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Structure and Space Charge Inhibition Performance of Reactive RF Magnetron Sputter Deposited Al_2O_3 Thin Film on the Fiber Surface

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***Abstract*—Cellulose insulation paper/pressboard has been widely used in HVDC transformers. In this study, the aluminum oxide function film was deposited on the cellulose insulation pressboard surface using reactive radio frequency (RF) magnetron sputtering. The sputtered thin film was characterized by the scanning electron microscopy (SEM) and the X-ray photoelectron spectroscopy (XPS). The influence of the deposited function film on the space charge injection/accumulation behavior was investigated. SEM and XPS results show that the nano-structured Al_2O_3 film was successfully fabricated on the fiber surface. The oil impregnated sputtered pressboard presents apparent space charge suppression effect. The obtained band gap energy (Eg) of Al_2O_3 is 7.39 eV. The nano-structured Al_2O_3 film introduces a new trap band which is helpful for the charge inhibition.**

Keywords-nano-structure Al_2O_3 film; magnetron sputtering; cellulose insulation pressboard; space charge; inhibition performance

I. INTRODUCTION

Pursuing high efficiency in electric power transmission and renewable energy has led to the rapid development in high voltage DC (HVDC) transmission systems. One of the most challenging issues in the HVDC insulation material development and insulation structure design is the space charge accumulation within the insulation material [1]. The formation of space charge in insulation system 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 leads to material degradation in the high electric field region and affect system reliability [2-5]. Therefore, the effective ways to suppress space charge accumulation have been regarded as the key foundation for the design and ensure safety of polymeric HVDC insulation material [1].

Most of the attempts at present for reducing the space charge concern the modification of insulation material by the dispersion of nano-fillers into polymer bulk [6-9]. Not exactly the same as the above method, sandwich-structured nano-composites (nano-LDPE/LDPE/nano-LDPE) was reported [10, 11]. The total charge accumulated in the sandwich-structured nanocomposites (nano-LDPE/LDPE/nano-LDPE) is less than that in neat LDPE and nano-LDPE nanocomposites, which indicates that the elaborate structure design can satisfy the inhibition of space charge. Therefore, in addition to adding nano-fillers to the material, it is worthwhile to further investigate the fabrication of a special functional structure on the surface of insulating material, which could have effective suppression function of space charge injection and accumulation, even better space charge suppression performance than the material with nanoparticles filled in the bulk.

In this study, considering the Al_2O_3 is a well-known insulator which has good mechanical, thermal, and chemical stability [9, 12], and being frequently used as a coating material [13, 14] and nano-fillers of insulation paper. The Al_2O_3 function thin film was deposited on cellulose insulation pressboard by using reactive RF magnetron sputtering under room temperature. The physical and chemical characterization of the as-prepared function film was presented. The influence of the Al_2O_3 function thin film on the space charge behaviour of the sputtered pressboard was investigated. The band gap energy of Al_2O_3 was calculated at last.

II. EXPERIMENTS

A. Sample Preparation

The cellulose insulation pressboard (thickness 0.5 mm) used for reactive RF magnetron sputtering derived from NARI Borui transformer factory. The insulation pressboard substrates were cut into 15 cm*10 cm. The JPGR-480 reactive RF magnetron sputtering device with 13.56 MHz was used in this experiment. The principle of the reactive RF magnetron sputtering in shown Fig. 1 [13]. For deposition of Al_2O_3 film, the aluminum target 99.999% purity was sputtered. The distance between the target and substrate sample was 10 cm. The vacuum chamber was pumped down to a base pressure of $4.0*10^{-3}$ Pa before sputtering. Deposition processes were performed by using 110 W of

forward power. Argon was used as working gas with a constant pressure 1.5 Pa. Oxygen was reactive gas with flow 20 sccm. The total pressure was constant during the whole sputtering procedure. The sputtering time is 60 min under 28 ^{o}C.

