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Effect of epitaxial crystallization on packet-like space charge characteristics in low-density polyethylene under multi-field coupling conditions

Ling Zhang [a], Yuanxiang Zhou [a, *], Chenyuan Teng [a, b], Yunxiao Zhang [a], Ming Chen [a], Zixia Cheng [b]

[a] State Key Laboratory of Control and Simulation of Power Systems and Generation Equipments, Department of Electrical Engineering, Tsinghua University, Beijing 100084, China
[b] School of Electrical Engineering, Zhengzhou University, Zhengzhou 450001, China

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ABSTRACT

In order to study the effect of epitaxial crystallization on charge transport in low-density polyethylene (LDPE) under multi-field coupling conditions, three typical epitaxial crystallizations, namely disorder (glass substrate), crossover (isotactic polypropylene substrate), and parallel (polytetrafluoroethylene substrate), were prepared and denoted as LD-G, LD-iPP, and LD-PT, respectively. Packet-like space charge through samples was analyzed by the pulsed electro-acoustic (PEA) method. It is shown that different microscopic surface morphologies appeared in the LDPE samples with different epitaxial crystallizations, which, however, do not change the crystalline structure of the bulk. Packet-like space charge phenomena were observed and the distortion field increased with the temperature which could be attributed to the larger amount of charge injection in a shorter period. The differences of the amount and injection rate of the space charge were explained and verified considering the typical chain alignment of epitaxial crystallization, which, in our opinions, contributes to the pass over of positive charge in LD-iPP samples.

1. Introduction

Polyethylene, which has excellent insulation properties such as low carrier mobility and high trap concentration, is the most common and development potential insulating dielectrics in the field of HVDC power cable [1,2]. In the past decade, researches on the extra/ultra HVDC extruded power cable have been a hot spot [3]. Moreover, triple jumps of cross-linked polyethylene (XLPE) insulated HVDC cable have been tested or successfully commissioned in HVDC transmission projects in China from ±160 kV, ±200 kV, to ±320 kV since 2012 [4]. With the continuous rise of DC voltage level, the designed operating field within the cable insulation could reach 20 kV/mm, or even higher. In high DC field, space charge formation, transport, accumulation, and dissipation will severely distort internal field distribution, influencing material degradation and electrical breakdown [5,6]. Except withstanding high DC field, HVDC cable insulation also experience temperature field caused by the conductor core in the actual operation [7]. Thus, systematically investigation on space charge effect under multi-field coupling conditions is indispensable to promote the technical reserve of HVDC cable.

Epitaxial crystallization phenomenon was firstly found in the natural minerals in 1817 and is defined as oriented growth of one type of materials on the other substrates surface [8]. Study on the polymeric epitaxial crystallization started from 80's in the last century [9]. And it has been proven that the mechanical, electrical, and photoconductive properties could be improved by the introduction of epitaxial crystallization in the polymeric matrix [10,11]. In the real manufacture of cable-used insulating materials, issue of epitaxial crystallization occurred in the interface between insulation layer and other materials. Zhou Y X preliminarily studied the epitaxial crystalline behavior and its relationship with space charge characteristics under room temperature [12]. Zheng C J selected mica and polyester separately as the substrate to prepare LDPE, and he believed the variation of space charge and conduction current should be attributed to the different crystalline structure on the surface of LDPE films [13].

* Corresponding author.
E-mail address: zhou-yx@tsinghua.edu.cn (Y. Zhou).

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This paper prepares LDPE film samples with three typical epitaxial crystallization system, namely parallel, crossover, and disorder. An improved pulsed electro-acoustic method (PEA method) system was adopted to investigate the space charge behavior under multi-field coupling conditions. The influence and the mechanism of epitaxial crystallization on the space charge formation and transport process in LDPE samples was discussed.

2. Experiment details

2.1. Material and sample preparation

LDPE pellets were purchased from Sigma-Aldrich (CAS 9002-88-4) with the density of 0.925 g/mL(25 °C), melt point of 116 °C, and melt index of 25 g/10min. According to the orientation relationship on the surface of the polymer, the system of epitaxial crystallization in polymer could be classified as disorder, crossover, and parallel alignment of chain segment. Thus, three types of basement material, namely glass(G), isotactic polypropylene(iPP), and polytetrafluoroethylene(PTFE), were chosen in order to form the epitaxial structure mentioned above. In the detailed preparation process, LDPE pellets were first wrapped in basement film and pre-heated for 3min in the hot mold in advance. Then, the hot press was performed under 145 °C and 10 MPa for 6min. After that, LDPE films with different epitaxial crystallizations were annealed in air with the thickness of 200 ± 15 μm, and denoted as LD-G, LD-iPP, and LD-PT.

