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# The Effects of Corrosive Sulfur in Bushing: Corrosion of Aluminum Foil and Its Effect of Oil-Paper Insulation

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**ABSTRACT** Failures of bushing caused by corrosive sulfur has gradually appeared in recent years. Apart from copper and sulfur, certain amount of aluminum was also found both in oil and paper after disassembling. The corrosion mechanism of the aluminum foil and its effect on oil-paper insulation were unknown. In this study, series of sulfur corrosion experiments were taken to investigate the sulfur corrosion of aluminum foil. The results showed the aluminum foil could certainly be corroded in oil containing corrosive sulfur (DBDS). Thermal products of insulting paper triggered the corrosion by removing the alumina film on the surface. Then, the contact corrosion occurred between aluminum and DBDS. In addition, the mechanisms of corrosion products generation both in oil and on paper were studied. The corrosion products in oil was DBDS-$Al_{(3,4)}$ and that on paper was $RCOOAl_n$. It caused the decrease of electrical properties of oil-paper. Particularly, the dielectric loss of oil and paper were dramatically enhanced.

**INDEX TERMS** Corrosive sulfur, aluminum foil, corrosion products, effect, oil-paper insulation.

## I. INTRODUCTION

Over the past decade, failures of power transformers and shunt reactors caused by corrosive sulfur have been taken seriously both by academics and industrial researchers [1], [2]. Much work has been done to investigate the generation of the $Cu_2S$ and its influence on oil-paper insulation, including failures which triggered by corrosive products [3]–[9]. Many effective methods have been used to avoid sulfur corrosion of copper winding in oil-immersed power transformers and reactors [7], [10]–[12]. However, failures of high voltage bushing, another key part of oil-immersed power equipment, caused by corrosive sulfur has gradually appeared in recent years [13]–[15]. Since 2012, abnormally large dielectric loss of 110kV bushings from multiple substations have presented successively in China. And the moisture in oil was very low. After disintegration, copper, aluminum as well as sulfur with a small amount were found both in oil and paper, which might result in the increasing of the dielectric loss. Actually, it is reported early in 1980s that many bushing failures were caused by corrosive sulfur in the former Soviet Union.[16]. With the development of AC and DC transmission, in 2017, the same situations occurred again in power Grid of China. But the work has barely been reported for the study of sulfur corrosion in bushing. Only several decades ago, the effect of the temperature and electrical field for sulfur corrosion in high voltage bushing was discussed [10].

Different from power transformer and shunt reactors, the interior of bushings is alternately by the aluminum foil and insulating paper on the copper conductive pole to obtain the required potential gradient. It contains much less oil than that in transformers and shunt reactors. Moreover, the aluminum has also been considered as a kind of metal with good corrosion resistance. In this study, series of experiments were performed to investigate the sulfur corrosion of aluminum foil. Meanwhile, the generation mechanisms of corrosion products in oil and on paper were studied. Then, its effect on oil-paper insulation was characterized as well.

## II. EXPERIMENTAL

### *A. SULFUR CORROSION EXPERIMENT*

Several methods of studying the corrosive sulfur in mineral oil have been standardized. The mineral oil sample prepared

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**TABLE 1. The experimental parameters and samples.**