B. Characterization Methods

The scanning electron microscopy (SEM, JSM-7800F, Japan JEOL company) was employed to characterize the surface morphology of the deposition film. The X-ray photoelectron spectroscopy (XPS, Thermo escalab 250Xi, US) was employed to investigate the chemical bonding state of the films. The PEA (Pulsed Electro-acoustic) method has been widely used by researchers to measure the space charge in solid dielectrics. The principle of the PEA can be seen in many literatures [2, 4]. The PEA system used has a pulse voltage 600 V with a width of 5 ns. The bottom electrode is made of 10 mm thick aluminum plate and the top electrode is the semiconducting polymer film. Before space charge measurement, the untreated pressboard, treated pressboard 60 min were put into three glass bottle, respectively. All the bottle with samples were put into a vacuum box and dried at 90 ℃ for 24 hours. Then the temperature of the vacuum box was adjusted to 40 ℃. The new mineral oil was infused into the vacuum box. The vacuum box was left for 48 hours for pressboard impregnation at 40 ℃ before cooled down to room temperature. After impregnation, the moisture content of the oil impregnated pressboard is 1.35% using Karl Fischer Titration method. The PEA measurement testing temperature is 28 ℃. The pressboard surface with only one side sputtered aluminum oxides thin film was attached to the cathode when the measurement was performed.

III. RESULTS AND DISCUSSIONS

A. SEM Analysis

The SEM images for un-treated pressboard, the pressboard surface prepared by magnetron sputtering of Al target for 60 min are illustrated in Fig. 1. Fig.1a shows the SEM image for untreated pressboard surface at 1000 magnification. The cellulose fibers are tied together and packed closely. There are some tiny holes where the fibers criss-cross between each other. The pressboard surface with magnetron sputtering treatment for 60 min at ×1000 magnification (Fig.1b) illustrates the formation of thin film which deposits on the fiber surface. It is obviously that a thin film was deposited on the fiber surface. Fig.1c is the surface morphology of the pressboard sputtered 60 min at ×40,000 magnification. There are dense, uniform distribution and tightly arranged particles deposited on the surface of fiber sputtered 60 min. Each particle can be clearly seen and the particle size of most particles is less than 100 nm in diameter. It is particularly emphasized that when the image is magnified to 100,000 times(Fig.1d), it is found that a large number of particles with much smaller particle size (<10nm) cling to the larger particles.

B. XPS Analysis

In order to investigate the chemical state of the aluminum oxides film fabricated on the fiber surface, the XPS measurement was performed. Fig.2 shows the compared XPS survey spectrum of the blank pressboard surface, the pressboard surface sputtered for 60 min. The deviation caused by the charging effect was calibrated using adventitious carbon referencing (C1s, 284.6 eV). The peaks observed at binding energies about 74eV, 119eV, 285eV, 532eV, 979 eV and 1228eV are associated with the chemical

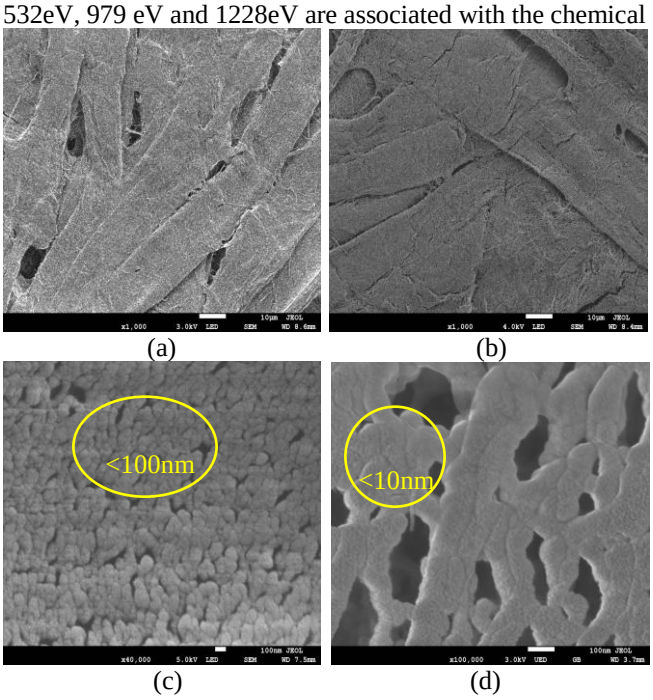

Figure 1. SEM images of the fresh pressboard surface at 1000 × (a), pressboard surface prepared by reactive RF magnetron sputtering of Al target for 60min at 1000 × (b), 40, 000 ×(c) and 100,000 ×(d).

