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Comparative Study of DC Breakdown and Space Charge Characteristics of Insulation Paper Impregnated with Natural Ester and Mineral Oil

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Abstract – Natural ester is a suitable substitute for mineral oil and has been widely used in AC transformer in many countries. In order to further application of natural ester in direct current (DC) equipment, it is needed to investigate its long term insulation property under DC condition. In this paper, a thermal ageing experiment was conducted for both mineral oil-paper and natural ester-paper insulation. The DC breakdown and space charge characteristics of insulation paper impregnated with natural ester and mineral oil was compared. Results show that the resistivity of the paper immersed in natural ester and mineral oil both increase as the ageing goes on. While insulation paper impregnated with natural ester has higher resistivity and DC breakdown voltage than the paper impregnated with mineral oil. The DC breakdown voltage for the oil impregnated insulation paper being DC pre-stressing is higher than that without pre-stressing. The average DC breakdown field strength difference between the test with pre-stressing and without pre-stressing clearly shows that there is an apparent enhancement effect for the homo-charge injection on the DC breakdown.

Keywords: Natural ester, Mineral oil, Oil impregnated insulation paper, DC breakdown, Space charge, Thermal ageing.

1. Introduction

Mineral oil is widely used in the transformers as insulation oil because of its preferable low price, good insulation quality and a relatively low condensation point [1]. However, with the consideration of the fact that it almost does not degrade which can cost the contamination of soil, water and our living environment [2, 3]. While on the other hand that fossil fuel would be a scarce resource for the 21st century, it became urgent for the researchers to find out its alternative. Natural ester drew the researchers' attention since the 1990s. It has the following advantages as insulation fluid. Firstly, natural ester has higher flash and fire point than mineral oil making it better suited for transformers and essential in environments where fire prevention is of high priority [4]. Secondly, natural ester is more biodegradable than mineral oil [5]. Thirdly, natural ester is nontoxic while mineral oil, on the other hand, contains light naphthenic petroleum distillates. The International Agency for Research on Cancer regards certain distillates as carcinogenic, however Nynas believes that very little of these distillates remain after refining [6]. Finally, natural ester is a renewable resource.

Natural ester has been used as the insulation fluid for transformers from the beginning of the century. According to literature [4], there have been developments in using esters in several large power transformers. A Brazilian company built a 138 kV 30 MVA mobile transformer and two 138 kV 40MVA substation transformers using natural ester [4]. Another Brazilian company revised its specification to use natural ester in all transformers and reactors rated up to 138kV [4]. In UK, the EDF and AREVA T&D companies have developed a natural ester filled 132 kV 90 MVA transformer [4]. Siemens applied the global first 420 kV vegetable-oil transformer in Bruchsal-Kändelweg substation successfully [7].

The working temperature for the DC equipment, such as converter transformer is relatively higher than that of the AC transformer. In addition to its excellent insulation performance, natural ester also has better thermal stability than mineral oil [8], which makes the natural ester may be a more suitable option for the DC equipment. While the formation of space charge in oil/paper insulation system under DC condition can result in distortion of the electric field distribution, i.e. enhanced electric field in one region and reduced electric field in the other. This may lead to material degradation in the high electric field region and affect system reliability [9]. In order to ensure the long term safety of the equipment using natural ester as insulating fluid under DC operation condition, it is necessary to understand its breakdown and space charge

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characteristics of the insulation paper impregnated in natural ester.

In this paper, a thermal ageing experiment had been conducted on the natural ester impregnated paper insulation. Samples were collected at different days and a series of analysis were performed. For the comparison purpose, there was a reference group using mineral oil instead. For these two groups of sample, the moisture, DC resistivity, DC breakdown voltage and the space charge characteristics was compared. Moreover, the relationship between samples' DC breakdown and space charge characteristic was analyzed.

2. Experiments

2.1 Thermal ageing experiment

The insulation paper was 0.12 mm thick and was cut into 4×4 cm square, which was provided by the NARI Borui transformer factory. The natural ester was originated from rapeseed oil. The K25 mineral oil provided by Chuanrun Lubricating Oil Co. Ltd. Was used. The main parameter of the oil is shown in Table 1. The main parameter of the insulation paper (without oil impregnation) is shown in Table 2.

The insulation paper was immersed into the natural ester and mineral oil using 500 mL ground-glass stopper flask. There are 400ml oil and 40g oil impregnated insulation paper in each glass bottle. A piece of 3 cm ×1 cm copper with the thickness 3 mm was added into each flask. Then the samples were put into the vacuum drying oven under 60℃ with vacuum air condition for 48 hours to form the oil-paper insulation system. After the vacuum impregnation, the samples were placed into the thermal ageing oven for the thermal ageing experiment under 130℃. According to literature [10], it is found out that the DP of the mineral oil impregnated paper dropped below 200 after 50 days at 130 ℃ thermal ageing, which means that the mineral oil impregnated paper has reached the terminal state of its life. Therefore, the thermal ageing time in this experiment was chosen to be 50 days, and the samples were collected on 0, 33 and 50 days standing for the new sample, medium-term aging sample and the end aging sample, respectively.

