

Kerr Electro-optic Field Mapping and Charge Dynamics in Impurity-doped Transformer Oil

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ABSTRACT

We measured dc electric field distribution in transformer oil mixed with different impurities and additives so as to simulate practical degradation conditions of transformer oil. The electric field in the oil doped with asphalt was reduced near the cathode and enhanced near the anode. On the other hand, electric field in oil with copper oleate was enhanced near both electrodes and reduced at the center between the electrodes. The space charge density was estimated from the measured electric field profile using a 1-dimensional form of Gauss law; negative ions occurred more than positive ions by 5 to 40 pC/cm³ in the oil with asphalt from 10 to 30 ppm. It was also found that heterocharges with 100 to 300 pC/cm³ existed near both electrodes in the oil with copper oleate. Consequently, the electric field and the charge distribution in the oil proved to change depending on the kind and content of impurities or additives and thus depending on the degradation of the oil.

1. INTRODUCTION

DIELECTRIC liquids are generally used for the insulation of electric power apparatus. Degradation of dielectric liquids proceeds by the influence of residual oxygen, moisture and so on during long-time service. The degradation substances dissociate under the electric field and behave as space charges. As a result, the degradation substances have an influence on the electric field distribution in dielectric liquids. Hence, it is important to investigate the effect of degradation substances on the electric field profile. It has been studied how impurity or additive affects the electric field in transformer oil using the Kerr electro-optic effect. Gäfvert *et al.* measured the field distribution in aged oil and transformer oil with tri-isoamylammonium picrate (TIAP) [1]. Nonaka *et al.* reported time dependence of distorted electric field and charge distributions in oil containing additives [2]. However, the charge dynamics in transformer oil for various phases of degradation is not sufficiently understood.

In this paper, we perform Kerr electro-optic field measurements in transformer oil with different impurities and additives dependent on the degradation process of the transformer oil. Furthermore, to clarify the influence of degradation substances in the oil on the electric field and charge profiles, we discuss charge dynamics in the oil from the measured results.

2. EXPERIMENTAL PROCEDURE

Diisopropylbenzene hydroperoxide (DIH) was used as an impurity, so as to simulate the initial stage of degradation of transformer oil [3]. Asphalt, oleate acid, and copper oleate were also added to the oil to simulate the final stage of degradation [3, 4]. We also doped 1,2,3-benzotriazole (BTA), which is used in practice for the suppression of streaming electrification [4, 5].

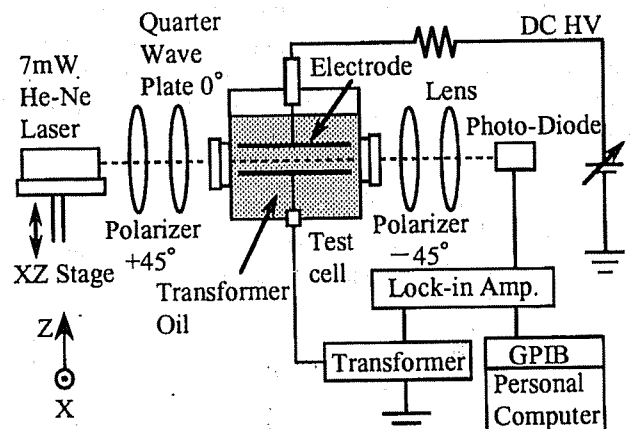


Figure 1. Experimental setup for measurement of electric fields in transformer oil using the Kerr effect.

Figure 1 illustrates the experimental setup for the measurement of electric field in transformer oil using the Kerr effect. The

Kerr constant B of transformer oil is as small as $3 \times 10^{-15} \text{ m/V}^2$, and we used a combined technique of ac electric field modulation and circularly polarized light incidence for the field measurement [6, 7]. The transformer oil was filled in a test cell made of stainless steel with glass windows. A pair of brass electrodes with length $l = 100 \text{ mm}$ and width 65 mm for the gap length $d = 10 \text{ mm}$, were placed in parallel with each other in the oil. To rule out the influence of oxygen and moisture, we removed gas from the oil with a vacuum pump and then enclosed dry nitrogen gas above the oil surface at atmospheric pressure. A dc voltage with a given magnitude was applied to the upper electrode while an ac voltage 200 V (peak) with a frequency of 1 kHz was superimposed on the lower electrode. Measurements started 1 h after dc voltage application to obtain the steady-state electric field distribution in the oil. The light source employed was a He-Ne laser with the wavelength 632.8 nm and output 7.0 mW . The laser was mounted on an $X - Z$ stage and scanned in the Z direction, that is, the gap length direction between electrodes.

