.150		0,68	GTL.T2.150		1,84
MO.T4.150		0,5	GTL.T4. 150		0,08
MO.T6.150		0,32	GTL.T6. 150		0,04
MO.T8.150		0,26	GTL.T8.150		0,02

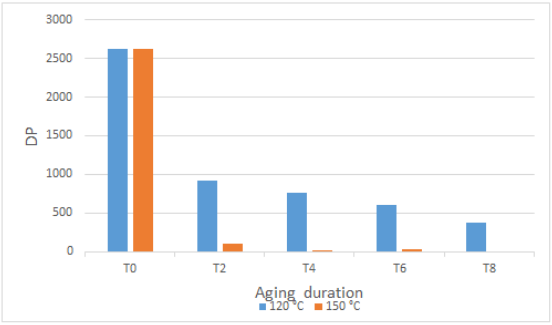
Based on thermal accelerated aging that has been done for 1344 hours, the result of degree of polymerization test are shown in table 4. The DP of new paper is typically greater than 2000. Due to aging the DP reduced due to the scission of polymer to form a smaller number of monomers in the polymer. The transformer lifetime end is reached when the DP of the insulating paper decreases and reaches approximately 250. DP value has decreased faster at 150° C than at 120° C as shown in figure 10. The decrease of DP affects the mechanical strength of the paper samples as indicated by the reduction of their tensile strength. The correlation of DP values and tensile strength are shown in figure 11.

Table 4. DP of paper samples aged in mineral and GTL oils at different temperature and aging time

Sample	DP	Sample	DP
MO.T0	2616	GTL.T0	2616
MO.T2.120	1332	GTL.T2.120	921
MO.T4.120	656	GTL.T4.120	759
MO.T6.120	593	GTL.T6.120	604
MO.T8.120	448	GTL.T8.120	381
MO.T2.150	80	GTL.T2.150	103
MO.T4.150	45	GTL.T4. 150	24
MO.T6.150	28	GTL.T6. 150	36



(a)



(b)

Figure 10. Degree of polymerization (DP)of paper samples aged in (a) mineral oil (b) gas-to-liquid oil

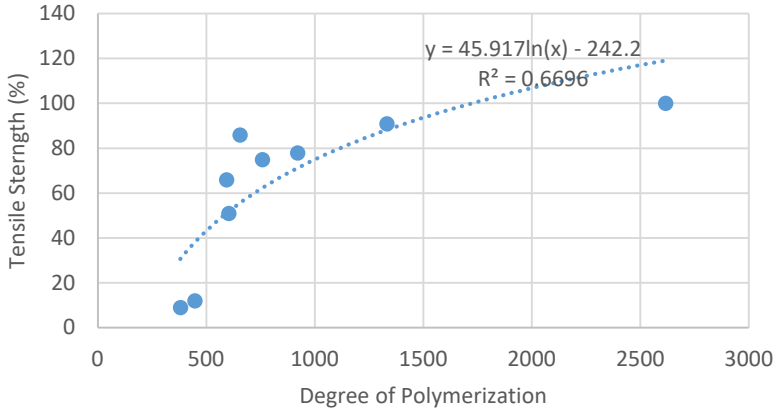


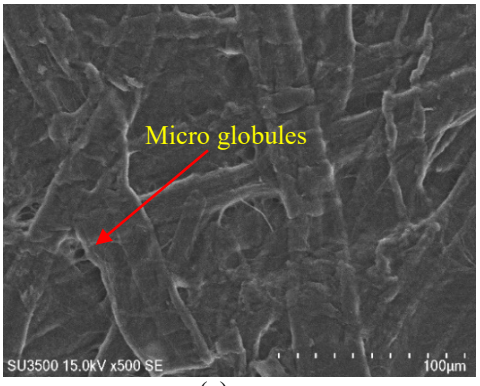
Figure 11. Degree of Polymerization DP and Tensile strength