Table 1. Parameters for mineral oil & natural ester

Property	Mineral oil	Natural ester
Density at 20°C (kg/m^3)	0.89	0.91
Kinematic viscosity 40°C (mm^2/s)	9.55	38.30
Relative permittivity(20°C, 50Hz)	2.19	3.14
Moisture content (ppm)	17	110

Table 2. Parameters for insulation paper

Property	Number
Thickness (μm)	120
Density(g/cm^3)	0.95
AC breakdown strength(kV/mm)	43
Resistivity(10^{13} Ω.m)	1.32
Permittivity (25°C)	3

2.2 Moisture content and resistivity test

Moisture content of the oil impregnated insulation paper was determined by the Karl Fischer coulometric titrator method according to IEC 60814. The setup used was Metrohm moisture meter 851 and oven 885. The moisture content for each sample was measured twice, and their average values was recorded. The DC resistivity was tested according to ASTM D257-07 with three electrodes system under 25 ℃. The Three electrodes system diagram for DC resistivity is shown in Fig. 1.

2.3 DC breakdown voltage test

In order to ensure natural ester's long term safety under DC conditions, the DC breakdown voltage test was conducted for both mineral oil-paper insulation and natural ester-paper insulation. The electrodes consisted two brass cylinders with radius to be 15 mm and the height to be 25 mm. The edges of the electrodes were rounded. The schematic diagram for DC breakdown test is shown in Fig. 2. In order to verify the influence of space charge on the breakdown voltage, samples were applied DC voltage for a period of time before increasing the voltage to breakdown.

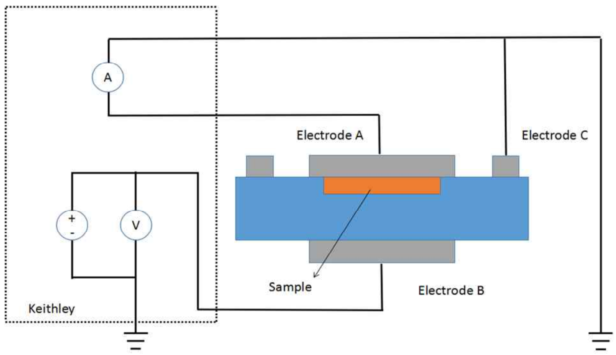


Fig. 1. Three electrodes system diagram for DC resistivity

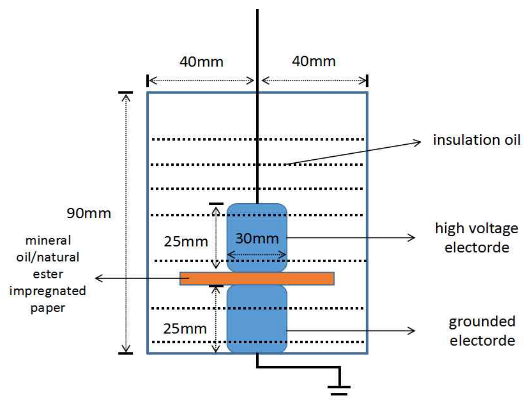


Fig. 2. Schematic diagram for the DC breakdown test

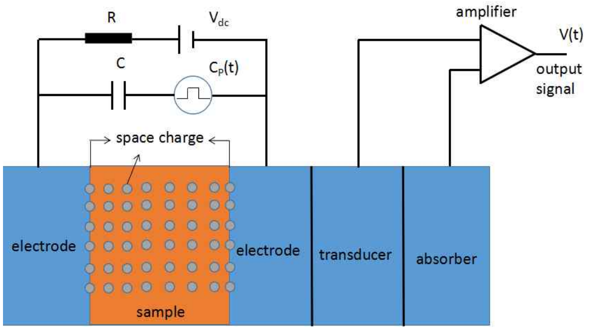


Fig. 3. Schematic diagram for PEA measurement

The DC voltage pre-stressing was 25 kV/mm for 5 minutes. After 5 minutes, the DC voltage was increased at 0.2 kV/s until the sample breakdown. The breakdown voltage was recorded. In addition, the DC breakdown of the samples without being DC voltage pre-stressing was also tested. Both experiment were carried out at 25℃.

2.4 Space charge characteristic test

The pulsed electro-acoustic (PEA) method is widely accepted by researchers to measure the space charge characteristics in solid dielectrics around the world. The principle of the PEA method can be found in many literatures [11-14]. In general, it consists of detecting acoustic waves generated by internal charges under the Coulomb force of a pulsed electric field. The waves are detected by an external piezoelectric transducer, which converts the acoustic signal to an electrical signal. Then the internal charge density is deduced by signal processing and mathematic treatment. The block diagram for PEA method is shown in Fig. 3 [14].

