



# Influence of hierarchy structure on electrical properties of gradient-distribution aluminum oxide/polyethylene nanocomposites

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## ABSTRACT

In the recent decades, the phenomena of space charge accumulation in the high voltage direct current (HVDC) insulation have attracted more and more attention. In this paper, the gradient-distribution multi-layered Al<sub>2</sub>O<sub>3</sub>/low density polyethylene (LDPE) nanocomposites were prepared via layer-by-layer melting blending and hot pressing method. Morphologies and electrical properties of these gradient-distribution structured composites were discussed. Results showed that this unique structure could largely enhance the breakdown strength of LDPE. The breakdown strength of the LDPE nanocomposites varied with nano-Al<sub>2</sub>O<sub>3</sub> concentrations, and it could reach to 200 kV/mm for the LDPE nanocomposites. Besides, space charge of the LDPE nanocomposites was measured by pulsed electro-acoustic (PEA) method. The charge profiles indicated that space charge suppression of the gradient-distribution multi-layered composites was better than that of the monolayer ones with general mean-distribution structure. Their excellent insulating properties were attributed to the trap which exists in the surface, and that could well inhibit the charge injecting into the materials.

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## 1. Introduction

Dielectric nanocomposites have been widely used in the high voltage direct current (HVDC) cables [1–3]. As we know, a large amount of space charges will accumulate when a dielectric is energized, resulted from the traps existing in the dielectric materials [4]. Generally, the presence of the charge accumulation will lead to the distortion of electric field distribution, thus resulting in the enhancement of electric field in certain region of the dielectrics. Then it will lead to the degradation, even premature failure of the

materials. Therefore, the suppression of charge is very important for long-term using of the dielectric materials [5–8].

Several mechanisms have been proposed to interpret the suppression of space charges in the nanocomposites. One is that the trap centers near the interface between the electrodes and the nanocomposites can capture the injected charges [9]. The captured charges can distort the electric field in the vicinity of the interface, which will decrease the charge injection, and the space charges will be suppressed [10,11]. Many researches demonstrated that the nanoparticles introduced into the dielectric materials could induce the deep trap. A certain amount of deep traps could inhibit the injecting of charges into the materials [12].

In this work, two kinds of insulation systems, namely the mean-distribution monolayered and gradient-distribution multi-layered nano-Al<sub>2</sub>O<sub>3</sub>/LDPE nanocomposites were respectively fabricated. Their space charge distributions with increasing time under *E*-fields were tested by the pulsed electro acoustic (PEA) method. Space charge characteristics at the interface of the composites and the effect of nano-Al<sub>2</sub>O<sub>3</sub> concentrations on the charge transport

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characteristics were investigated. The results are useful to understand the interface effect and the transport of carriers in the dielectric composites.

## 2. Experimental

### 2.1. Materials

The additive-free LDPE was chosen not only because of its relatively simple chemical configuration but also because it can minimize the influence of impurities and additives. The LDPE (LD200BW) with the density of  $0.922 \text{ g/cm}^3$  and melt flow rate of  $2.3 \text{ g/10 min}$  was purchased from Sinopec Beijing Yanshan Co. (China). The  $\text{Al}_2\text{O}_3$  particles with diameter of  $\sim 100 \text{ nm}$  were prepared using low-temperature plasma method by Institute of Process Engineering Chinese Academy of Sciences (China), and their surface was treated using vinyltrimethoxysilane (VMES) prior to use.