A.2. Scaninng Electron Microscopy (SEM) Analysis

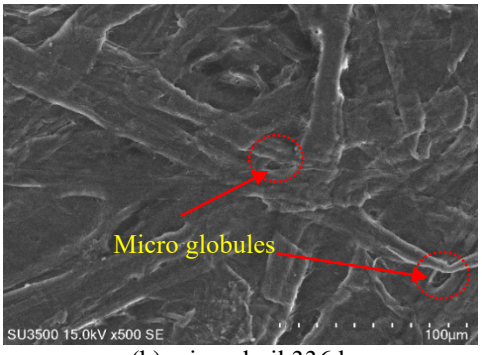
The morphology of paper surface was identified by using SEM while the chemical element of paper was identified by using EDX. The result of energy dispersive x-ray spectroscopy (EDS) shows that C and O elements are the main contents of the insulating paper which is consistent with its chemical formula $C_6H_{10}O_5$. Cellulose as compound of paper is contain a polysaccharide with a linear chain of several hundred to many thousands of β linked D-glucose units. Figure 12 shows the SEM results of new paper sample and paper samples after thermal aging in both mineral oil while figure 13 in GTL at aging temperature of 120°C and 150°C. Table 5 shows the maximum diameter of micro globules detected on the paper surface before and after aging. The table indicates that the size of micro globules increased with aging time in both mineral and GTL oils. Larger increase was observed in higher aging temperature. Even though the behavior performance of aged papers immersed in mineral oil and GTL oil showed the similar behavior, but diameter of micro globules found in mineral oil was slightly bigger than in GTL oil.

Table 5. Maximum diameter of micro globule of paper samples

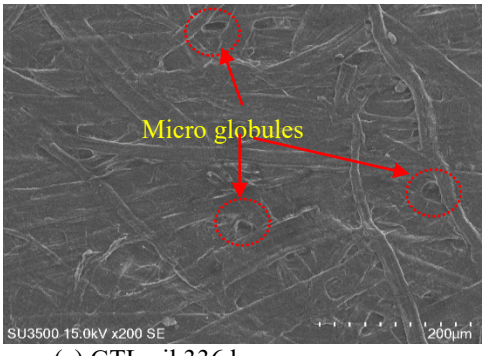
Sample	Maximum Diameter (µm)	Sample	Maximum Diameter (µm)
MO.T0	5	GTL.T0	5
MO.T2.120	7	GTL.T2.120	3,5
MO.T4.120	9	GTL.T4.120	7
MO.T6.120	10	GTL.T6.120	10
MO.T8.120	15	GTL.T8.120	11
MO.T2.150	10	GTL.T2.150	10
MO.T4.150	15	GTL.T4. 150	15
MO.T6.150	20	GTL.T6. 150	16
MO.T8.150	24	GTL.T8.150	20