The PEA platform uses an aluminum plate as its bottom electrode and the top electrode is semiconducting polymer film. The PEA platform in this experiment has a pulse amplitude ranging from 0 to 1 kV while the pulse width can be adjusted from 0 to 10 ns with the frequency of 50 Hz. The space charge sensitivity is 0.2 C/m^3. The oil impregnated insulation paper sample was placed into the PEA platform with the pulse amplitude to be 500V and the width to be 5 ns. In the space charge characteristic test, samples were stressed at two different dc voltages (25 kV/mm and 40 kV/mm) at 25℃. A suitable 3.3 kV/mm voltage was selected for the natural ester-paper sample as its calibration signal. And a 2.5 kV/mm voltage was chosen as the calibration signal for the mineral oil-paper group. The applied DC voltage was negative (semiconducting polymer as the cathode while the aluminum plate as the anode).

3. Result and Analysis

3.1 Moisture content and resistivity

The moisture content of samples with different ageing condition is shown in Table 3. For the mineral oil-paper insulation group, the moisture content of the sample ageing 0 day was 1.95%, then the moisture content raised to 2.11% for the sample ageing 33 days. However, the moisture for the sample ageing 50 days dropped to 1.79%. According to literature [8] and [15], the moisture content of the oil-paper insulation increases because of its deterioration generating moisture. However, when the oil and the air inside the bottles being relatively drier than the moisture condition of paper, there is always a migration of moisture from the paper to the oil and then to the air. Thus the moisture content of the oil impregnated insulation paper showed a decline.

Table 3. The moisture content of samples with different aging days

Ageing	Mineral Oil-Paper	Natural Ester-Paper
0 day	1.95%	1.97%
33days	2.11%	2.15%
50 days	1.79%	2.28%

Table 4. The DC resistivity of samples with different aging days

Ageing	Mineral Oil-Paper	Natural Ester-Paper
0 day	$1.037\times10^{14}\Omega$/cm	$3.39\times10^{14}\Omega$/cm
33days	$2.754\times10^{14}\Omega$/cm	$4.51\times10^{14}\Omega$/cm
50 days	$3.289\times10^{14}\Omega$/cm	$4.46\times10^{14}\Omega$/cm

For the natural ester-paper insulation group, the moisture content of the sample increased from 1.97% to 2.28%. According to literature [8] and [15], the natural ester-paper insulation deterioration will also generate moisture during thermal ageing process. Due to the higher absolute moisture content of new natural ester and the great moisture affinity of kraft paper, the paper may absorb moisture from natural ester in order to keep moisture equilibrium. This may be the reason that there was a steady increase for the moisture content of the paper in natural ester-paper insulation system in the process of 50 days ageing.

The DC resistivity for both types of oil impregnated insulation paper aged different days is shown in Table 4. It can be observed that for mineral oil-paper insulation group, the resistivity was 1.037×10^{14} Ω/cm for the new sample, then for samples aged 33 and 50 days, the resistivity was $2.754\times10^{14}\Omega$/cm and $3.289\times10^{14}\Omega$/cm respectively. It can be observed that with the increase of ageing time, the resistivity for mineral oil-paper insulation group increases. The resistivity of the sample at 50 days is almost 3 times as that of the new sample.

The resistivity of new natural ester-paper insulation was $3.39\times10^{14}\Omega$/cm, the resistivity of natural ester-paper insulation aged for 33 days and 50 days was 4.51×10^{14} Ω/cm and $4.46\times10^{14}\Omega$/cm respectively. The resistivity's difference of natural ester-paper insulation ageing 33 days and 50 days is within 1%. It is noticeable that the DC resistivity of new natural ester-paper insulation is even slightly higher than the DC resistivity of mineral oil-paper insulation in the process of 50 days ageing.

3.2 DC Breakdown voltage

The DC breakdown field strength for both types of samples at different ageing days without pre-stressing is shown in Fig. 4. And the average DC breakdown field strength is shown in Fig. 5. From Fig. 4 and Fig. 5, it can be observed that the DC breakdown field strength for the natural ester-paper insulation is slightly higher than the mineral oil-paper insulation without pre-stressing. The DC breakdown for both types of samples after being 5 minute pre-stressing is shown in Fig. 6 and Fig. 7. It can be observed that the breakdown field strength for natural ester-paper insulation is significantly higher than that of the mineral oil-paper insulation after being pre-stressing.

