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Insulating Properties of Low Density Polyethylene/Alumina Nanocomposites

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Abstract: High-Voltage Direct Current (HVDC) cable has become an important factor in the development of HVDC transmission. However, there are still difficulties such as space charge distribution in the development of HVDC cables. In this study, Low Density Polyethylene (LDPE) nanocomposites filled with nano alumina (nano-Al_2O_3) particles with or without modification were prepared by employing melting blend method. The results show that the modified nano-Al_2O_3 particles with vinyl silane coupling can be homogeneously dispersed in LDPE matrix. Space charge of the LDPE nanocomposites was tested by Pulsed Electro-Acoustic (PEA) method. The PEA spectrum indicated that decay charge effect of the LDPE/Al_2O_3 nanocomposites is better than that of neat LDPE. The space charge accumulated in the nanocomposites can be effectively decayed. The *J-V* curves show the nano-Al_2O_3 decrease the carrier mobility. The excellent insulation properties of the LDPE nanocomposites were attributed to the better interfacial adhesion between the surface-treated nano-Al_2O_3 particles and the matrix.

Keywords: Polyethylene, Al_2O_3, Composites, Space Charge, Decay, *J-V* Curves

Introduction

Low Density Polyethylene (LDPE) is widely used as bulk insulation in extruded cables, but under High-Voltage Direct Current (HVDC) the space charge accumulated in the LDPE can greatly distort the electrical field and then influence the aging property. The reason is probably that the physical and chemical defects existed in LDPE could lead to the increasing local electric field so that the insulating properties would decrease. To solve the problem, the inorganic nanoparticles and polarity substance have been added into PE. The effect of nano-sized particle additives on the electrical properties of dielectrics is currently of considerable interest (Olthuis and Bergveld, 1992; Liu and Chen, 2013; Wu *et al*., 2014; Kim *et al*., 2013; Fleming *et al*., 2011; Ju *et al*., 2014; Takada *et al*., 2008; Hosier *et al*., 2010).

In this study, LDPE nanocmposites filled with the nano-Al_2O_3 particles with and without surface modification were prepared. Space charge characteristics in LDPE nanocomposites were measured using the Pulsed Electro Acoustic technique (PEA). The charge decay in both types of the samples was also tested. Traps and carrier mobility of LDPE nanocomposites were also discussed.

Experimental

Materials

LDPE (LD200BW) with density of 0.922 g/cm^3 and melt flow rate of 2.3 g/10 min was purchased from Sinopec Beijing Yanshan Co., China. The nano-Al_2O_3 particles were supplied by Institute of Process Engineering Chinese Academy of Sciences (China) and they were modified by vinyltrimethoxysilane (VMES, V-Al_2O_3) prior to use. The reaction route is presented in Fig. 1.

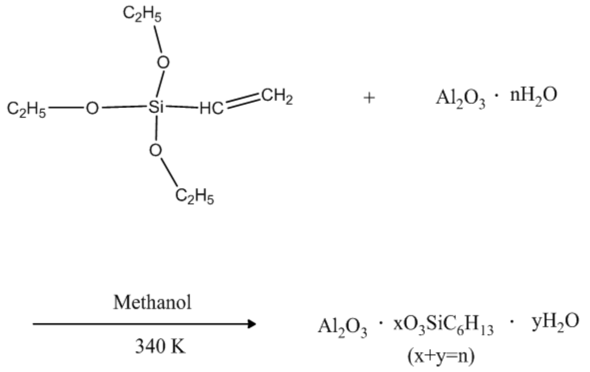


Fig. 1. Schematic of the reactions between the original Al_2O_3 and the silane.

The different contents (0.1, 0.2, 0.5, 1 wt%) of nano-Al_2O_3 particles were mechanically mixed with LDPE granules using the HAAKE PolyLab mixer (HAAKE Rheomix 600, Germany) with the processing temperature of 130°C. The neat LDPE and Al_2O_3/LDPE nanocomposite films were prepared by a hot-press method at the temperature of 150°C and the pressure of 15 MPa. The prepared films were placed in a vacuum oven at 80°C for 24 h and then cooled down to room temperature to eliminate thermal history. To banish the remainder charge, the films were put between two polished copper plates in a vacuum oven at 80°C for 48 h short-circuiting.