3. PRINCIPLE OF KERR ELECTRO-OPTIC FIELD MAPPING

In this paper, the axis of the polarizer was set at 45° with respect to the vertical axis (Z axis), i.e. the direction of the electric field formed in the oil. We also set the fast axis of the quarter wave plate perpendicular to the direction of the electric field. The axis of the analyzer was set at -45° [7]. We use the ratio of the first harmonic component $I_{1\omega}$ of the light intensity to the dc component I_{dc}

$$\frac{I_{1\omega}}{I_{dc}} = 2\pi \frac{E_{dc} E_{ac}}{E_m^2} \quad (1)$$

where E_{dc} and E_{ac} are dc applied field and superimposed ac electric field, respectively. E_m is the electric field strength when I is maximum,

and defined as

$$E_m = \frac{1}{\sqrt{2B}} \quad (2)$$

These equations allow the electric field to be derived from the measured light intensity.

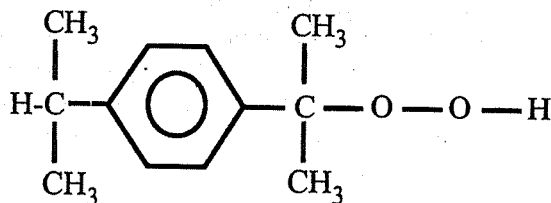


Figure 2. Molecular structure of diisopropylbenzene hydroperoxide (DIH).

4. RESULTS

4.1. ELECTRIC FIELD DISTRIBUTION IN OIL WITH HYDROPEROXIDE

It is known that hydroperoxide plays a crucial role in the initial stage degradation of transformer oil [3]. We used DIH with

its molecular structure $\text{C}_{12}\text{H}_{17}\text{OOH}$ as given in Figure 2 to simulate the substances for initial degradation in transformer oil. The molecular weight of mineral oil is as high as 250, so that we selected DIH which has the largest molecular weight (194) among available hydroperoxides. Figure 3 displays the normalized electric field distribution in the oil mixed with DIH with the value of peroxide of 10 ppm for different V_{dc} . Note that the value of peroxide represents the degree of oxygen activity in hydroperoxide. It is found in this Figure that the electric field distribution in the oil is almost constant regardless of the magnitude of applied voltages. Additional measurements showed that the resistivity and $\tan \delta$ of the contaminated oil was the same as those of fresh oil. With the above results considered, there is a possibility that the hydroperoxide in the oil did not exhibit active properties within the limit of the present experimental conditions.

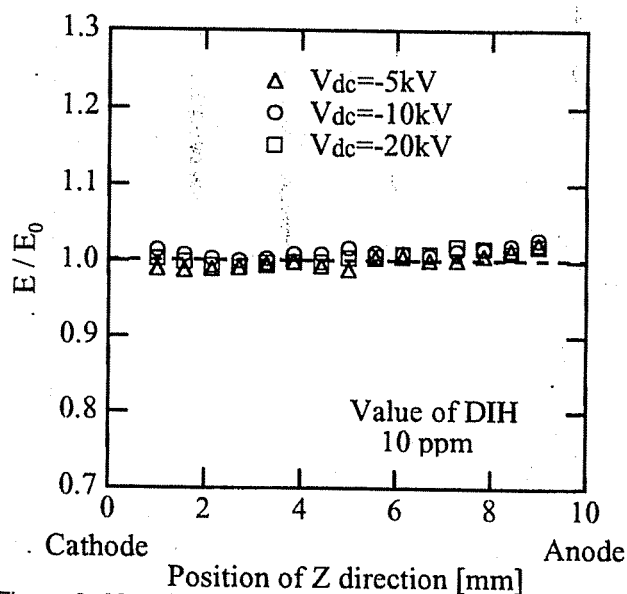


Figure 3. Normalized electric field distribution in the oil mixed with DIH with the value of peroxide of 10 ppm for different V_{dc} .