| Samples | Oil + Al foil | Paper (g) | DBDS (mg/kg) | Acid value (mgKOH/g) | |
|---|---|---|---|---|---|
| | | | | formic acid | oleic acid |
| Series I: Corrosion of aluminum foil | | | | | |
| OA | Al foil: 9×4.5cm$^2$; Oil: 675ml | -- | -- | -- | -- |
| OAD | | -- | 500 | | |
| OAP | | 45 | -- | | |
| OAPD | | | 500 | | |
| Series II: Investigation of corrosion products | | | | | |
| OAPD-0 | Al foil: 9×4.5cm$^2$; Oil: 675ml | 45 | 0 | -- | -- |
| OAPD-200 | | | 200 | | |
| OAPD-500 | | | 500 | | |
| OAPD-1000 | | | 1000 | | |
| Series III: Mechanism of sulfur corrosion | | | | | |
| OA-fa-0.1 | Al foil: 9×4.5cm$^2$; Oil: 675ml | -- | 0 | 0.1 | -- |
| OA-fa-0.2 | | | 0 | 0.2 | |
| OA-fa-0.3 | | | 0 | 0.3 | |
| OA-fa-0.4 | | | 0 | 0.4 | |
| OAD-fa-0.1 | | | 500 | 0.1 | |
| OAD-fa-0.2 | | | 500 | 0.2 | |
| OAD-fa-0.3 | | | 500 | 0.3 | |
| OAD-fa-0.4 | | | 500 | 0.4 | |
| OA-oa-0.1 | Al foil: 9×4.5cm$^2$; Oil: 675ml | -- | 0 | -- | 0.1 |
| OA-oa-0.2 | | | 0 | | 0.2 |
| OA-oa-0.3 | | | 0 | | 0.3 |
| OA-oa-0.4 | | | 0 | | 0.4 |
| OAD-oa-0.1 | | | 500 | | 0.1 |
| OAD-oa-0.2 | | | 500 | | 0.2 |
| OAD-oa-0.3 | | | 500 | | 0.3 |
| OAD-oa-0.4 | | | 500 | | 0.4 |

***O-oil, A- aluminum, D- DBDS, P- paper, fa- formic acid, oa - oleic acid**

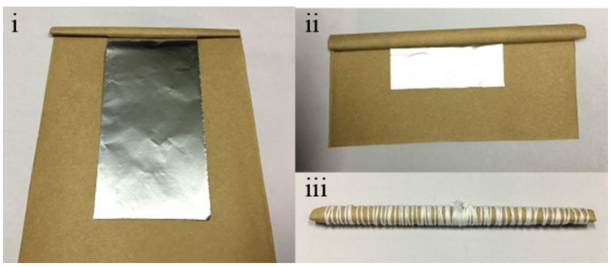


**FIGURE 1. Preparation of the aluminum-paper model (i: Fold: Putting the Al foil on the paper, ii: Roll: Rolling up the insulating paper and Al foil, iii: Fasten: Wrapping the insulating paper with cotton).**

for experiments were provided by Chongqing ChuanRun Petroleum Chemical Ltd in China, which meets the requirement of IEC 60296 standard. The aluminum foil was supplied by retailers. In this paper, the corrosion experiment was conducted according to the IEC 62535 standard, and dibenzyl disulfide (DBDS) was added as the typical corrosive sulfur in oil. In presence of DBDS, paper or some acid, aluminum foil was added in insulating oil and then the samples were aged for 72h at 150$^{\circ}$C. According to the structure of bushing, aluminum foil was rolled in insulating paper. Figure 1 is the aluminum foil-paper model for corrosion. Actually, the oil to paper ratio is 1ml to 1.5g in bushings. To ensure the integrity of the experimental data, the amount of insulating oil was increased. The experimental parameters are shown in table 1. Three series of experiments were performed to find out if the aluminum foil was corroded. In order to investigate the sulfur corrosion of aluminum foil, experiments of series I were done on the aluminum foil with or without wrapping insulating paper in oil. In Series II, the corrosion experiments of aluminum foil were conducted by adding DBDS with different concentrations in oil. The corrosion products in oil and paper were studied. Then, in Series III, the sulfur corrosion mechanism of aluminum foil was studied. Various acids with different concentrations were considered vital factors for corrosion.

