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INFLUENCE OF PHTHALIC ACID ON THE PROCESS OF DENDRITE DEVELOPMENT IN LOW-DENSITY POLYETHYLENE DURING ELECTRICAL BREAKDOWN

Sh.A. Zeynalov[a], H.N. Vezirov[b], F.Sh. Kerimov[a], S.I. Safarova[a], K.J. Gulmamedov[a], A.S. Alekperov[c,d,*]

[a] *Azerbaijan Technical University, Baku, AZ-1073, Azerbaijan*
[b] *Institute of Physics, Ministry of Science and Education Republic of Azerbaijan, Baku, AZ-1143, Azerbaijan*
[c] *Azerbaijan State Pedagogical University, Baku, AZ-1000, Azerbaijan*
[d] *Western Caspian University, Baku AZ-1001, Azerbaijan*
**Corresponding Author e-mail: aydinalekperov@yahoo.com*

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The presented work presents the results of a study on the effect of small amounts of phthalic acid additives on dendrite formation in low-density polyethene (LDPE). Based on the results obtained, it is shown that the dendrite resistance of LDPE, as expected, increases with the introduction of 0.05 wt% phthalic acid. The established increase in dendrite resistance of LDPE with the introduction of phthalic acid can primarily be explained based on a decrease in inhomogeneities in the form of air pores as a result of accelerated structure formation and the emergence of a more homogeneous supramolecular structure. It was revealed that an increase in dendrite resistance correlates with an improvement in the dielectric characteristics of the composition. The influence of mechanical load on the development of dendrites in polymer dielectrics has been studied. As a result of studying the growth of dendrites in LDPE samples and its optimal composition subjected to unilateral stretching, it was found that under the influence of mechanical tensile stresses, the shape of the surface delimiting tree-like shoots changes, this surface is flattened in the direction of stretching. It has been shown that the rate of dendrite growth increases as mechanical tensile forces increase.

Keywords: *LDPE; Dendrites; Tension; Supramolecular structure; Phthalic acid*
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1. INTRODUCTION

Extensive research is being conducted to study functional materials' structure and dielectric, electrical, thermal and optical properties. These studies are very important both from the point of view of fundamental physics and for determining the possibilities of using materials. Therefore, research in this direction has continued in recent years [1-5]. Polymer and polymer-nanocomposite materials occupy a special place among functional materials. It has been established that, depending on the properties of the nanoparticles included in the composition, interesting physical properties can be observed in these materials [6-10]. The most studied material among polymers is polyethene. Polyethene is widely used as high-voltage insulation, particularly in cables, which raises interest in studying the processes of electrical ageing leading to its breakdown. These processes are associated with the appearance of irreversible changes, so-called dendrites or trees, which are a system of micron-diameter and millimeter-length cavities [11]. It is established that electrical dendrites in polymers are hollow tubes with a diameter of 1 μm gradually tapering at the end. Depending on the growth conditions, dendrites can take various forms such as tree-like, bush-like, or cavity forms [12]. According to the literature [11-13], dendrite channels cannot be considered as a continuation of the needle. In [14], the nature of the development of dendrites in epoxy resin under the influence of high voltage is analysed.

Based on photographic recording of flashes during partial discharge, the authors conclude that dendrite branches can be both conductive and non-conducting. So in the case of a bush-shaped dendrite, a flash during partial discharges fills several branches of the dendrite and originates from the needle. This is a dendrite with conducting branches. It reflected light photographs given in show that the non-conducting branches appear white and the conductive branches appear black, apparently due to carbonation. In [15], it is noted that in polyethene, at some stage of dendrite growth, the phenomenon of the transition of its branches from a non-conducting to a conducting state is observed [16]. It should be noted that in recent years, a large number of works have appeared devoted to the study of pre-breakdown phenomena in polymers in strong electric fields; electroluminescence that occurs before the appearance of dendrites has been discovered [17,18], the occurrence of pores and cracks in polymers in the zone of high electric field strengths when exposed to a sample has been established series of pulses.

2. OBJECTIVE

This study aimed to develop new polymer modifications to increase dendrite resistance, which can be achieved by introducing certain organic and inorganic low-molecular-weight additives. Studying dendrite growth is of great practical interest since dendrites are found in the insulation of cables that have been in operation for a long time. The influence of mechanical stress on the process of dendrite development in the polymer was also investigated.