4.2. OIL MIXED WITH OLEIC ACID

Next, we deal with an electric field measurement in oil with oleic acid. It is well known that transformer oils are composite consisting of a large number of complicated hydrocarbon molecules. Thus, a considerable amount of acid with a normal chain would be produced in the degraded oil. Due to this consideration, we used oleic acid to simulate the final degradation of transformer oil. Figure 4 displays normalized electric field in oil with different contents of oleic acid as a function of position in the Z direction for aluminum cathode and brass anode when $V_{dc} = -10 \text{ kV}$. The result of fresh oil is also given in the Figure. It is seen from the Figure that the electric field distribution is almost uniform, irrespective of oleic acid content within the present experimental conditions. The results also indicate that oleic acid does not contribute to an excess charge to form space charge.

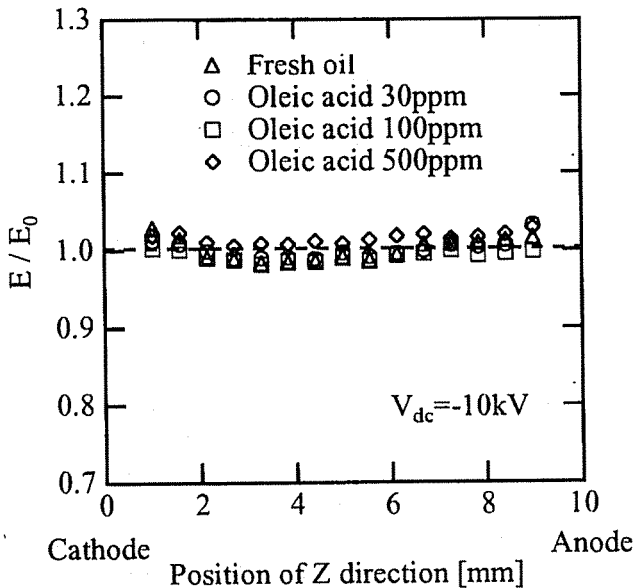


Figure 4. Normalized electric field in oil with different contents of oleic acid as a function of position of Z direction for aluminum cathode and brass anode when $V_{dc} = -10$ kV.

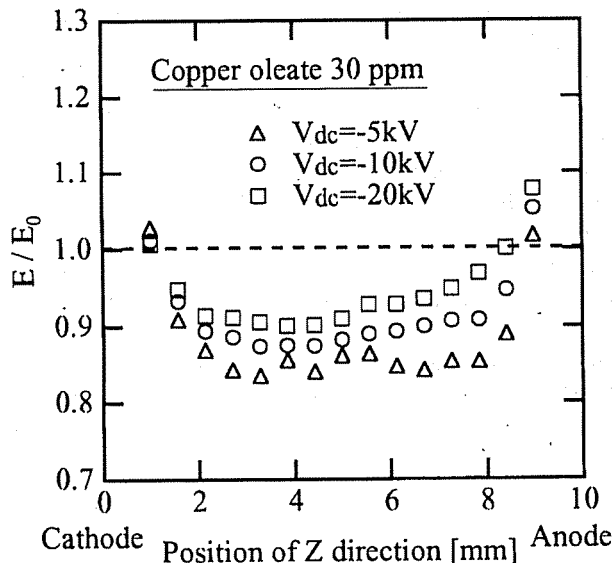


Figure 5. Normalized electric field by E_0 in the oil with 30 ppm copper oleate as a function of Z direction for different negative dc voltages.