Fig. 8 is the average DC breakdown field strength difference between the test with pre-stressing and without pre-stressing. Differences *D* was calculated based on Eq. (1). Where u_s stands for breakdown field strength with pre-stressing, while u_0 stands for the breakdown field strength without pre-stressing. For the new mineral oil-paper insulation, the pre-stressing process increases the breakdown field strength 2.6%, while for new natural ester-paper insulation, this process increases the breakdown field strength about 5.8%. For mineral oil-paper insulation aged for 33 and 50 days, the pre-stressing process increases its breakdown field strength 8.3% and 15.7%, respectively. For natural ester-paper insulation aged for 33 and 50 days, it increases the breakdown field strength 7.6% and 19.5%, respectively. From Fig. 8, it can be observed that the DC pre-stressing process increases the breakdown field strength for both types of samples compared with the samples without pre-stressing. For both types of samples, with the increase of thermal ageing time, the pre-stressing process would have greater influence on the DC breakdown field strength.

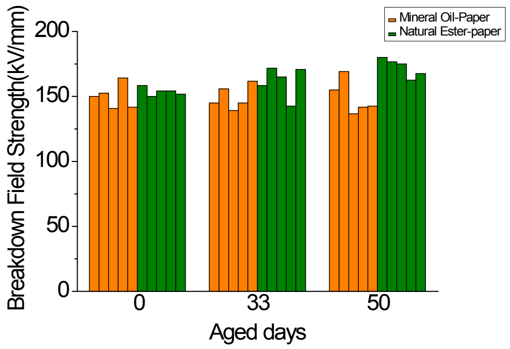


Fig. 4. DC breakdown field strength for both types of samples without pre-stressing

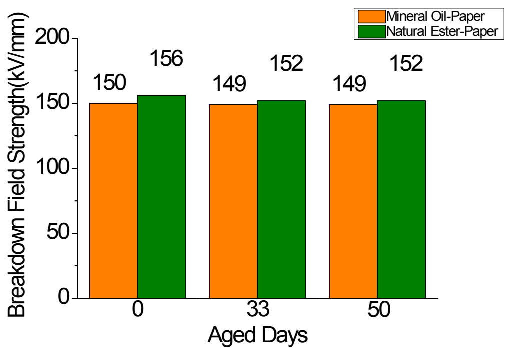


Fig. 5. Average DC breakdown field strength for both types of samples without pre-stressing

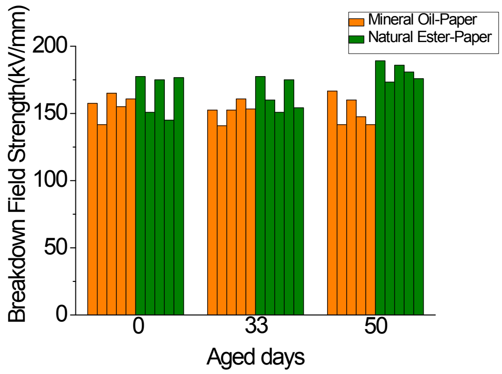


Fig. 6. DC breakdown field strength for both types of samples with 5 minute pre-stressing

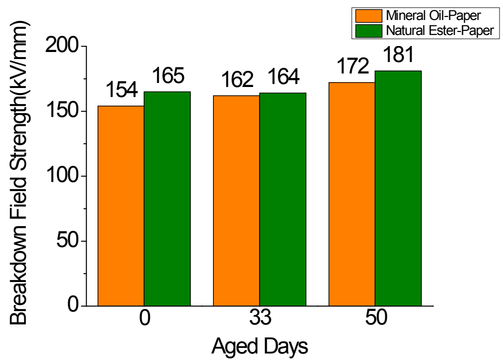


Fig. 7. Average DC breakdown field strength for both types of samples with 5 minute pre-stressing

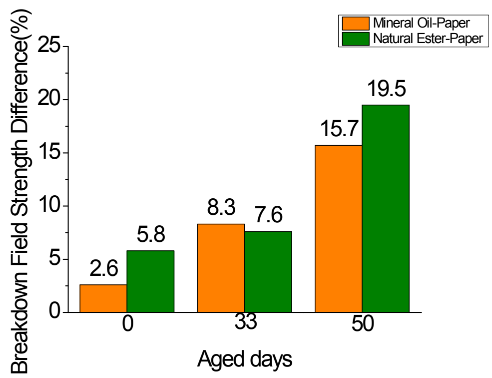


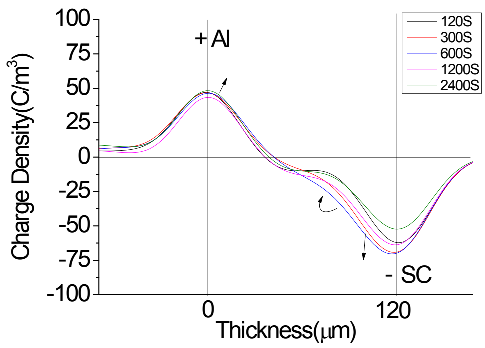
Fig. 8. Average DC breakdown difference between test with pre-stressing and test without pre-stressing