### 2.2. Preparation of the nanocomposites

Different concentrations of nano- $\text{Al}_2\text{O}_3$  (0.1 wt%, 0.2 wt%, 0.5 wt% and 1 wt%) were introduced into LDPE matrix using HAAKE PolyLab mixer (HAAKE Rheomix 600, Germany). The composites with 0.1 wt% nano- $\text{Al}_2\text{O}_3$  is named as 0.1%N, and for other concentrations, they are 0.2%N, 0.5%N and 1%N, respectively. Both the pure LDPE and  $\text{Al}_2\text{O}_3$ /LDPE nanocomposites were pressed using panel vulcanizer at  $120 \text{ }^\circ\text{C}$ , and the thin films with the thicknesses of about  $0.16 \text{ mm}$  were obtained. Then the films as one layer were further co-pressed into the three-layer stacks with  $\sim 0.48 \text{ mm}$  thickness, under a pressure of  $10 \text{ MPa}$  at  $130 \text{ }^\circ\text{C}$  for  $40 \text{ min}$ . The schematic of preparing process of the multi-layered nanocomposites is shown in Fig. 1. The three kinds of multi-layered dielectric, i.e., 1%N/0.1%N/LDPE, 1%N/0.2%N/LDPE, and 1%N/0.5%N/LDPE nanocomposites were prepared.

Before testing, all the samples were placed in a vacuum oven at  $80 \text{ }^\circ\text{C}$  for  $24 \text{ h}$ , and then cooled down to room temperature to eliminate their thermal histories. To banish the residual charges, the films were put into two polished copper plates in a vacuum oven at  $80 \text{ }^\circ\text{C}$  for  $48 \text{ h}$  short-circuiting.

### 2.3. Characterization

Space charge distribution of the samples was tested by pulsed

electro-acoustic measurement (PEA, Shanghai Jiao Tong University) under an electric field of  $30 \text{ kV/mm}$  for  $60 \text{ min}$  at  $25 \text{ }^\circ\text{C}$ . During the measurement, the samples with the size of  $500 \text{ } \mu\text{m} \times 7 \text{ cm} \times 7 \text{ cm}$  were sandwiched between an aluminum electrode with diameter of  $\sim 12 \text{ cm}$  and one semiconductive polymer electrode with diameter of  $\sim 2 \text{ cm}$ .

In order to analysis the effect of the additive and gradient-distribution structure on the crystal forms of LDPE matrix, the samples were measured by differential scanning calorimetry (DSC-60, SHIMADZU Company). The weights of the samples were approximately  $4 \text{ mg}$ . The experiments were performed in the temperature range from  $50 \text{ }^\circ\text{C}$  to  $200 \text{ }^\circ\text{C}$  under  $\text{N}_2$  atmosphere with the rate of  $10 \text{ }^\circ\text{C/min}$ , and then decreased to  $50 \text{ }^\circ\text{C}$ .

Direct current (DC) breakdown strength of the samples was measured using one dielectric strength tester (Ji Lin Province Huayang Equipment Co., Ltd., China). The breakdown strength was determined by a progressive electrical stress test. The chosen rate of voltage ramp was  $1200 \text{ V/s}$ , corresponding to reaching  $50 \text{ kV DC}$  voltage in  $50 \text{ s}$ . The diameter of plate electrode was kept at  $6 \text{ cm}$  with edge radius of  $1 \text{ cm}$ . The whole setup was immersed in silicone oil. Weibull distribution was employed to fit the experimental data and determined the characteristic of DC dielectric breakdown strength according to the IEEE Standard 930-2004 [13].

## 3. Results and discussion

### 3.1. Microstructure

Fig. 2a shows SEM morphologies of the cross section of the multi-layered 1%N/0.1%N/LDPE nanocomposites with 2000 multiples. It can be seen that the content of nano- $\text{Al}_2\text{O}_3$  in the top layer is larger than that in the middle layer. From the gradual change of the  $\text{Al}_2\text{O}_3$  concentrations, the gradient-distribution structure can be clearly observed. And the interface between the adjacent layers is not obvious, which illustrates that the melting between layers is well.