4.3. OIL WITH COPPER OLEATE

It should be noticed that copper exists as a conductor in a transformer, so that after degradation of oil proceeds, metal salts such as copper oleate may appear as degradation products. Thus, copper oleate with known molecular structure was mixed in transformer oil to simulate the final degradation stage of the oil. Because of being a metal salt, copper oleate is likely to dissociate in the oil at room temperature and is possibly present in degraded oil. Thus, it is suitable for use as the degradation substance in the oil. The molecular formula of copper oleate is $Cu(C_{17}H_{33}COO)_2$ and the molecular weight is 626.46. Figure 5 depicts the electric field distribution in oil with 30 ppm copper oleate for different

applied dc voltages. It is obvious in this Figure that the electric field in the oil is enhanced near the both electrodes. On the other hand, the electric field is reduced by 10 to 20% of E_0 at the center between the electrodes; this result indicates that heterocharges exist near both electrodes.

Consequently, the difference in the electric field profile between the oil with oleic acid and that with copper oleate is probably associated with the different kind of ions: copper ions for the former and H^+ for the latter.

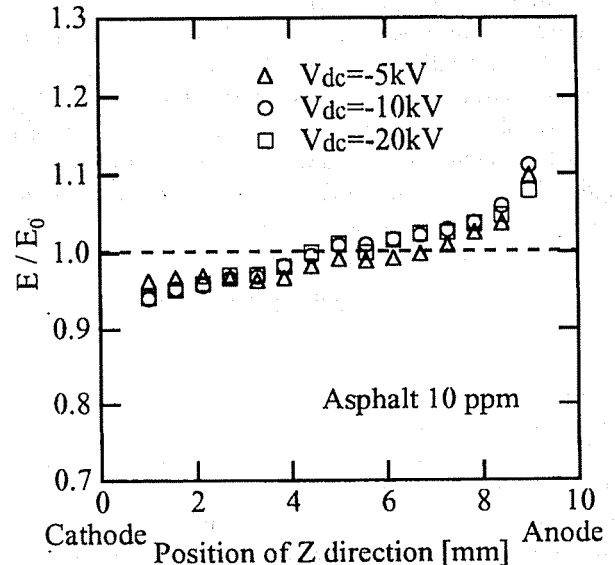


Figure 6. Normalized electric field by E_0 in the oil with 10 ppm asphalt as a function of Z direction for different negative dc voltages. Both electrodes are made of brass.

4.4. OIL WITH ASPHALT

Asphalt was mixed in transformer oil to simulate the degraded oil. Asphalt is the substance which remains after a refinement of crude oil and contains many ionic and polar substances. Hence, it can simulate the substance measured in degraded oil after a long use. Figure 6 shows the electric field distribution in the oil with 10 ppm asphalt. The vertical axis is the normalized electric field with E_0 , the value divided applied dc voltage by the gap length d . The horizontal axis shows the position between electrodes: 0 mm at the cathode, and 10 mm at the anode. In this Figure, the electric field in the oil is reduced by 5% of E_0 near the cathode, while enhanced by 10% near the anode, irrespective of applied voltage. The distortion of the electric field distribution in the oil with asphalt indicates that more negative ions exist in the oil than positive ions.

The origin of the negative space charge is possibly attributed to electrons injected from the cathode. Thus, we investigated the influence of electrode material on the electric field distribution. As cathode, we used aluminum whose work function is less than that of brass; *i.e.* aluminum is more likely to inject electrons than brass. Figure 7 shows the electric field profile in oil with 10 ppm asphalt for aluminum cathode and brass anode for different V_{dc} . Note that all the measurements used brass as both cathode and