### B. CHARACTERIZATION

After the corrosion experiments, the corroded condition of aluminum foil was investigated. First, the quantitative analysis of aluminum in oil-paper was performed by inductively coupled plasma atomic emission spectrometry (PERKINELMER, OPTIMA8000). Then the micromorphology of the surface, especially the corrosion pits on aluminum foil was characterized by scanning electron microscope (JEOL, JSM-7800F). GC-MS (agilent, 7890A/5975C) was used to detect the corrosion products in oil, and its effect on oil-paper was subsequently studied. The dielectric loss of oil was measured by insulating oil comprehensive test instrument (Baur, DTLC), as well as the resistivity. Also, kinematic viscosity of oil was tested by rheometer (TA, Discovery). Titration was utilized for acid value of oil according to GB264-1983. Moreover, the

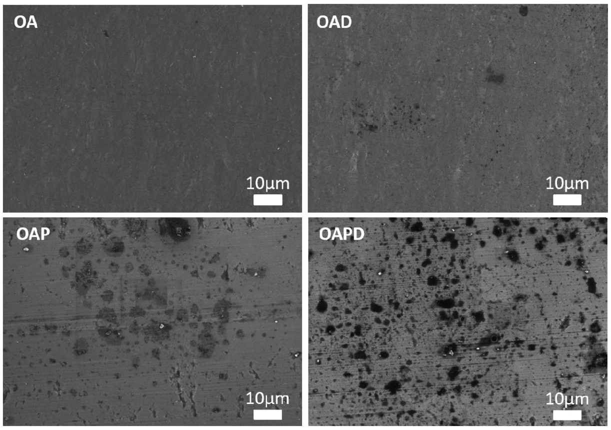


**FIGURE 2.** The micromorphology of aluminum foil.

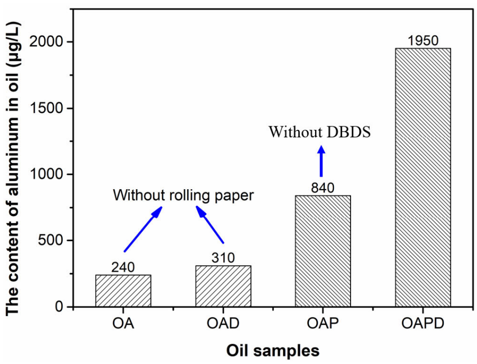


**FIGURE 3.** The content of aluminum in oil.

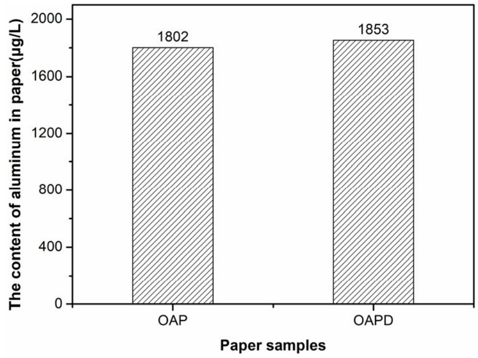


**FIGURE 4.** The content of aluminum in paper.

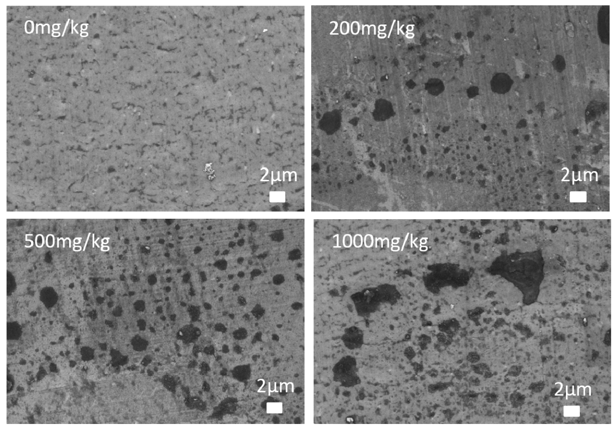


**FIGURE 5.** The micromorphology of aluminum foil in different concentration of DBDS.

broadband dielectric spectroscopy (Novocontrol, concept80-3uHz-20MHz) was applied to gain the dielectric properties of insulating paper. Breakdown-voltage of oil-paper was tested by breakdown-voltage tester.