Fig. 2b shows SEM morphologies of the cross section of multi-layered 1%N/0.1%N/LDPE nanocomposites with 20000 multiples. These results indicate that the surface modification of  $\text{Al}_2\text{O}_3$  effectively improves their interfacial adhesion with LDPE. The interfacial interactions between surface of  $\text{Al}_2\text{O}_3$  and vinyl groups of grafted chains increase the compatibility. It may be due to that the LDPE molecule has almost the same basic repeating unit vinyl

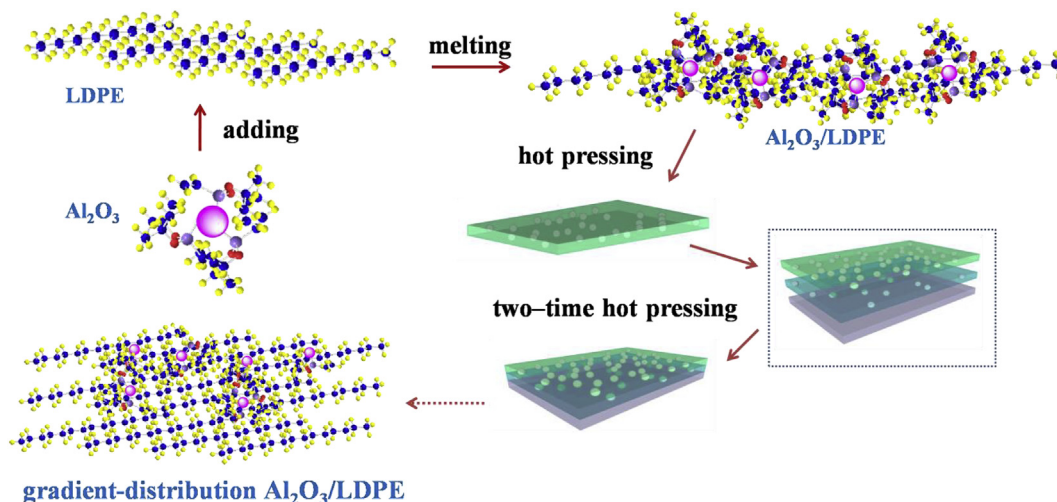


Fig. 1. Schematics of preparing process of the multi-layered nanocomposites.

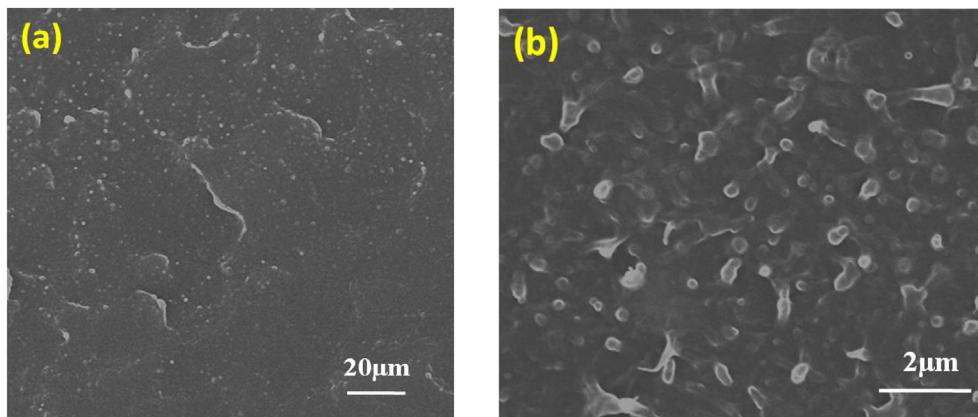


Fig. 2. SEM images of the cross-section of 1%N/0.1%N/LDPE nanocomposites with different amplifications (a) 2000 multiples (b) 20000 multiples.

group with that of VMES, which possibly improve the compatibility of  $\text{Al}_2\text{O}_3$  nanoparticles with LDPE.

### 3.2. Crystallization

Fig. 3 shows the non-isothermal crystallization curves of the multi-layered LDPE nanocomposites at the temperature rate of  $10^\circ\text{C}/\text{min}$ . Generally, the crystallization process of polymers includes the formation of crystal nuclei and the growth of crystal grain. The growth of crystal grains mainly depends on the crystallization temperature. It is known that the temperature corresponding to the crystallization peak is usually proportional to the crystal growth rate [14].