$$D=\frac{u_s-u_0}{u_0}*100\% \quad (1)$$

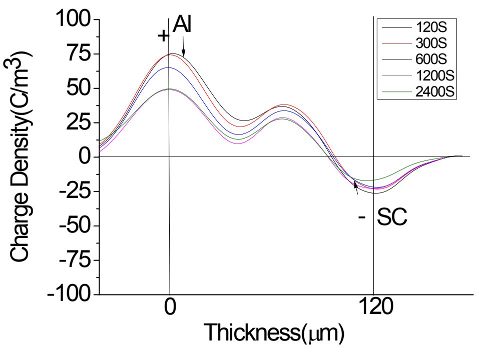
3.3 Space charge characteristics

The space charge characteristics for mineral oil-paper insulation and natural ester-paper insulation under 25 kV/mm is shown in Fig. 9 and Fig. 10. For the mineral oil-paper insulation, the space charge injection is homo-charge injection. The new mineral oil-paper insulation's charge density on both positive and negative sides increased with the increase of time. For new mineral oil-paper insulation, the charge that moved to the middle of the sample was negative, while for the aged mineral oil-paper sample, the charge that moved to the middle was positive. What is more, most of the positive charge moved from the interface between anode and oil-paper insulation to the middle part of the sample and some of the positive charge got neutralized with the negative charge in the middle of the sample. This caused the charge density in the middle part increased at first and then decreased as the time passed by.

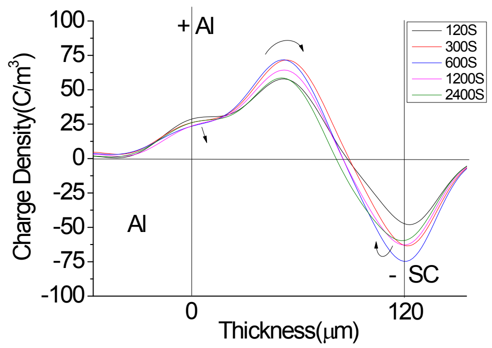
It can be seen from Fig. 10 that the space charge injection for natural ester-paper insulation is also homo-charge injection. For both positive and negative electrode sides, the space charge density in the electrode-paper interface increased first, then the charge density decreased as the time passed by. For the new natural ester-paper insulation,



(a) mineral oil-paper aged for 0 day

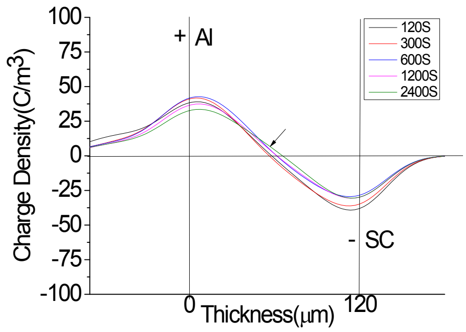


(b) mineral oil-paper aged for 33 days

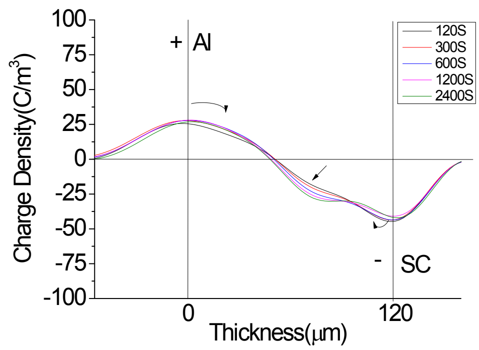


(c) mineral oil-paper aged for 50 days

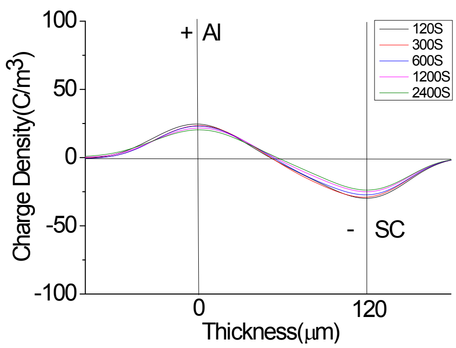
Fig. 9. Mineral oil-paper's space charge characteristics under 25kV/mm



(a) natural ester-paper aged for 0 day



(b) natural ester-paper aged for 33 days



(c) natural ester-paper aged for 50 days

Fig. 10. Natural ester-paper's space charge characteristics under 25kV/mm

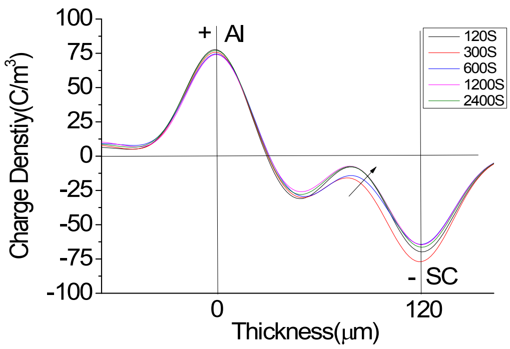
the space charge injection in the middle of the sample is not apparent. But for the sample aged for 50 days, the space charge moved to the middle of the sample more apparently. It is also noticeable that the charge density near electrodes was around 45 C/m^3 for new natural ester-paper insulation.

