Effect of surface topography and morphology on space charge packets in polyethylene

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Abstract. Polyethylene (PE) is a major kind of internal insulating material. With great progresses of space charge measurement technologies in the last three decades, lots of researches are focused on space charge in PE. The heat pressing and annealing condition of polyethylene affect its morphology obviously. During the heat pressing, the surface of PE forms different surface topographies because of different substrate materials. Surface topography has great relation to the epitaxial crystallization layer and influences the space charge characteristic of PE dramatically. This paper studied the formation process of different surface topographies and their micrographic characters in low density polyethylene (LDPE). pulsed electro-acoustic (PEA) method was used to measure the space charge distribution of samples with different surface topographies and morphologies in LDPE. The effect of surface topography and morphology to space charge packet were studied. The surface topography has great influence on space charge packet polarity and morphology has influence on both movement speed rate and polarity of space charge packet.

1. Introduction

Polyethylene is widely used in electrical insulation because of its outstanding insulation performance and mechanical properties. Space charge always plays a very important role in electrical properties of dielectrics. Space charge is well known to distort the local electrical fields, and to affect conduction, breakdown and aging processes [1-3], and thus affect the properties and service life of dielectrics [4]. The space charge packet phenomenon, which appears under higher electrical fields, distorts the local electrical field even more seriously [5-7], and affects the properties of insulating dielectrics more obviously.

The annealing method affects the morphology of polyethylene significantly [8]. During the heat compression moulding process, the surface of the samples forms different epitaxial crystallizations with different substrate materials [9-11]. The epitaxial crystallization affects the space charge properties remarkably according to the research in this paper.

Epitaxial crystallization is the oriented growth of one material on another substrate material, it is a kind of Surface-Induced oriented crystallization. The research of polymer epitaxial crystallization on inorganic salts started during the 1950s, and the epitaxial crystallization between polymers is a new area started during the 1980s [9]. There is research which proves that the cooling rate affects the epitaxial crystallization [12].
It is proved that the epitaxial crystallization affect the mechanical properties of materials [9, 10, 13]. In this paper, the low-density polyethylene (LDPE) was hot pressed with different cooling rate on two different substrate materials to get samples with different epitaxial crystallizations and morphologies, and then observed the space charge packet movement in different sample to research the effect of epitaxial crystallization and morphology on space charge packet.

2. Samples and experiments

2.1. Samples
The raw material used in this paper was pellet LDPE. Its characteristics are as follows: density 0.92 g/cm³, melt index 4.0 g/10 min. Pellet samples were laminated between two pieces of Polytetrafluoroethylene (PTFE) or two glass plates in an oven preheated at 180 °C. They were pressed using a 5 kg steel block, and kept at this temperature for 30 min so as to make the samples melt sufficiently. Then LDPE samples were cooled at a low rate (0.03 °C/s, cooled with hot steel block together), a normal rate (10 °C/s, cooled in air) and a high rate (50 °C/s, cooled in ice water), respectively. The samples are about 100 µm in thickness and about 4 mm in diameter. The samples pressed with PTFE were named as LDPE-F and the samples pressed with glass plates were named LDPE-G.

2.2. The morphology of LDPE
Table 1 shows the crystallinity of the three kinds of samples calculated by FTIR spectrophotometry. The results indicate that different cooling rates result in different morphology in LDPE: lower cooling rate results in larger spherulite and higher crystallinity; and higher cooling rate results in smaller spherulite and lower crystallinity [8]. However, the different substrate materials do not affect the size of spherulite and the crystallinity.

The degree of crystallinity X in % can be obtained by applying the following relation to the infrared spectrum [14]:

\[ X(\%) = \frac{337}{(A_a/A_c) + 3.37} \]  \hspace{1cm} (1)

Where \( A_a \) is the absorption of the amorphous band at 1303 cm\(^{-1}\) and \( A_c \) is the absorption of the crystalline band at 1901 cm\(^{-1}\).

2.3. SEM of the surface topography
This paper used the SEM to analysis the surface topography. Figure 1 is the SEM pictures of LDPE-F and LDPE-G samples with different cooling rates. LDPE-G samples have smooth surface, and the LDPE-F samples have rough surface which is epitaxial crystallization layer. Polyethylene forms parallel chain epitaxial crystallization on PTFE substrate, this phenomenon is the oriented crystallization of polymers to the substrate materials [10].

2.4. Space charge measurement of LDPE
Space charge in samples was measured by PEA method [15-17]. DC electric fields of -150 kV/mm were applied to the sample and the the space charge packet movement was observed. The high voltage plate electrode is made of semiconductor and the earth electrode is aluminium. The semiconductor material was polyethylene with carbon black. The earth electrode was an aluminium plate with smooth surface [16]. In this paper the space charge distribution was measured every 5 s.
3. Space charge packet characteristic of different morphology and surface topography

3.1. Space charge packet of LDPE-F with different morphology

The LDPE-F samples with surface epitaxial crystallization layer were hot pressed by PTFE substrate material. In this paper a -150 kV/mm were applied to the sample and the space charge packet movement was observed.

Figure 2. The space charge distribution of high cooling rate LDPE-F sample under a -150 kV/mm field.