## III. RESULT AND DISCUSSION

### A. CORROSION OF ALUMINUM FOIL

This study focused on the corrosion of aluminum foil. After the corrosion experiments (Series I), the micromorphology of aluminum foil was characterized and presented in figure 2. From this figure, obvious corrosion pits are exhibited on the aluminum foil rolled with insulating paper (OAP and OAPD). And the amount and size of corrosion pits on OAPD sample were also increasing with the addition of DBDS. Without paper, corrosion pits on aluminum foil (OA and OAD) barely appeared, even in the oil containing DBDS. In addition, the contents of aluminum in oil were investigated and presented in figure 3. Without the existence of paper, the contents of aluminum in oil were very low. A tiny growth of the aluminum content presented in oil with DBDS. On the contrary, the contents of aluminum dramatically increased in oil, when paper existed. The content of aluminum reached 840ug/L in oil even without DBDS. Compared with it, a sharp rise of the aluminum content reached 1950ug/L, simultaneously showed in the condition containing both paper and DBDS. The result was in according with the micromorphology of aluminum foil that insulating paper played a key role in corrosion process. Then, as shown in figure 4, the contents of aluminum on paper seemed almost the same, whether DBDS was in oil or not. It indicated that the aluminum foil could be obviously corroded in oil by the influence of paper. When the paper triggered the corrosion, DBDS could certainly has aggravated it.

### B. CORROSION PRODUCTS AND MECHANISM

It was found that the content of corrosion products in paper were almost the same, whether DBDS was in oil or not. The corrosion experiments of aluminum foil were performed in oil containing paper and various concentrations of DBDS (Series II). Figure 5 presented the micromorphology of aluminum foil corroded in different concentration of DBDS. With the increasing of the concentration of DBDS, the corrosion pits were gradually expanded and deepened. And in figure 6, the content of aluminum also exhibited an obvious increasing trend in the oil. But still with the increasing of the DBDS concentration, the content of aluminum on paper were nearly the same. It meant that the main corrosion products of DBDS were in oil, while another corrosion product was on paper.

**TABLE 2. The charge-mass ratio of molecular fragments in oil.**

| Sample | Charge-mass of molecular fragments | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | Same peak | | | | | | | | | Extra peak | |
| before corrosion | 29.1 | 57.1 | 81 | 105 | 145.1 | 177.1 | 205.2 | 238.2 | 280.2 | 315.3 | -- | -- |
| after corrosion | 29.1 | 57.1 | 81 | 105 | 145.1 | 177.1 | 205.1 | 234.2 | 281.0 | -- | 326.8 | 354.9 |