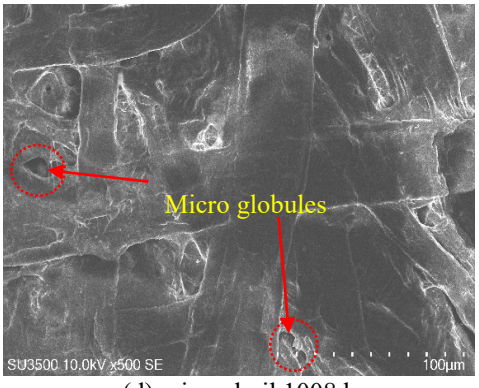
(a) new



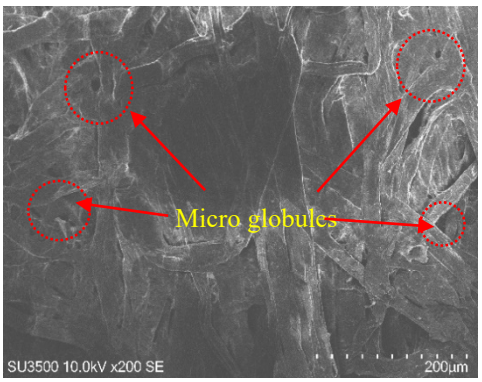
(b) mineral oil 336 h



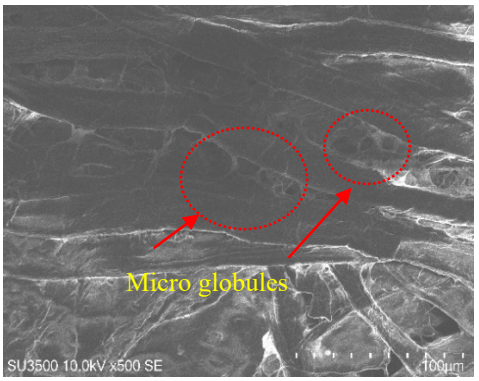
(c) GTL oil 336 h



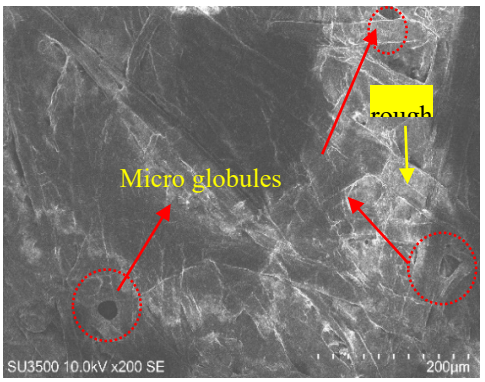
(d) mineral oil 1008 h



(e) GTL oil 1008 h

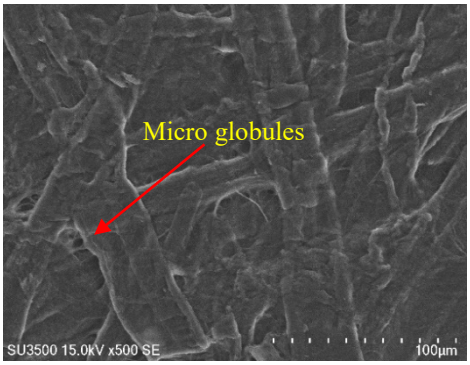


(f) mineral oil 1334 h

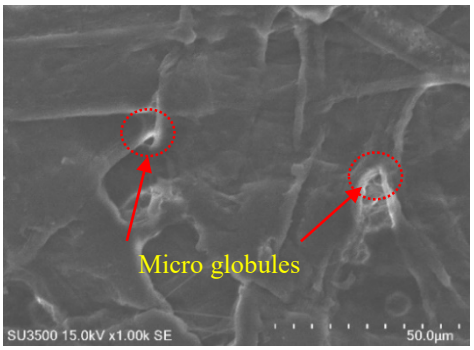


(g) GTL oil 1334 h

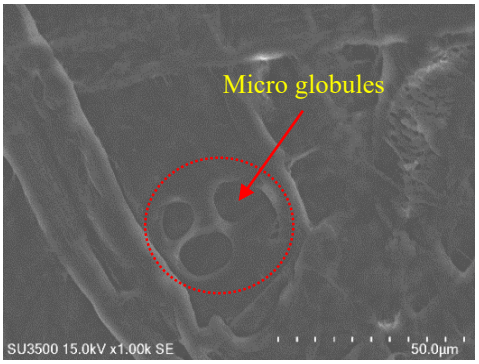
Figure 12. SEM result for paper samples aged in mineral oil at 120ºC and 150ºC (a) T0 (b) MO.T2.120 (c) MO.T2. (d) MO. T6.120 (e) MO.T6.150 (f) MO.T8.120 (g) MO.T8.150