Characterization

Carrier mobility of the samples was tested with a Keithley electrometer model 6517 B. The samples with the diameter of 54 mm and the thickness of 0.4 mm were prepared for testing. The polarization current was obtained after short-circuiting for 5 min. The polarization voltage was set at 1 kV.

The space charge distribution was tested by PEA (Shanghai Jiao Tong University) carried out under an electrical field of 30 kV/mm for 60 min at 25°C. During the measurement, the samples with the size of 400 μm ×7 cm ×7 cm were sandwiched between an aluminum electrode and a semi-conductive polymer electrode (the diameter is 12 cm and 2 cm, respectively).

Results and Discussion

Figure 2 shows polarization currents measured for 5 min. The current decrease exponentially with time increasing and subsequently reaches an equilibrium value. The polarization currents curves of LDPE reach the equilibrium value at 100 s, while the Al_2O_3/LDPE nanocomposites filled with 1 wt% nano-Al_2O_3 only at 50 s. The equilibrium value of polarization current of LDPE is much larger than that of 1 wt% Al_2O_3/LDPE nanocomposite. It could be indicated that a large amount of charges inject into the LDPE. However, the nano-Al_2O_3 can inhibit the charges to inject into the interior of nanocomposite and prevent most of charges gather on the surface of the samples (Nelson and Hu, 2005).

The current density-voltage (*J-V*) curves obtained from the equilibrium value of polarization current at different voltages were shown in Fig. 3. The *J-V* curves are consistent with the well-known Mott and Gurney equation (Meunier *et al.*, 2001). At low applied voltages the *J-V* characteristics may follow Ohm's law:

$$J = qp_0\mu_p \frac{V}{d} \tag{1}$$

Where:
p_0 = The density of thermally generated free carriers inside the sample
μ_p = The carrier mobility
d = The thickness of the sample. The slope of the Equation 1 was given as follow Equation 2:

$$k_1 = qp_o\mu_p \tag{2}$$

Due to the increased space charge, quadratic relationship of *J-V* curves fitting is obtained at high applied voltages as shown by Equation 3:

$$J = \frac{9}{8}\varepsilon_r\varepsilon_0\mu_p \frac{V^2}{d^3} \tag{3}$$

where, ε_r is the relative permittivity of the material and $\varepsilon_0 = 8.854\times10^{-12}$ F/m, is the permittivity of vacuum. Then the carrier mobility μ_p are calculated from the fitting constants k, which have given as follows Equation 4:

$$k = \frac{9}{8d^3}\varepsilon_r\varepsilon_0\mu_p \tag{4}$$

The carrier mobility of Al_2O_3/LDPE nanocomposites calculated by the above equations is 1.54×10^{-11} cm^2/ (V·s) and which of the V-Al_2O_3/LDPE nanocomposites is 9.97×10^{-12} cm^2/ (V·s), which is lower than the published values of LDPE (Meunier *et al.*, 2001). The addition of Al_2O_3 decreases the carrier mobility of composites, especially the Al_2O_3 modified by silane coupling agent, about an order of magnitude.

Figure 3 shows the Al_2O_3/LDPE nanocomposites follow Ohm's law at low applied voltages (0-200 V) in the *J-V* curves. It can be seen that the area of the Ohm's law of the V-Al_2O_3/LDPE nanocomposites is larger than that of Al_2O_3/LDPE nanocomposites without modification, indicating that the V-

Al_2O_3/LDPE nanocomposites have better insulation. The *J-V* curves of Al_2O_3/LDPE nanocomposites and V-Al_2O_3/LDPE nanocomposites are similar to schottky curves. It demonstrates that the main mechanism of charge injected into from electrode to dielectric interface is Schottky injection.