Figure 2 shows the space charge movement process in high cooling rate LDPE-F sample. After the voltage was applied, a positive space charge packet formed immediately at the anode side of the sample and then moved toward the cathode side, meanwhile, some negative space charges
accumulated at the cathode side of the sample. The positive packet decayed during the movement and finally neutralized with the negative packet at the cathode. A part of the negative charges came out at the back side of the positive packet.

Figure 3 and Figure 4 shows the space charge packet movement process of normal cooling rate and the low cooling rate LDPE-F samples. There were also obviously space charge packet accumulation and movement as the high cooling rate sample. The polarities of these three kinds of LDPE-F samples were positive mainly.

![Figure 3](image1.png)

**Figure 3.** The space charge distribution of normal cooling rate LDPE-F sample under a -150 kV/mm field.

![Figure 4](image2.png)

**Figure 4.** The space charge distribution of low cooling rate LDPE-F sample under a -150 kV/mm field.

The positive space charge packet phenomenon, which forms at the anode side and then moves towards the cathode, was also reported by [18] and [19].

3.2. Space charge packet of LDPE-G with different morphology
The LDPE-G samples without surface epitaxial crystallization layer were hot pressed by glass plates as the substrate material. Figure 5 shows the space charge distribution of high cooling rate LDPE-G sample under a -150 kV/mm field. The positive and negative space charge packets formed at the anode and cathode respectively. The positive packet kept the amplitude at the beginning and then decayed when it moved nearby the negative packet and neutralized with each other. The positive packet decayed to zero when it moved to the cathode. The negative packet moved much slower than the positive packet and did not move near to the anode but stayed in the middle of the sample.

![Figure 5. The space charge distribution of high cooling rate LDPE-G sample under a -150 kV/mm field.](image)

During the process described above, the obvious negative space charge packet process was observed. Researches already proved that it is easier to form negative space charge by ionization at the inner side of the cathode without the epitaxial crystallization layer [20]. It accorded with the phenomenon that LDPE-G sample was easier to form negative packet than LDPE-F sample.

Figure 6 and Figure 7 shows the space charge packet movement process of normal cooling rate and the low cooling rate LDPE-G samples. There ware only negative space charge packet appeared in the sample. The negative packet moved towards the anode and the amplitude increased during the movement. The negative packet accumulated at the inner side of the anode finally.

Figure 8 shows the electrical field distribution calculated by the space charge of Figure 7. The negative space charge packet accumulated near the anode and distorted the local field seriously. The field was up to 150% of the applied field. This situation is a severe test to the insulating property of material.

3.3. Discussions of the results
The movement characteristics of positive packet and negative packet have significant differences. The negative packets only appeared in the LDPE-G samples, and the positive packets appeared mainly in LDPE-F samples. The positive packets moved faster than the negative ones and decayed to zero when they moved near to the cathode. The negative packets moved slower, and the amplitude increased during the movement. The negative packets stayed near the anode side of the sample and distorted the local field seriously. The positive packet always repeated several times one after another in a same sample, each packet appeared after the former one decayed to zero. The negative packet only generated once during every experiment. These differences are caused by the morphology and the surface topography.
The morphology affects the space charge packet phenomenon significantly. The three kinds of LDPE-F samples with different cooling rates show that the space charge packet movement velocity got slower when the cooling rate got slower. When the cooling rate gets slower, the crystallinity will get higher, and the spherulites of the sample will get larger. The spherulites block the transportation of the positive carriers. Some related calculation also showed that the ion mobility decreases while the crystallinity of polymers gets higher [21]. The morphology also affect the polarity of the packets. In LDPE-F samples, when the crystallinity is lower, both of the two kinds of polarity packets exist, however, when the crystallinity goes higher, only positive space charge packet remains.
The epitaxial crystallization layer has affection to the space charge packet characteristic. It is comparatively difficult to ionize at the electrode with epitaxial crystallization layer as LDPE-F samples. The space charge packets were form by the positive charge injection of the anode, and the negative space charge accumulation was limited. It is easier to ionize at the electrode when the sample have no epitaxial crystallization layer at the surface as the LDPE-G samples. The space charge packets were formed by the negative carriers which were ionized at the cathode.

4. Conclusions
This paper prepared samples with different morphology and surface topography by changing the substrate material and adjusting the cooling rate. The space charge packet characteristics of these samples were studied and the conclusions are as following:

(1) The cooling rate affects the morphology. The higher the cooling rate is, the lower the crystallinity is. The crystallinity is only affected by the cooling rate and has no relationship to the substrate material.

(2) The substrate material affects the epitaxial crystallization layer of LDPE surface. There is significant difference of surface topography between the samples hot pressed by PTFE and glass plates. This is because that an epitaxial crystallization layer generates on the surface of LDPE samples when it is hot pressed by PTFE.

(3) The morphology affects the movement velocity and the polarity of space charge packet. When the crystallinity is higher, the packet movement velocity is lower, and it is easier to form positive packets in LDPE-F samples while negative packets in LDPE-G samples.

(4) The surface topography has affection to the space charge packet characteristic. The positive space charge packets were form by the positive charge injection at the anode in LDPE-F with epitaxial crystallization layer, while the negative space charge packets were formed by the negative charge ionized at the cathode on LDPE-G without epitaxial crystallization layer.

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