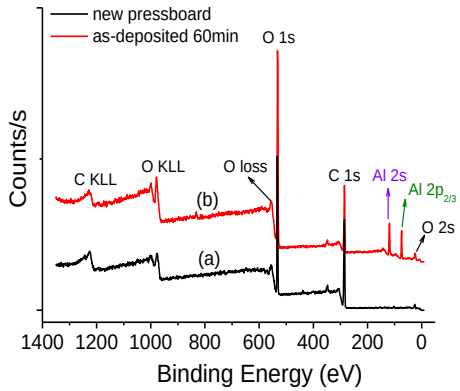

Figure 2. The X-ray photoelectron spectra of fresh pressboard (a) and pressboard sputtered for 60min (b).

element states of Al 2p, Al 2s, C 1s, O KLL and C KLL respectively. Compared with the fiber surface without sputtering, the new element peaks of Al 2p, Al 2s were detected from the sputtered fiber surface. The appearance of

new element peak Al by XPS measurement also indicates that the aluminum oxides film has been fabricated on the pressboard surface.

In order to elucidate the chemical state of the aluminum oxides film, resolution analysis of O 1s and Al 2p was carried out, as presented in Fig.3. Fig.3a, Fig.3b clearly show that the O 1s XPS spectra of the sputtered pressboard surface is much different from the un-treated pressboard. In addition to the intensity changing of the C-O and C=O peaks, a new peak located at 531.2eV originated from O-Al bond could be seen for the sputtered pressboard surface. Fig.3c, Fig.3d show that a new peak has risen at 74.4 eV originated from Al-O in the Al 2p XPS spectra of the sputtered pressboard surface. As reported in [15], the binding energy of O-Al 531.2 eV shown in O 1s spectra and the binding energy of Al-O 74.4 eV shown in Al 2p spectra are attributed to A12O3. The binding energy difference between Al 2p and O 1s is 458 eV for samples sputtered 60 min, which is similar to literature [15]. According to XPS analysis results, the atomic content ratio of Al/O is 0.65, which is close to the stoichiometric composition of Al_2O_3 (Al/O=0.66) [15]. Therefore, Fig.1 and Fig.3 presents that the thin film fabricated on the pressboard surface is nano-structured Al_2O_3.

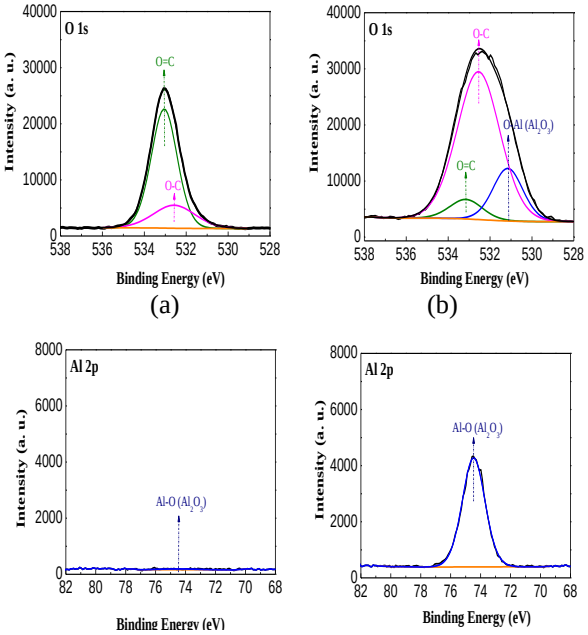

Figure 3. The O 1s and Al 2p XPS spectra of fresh pressboard surface (a, c), pressboard surface sputtered for 60 min (b, d).