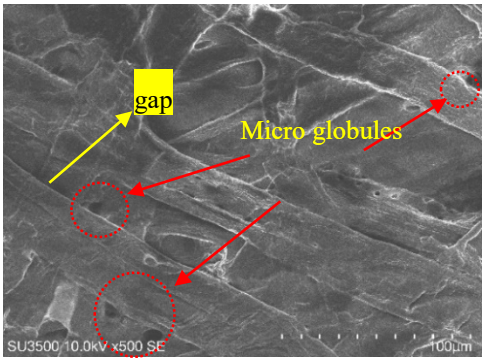
(a) T0



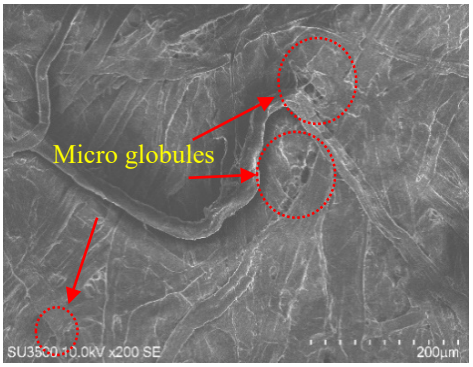
(b) mineral oil 336 h



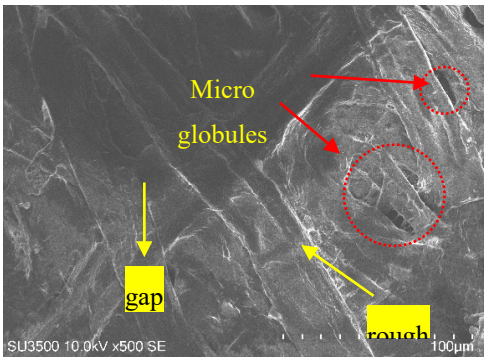
(c) GTL oil 336 h



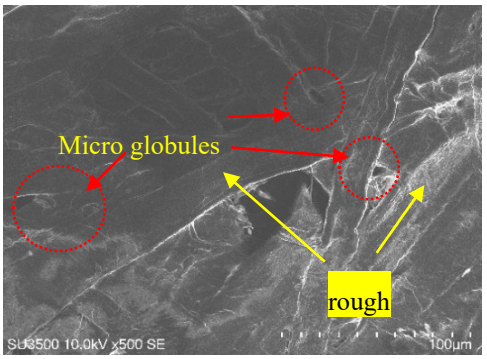
(d) mineral oil 1008 h



(e) GTL oil 1008 h



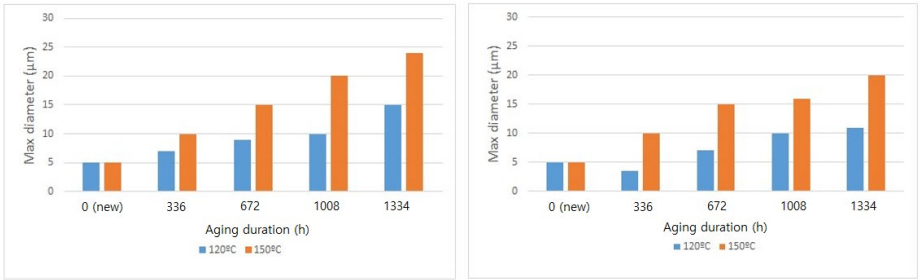
(f) mineral oil 1334 h



(g) GTL oil 1334 h

Figure 13. SEM result for paper samples in GTL oil (a) T0 (b) GTL.T2.120 (c) GTL.T2.150 (d) GTL.T6.120 (e) GTL.T6.150 (f) GTL.T8.120 (g) GTL.T8.150

Figure 14 shows the relation between maximum diameter of micro globule of paper samples and aging duration. It is clearly seen that micro globule diameters increased with time and aging temperature. The increase of micro globule affects the mechanical properties like tensile strength. The breakage of hydrogen bonds between cellulose molecules causes the diameter getting bigger [39].