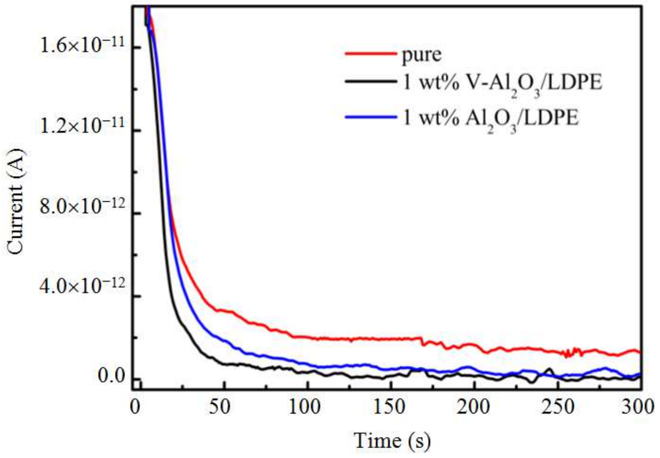


Fig. 2. Polarization currents curves of Al_2O_3/LDPE and V-Al_2O_3/LDPE nanocomposites at the applied of 1 kV

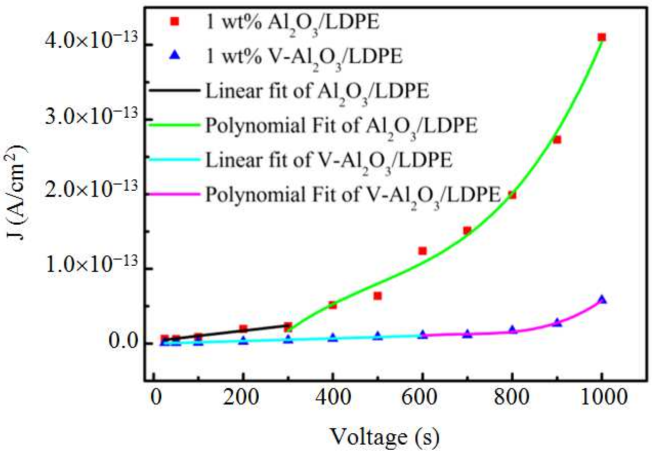
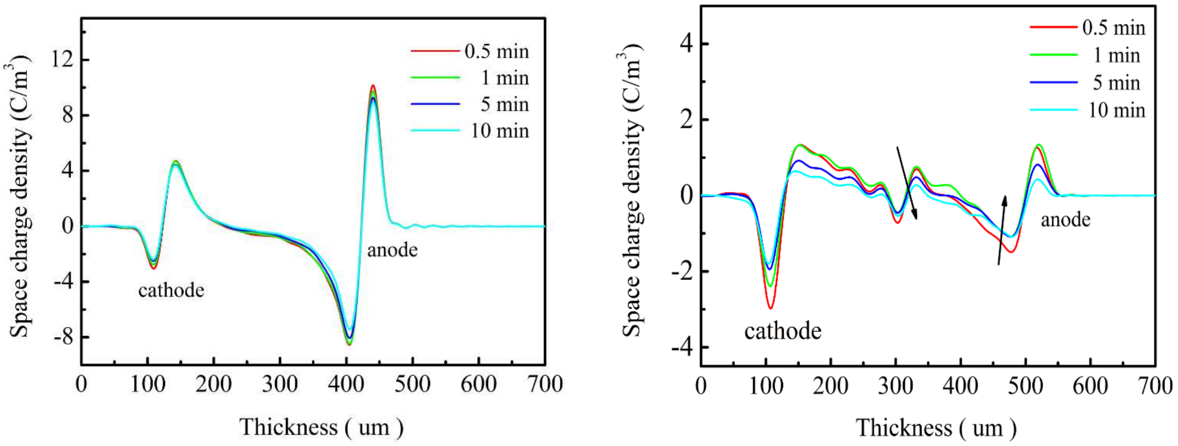
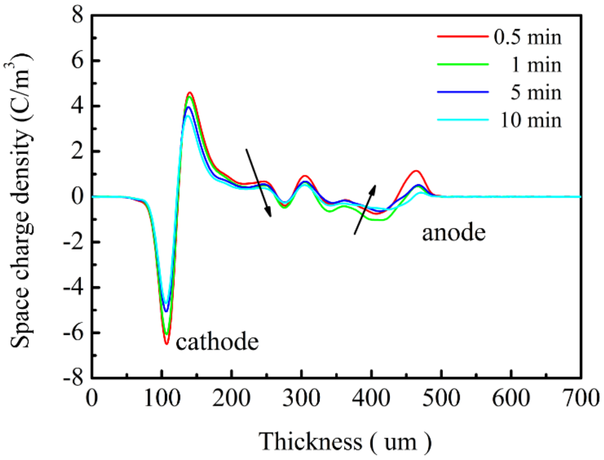