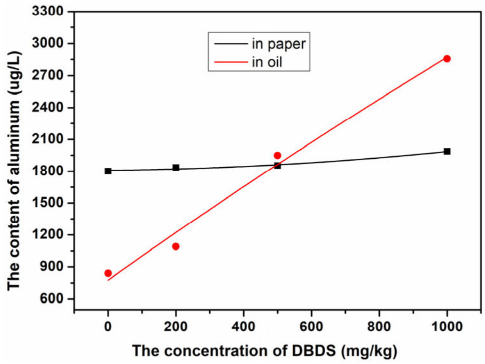


**FIGURE 6. The content of aluminum in oil-paper.**

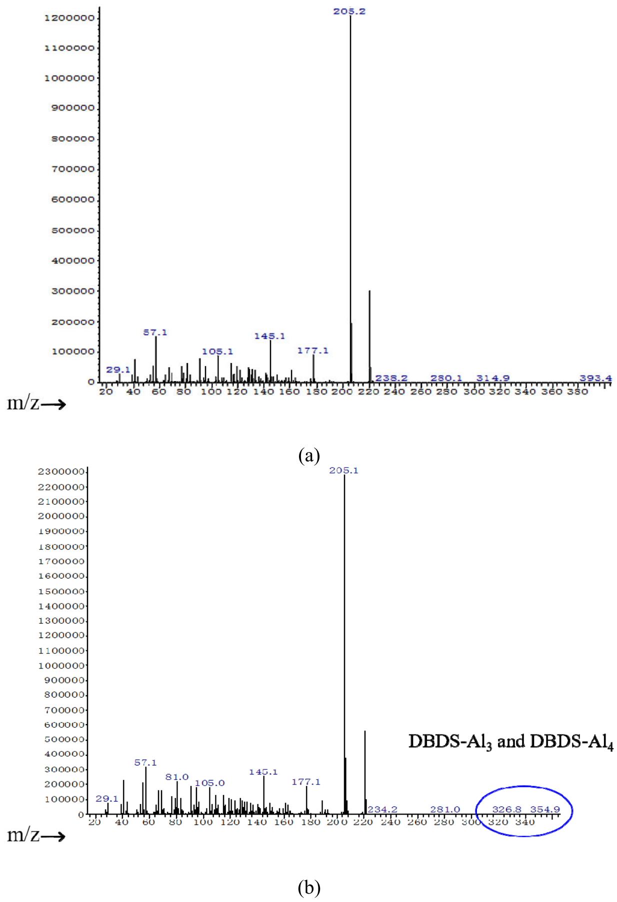


**FIGURE 7. The charge-mass ratio of molecular fragments in oil: (a) before corrosion; (b) after corrosion(200mg/kg).**

Then the corrosion products in oil were studied. GC-MS was used to investigate the corrosion products in oil. Figure 7 showed the charge-mass ratio of molecular fragments in oil, and table 2 showed the data of charge-mass ratio of molecular fragments in figure 7. From the figure and the table, the oil after corrosion possessed more peaks of molecular fragments than those of oil without corrosion. The intensity of the peaks of the fragments was very small for the concentration of corrosion products in oil was really low. In the same condition, the corrosion products of copper in oil were DBDS-$Cu_{(n)}$ [8]. It was a kind of complex compound made from copper, a kind of metal with unoccupied orbitals and DBDS, a compound with lone pair electrons. Similarly, aluminum was a kind of metal with unoccupied orbitals. Another kind of complex compound could be produced by reaction between aluminum and DBDS. Compare with figure 7(a), extra fragments with molecular weight of 326.8 and 354.9 in figure 7(b) indicated that the corrosion products in oil were DBDS-$Al_3$ and DBDS-$Al_4$, whose molecular weight were about 327 and 354 respectively.

As a matter of fact, aluminum is a kind of metals with good corrosion resistance for the naturally formed aluminum passivating film. Without paper, aluminum foil could not be corroded by DBDS in former experiments. On the contrary, with the existence of paper, aluminum foil appeared obvious corrosion phenomena even without DBDS. Then, acid, one of the thermal aging products of oil-paper insulation, was considered to be a key factor for corrosion. In experiment Series III, formic acid, as a typical thermal aging product of paper, and oleic acid, as well as that of oil were added respectively in oil for corrosion. The aluminum foil was corroded in the oil without paper. Figure 8 showed the micromorphology of aluminum foil in oil containing acid after corrosion. Completely different corrosion

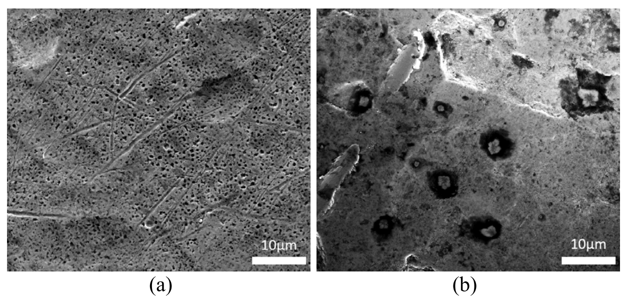


**FIGURE 8. The micromorphology of aluminum foil in oil containing acid with DBDS: (a) formic acid (0.4 mgKOH/g); (b) oleic acid (0.4 mgKOH/g).**

phenomenon appeared. The micromorphology of aluminum foil with formic acid showed a number of deep pits with small size. In the microstructure of aluminum foil with oleic acid, the shallow corrosive pits with large size were sparse. Both alumina and aluminum could react with formic acid.