(a) (b)

Figure 14. Relation of maximum diameter of micro globule of paper samples of and aging duration; (a) Paper samples of mineral oil (b) Paper samples of GTL oil

A.3. X-ray Diffraction (XRD)

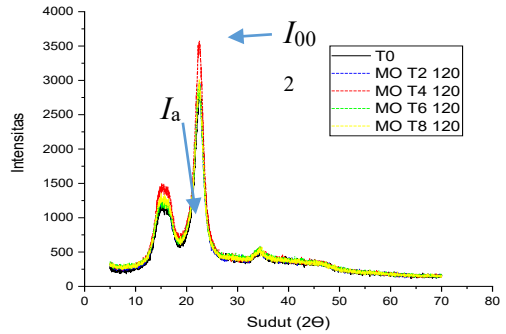
XRD analysis is utilized to estimate crystallinity of paper samples. Figure 15 shows the XRD pattern of the paper samples aged in mineral and GTL oils. There are 2 peaks which is 15° and 22°, which corresponds to crystalline structure. From all samples of the XRD diffraction spectrum, the crystalline peaks are similar. This indicates that the crystal type has not changed during the thermal aging [30]. Even though the peak positions are the same, the crystallinity of paper increased. The insulating paper consists largely of cellulose fibers. There are about 70-80 percent crystalline region and 20-30 percent amorphous region in the cellulose fibers. The amorphous region is firstly destroyed during the aging process. The degraded cellulose molecules then will be no longer constrained by the covalent bond and they will engage in recrystallization. That will cause the increase of the crystallinity index. In this experiment, we used the method suggested by Segal to calculate the crystallinity index[30]. The crystallinity index was

$$C_r = \frac{I_{002} - I_{am}}{I_{002}} \; x \; 100 \qquad (2)$$

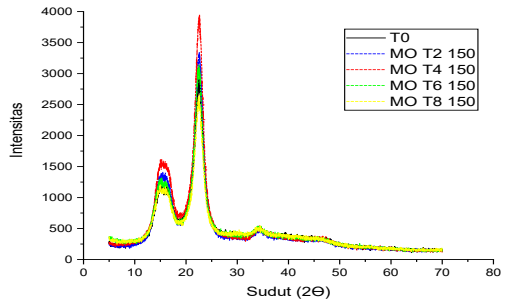
Table 6. Relative Crystallinity and Amorphous of paper samples

Sample	Crystallinity (%)	Amorphous (%)	Sample	Crystallinity (%)	Amorphous (%)
MO.T0	80,5	19,5	GTL.T0	80,5	19,5
MO.T2.120	79,1	20,9	GTL.T2.120	78,3	21,7
MO.T4.120	81,4	18,6	GTL.T4.120	78,7	21,3
MO.T6.120	79,1	20,9	GTL.T6.120	80,8	19,2
MO.T8.120	79,6	20,4	GTL.T8.120	80,2	19,8
MO.T2.150	82,3	17,7	GTL.T2.150	82,2	17,8
MO.T4.150	83,4	16,6	GTL.T4.150	82,2	17,8
MO.T6.150	81,7	18,3	GTL.T6.150	82,9	17,1
MO.T8.150	77,8	22,2	GTL.T8.150	80,4	19,6

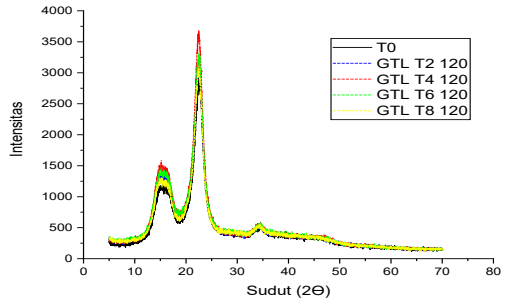
C_r is defined as the crystallinity index, I_{002} is the diffraction intensity at the 002 peak and I_{am} is the diffraction intensity at about 18°. Table 6 shows the crystallinity and amorphous for all paper samples. The crystallinity index increases with the increase of aging time in general as shown in Figure 16. This increase of the percentage of crystallinity (decrease of the percentage of amorphous) is due to the decomposition of some hemicellulose. This is discussed in In the 120°C samples of mineral oil and GTL oil, the relative crystallinity declines sometimes but not much, and in the 8 weeks the relative crystallinity decrease slightly [31]. In the 150°C samples of both paper from mineral oil and GTL oil shows the same behavior, the crystallinity index in the 2 weeks, 4 weeks and 6 weeks increase and in the 8 weeks the crystallinity index decrease lower than the initial condition sample. Both paper of mineral oil and kraft paper shows the same behavior of performance, however the crystallinity index is slightly higher in the samples of paper immersed in GTL oil especially in the 150°C samples.