Then the charge density near electrodes for natural ester-paper insulation aged for 33 days dropped to about 25 C/m^3, and the charge density near electrodes for natural ester-paper aged for 50 days was also about 25 C/m^3.

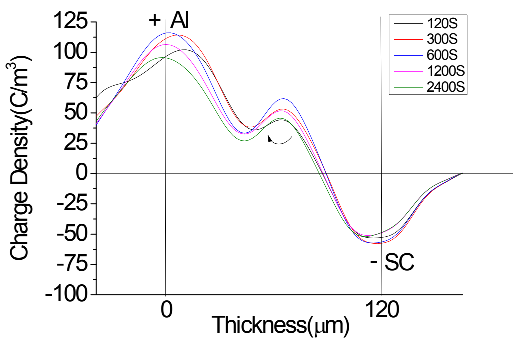
For the mineral oil-paper insulation group under 40 kV/mm shown in Fig. 11 the charge density on the electrode sides increases significantly. For mineral oil-paper sample aged for 50 days, the space charge injection was still very apparent. However, the charge density in the middle part of the sample did not increase significantly with the increase of applied voltage, which is around 75C/m^3. In Fig. 12, for natural ester-paper insulation under 40kV/mm, the charge injection became more significant.

For two samples under 25 kV/mm and 40 kV/mm, as the ageing days increase, the charge injection in the middle of the sample became more apparent. This phenomenon can be explained by that with the proceeding of thermal ageing, the micro structure of the oil-paper insulation was undermined, making it easier for the space charge injection, especially the positive charge injection for the aged mineral oil-paper insulation.

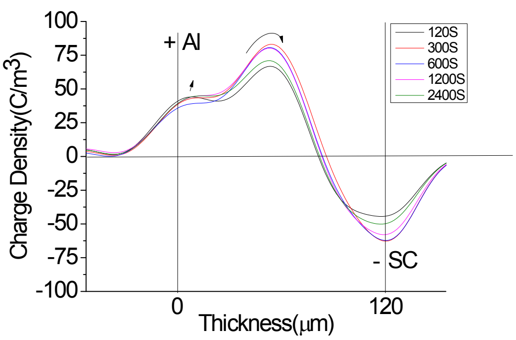
The microscopic structure of the mineral oil impregnated



(a) mineral oil-paper aged for 0 day

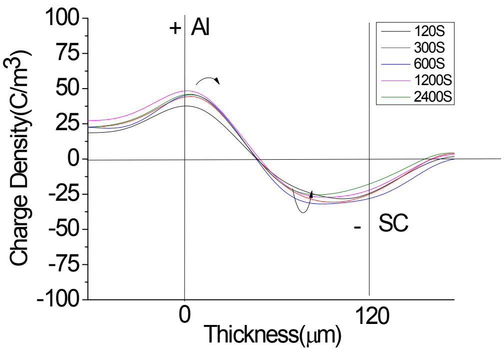


(b) mineral oil-paper aged for 33 days

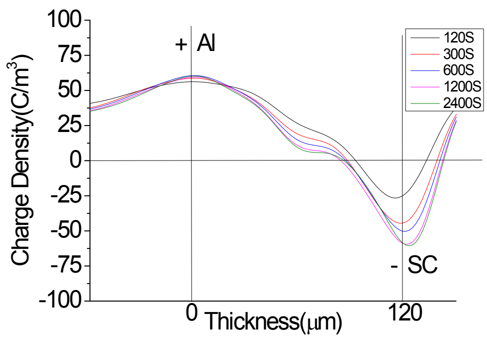


(c) mineral oil-paper aged for 50 days

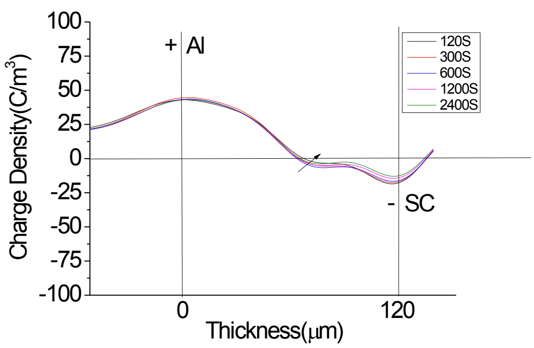
Fig. 11. Mineral oil-paper's space charge characteristics under 40kV/mm



(a) natural ester-paper aged for 0 day



(b) natural ester-paper aged for 33 days



(c) natural ester-paper aged for 50 days

Fig. 12. Natural ester-paper's space charge characteristics under 40kV/mm

insulation paper and the natural ester impregnated insulation paper aged in the oil-paper insulation samples for 0 and 50 days were measured using the scanning electron microscopy (SEM, JSM-7800F, JEOL, Japan). The results are shown below in Fig. 13. From the SEM results of the oil impregnated insulation paper at 500×, it could be seen that the after thermally aged for 50 days, the thermal ageing seriously deteriorated the microstructure of the mineral oil impregnated insulation paper. The natural ester impregnated insulation paper has better microscopic structure property than the mineral oil impregnated insulation paper in the process of the thermal ageing due to its slower ageing rate [18].

