Results in Physics 11 (2018) 52–57

Contents lists available at ScienceDirect

Results in Physics

journal homepage: www.elsevier.com/locate/rinp

Effect of TiO_2 size factor on the electrical properties of polyethylene matrix dielectrics

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ARTICLE INFO

Keywords:
TiO_2
LLDPE
Breakdown field strength
Space charge
Size effect

ABSTRACT

In this study, the TiO_2 nanoparticles and TiO_2 nanofibers were chosen as inorganic fillers to dope into the linear low density polyethylene (LLDPE). The TiO_2 nanofibers were prepared by the electrospinning method, the diameter and length of TiO_2 nanofibers were about 100 nm–200 nm and 3 μm–4 μm, respectively, and the size of the purchased TiO_2 nanoparticles was about 100 nm–200 nm. The microstructure of TiO_2 fillers and their composites was analyzed by the X-ray diffraction (XRD) and scanning electron microscopy (SEM). The breakdown field strength and space charge characteristics of the LLDPE/TiO_2 nanoparticles and LLDPE/TiO_2 nanofiber composites with different sizes of inorganic fillers were investigated. The results showed that the LLDPE/TiO_2 nanofiber composites had higher breakdown strength than LLDPE/TiO_2 nanoparticle composites at the same doping content. The breakdown strength of 0.5 wt% LLDPE/TiO_2 nanofiber composites (237 kV/mm) was higher than that of pure LLDPE (223 kV/mm) and 0.5 wt% LLDPE/TiO_2 nanoparticle composites (205 kV/mm). In addition, the space charge accumulation and electric field distortion in the composites were significantly relieved by doping TiO_2 nanofibers compared with that of doping TiO_2 nanoparticles.

Introduction

High voltage DC transmission has higher transmission capacity, lower loss, longer transmission distance, lower wiring cost, and stable system than the AC transmission. So that it has attracted the attention of scholars. As the voltage level of HVDC transmission increases, the insulation requirements also enhance, so it is necessary to find an insulation material that can withstand high voltage [1,2]. At present, polyethylene is the main insulating material for HVDC transmission. Linear low density polyethylene has the characteristics of stable chemical properties, high melting temperature and easy processing, which can be used as insulated cables. However, the existence of space charge limits the development of polyethylene insulated cables. The injection of space charge makes the internal electric field of the cable distorted. When insulated cables working under high voltage for a long time, it easily causes the dielectric materials to age, affects its service life, even causes the electric breakdown [3–6]. As early as 1994, T J Lewis proposed the concept of nano-dielectrics. Adding a small amount of nanoparticles in the polymer could effectively improve the electrical properties of composites [7]. Tanaka later proposed a multi-core model to analyze the various phenomenon of polymer-based nanocomposites, the “interaction zones” between the interface of fillers and polymer matrix played an important role on the resistance of the nanocomposites [8]. Takada et al. proposed that under high-field strength, nanoparticles filled in a polymer induced dipoles, formed potential wells, and restricted the free movement of carriers [9]. He et al. found that LDPE/MgO interface has a significant effect on the electrical properties of nanocomposites by adding MgO nanoparticles to polyethylene. The addition of MgO nanoparticles was available to suppress the production of space charges and enhance the DC breakdown strength. Thermally stimulated currents of composites reveal strong correlation between the traps and electrical properties of composites [10]. Wang et al. prepared XLPE/SiC composites with different concentrations. The test results show that the composites with concentrations of 1 wt% and 3 wt% have higher DC breakdown field strength than neat XLPE. It was also reported that nano-Al_2O_3/polyimide composites exhibited excellent

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https://doi.org/10.1016/j.rinp.2018.08.024
Received 5 June 2018; Received in revised form 25 July 2018; Accepted 13 August 2018
Available online 18 August 2018

electrical performance due to the deeper trap level and higher trap density due to introduced nano-fillers [11–13]. Based on the space charge behavior, all composites could suppress space charge accumulation, but the composite with a concentration of 1 wt% exhibited the best effect [14]. A series of studies have shown that adding a small amount of inorganic fillers into the polymer can significantly improve the electrical properties of the dielectric composites [15–19].

In this work, TiO_2 nanoparticles (TiO_2-P) and nanofibers (TiO_2-F) were used to study the effect of TiO_2 size factor on the electrical properties of polyethylene composites. The microstructure of the composites was characterized by XRD, TEM and SEM. At the same time, the breakdown field strength, space charge accumulation, and electric field distortion of the dielectric composites were investigated.

Experimental section

Materials

Linear low-density polyethylene resin (DFDA-7042) was purchased from China National Petroleum Corporation. Titanium dioxide (TiO_2) nanoparticles, tetrabutyltitanate ($C_{16}H_{36}O_4Ti$), ethanol and acetic acid were supplied by Sinopharm Chemical Reagent Co., China., polyvinylpyrrolidone (PVP, Mw = 1,300,000) was supplied by Alpha Corporation. Dopamine hydrochloride ($C_8H_{11}NO_2$·HCl) and tris-(hydroxymethyl)-aminomethane ($C_4H_{11}NO_3$) were provided by Aladdin. All other chemicals were obtained as analytical grade products and used without further purification.