2.2. Material characterization

The surface morphology of LDPE samples was examined using Bruker Dimension Icon Atomic Force Microscope(AFM) performed in tapping mode. Both height and phase images were simultaneously collected under ambient condition at a scan rate of 1 Hz.

A Rigaku SmartLab X-ray diffraction(XRD) system was adopted to test the crystallinity and crystal cell parameters of LDPE samples. The wave length was set as 1.5418 Å, the voltage as 40 kV, and the current as 40 mA. XRD was performed over angles ranging from 20° to 50° with the scan rate of 4°/min.

2.3. Space charge measurement

Space charge behavior under multi-field coupling conditions were tested using pulsed electro-acoustic (PEA) method [14]. A 9 μm gold-coated β-polyvinylidene fluoride(PVDF) film was adopted as the piezoelectric sensor, which possesses the operating frequency range of 0–500 MHz. The temperature of the PEA unit was conditioned via an oven up to 70 °C. The repetitive frequency, voltage amplitude, and pulse width of solid-state high voltage pulse generator were 400 Hz, 400 V, and 4.5 ns, respectively. In this work, due to the applied DC fields were chosen as −100 kV/mm, the PEA system has relatively good signal-noise ratio and high system spatial resolution of 15 μm. LabView software platform was chosen as the interactive platform among PC, oscilloscope, and programmable power supply. Details of the PEA system could refer to Fig. 1. The polarization time was 60min and the depolarization time was 10min with the acquisition interval of 3s. The temperature was selected as 20 °C, 40 °C, and 60 °C. Each group was tested at least twice to establish the reproducibility.

3. Results

3.1. Material characterization results

Fig. 2 shows the AFM images of the microscopic surface morphology of LDPE samples. And from Fig. 2c, parallel oriented lamella occurs on the surface of LD-PT film with the size of micrometer level. As for Fig. 2b, mass overlapping lamella occurs on the surface morphology of LD-iPP with different heights and direction. Fig. 2a is the surface morphology of LD-G group, which has no induction relationship between substrate and polymer.

Fig. 3 and Table 1 present the XRD curves, crystal cell parameters and crystallinity of LDPE samples, and no obvious difference could be seen directly from the figure, which might mean that the macroscopic internal crystalline structure of LDPE samples was not altered even with the introduction of epitaxial crystallization. Moreover, it is worth mentioning that the detection depth of the XRD method could be as deep as tens of micrometers, but the thickness of the epitaxial crystallization is just tens of nanometers and up to a hundred nanometers, from which it could be concluded that the XRD method is not sensitive enough for the characterization of epitaxial crystallization.

3.2. Space charge test results

The space charge dynamic evolution process of LDPE samples was vividly presented in the 2-dimention colorful plot in Fig. 4. From Fig. 4a–c, it is quite different for the space charge polarity, formation, transport, and accumulation process under −100 kV/mm at 20 °C. In LD-G samples, positive charges injected at the first 10min and then they accumulated in the vicinity of the cathode which last almost 30min. In LD-iPP samples, positive charge accumulation appeared earlier and just last for 10min, then a huge amount of negative charge stayed near the anode since 20min. As for LD-PT group, the injection of both positive and negative packet-like space charge was observed after the polarization and recombination process happened in the bulk of the samples.

During the rise of the temperature, space charge evolution is not only accelerated but also encountered different influences due to the diversity of charge carriers. In the case of the results at 40 °C, negative charge quickly injected and transported to the anode and then it seemed to retreat as the advance of positive charge front in LD-G samples. In LD-iPP group, only small negative packet-like space charge moved fast towards the anode repetitively. In LD-PT samples, only positive packet-like charge was observed within 60min polarization and negative charge in the vicinity of the cathode appeared to recombine positive ones. Fig. 5 gives out transient charge behavior within 60 s at 40 °C for the three groups. It could be clearly seen that negative charge already accumulated in the vicinity of the anode in both LD-G and LD-iPP groups at the very beginning of 3s. Then, positive packet-like space charge formed in the anode and move towards the cathode. However, in the LD-PT group no negative charge could be observed within 60s though positive packet-like space charge still appeared. In Fig. 4c1 to 4c3, fierce space charge dynamic evolution process ended in a very short time and then charge distribution entered into a relatively steady state at 60 °C.

4. Discussion

4.1. Effect of temperature on the field distortion

Study on the high field charge transport properties in polymers is of great interest and the reason is that charge transport is strongly associated with electric field distortion, power dissipation, electrical aging, and dielectric breakdown. The conductivity of polymeric dielectrics is sensitive to the temperature and electric field. Thus, the associated internal field variation in LDPE samples is crucial. Fig. 6 presents the maximum distortion field varied with polarization time at different temperatures. Combined with the