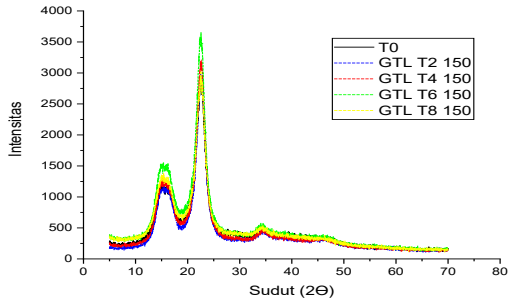
(a)



(b)

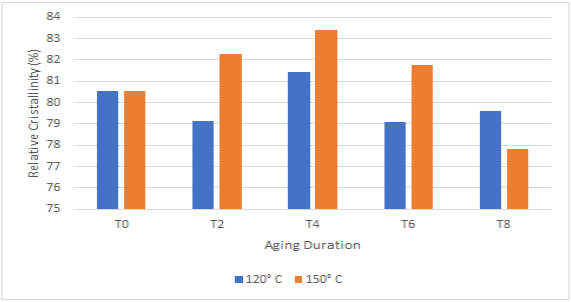


(c)

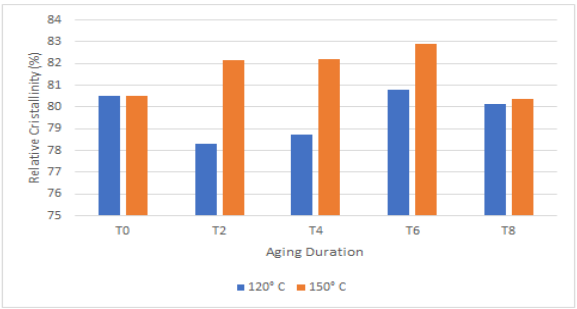


(d)

Figure 15. XRD Spectrum for samples (a) mineral oil with aging 120° C (b) mineral oil with aging 150° C (c) GTL oil with aging 120° C (d) GTL oil with aging 150° C



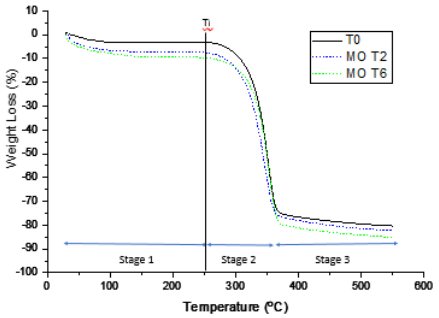
(a)



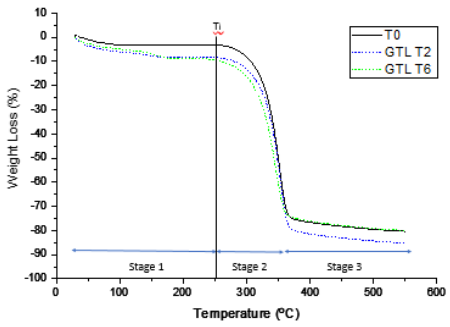
(b)

Figure 16. Crystallinity of kraft paper (a) mineral oil, (b) gas-to-liquid oil

A.4. Thermogravimetric (TG) analysis



(a)



(b)

Figure 17. TG Curves for samples at aging temperature of 120° C ini
(a) mineral oil (d) GTL oil

Figure 17 shows the result of TGA of kraft paper from mineral oil and GTL oil. The horizontal axis is the temperature (deg.) and the vertical axis is weight loss. The figure shows that aged paper samples have larger weight losses at low temperature.