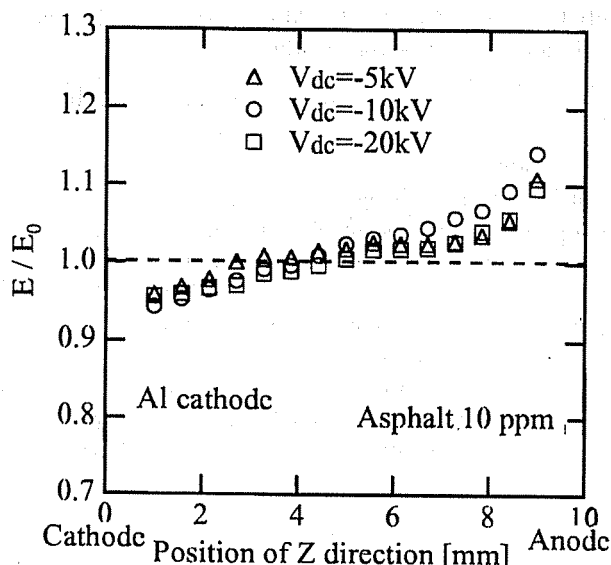


Figure 7. Normalized electric field profile in oil with 10 ppm asphalt for aluminum cathode and brass anode.

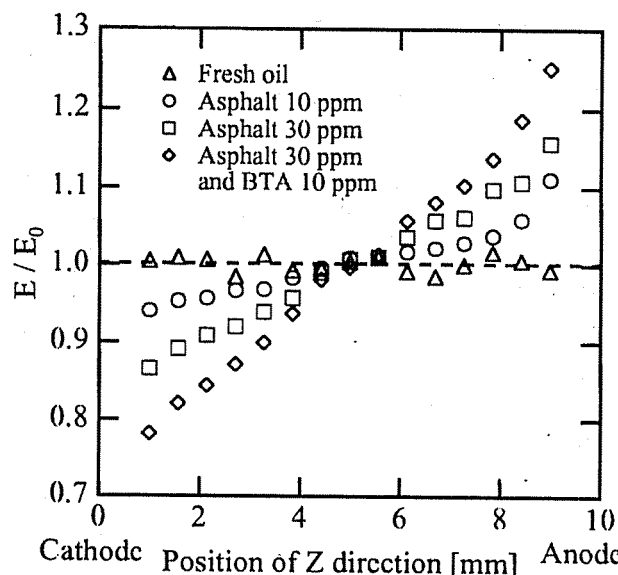


Figure 9. Normalized electric field distribution in the oil containing both 30 ppm asphalt and 10 ppm BTA for $V_{dc} = -10$ kV. The results for fresh oil and oil with different asphalt contents are also shown in the Figure.

anode except the results given in Figure 7 which shows the electrode material dependence of the field distribution. Namely, the measurements in Figure 7 alone used aluminum as the cathode and brass as the anode. As can be seen, the electric field in the oil is reduced by 5% of E_0 near the cathode, whereas enhanced by 10% near the anode. The results in Figure 7 are seen to be almost the same as those given in Figure 6; the distortion is independent of the electrode material. Consequently, it is considered that the effect of charge injection from the cathode does not play a major role in the space charge formation in the measurements.

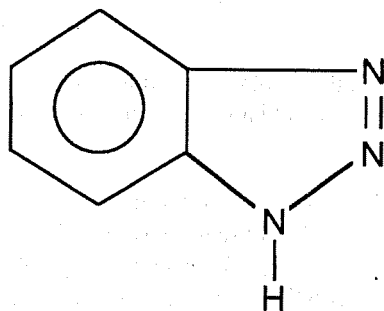


Figure 8. Molecular structure of BTA.

4.5. OIL MIXED WITH ASPHALT AND BTA

BTA is known as an antistatic agent for streaming electrification and used to dope transformer oil [4, 5]. Figure 8 shows the molecular structure of BTA. It is reported that the principle of the prevention of streaming electrification by adding BTA in transformer oil arises from adsorption of BTA on the insulating paper, so as to relax the surface potential [4, 5].

Figure 9 shows the electric field measurement in the oil containing both 30 ppm asphalt and 10 ppm BTA for $V_{dc} = -10$ kV. The results for fresh oil and oil with different asphalt contents are also shown in the Figure. It is evident that the further addition of 10 ppm BTA into the oil mixed with 30 ppm asphalt causes field

distortion by at most 22% of the average field; i.e. the degree of the distortion is larger by 7% compared with that for oil with asphalt alone. It implies that the ratio of the number of negative ions to that of positive ones increases by adding BTA in the oil.