It can be observed from Fig. 3 that the melting points of the multi-layered LDPE nanocomposites are higher than that of 1%N nanocomposites. The reason is that the interactions between the molecule chains are more obvious. The interfacial region always increases the area of molecule movement, and it decreases the inhibition of the growth of crystals. The crystallinity ( $X_c$ ) is related to the melting enthalpy ( $\Delta H$ ), and it can be calculated as follows:

$$\chi_c = \frac{\Delta H}{(1 - \phi)\Delta H_{100}} \times 100\% \quad (1)$$

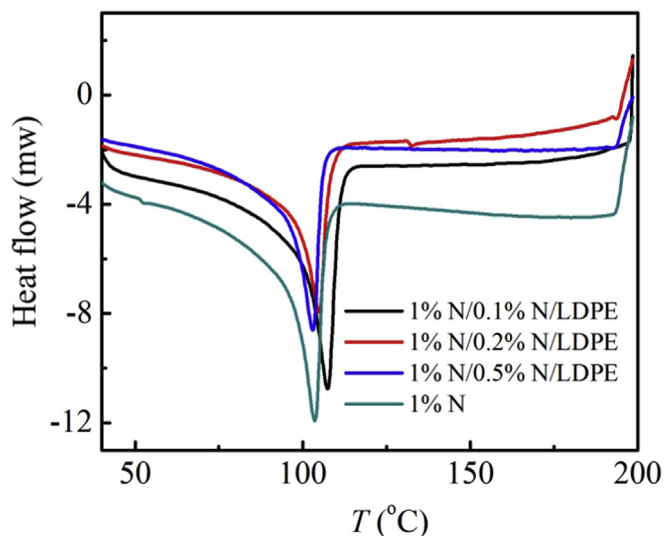


Fig. 3. DSC curves of the multi-layered LDPE nanocomposites.

here,  $\phi$  is the weight percent of  $\text{Al}_2\text{O}_3$ ;  $\Delta H$  is the integrated melting enthalpy;  $\Delta H_{100}$  is the enthalpy of LDPE when the crystallinity is 100% ( $287.3 \text{ J/g}$ ) [15,16]. From the results listed in Table 1, it can be seen that the crystallinity of 1%N/0.1%N/LDPE nanocomposites is higher than that of other gradient-distribution structured and mean-distribution ones. Two reasons could explain it. Firstly, the  $\text{Al}_2\text{O}_3$  particles can provide more nucleation centers of microcrystal and increase the nucleating rate of macromolecules; Secondly, the interfacial interactions between the surface of  $\text{Al}_2\text{O}_3$  and vinyl groups of grafted chains impeded the motions of molecules and lowered the growing rate [17]. Obviously, the larger amount of  $\text{Al}_2\text{O}_3$  is filled into the LDPE matrix, which could easily limit the movement of polymer molecular chains and little space is left for the additional crystallization, resulting in the decrease of crystallization. Thus, the  $X_c$  of 1%N/0.1%N/LDPE nanocomposite is higher than that of the other gradient-distribution structured nanocomposite.

Due to the effect of molecule force, LDPE molecule tends to form stable conformation and the molecule chain is more ordered, which is conducive to form the small spherulites. The gradient-distribution structured nanocomposites with twice hot-pressing tend to the more stable conformation, so the multi-layered LDPE nanocomposites with larger concentration of  $\text{Al}_2\text{O}_3$  have higher crystallinity. The increase of the nucleation rate will make the size of spherulite smaller, and the smaller dispersed spherulites may induce the deep traps in the bulk of LDPE [18].

### 3.3. Breakdown strength

Dielectric breakdown strength of the LDPE nanocomposites is analyzed within the framework of Weibull statistics. Weibull statistical distribution is one important method to analyze the breakdown strength of materials, which reflects the probability of the material at certain field strength ( $E$ ) or the probability of failure or breakdown at a certain time ( $t$ ). Weibull cumulative distribution breakdown field strength can be described as follows.