C. Space Charge Performance Analysis

In order to confirm the nano-structured Al_2O_3 film has the function of restraining charge injection and accumulation, the space charge distribution of fresh pressboard and pressboard sputtered Al_2O_3 films impregnated with new mineral oil is shown in Fig. 4. In this experiment, the applied DC electric field strength is 15 kV/mm. The pressboard surface with only one side sputtered Al_2O_3 film was attached to the cathode, and the another surface without treatment was attached to the anode. In the process of voltage applied, the charge density on the anode/cathode and the total amount of charges trapped in the samples was calculated and presented in Fig. 5. The total amount of charges trapped in the samples was calculated according to the equation (1). Where ρ(x, t) is the charge density at positon x (C/m-3), S is the electrode area (m2), and d is the thickness of the sample (m).

$$Q(t) = \int_0^d \rho(x,t)\,S\,dx \tag{1}$$

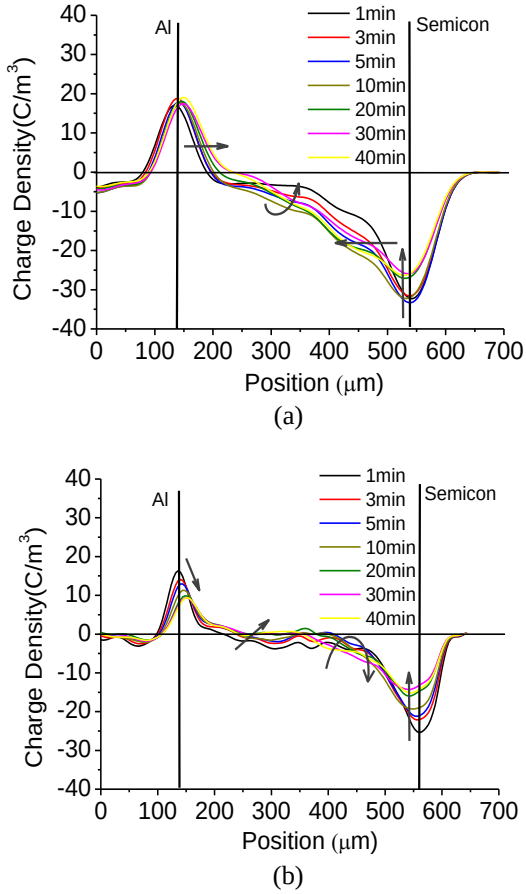

Figure 4. The space charge behaviour of the oil impregnated fresh pressboard (a), the oil impregnated pressboard surface sputtered Al_2O_3 film for 60min (b) \under DC 15kV/mm.

In Fig. 4, the black vertical real line is the position of the anode and cathode. The distance between the anode and the cathode means the thickness of the sample. The arrow direction represents the direction of charge movement. As shown in Fig. 4, homo-charge injection takes place quickly after the voltage is applied for the fresh pressboard and pressboard sputtered by Al_2O_3 films. For the fresh pressboard sample (Fig.4a), the charge density lines of positive charges near to the anode and the negative charges near to the cathode both move toward inner of the pressboard. More obvious negative charges injection was observed, which makes the density of negative charges on the cathode decreases with time until stable, while the positive charges

on the anode nearly no change after applying voltage for 5 min (Fig.5a). In the middle part of the sample, there are mainly negatives charges. Its density increases in the process of 0-10min voltage applying, then decreases with time until approximately reaches to stable after 30 min. The amount of charges trapped in the fresh pressboard increases firstly and then decreases until close to stable, as shown in Fig.5b.

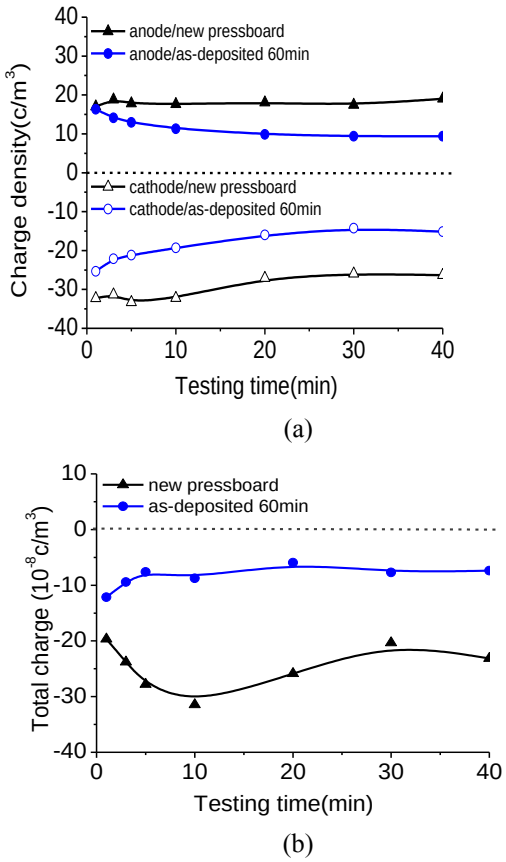

Figure 5. The charge density on the anode and cathode (a), the charge density ratio of treated samples to untreated samples (b) under DC 15kV/mm.