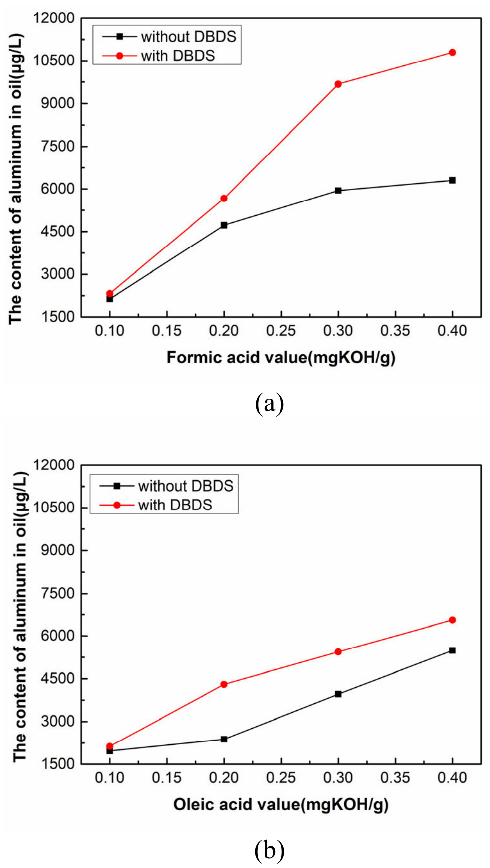


**FIGURE 9.** **The content of aluminum in oil containing acid: (a) formic acid; (b) oleic acid.**

When alumina film was corroded, formic acid immediately corroded the aluminum below. Meanwhile, DBDS also began to corrode aluminum. Differently, oleic acid was weak acid, which could only react with alumina. When the alumina film was corroded, the corrosion reaction could not be downwards, only extending around. Yet corrosion between aluminum and DBDS occurred as before. Simultaneously, the content of aluminum in oil was investigated. Figure 9 possessed the content of aluminum in oil containing acid. From the figure, it could be seen that the content of aluminum in oil arose with the increasing of acid value ranging from 0.1 to 0.4 mg KOH/g. The content of aluminum in oil with DBDS was apparently higher than that without DBDS. Moreover, the effect of formic acid on corrosion was more obvious than that of oleic acid. Considering the experiment Series I and III, the concentration of the thermal products also played a vital role in the corrosion process. When the aluminum foil was rolled in insulating paper (OAP and OAPD), the concentration of thermal products including formic acid, was much higher in oil-gap between insulating paper and aluminum foil than that in sample OA and OAD without paper. Therefore, the corrosion situation of the samples (OAP and OAPD) was much more serious. No matter which acid it contained, the thermal products of oil-paper triggered the corrosion between aluminum and DBDS by removing the alumina film.