Table 1  
The melting point and melting heat of the multi-layered nanocomposites.

Samples	$T_g$	$\Delta H$ (J/g)	$X_c$ (%)
1%N/0.1%N/LDPE	107	140	49
1%N/0.2%N/LDPE	105	120	42
1%N/0.5%N/LDPE	104	107	37
1%N	103	99	34

$$P = 1 - \exp \left[ - \left( \frac{E}{\alpha} \right)^\beta \right] \quad (2)$$

here,  $P$  is the cumulative probability of the dielectric breakdown;  $E$  is the breakdown strength for testing,  $\beta$  is the shape factor related to the dispersion of sample, and  $\alpha$  is the characteristic breakdown strength during the cumulative probability of the dielectric breakdown is 63.2%. After twice taking the logarithm, Equation (2) can be described as follows:

$$\log[-\ln(1 - P)] = \beta \log(E) - \beta \log(\alpha) \quad (3)$$

this point  $\log[-\ln(1 - P)]$  and  $\log(E)$  form a linear relationship in the Cartesian coordinate system. According to IEEE Standard 930-2004, when the number of samples is less than 25,  $P$  should be computed by the formula as follows.

$$P_i = \frac{i - 0.44}{n + 0.25} \times 100\% \quad (4)$$

where  $n$  is the breakdown times or voltages in order from smallest to largest and assign them a rank from  $i = 1$  to  $i = n$ .

The Weibull parameters  $\alpha$  and  $\beta$  are shown in the insert table in Fig. 4.  $\alpha$  is found to be strongly affected by the filler modification in the nanocomposites and the voltage ramping rate. The breakdown strength of 1%N/0.5%N/LDPE nanocomposites is lower than that of other composites. The breakdown strength of 1%N/0.1%N/LDPE nanocomposites reaches to 200 kV/mm, which is higher than that of 1%N nanocomposites with the 120 kV/mm. For the gradient-distribution structured nanocomposites, the breakdown strength of 1%N/0.1%N/LDPE is higher than that of the other ones. The reason may be that the amount of  $\text{Al}_2\text{O}_3$  nanoparticles affects the breakdown strength data. When the nanoparticles amount reaches a certain value, the conductivity path will be formed. The  $\text{Al}_2\text{O}_3$  contents of 1%N/0.2%N/LDPE and 1%N/0.5%N/LDPE has over the threshold.

In the theory of electrical strength, usually referred to as free-volume theory, breakdown is initiated by free electrons which are accelerated by electric field in the largest of the holes present in the amorphous phase of all polymers. Breakdown starts when the

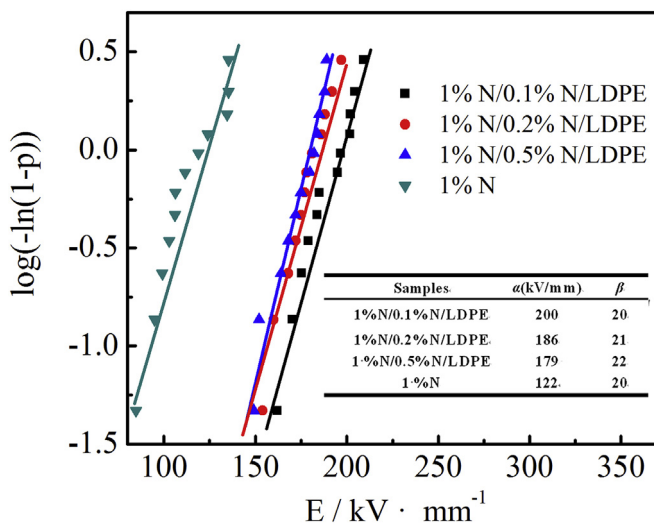


Fig. 4. Weibull-distribution plot of the multi-layered LDPE nanocomposites with the thickness of 480  $\mu\text{m}$ . The insert is the characteristic breakdown strength ( $\alpha$ ) and shape factor ( $\beta$ ).