3.4 Total charge

The total charge amount of charges Q trapped in the oil impregnated insulation paper in the process of voltage applied process was calculated based on Eq. (2). In this equation, S stands for the area of the sample, while l stands for the thickness of the sample, $q(x)$ means the charge density at positon x, $0 \le x \le l$.

$$Q = S\int_0^l q(x)dx \tag{2}$$

From Fig. 14(a), it can be observed that under 25kV/mm, the total charge amount for mineral oil-paper increases at first, then from 600 seconds, the total charge amount has a slightly drop then almost unchanged. Before 210s, the total charge amount for the aged samples is higher than the new sample. While after that, the sample aged for 30days shows decreases in the total charge amount compared with the new sample. As for the natural ester paper insulation presented in Fig. 14(b), it can be observed that the total charge amount of all three samples changed slightly with the increase of time. With the further degree of thermal ageing, the total charge amount of samples decreases. The total charge amount for natural ester paper insulation aged 0 day at 2400s is about 0.8C, while the total charge amount for natural ester paper ageing 33 days and 50 days is 0.48C and 0.52C, respectively. From Fig. 14, it presents that the charges trapped in the insulation paper impregnated with natural ester is less than the insulation paper impregnated with mineral oil under 25kVmm after being the same ageing time.

Compared to the results under 25 kV/mm. the steady total charge amount of charges in the aged samples increases under 40kV/mm, as shown in Fig. 15. For the samples aged 0 day and 50 days, the steady total charge amount of charges in the mineral oil impregnated insulation paper is about 2~3 times higher than that of the natural ester impregnated insulation paper. While for the sample aged 30 days, the natural ester impregnated insulation paper has

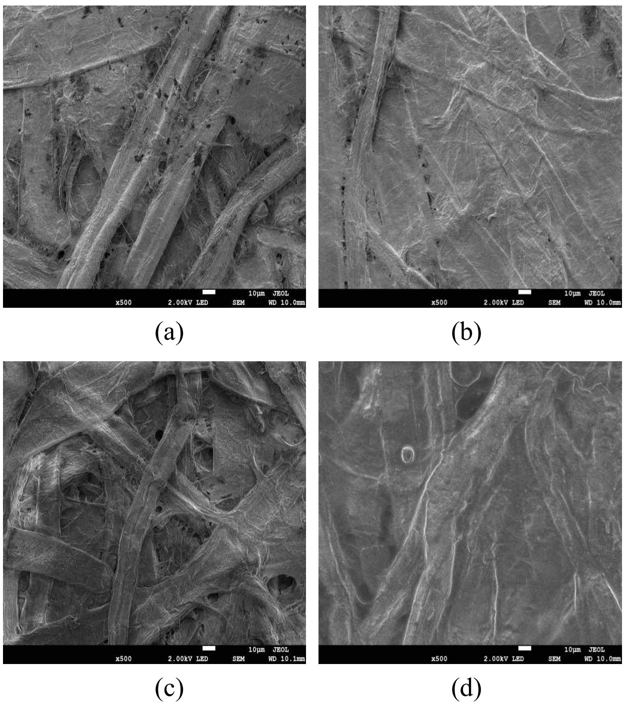
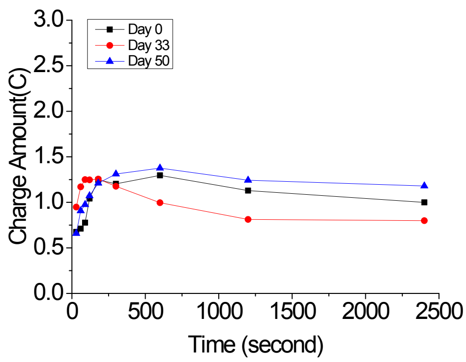
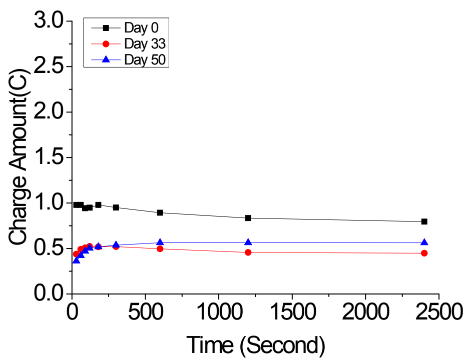