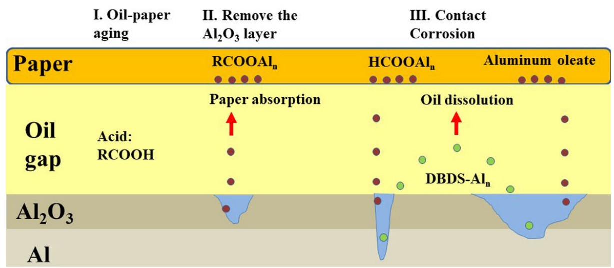


**FIGURE 10.** **The mechanism of the corrosion.**

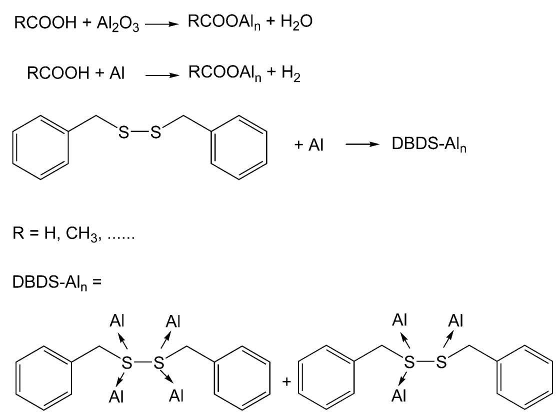


**FIGURE 11.** **The chemical equation of corrosion process.**

Then, the mechanism of the corrosion for aluminum foil was proposed. Figure 10 showed the mechanism of the corrosion. First, the oil-paper insulation aged. The thermal aging products removed the alumina film. Then the contact corrosion occurred between DBDS and aluminum blew, in which process two kinds of corrosion products were generated. One was polar compound, such as $RCOOAl_n$, absorbed in paper, which was obtained by the reaction between alumina and acid; another was DBDS-$Al_{(3,4)}$ in oil. Figure 11 exhibited the chemical equation of the corrosion process.

### C. EFFECT FOR OIL-PAPER

The effect of the corrosion products (experiment series I) for oil-paper was discussed. Figure 12 contained the result of the dielectric loss of insulating oil after corrosion. Actually, the corrosion products were considered to be impurities in oil. The polarity and electrical conductivity of the corrosion products were positive. From this figure, it is found that the dielectric loss of insulating oil samples obviously increased with generation of corrosion products in oil. Compared with samples without paper, the dielectric loss of samples OAP and OAPD showed clear enhancement. Both of the tg$\delta$ data (0.28% and 0.4%) were in excess of the particular standard value (0.2%). Meanwhile, the resistivity of the oil after corrosion was studied. In figure 13, sharp reduction of the resistivity of the oil samples exhibited for the corrosion products in oil. The presence of paper or corrosive sulfur (DBDS) could evidently affect the performance of insulating oil by accelerating the formation of corrosion products in oil.