energy gained by free electrons in field  $E$  on free path of length  $l$  attains with probability  $P = 1$  the barrier energy  $E_u$ . Then all electrons with this energy hop over  $E_u$ , separating the traps in the compact structure [19,20]. The relationship is shown in Equation (5). The molecule motion will make the molecule rearrangement towards to the more stable conformations when the polymer is at the melt point. The distance between the chain branches could increase, due to the decrease of the molecule force. The schematic diagram of hot motion is shown in Fig. 5. The  $l$  becomes shorter after the molecule rearrangement, which result to the  $E_b$  higher. The hot motion of molecule in multi-layers nanocomposites will be easier completed with two-time hot pressing. So the breakdown strength data of multi-layered LDPE nanocomposites is higher than that of single-layer LDPE nanocomposites.

$$E_b = \frac{E_u}{el_e} \quad (5)$$

### 3.4. Space charge

Fig. 6 shows the space charge distribution of LDPE nanocomposites polarized at the electric field of 30 kV/mm for 120 min. Although the interface is not clear from the SEM figure (as shown in Fig. 2a), the interface charge is distinct. With the different concentrations of nano- $\text{Al}_2\text{O}_3$  in the middle layer, the space charge distribution is obviously different. It can be observed from Fig. 6a that the large amount of space charges injected into LDPE materials from the anode and cathode. The space charges aggregated near the electrodes and interface. For the 1%N/0.1%N/LDPE nanocomposites, the injection of space charge from the cathode was inhibited. From Fig. 6b, we can see that the interface charge aggregates between the layers. With the time increasing, the interface charge is larger as shown in Fig. 6c. Fig. 6d is the space charge distribution of 1%N nanocomposite with the single-layer film. The charge injected into the materials is from the anode and cathode. The effect of inhibition for the 1%N nanocomposites is better than that of LDPE [21].

The charge amount in the materials is based on charge density and the thickness of the materials. The charge amount  $Q$  represented in Fig. 6a–d is derived from the experimental results according to Equation (6).

$$Q = \int_0^d |\rho(x)| S dx \quad (6)$$

where  $Q$  is the total trapped charge amount,  $\rho(x)$  is the charge density,  $S$  is the electrode area, and  $d$  is the thickness of the sample. The results are plotted in Fig. 7. The total charge in the materials is increasing as time increased. The rate of charge accumulation in the 1%N/0.2%N/LDPE and 1%N/0.5%N/LDPE nanocomposites is faster than the other composites in 60 min. The total charge accumulated in the 1%N/0.1%N/LDPE nanocomposites is less than those in 1%N/0.2%N/LDPE and 1%N/0.5%N/LDPE nanocomposites at 120 min. The amount of charge accumulated in the 1%N is the least. The possible reason is that the nanoparticles added into the composites improve the energy level of energy band [22], which inhibits the injection of charge. The charge in the materials was induced to the polarity at the beginning time. As the stressing time increased, the charge amount grows with exponential. That may be the injection charge in the materials. The charge of the 1%N/0.2%N/LDPE is more than that of 1%N/0.2%N/LDPE. The reason is that the certain content nanoparticles added into composites could inhibit the injection of charge. When the nano- $\text{Al}_2\text{O}_3$  content is over 1%, the inhibition