Fig. 3. Current density-voltage (*J-V*) curves from the equilibrium value of polarization currents at 25-1000 V of Al_2O_3/LDPE and V-Al_2O_3/LDPE nanocomposites



(a) (b)



(c)

Fig. 4. Space charge decay after removal of 30 kV stressing in 120 min on LDPE sample (a) neat LDPE (b) Al_2O_3/LDPE nanocomposites (c) V-Al_2O_3/LDPE nanocomposites

The space charge decay for both types of LDPE samples are shown in Fig. 4. Both positive and negative charges were introduced into the neat LDPE near the cathode and anode, respectively, was observed in Fig. 4a. Many researchers believe that electrons were injected into the samples from the two electrodes. Fig. 4b shows the negative charges in the samples, migrated in the bulk, reducing the height of the positive peak. After 10 min, the negative peak totally disappears at the anode and negative charges are now accumulated in the bulk of samples. The electrons in Al_2O_3/LDPE composites have a higher mobility than the neat LDPE, they may disappear very quickly. The reason is that the charges move from the anode to the cathode by negative trapped charges. It is likely that a potential energy barrier exists for charges moving from sample to the anode.

As a result, the charges gather near the electrode and some of them get retrapped near the cathode. As can be seen from Fig. 4c, the injected charge in the cathode is fairly smaller than the Al_2O_3/LDPE composites. It may due to that the silane coupling can enhance the compatibility between LDPE and nano-particles and decrease the defects in the interface.

It makes the nano-Al_2O_3 become effective trapping site to fix carriers and decrease the carrier mobility. A charges injected from electrodes generate in the deep trap induced by the nano-Al_2O_3 after electric voltage applied. The charges are difficult to move and accumulate near the electrodes to produce an additional electric potential reversed to the applied voltage. It inhibits the dissociation of small molecule and prevents further injection of the charges. Therefore, the space charge inside the sample is reduced.

Conclusion

In summary, the LDPE nanocomposites filled with the unmodified Al_2O_3 and V-Al_2O_3 nanoparticles were prepared by using melt mixing and hot press methods, respectively. The main conclusions are as follows.

Modified nano-Al_2O_3 particles show good dispersion in LDPE matrix compared with the unmodified nano-Al_2O_3 particles.

The addictive nanoparticles bring more deep traps and shallow trap, resulting in the effect of suppressing space charge. The suppressing effect of V-Al_2O_3/LDPE nanoparticles is better than that of unmodified one.

The additives of V-Al_2O_3 could decrease the carrier mobility. The V-Al_2O_3/LDPE nanocomposites show excellent decay charge effect.

Acknowledgment

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Author's Contributions

Si-Jiao Wang participated in all experiments and contributed to the writing of the manuscript. Jun-Wei Zha designed the research plan, organized the study, coordinated the data-analysis and contributed to the revising of the manuscript. Yun-Hui Wu and Hong-Da Yan participated in the experiments. Zhi-Min Dang coordinated the data-analysis and contributed to the revising of the manuscript.

Ethics

The authors declare no competing financial interest.

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