The above results can be interpreted as follows: It is generally said that since insulating paper used in a transformer selectively adsorbs negative ions existing in the oil, the oil is charged positive [4, 5]. It is also reported that BTA can suppress the streaming electrification because nitrogen atoms having the isolated electron pair in BTA attracts positive ions in the oil [4, 5]. The above mentioned reports support the present experimental results; BTA draws positive ions in the contaminated oil, resulting in more excessive negative ions with a consequence of further electric field distortion.

5. DISCUSSION

5.1. ESTIMATION OF SPACE CHARGE DENSITY

All the present experimental results revealed that the electric field distribution in the oil largely depends on the kind of impurity or additive. It was observed that the electric field in oil mixed with asphalt was reduced and enhanced near cathode and anode, respectively. Adding copper oleate in the oil enhanced the electric field near both the cathode and anode. In this Section, we investigate charge dynamics and estimate space charge density from the measured electric field profile using Gauss law.

Since the curve of the field distribution in the oil with asphalt was almost linear, as shown in Figure 9, it is reasonable to assume that space charge spreads uniformly in the oil. Figure 10 shows a relationship between the space charge density estimated from the measured profile using Poisson's equation and the conductivity of the oil for different V_{dc} and electrode materials. Note that the

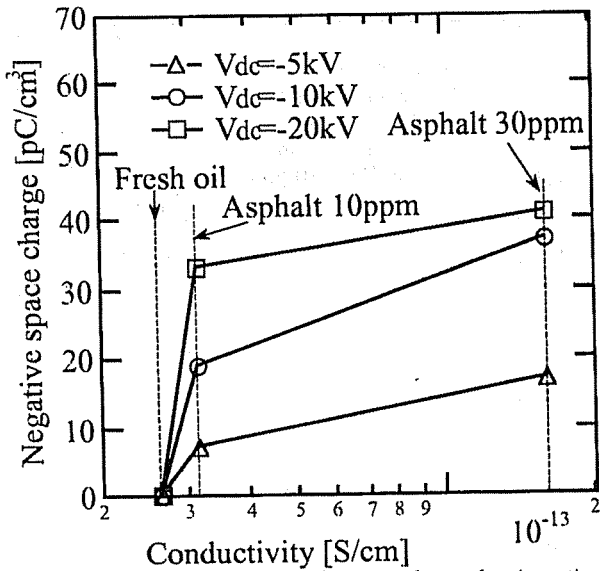


Figure 10. Relationship between the space charge density estimated from the measured profile and the conductivity of oil for different V_{dc} .

conductivity k of the oil with different asphalt contents was also measured. It was also found that the conductivity of the fresh oil sometimes was as high as 0.27×10^{-13} S/cm, as shown in Figure 10. The high value of k may be due to relatively high moisture content or existence of impurities in this fresh oil. It is obvious from Figure 10 that the space charge density q is zero for fresh oil, indicating that there is no difference in number between positive and negative ions in fresh oil; whereas the density q of negative ions is estimated to be -5 to -40 pC/cm³ for asphalt of 10 to 30 ppm in oil.

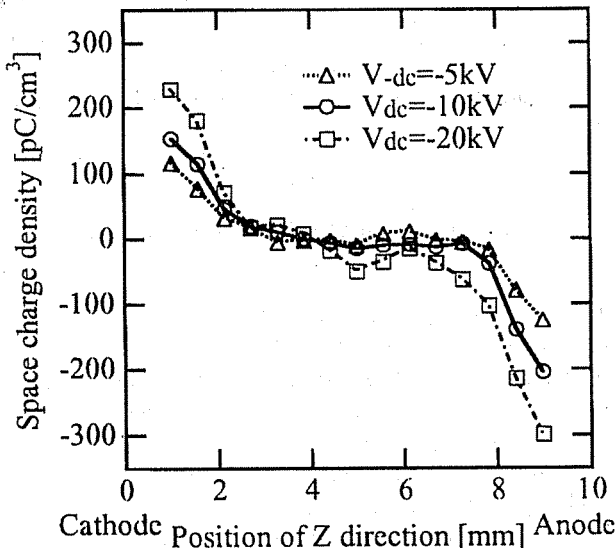


Figure 11. Space charge distribution in the oil with 30 ppm copper oleate for different V_{dc} .