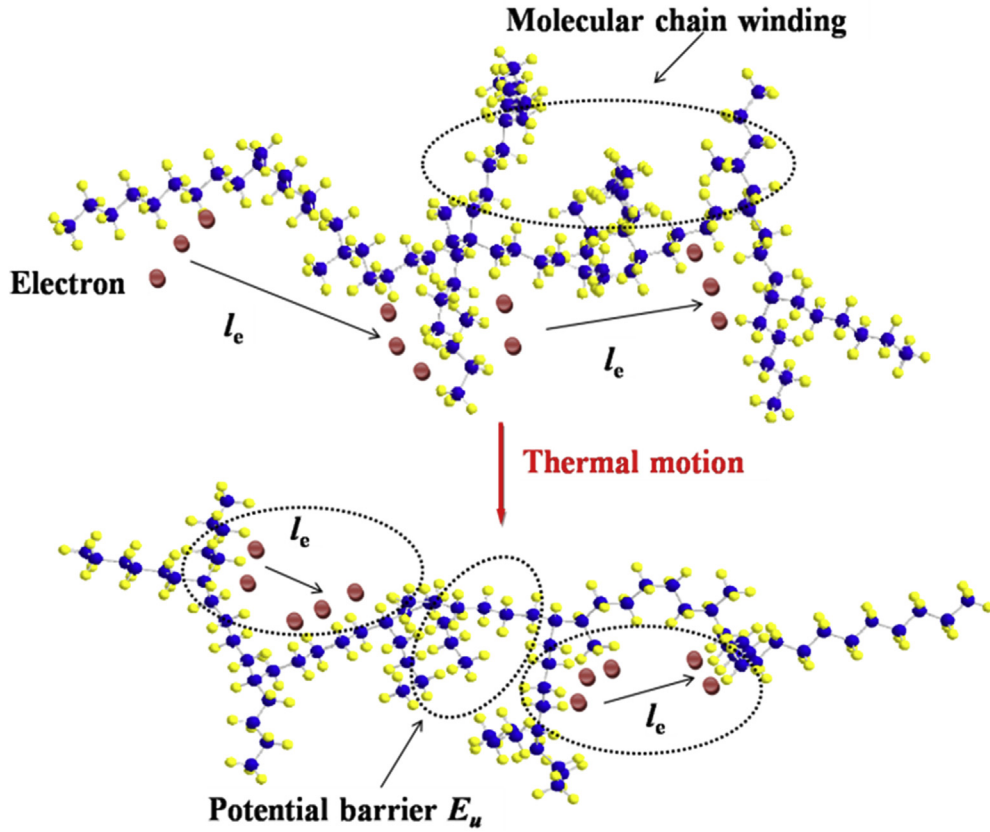


Fig. 5. Schematic diagram of thermal motion of the molecular chains.