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2. RESEARCH METHODOLOGY

Low-density polyethylene grade 15313-003 was chosen as the research object, and organic compounds such as phthalic acid (chemical formula $C_6H_4(CO_2H_2)$ were used as additives. Additives were introduced into the initial LDPE raw material by mechanical mixing in amounts of 0.01-0.1 wt%. Samples for the study were made in films with a thickness of 50-70 μm by pressing on a PG-60 hydraulic press. Before introducing the additives into LDPE, its dispersion was achieved using sieve analysis on a grain size determination device. The particle size was less than 50 μm.

The effect of mechanical loads on the change in τ with the electric field intensity of polymer materials was measured in a test cell (Fig. 1).

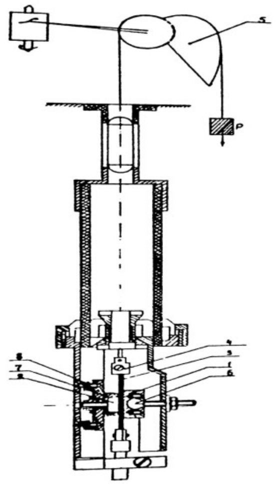
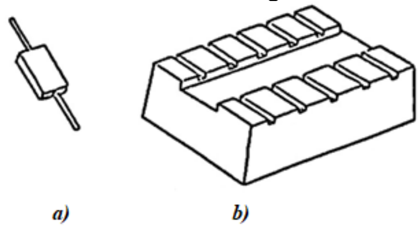


Figure 1. Installation for determining the electrical and mechanical strength of polymer film materials: 1, 2 – electrodes; 3 – sample; 4 – clamps; 5 – lever device; 6 – ball; 7 – security ring; 8 – springs

Dendritic resistance was determined using the method of 2 needles at a frequency of 50 Hz and room temperature on the high-voltage setup AII-70.

Steel needles were used as electrodes; the grounded electrode had a diameter of 80 μm. To obtain standard sizes, the tips of the steel needles for high-voltage electrodes were electrolytically etched in a 2% NaOH solution until the tip diameter was rounded to 4±0.5 μm. The current magnitude was selected experimentally, depending on the number of needles etched simultaneously. In our case, it ranged from 10-15 A during the simultaneous etching of 70 needles. The diameter of the needle rounding was determined using an MHP microscope equipped with a projection screen (magnification 500×).

The samples had the shape of a bar with dimensions of 20×10×4 mm (Fig. 2). To prevent the formation of cracks during the preparation of the sample when inserting the electrodes, the samples and electrodes were heated for 15 minutes in a thermostat close to the melting temperature (at 105K). When inserting the electrodes, the samples were placed in a groove of the device (Fig. 2), and the needles were inserted through the slots.



a) *b)*

Figure 2. Sample forms (a) and device with a groove (b) for positioning the samples

The distance between the ends of the electrodes was 4±0.1 mm, which was controlled under a microscope.

The electrode device was placed in a desiccator filled with transformer oil to eliminate surface discharges (with an electric strength greater than 25 kV/mm).

Simultaneously, 10 samples were tested. A certain voltage U was applied to the samples for 1 hour. Then, the number of samples in which dendrites grew at the given voltage was determined. For each batch of polymer, several (at least 3) series of samples (10 each) were tested at different voltages.

Based on the experimental data, the dependence of the number of samples with dendrites on the voltage at which they form was plotted. The dependence n=f(u), in the first approximation over a narrow range of variation, can be represented as a straight line. From the graph U_d, the value corresponding to n=5, was found and taken as the measure of dendritic resistance. The relative error did not exceed 5%.

3. RESULTS AND DISCUSSIONS

Fig. 3 presents the experimental results obtained using the described method for the film made of LDPE (without additives). As can be seen, the numerical value U_d for the tested samples is 6.8 kV.

However, as shown by the data (Fig. 4), the introduction of phthalic acid (PhAc) into LDPE in an amount of 0.05wt% leads to an increase in U_d to 9.5 kV.