Table 7. Initial weight losses and initial temperature for the second stage in mineral and GTL oils at aging temperature of 120ºC.

Aging time (h)	Mineral oil		GTL Oil	
	Initial stage weight loss W_i(%)	Initial temperature for the second stage T_i (ºC)	Initial stage weight loss W_i (%)	Initial temperature for the second stage T_i (ºC)
0 (new sample)	2.8	250	2.8	250
336	7.2	241	6.8	248
1008	9.4	240	8.3	228

Thermogravimetric method is used to identify the residue, mositure and to estimate the oxidation, vaporization and thermal stability of a polimer [32]. TG curve shows the temporal change weight loss percentage as a function of temperature. Based on TGA and DTG curve, generally kraft paper degradation consist of three stages. The first stage occurs between 30-250° C. Water loss is observed around 100° C, and the first thermal degradation begin with water vaporization. In this stages the degradation of mainly due to hemicellulose which typically decomposed at around 150-350° C at lower starting temperature from those of cellulose which starts at around 250ºC. The second stage occurs between around 220-370° C which the main degradation of cellulose occurs. The final stage or the third stage occurs between around 370-550° C mainly due to lignin decomposition. Lignin degradation occurs between 250° C and 500° C. For comparison among samples it is important to see the mass losses during 1st stage and the initial temperature of the second stage. The values for mineral oil and GTL oil are shown in table 7.

Table 7 Shows the initial stage weight losses W_i (%) and initial decomposition temperature at the second stage (ºC). Wi indicates the weakness of paper at the early degradation with temperature of less than 250ºC. In an actual transformer it reflects the situation of short term local overheating for example due to electrical fault. The larger the Wi is the weaker the paper. For paper samples in mineral oil, the W_i increased from 2.8 % at the beginning to 7.2 % for sample aged for 336 hours and then increased further to 9.4 % for sample aged for 1008 hours. Similar pattern was observed for paper samples in GTL oil. The W_i increased from 2.8 % at the beginning to 6.8% for sample aged for 336 hours and then increased further to 8.3 % for sample aged for 1008 hours. In this initial losses point of view paper in GTL oil is slightly better than in mineral oil. The Ti values for paper samples in mineral oil decreased from 250ºC for new sample to 241ºC after 336 hours aging and 240ºC after 1008 hours aging. While for paper samples in GTL oil, the Ti values dropped from 250ºC at the beginning to 228ºC after aging period of 1008 hours.

4. Conclusions

Ageing performance of kraft paper immersed in GTL oil was compared with kraft paper immersed in mineral oil through an accelerated laboratory ageing experiment. SEM and EDS measurement shows that the behavior performance of paper immersed in mineral oil and GTL oil shows the same, but the bigger diameter of micro globules found in mineral oil slightly bigger than the mineral oil paper. From XRD measurement, we found there are 2 peaks which is 15° and 22°. Though the crystallinity index declines sometimes especially when the aging time is relatively long, it is still much larger than that of new insulating papers. Thermogravimetric analysis indicated that there are three stages thermal degradation. The first stage occurs between 30-250° C. Water loss is observed around 100 C, and the first thermal degradation begin with

water vaporization. The degradation of hemicellulose at around 150-350° C. The second stage occurs between around 250-370° C the main degradation of cellulose occurs. The final stage or the third stage occurs between around 370-550° C. Higher mass loss was found in temperature 150°C paper samples than samples in temperature 120°C due to condition of the paper samples in the 150°C were severely damaged. The result of DP and TS tests show that both DP and TS decreased with the aging time and temperature of aging both in mineral oil and GTL. Both papers in mineral oil and GTL show the same behavior, even though DP and tensile strength of mineral oil paper slightly better than in GTL Nevertheless overall result from the accelerated ageing experiment suggest that ageing performances Kraft paper in GTL oil and mineral oil are comparable

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