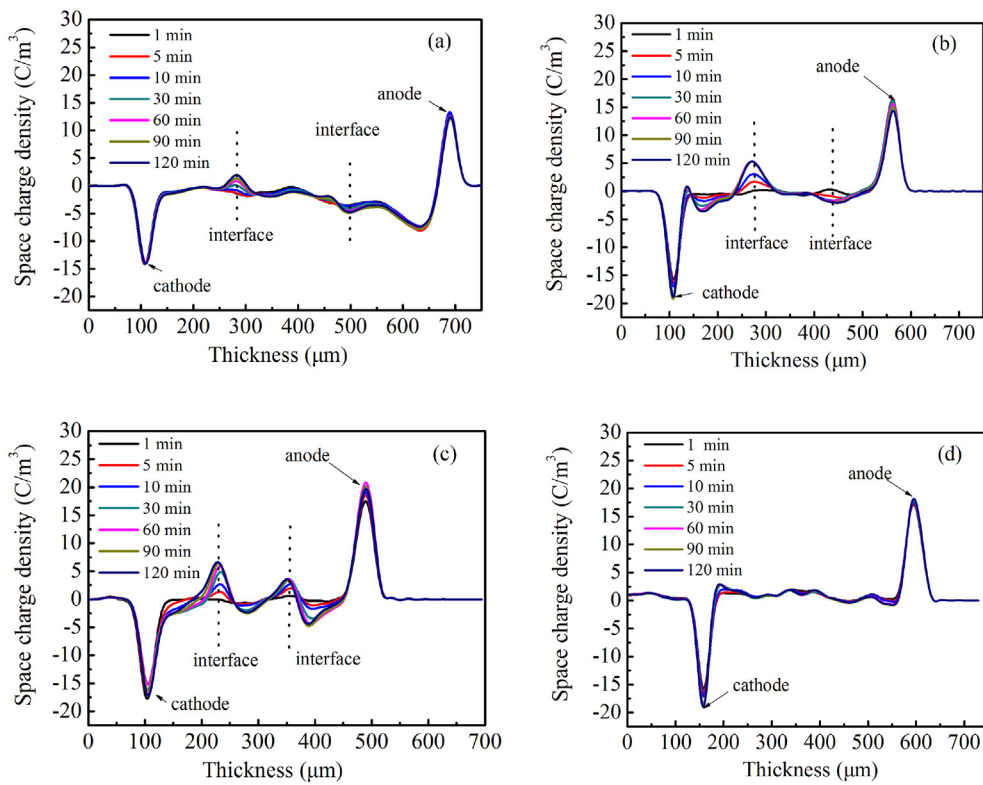


Fig. 6. Space charge distribution plot of multi-layered LDPE nanocomposites with the thickness of 450  $\mu\text{m}$  (a) 1%N/0.1%N/LDPE, (b) 1%N/0.2%N/LDPE, (c) 1%N/0.5%N/LDPE, and (d) 1%N.

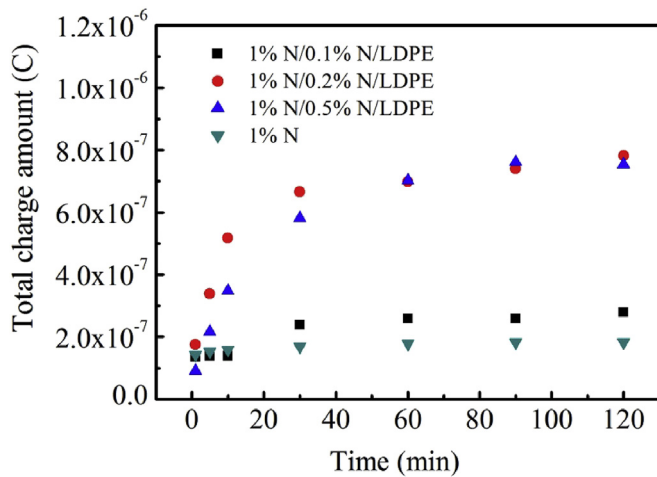


Fig. 7. Total charge amount of the samples.

effect will be decreased, that had been reported in the referents [23–25]. The multi-layered nanocomposites with 1%N/0.1%N/LDPE are benefit to suppress the charge injection. Compared the total charge in the 1%N/0.1%N/LDPE nanocomposites with 1%N, the charge is similar.

#### 4. Conclusions

The gradient-distribution  $\text{Al}_2\text{O}_3$ /LDPE nanocomposites with multi-layered structure were prepared in this paper. Compared with the general mean-distribution structured monolayer composites, these composites with unique morphology have excellent electrical properties as follows. For the gradient-distribution structured LDPE nanocomposites, there was no clear interface between the adjacent layers, though they were prepared through two-time hot pressing progress. This continuous morphology can well avoid the air layer. Meanwhile, the polymer molecules in thermal motion during the hot pressing process, makes the whole molecule rearrangement. This rearrangement of molecule chains contributes to its forming stable conformation, which has the minimum molecular force. Space charge amount of the gradient-distribution structured LDPE nanocomposites is less than that of the mean-distribution structured ones as the trap near the composites surface can capture the injected charges. The captured charges can distort the electric field in the vicinity of the interface, which will decrease the charge injection. Thus, the space charges can be suppressed. Meanwhile, the nano- $\text{Al}_2\text{O}_3$  tends to distribute in the interface between the neighbouring layers. It suggests that the carriers prefer to conduct through this interaction zone, then causing the space charge suppression.

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