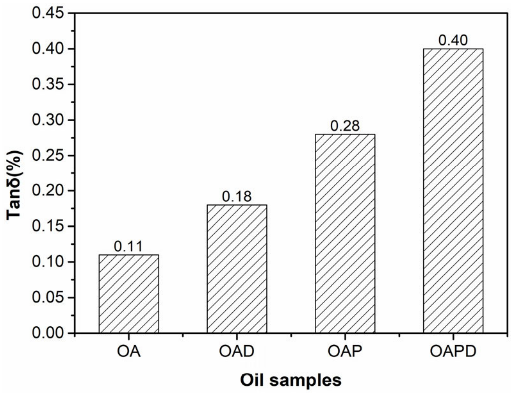


**FIGURE 12.** The dielectric loss of the oil after corrosion.

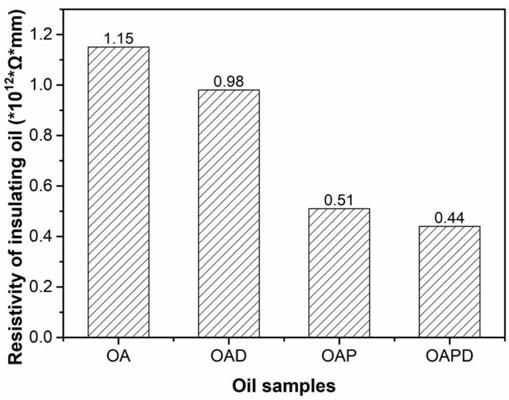


**FIGURE 13.** The resistivity of the oil after corrosion.

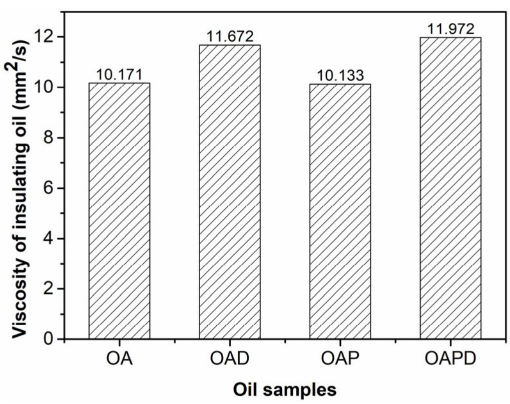


**FIGURE 14.** The viscosity of the oil after corrosion.

Figure 14 contained the viscosity of the oil after corrosion. The oil samples possessing DBDS showed higher viscosity than that without corrosive sulfur. But the viscosity of oil samples (OAP and OAPD) with much more corrosion products, exhibited no obvious change. That meant the corrosion process almost could not influence the viscosity of insulting oil. In addition, the acid value of the oil after corrosion experiment was investigated. Similar to the viscosity, the acid value of the oil samples with DBDS increased a little. The influence of paper was presented too. The acid value of the

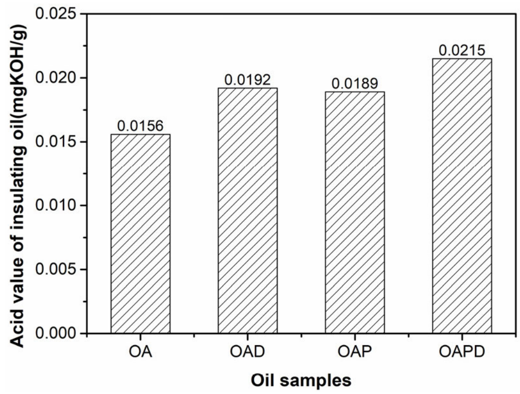


**FIGURE 15.** The acid value of the oil after corrosion.

**TABLE 3.** The dielectric properties of the insulating paper.

| Samples | New paper | Paper aged | OAP | OAPD |
|---|---|---|---|---|
| Dielectric constants (50 Hz) | 3.88[17, 18] | 4.36[17, 18] | 4.77 | 5.48 |
| tgδ (%) (50 Hz) | 0.00641 | 0.00914 | 0.0118 | 0.0179 |

oil samples corroding with paper were higher as well. The effect of aging was greater than corrosion in the system.

The dielectric properties of the paper are studied. Table 3 contained the dielectric constants and dielectric loss at 50 Hz. As shown in the table, compared with the new paper and aged paper, the dielectric constants of the paper samples after corrosion raised obviously, especially the dielectric constant of the paper samples which corroded in oil containing DBDS reaching 5.48. This was a result of the generation of corrosion products on paper and the accelerated aging by corrosion products in oil-paper. Concurrently, the dielectric loss presented the same trend with the dielectric constants. The corrosion products could certainly exacerbate the dielectric loss. The dielectric loss of paper after corrosion in oil with DBDS dramatically reached 0.0179.

## IV. CONCLUSION

In this study, series of sulfur corrosion experiments were performed to investigate the sulfur corrosion of aluminum foil. Meanwhile, the mechanism of the generation of corrosion products both in oil and on paper were studied. Then, its effect in oil-paper insulation was also characterized. We could draw conclusions as follows:

(1) The aluminum foil could certainly be corroded by DBDS, and the existence of the paper triggered the corrosion reaction.

(2) The mechanism of the corrosion for aluminum foil was proposed. First, the oil-paper insulation aged. The thermal aging products (acid) removed the alumina film. Then the contact corrosion occurred between DBDS and aluminum blew. Two kinds of corrosion products were generated in

the corrosion process. One was polar compound $RCOOAl_n$ absorbed by paper, which was produced by alumina and acid; another was DBDS-$Al_{(3,4)}$ in oil generated by reaction between DBDS and aluminum.

(3) The electrical properties of oil-paper insulation could be influenced by corrosion products. The dielectric properties of the oil were seriously affected as well as that of paper.

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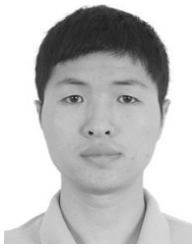


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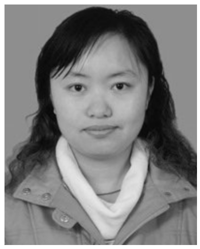


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