Fig. 13. SEM images of the oil impregnated insulation paper at 500×. (a) new mineral oil impregnated insulation paper. (b) new natural ester impregnated insulation paper. (c) mineral oil impregnated insulation paper aged for 50 days. (d) natural ester impregnated insulation paper aged for 50 days

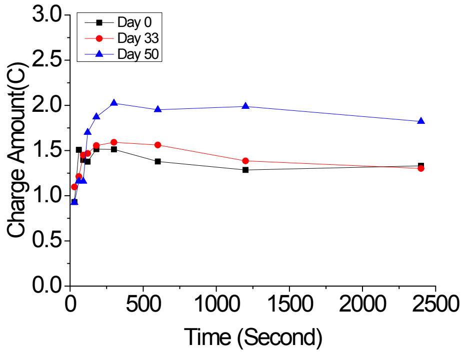


(a) total charge amount-mineral oil-paper

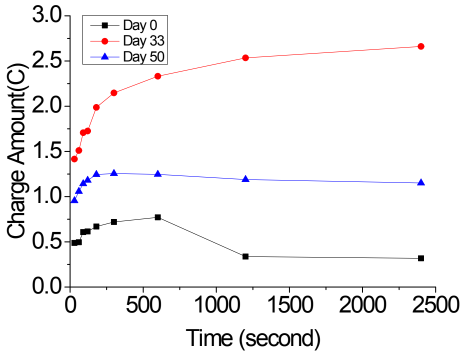


(b) total charge amount-natural ester-paper

Fig. 14. Total charge amount with different ageing days under 25kV/mm



(a) total charge amount-mineral oil-paper



(b) total charge amount-natural ester-paper

Fig. 15. Total charge amount with different ageing days under 40kV/mm

higher total charge amount of charges than the mineral oil impregnated insulation paper. The ageing is a complex process, which may lead to the difference above.

3.5 DC breakdown results discussions

From Fig. 4-5 it can be seen that the DC breakdown values for the aged samples without DC pre-stressing is nearly no change compared with the new sample. This results is consistent with the results shown in [18] that the breakdown voltage of oil-paper samples without pre-stressing remains almost constant with thermal degradation.

However, Fig. 6-7 presents that the DC breakdown values for the aged samples being DC pre-stressing is a little higher than the new sample. In addition, the DC breakdown values for all the samples being DC pre-stressing is higher than the samples without pre-stressing, especially for the aged samples. Overall, the natural ester impregnated insulation paper has higher DC breakdown values than the mineral oil impregnated insulation paper, especially for the natural ester impregnated insulation paper being DC pre-stressing. The DC breakdown values for the oil-paper insulation does not decreases with the aggravation of aging degree.

The increasing DC breakdown values for the samples being DC pre-stressing is mainly related to the homo charges injection, which leads to the field strength near the electrode being weakened [16-18]. Therefore, it is needed higher DC voltage to cause the sample to breakdown [16-18]. The aged samples being DC pre-stressing has higher DC breakdown values than the aged samples without pre-stressing. Aging has caused damage to the micro-structure of fiber, the trap density increases with the aging degree of oil-paper insulation, enhancing the space charge effect and breakdown voltage [18]. The space charge is more easier to inject into the samples, there are more charges trapped near the electrode region after being 5min DC pre-stressing. Because the charges injected is homo-charge, which could reduce the field strength near the electrode. The more charges would lead to a greater reduction. The average DC breakdown field strength difference between the test with pre-stressing and without pre-stressing clearly shows the apparent enhancement effect of the homo-charge injection on the DC breakdown. In addition, the tested DC resistivity for the aged samples increases in the process of ageing. This is also contributes to the increasing of the DC breakdown values.

The aged nature ester impregnated insulation paper has higher pre-stressing DC breakdown voltage than the aged nature ester impregnated insulation paper. As shown in Figure 13, the better microscopic structure property could restrict the formation and development of discharge channels [18], which is also one reason related to the aged nature ester impregnated insulation paper has higher pre-stressing DC breakdown voltage than the aged nature ester impregnated insulation paper.

5. Conclusion

From the experiment and analysis above, some conclusions can be drawn as following.

The natural ester impregnated insulation paper has higher DC breakdown values than the mineral oil impregnated insulation paper in the process of thermal ageing, especially for the samples after being DC stressing.

The DC breakdown values for the oil-paper insulation does not show decrease with the aggravation of aging degree. The DC pre-stressing could increase the DC breakdown values of the mineral oil impregnated insulation paper and the natural ester impregnated insulation paper. This is mainly related to the apparent enhancement effect of the homo-charge injection on the DC breakdown.

The better microscopic structure property of the aged nature ester impregnated insulation paper is also helpful to keep its higher pre-stressing DC breakdown voltage than the aged nature ester impregnated insulation paper.

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