Using Gauss law in the same way as above allowed the estimation of space charge distribution in the oil with copper oleate from the measured electric field distribution. Figure 11 shows the space charge distribution in the oil with 30 ppm copper oleate for

Table 1. Features of the electric field and space charge distributions for oil with different kinds of impurities.

Additive	Field distrib.	Charge distrib.
hydro-peroxide	no change	—
oleic acid	no change	—
copper-oleate	enhancement at electrodes	heterocharge at electrodes, 100 to 300 pC/cm ³
asphalt	reduction at cathode, enhancement at anode	uniform pos.ions < neg.ions, 5 to 40 pC/cm ³
asphalt and BTA	further reduction at cathode, further enhancement at anode	uniform pos.ions << neg.ions

different V_{dc} . As can be seen in the Figure, positive space charge with 100 to 230 pC/cm³ is near the cathode, while negative charge with -120 to -230 pC/cm³ is near the anode and there is almost no space charge in the region of center of the oil gap. It is also found that as the magnitude of V_{dc} increases, q of the heterocharges near both electrodes grows. These results can be interpreted in terms of an increase in dissociation energy with enhancing the magnitude of V_{dc} , resulting in an increase in the number of positive and negative ions generated in the oil. The reason for the existence of the heterocharges is also explained as follows: When dissociated, one molecule of copper oleate $\text{Cu}(\text{C}_{17}\text{H}_{33}\text{COO})_2$ is separated into a positive and two negative ions: Cu^{2+} and two $\text{C}_{17}\text{H}_{33}\text{COO}^-$ ions. These ions drift in opposite direction from each other to form heterocharge near the electrodes.

To summarize all the obtained results, Table 1 summarizes the feature of the electric field and space charge distributions for oil with different impurities.

Next, we will estimate the total amount q_t of negative and positive ions. It was found that the conductivity κ varied from 5×10^{-15} to 1.5×10^{-13} S/cm for oil with asphalt content of 0 to 30 ppm. For rough estimation of q_t , let us assume that the drift mobility μ_+ of positive ions in oil is 10^{-5} cm²/Vs [1] and equals μ_- of negative ions, and q_t is given by

$$q_t = ne = \frac{\kappa}{\mu_+ + \mu_-} \quad (3)$$

where n is the number of positive and negative ions, and e the electronic charge. Using Equation (3) together with the measurement results allowed the estimation of q_t to be 0.5 to 15 nC/cm³. As a result, it can be said that under the dc field application, excess negative ions of 5 to 40 pC/cm³ exist in the oil contaminated with asphalt for the total $q_t = 0.25$ to 7.5 nC/cm³. In other words, 0.02 to 2% of the total number of ions contributes to the negative space charge leading to the electric field distortion in the contaminated oil.

5.2. CHARGE DYNAMICS

Gäfvert *et al.* showed that heterocharges emerge in transformer oil with $\kappa = 4.8 \times 10^{-13}$ S/cm [1]. To explain the results, they discussed the charge dynamics in the transformer oil using the current transport equations, continuity equations and Poisson's equation. In general, it is very difficult to solve the above set of differential equations with suitable boundary conditions. Hence, they also tried to interpret the heterocharge existence in terms of the ratio of the transit time τ_t for ions to travel through the gap to the dielectric relaxation time τ_r of the oil

$$\tau_t = \frac{d}{\mu E} \quad (4)$$

$$\tau_r = \frac{\varepsilon}{\kappa} \quad (5)$$

where ε is the permittivity, and κ the conductivity of the oil.