For the pressboard sputtered Al_2O_3 film for 60 min shown in Fig.4b, it could be seen that the charge injection phenomenon is not obvious compared to the fresh pressboard. The density of negative charges on the cathode and the positive charges on the anode both decreases with time until reaches stable. Due to the Al_2O_3 film is attached to the negative electrode surface, it could be seen that there are not many negative charges trapped in the bulk of the sample like the fresh pressboard. The negative charge injection could be seen effectively suppressed. The charge density on the anode and cathode (Fig.5a), as well as the amount of charges trapped in the pressboard sputtered Al_2O_3 film for 60 min (Fig.5b), is much lower than the fresh pressboard in the process of applying voltage. It is particularly noteworthy from Fig.5that the charge density value on the anode and cathode of the pressboard sputtered 60 min is only 49% and 57% of the untreated pressboard when the charge injection almost becomes stable (applying voltage 40min). What's more, Fig.5b shows that the total amount of charges trapped in the pressboard sputtered 60 min is only 31% of the untreated pressboard (applying voltage 40min). This may be the nano-structured Al_2O_3 film sputtered on the fiber surface could act as a functional barrier layer for suppression the charge injection.

According to the above analysis, we can deduce that the Al_2O_3 nano-structured film sputtered on the fiber surface could act as a functional barrier layer for charge injection. The band gap energy (Eg) of Al_2O_3 can be derived from the energy difference between elastic peak energy (E O1s) and onset of inelastic loss energy (E loss); Eg = E loss-E O1s [16-18]. As shown in Fig.6, The high resolution O 1s spectrum shows the energy of elastic peak (E O1s) is 532.08 eV. The energy of onset of inelastic losses (E loss), which is measured from O 1s spectrum by intersecting the linear-fit line and the background zero level, is 539.47 eV. Therefore, the obtained band gap energy (Eg) of Al_2O_3 is 7.39 eV. The measured band gap energy of Al_2O_3 is lower than the reported value of Al_2O_3 (8.8 eV) [17], which is consistent with the results shown in [16, 18]. This may be attributed to the defect structure of Al_2O_3 and required further investigation [16-18]. The nano-structured Al_2O_3 film introduces a new trap band which is helpful for the charge inhibition.

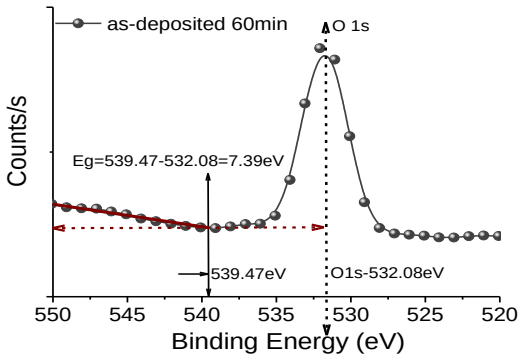

Figure 6. The band gap of Al2O3 from the O 1s peak obtained by high resolution XPS.

IV. CONCLUSION

The nano-structured Al_2O_3 film was successfully fabricated on the cellulose insulation pressboard surface by reactive radio frequency (RF) magnetron sputtering. SEM results show that there are dense, uniform distribution and tightly arranged Al_2O_3 particles with nano-meters in diameter or length. XPS analysis confirmed the deposited film is Al_2O_3. Compared with the fresh pressboard, the pressboard sputtered Al_2O_3 film 60 min shows obvious space charge suppression effect. Under DC 15kV/mm, the charge density value on the anode and cathode of the pressboard sputtered 60 min is only 49% and 57% of the untreated pressboard, and the total amount of charges trapped in the sputtered pressboard is less than 50% of the untreated pressboard. The obtained band gap energy (Eg) of Al_2O_3 is

7.39 eV. The nano-structured Al_2O_3 film introduces a new trap band which is helpful for the charge inhibition.

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