Preparation of inorganic nanofibers

TiO_2 nanofibers were prepared by electrospinning with a typical synthesis procedure. Firstly, 2 g PVP was dissolved in 20 mL ethanol to form PVP alcohol solution A (Sol A). Secondly, 4.66 mL ethanol, 4 mL acetic acid and 2 mL $C_{16}H_{36}O_4Ti$ were slowly dripped into the beaker B and stirred for 2 h to form a homogeneous and stable solution B (Sol B). Then put Sol B into Sol A and stirred for 2 h to form a spinning precursor. The precursor was electrospun at 1.5 kV/cm and the injection rate was 0.3 mm/min. Next, the fibers were calcined at 500 °C for 4 h with the raising temperature rate of 2 °C/min to obtain TiO_2 nanofibers.

In order to improve the interfacial compatibility between the inorganic filler and the polyethylene matrix, all inorganic fillers were surface-modified. The mass ratio of dopamine (PDA) to inorganic fillers was 1:10. Firstly, inorganic fillers were added into 2 g/L $C_4H_{11}NO_3$ standard buffer. Secondly, the pH value of the solution was adjusted to 8.5 by the addition of dilute hydrochloric acid, followed the PDA was added. Then, PDA modified inorganic fillers (TiO_2-P and TiO_2-F) were washed with deionized water and then dried in a vacuum oven at 80 °C for 12 h.

Preparation of composites

The composites were prepared as followed. The two roll mill was heated and kept at 125 °C. The LLDPE/TiO_2 mixture was poured into the two rolls with a certain ratio and each of the components was mixed for 20 min. After being uniformly dispersed, they were placed in a mold of different thickness, such as 100 μm and 280 μm, and the samples of different thickness were prepared by hot pressing with a flat plate vulcanizer and pressurized by a gradient. The pressure was held for 5 min each time and finally the samples were obtained.

Characterization

The crystalline structures of TiO_2 and LLDPE-based composites were performed by an X-ray diffractometer (PANalytical Empyrean XRD). Transmission electron microscopy (TECNAI Type 2–12 produced in the Netherlands) was used to observe the dopamine encapsulation. The size and distribution of fillers in the matrix were observed by scanning electron microscopy (SEM).

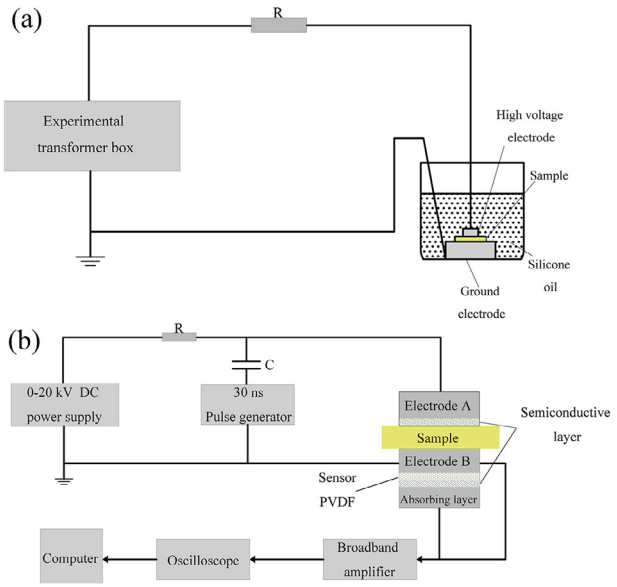

Fig. 1. Schematic diagram of the measuring device. (a) DC breakdown experimental, (b) PEA method.

DC breakdown

For DC breakdown strength measurement, the sample with average thickness of 100 μm was immersed in transformer oil to inhibit surface flashover discharging under flat electrode at room temperature. The DC voltage was continuously increased at the rate of 3 kV/s until the sample broken down. The two-parameter Weibull distribution was used to analyze the DC breakdown characteristics of the samples. Schematic diagram of DC breakdown measurement was shown in Fig. 1(a).

Space charge behavior

The pulsed electro-acoustic (PEA) method was used to measure space charge. Before testing the space charge, the sample was coated with the aluminum electrodes by a coating machine (ZHD-400 manufactured by Beijing Science and Technology Co., Ltd.). The electrode diameter was 25 mm. Then, the sample was short-circuited at 80 °C in a vacuum circumstance for 24 h to remove charges. The thickness of the samples was in the range from 260 μm to 300 μm. The charge density was measured at a electric field of 40 kV/mm. Schematic diagram of the measuring device for PEA method was shown in Fig. 1(b).

Results and discussion

The XRD is always used to evaluate the crystallization structures and phase structures of materials, especially for the composite materials. The crystalline structures of inorganic filler phase and the LLDPE-based composites are shown in Fig. 2. As shown in Fig. 2(a) and (b), the characteristic diffraction peaks of TiO_2-P and TiO_2-F can be indexed to the standard card (JCPDS No. 89–4921), indicating that TiO_2-F produced by electrospinning technology does not have any other impurity phase, TiO_2-P and TiO_2-F are anatase crystal forms. The characteristic diffraction peaks of LLDPE at $2\theta = 21.5°$, 23.7°, and 36°. At the same time, in the LLDPE/TiO_2-P and LLDPE/TiO_2-F dielectric composites, the characteristic peak of TiO_2 is found at $2\theta = 25.4°$, with the increase of doping content, the intensity of the characteristic diffraction peak increasing, the results indicate that the addition of inorganic fillers does not destroy the structure of the LLDPE matrix and does not react with