Next, we try to explain our present results using the above model, and proceed the discussion by assuming $\mu \sim 10^{-5}$ cm²/Vs. When $\tau_t < \tau_r$, the generated ions can reach the electrode, *i.e.* the ions are swept out before they relax, resulting in formation of a uniform electric field distribution. On the contrary, when $\tau_t > \tau_r$, *i.e.* the relaxation time is faster than the transit time, ions remain in the oil gap, leading to enhancement of the electric field near the electrode. It was found that the conductivity of the pure oil varied from 10^{-16} to 10^{-14} S/cm. Here, let us take $\kappa = 2.5 \times 10^{-16}$ [S/cm] for fresh oil on the basis of the conductivity measurements. When $d = 10$ mm and $V_{dc} = 10$ kV as in our present experimental conditions, the calculation using Equations (4) and (5) with known κ and μ gives $\tau_t = 10$ s and $\tau_r = 780$ s for fresh oil. In this case, the transit time is shorter than the relaxation time, so that a uniform electric field distribution appears. On the other hand, $\tau_r = 37$ s for the oil with copper oleate; note that τ_r is in the same order of τ_t . It means that ions are left behind before they relax, resulting in space charge formation in oil. Consequently, the above consideration is consistent with the observed hetero space charge formation near both electrodes. In other words, the electric field profile observed in the oil with copper oleate can be interpreted in terms of the ion drift model.

It was also found that the conductivity of the fresh oil sometimes was as high as 0.27×10^{-13} S/cm, as pointed out from Figure 10 (measurements for oil with asphalt). The high value of κ may be due to relatively high moisture content and existence of some impurities in this fresh oil. Note that such a high conductivity of 0.27×10^{-13} S/cm leads to the estimation of the relaxation time in the order of 10 s, when $\tau_r \sim \tau_t$; the data scatter in the conductivity measured in the oil will have a slight influence on the above discussion. Hence, further investigation is needed on the quantitative interpretation of the results for oil with copper oleate in terms of the ion drift model.

Next, we discuss the charge dynamics corresponding to the electric field profile observed for the oil with asphalt and the reason why such an excess negative space charge is formed. The observed electric field profile, *i.e.* the electric field increases monotonically with the distance along the gap direction is similar

to what Gäfvert *et al.* [1] obtained for transformer oil with triisomyllammonium picrate. He states that the cause of such an electric field profile is attributed to electron injection from the cathode. On the other side, Yamada *et al.* [8] estimated the electric field distribution in an insulating oil gap by assuming that thermionic electrons emitted from a cathode form negative ions and they drift toward the anode. The calculated results showed the electric field was reduced near the cathode and enhanced near the anode. The present experimental results, however, did not support the electron injection effect from the cathode; note that the observed electric field distribution was independent of the cathode electrode material and configuration. Accordingly, to elucidate the charge dynamics in the asphalt-doped oil, different mechanisms from the electron injection should be considered and further investigation is required on the issues.

6. CONCLUSIONS

WE performed Kerr electro-optic field measurements in transformer oil with different impurities and additives under dc voltage application. Additives added to the oil were asphalt, oleic acid, DIH and copper oleate to simulate practical degradation substances emerging in the initial and final degradation stages in the oil.

The experimental results revealed that the addition of asphalt in the oil enhanced and reduced the electric field near the anode and cathode, respectively. As a result, excess negative ions, *i.e.* negative space charge, existing in the oil contaminated with asphalt and was estimated to be -5 to -40 pC/cm³ for the total ionic charge $q_t = 0.25$ to 7.5 nC/cm³ under dc field application. It was also found that further addition of 10 ppm BTA into the 30 ppm asphalt-doped oil caused more field distortion. The existence of more negative space charge was interpreted in terms of the property of nitrogen atoms in BTA which are more likely to attract positive ions.

On the other hand, the electric field distribution in the oil with 30 ppm copper oleate proved to be enhanced near both electrodes, and reduced by 10 to 20% of the average electric field at the center between electrodes; the results indicated hetero-charges existing near both electrodes. The results were successfully explained by the ion drift model.

Moreover, it was found that the addition of DIH and oleic acid did not contribute to the electric field distortion, *i.e.* gave no space charge in the oil. Consequently, the electric field and space charge distributions proved to differ, depending on the kind of impurities.

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