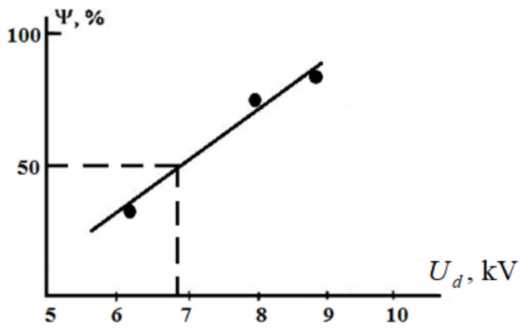


Figure 3. Dependence of LDPE dendritic resistance on electrical voltage

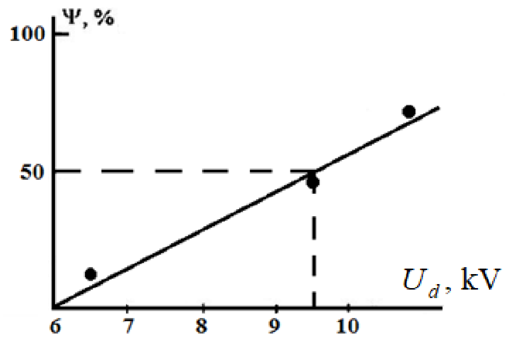


Figure 4. Dependence of dendritic resistanceof the LDPE+0.05wt.% PhAc composition on electrical voltage

This means that the organic additive PhAc in a small amount affects the dendritic resistance of LDPE. The observed increase in dendritic resistance of LDPE with the introduction of a small amount of PhAc can primarily be explained by the reduction of inhomogeneities in the form of air pores, resulting from the acceleration of structure formation and the development of a more uniform supramolecular structure.

The development of dendrites in polymers is also influenced by mechanical stresses in the insulation [19,20]. Studies on the growth of dendrites in polyethene samples subjected to unidirectional tension, with needle electrodes inserted in such a way that the electric field was applied perpendicular to the direction of tension, have shown that under the influence of mechanical tensile stresses, the shape of the surface limiting the tree-like branches changes; this surface flattens in the direction of the tension.

Based on the above, the observed increase in dendritic resistance of LDPE with the introduction of the proposed additive PhAc in the optimal amount (0.05wt.%) should contribute to the improvement of the mechanical strength and dielectric properties of the polymer composition.

We studied the effect of mechanical loads on the change in lifetime τ (the time elapsed from the moment the electrical voltage is applied to the sample until breakdown) of films of the original and modified low-density polyethene (LDPE).

Fig. 5 shows the dependence of the logarithm of the lifetime $lg\tau$ of the original and loaded polymer films LDPE and LDPE + 0.05wt% PhAc on the electric field strength E at 293 K.

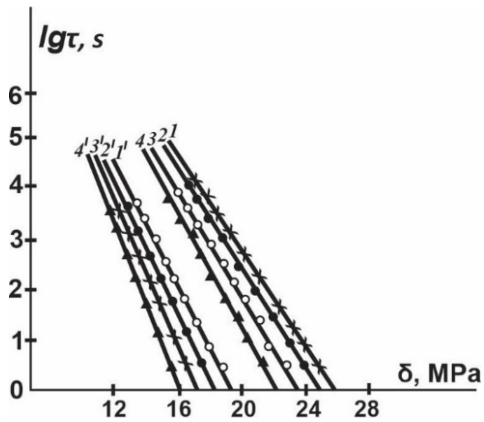


Figure 5. Dependence of the lifetime of polymer films LDPE (1'-4') and LDPE + 0.05wt.%PhAc (1-4) on the electric field strength under the simultaneous influence of different values of mechanical load ($\sigma,\ MPa$):
$1-0;\ 2-2.5;\ 3-5;\ 4-7.5;\ 1'-0;\ 2'-2.5;\ 3'-5;\ 4'-7.5.$

From Fig. 5, it can be seen that at a constant temperature, the presence of a continuously acting mechanical load does not disrupt the linear nature of the dependence of $lg\tau$ on E. That is, $\tau_E = f(E)$, can be described by the empirical formula:

$$\tau_E = B\exp(-\beta E), \tag{1}$$

where the parameters B and β depend on the nature and temperature of the test.

From Fig. 5, it can be seen that the introduction of phthalic acid into LDPE leads to a significant increase in its electrical strength compared to the original LDPE.

As follows from the obtained results, the process of electrical breakdown of the tested samples is observed even at relatively small values of mechanical stress. However, as the mechanical stress increases, the lifetime of the samples decreases, i.e., the rate of dendrite growth increases with the increase in mechanical load.

The slowdown in the development of the electrical breakdown process with the introduction of the optimal amount of PhAc additive into LDPE (curves 1-4), even in the case of a mechanically stressed state compared to the original LDPE (curves $1^{/} - 4^{/}$ can be explained by the fact that the accumulation and growth of dendrites in polymer dielectrics under the combined influence of the aforementioned factors, under otherwise identical conditions, also depends on structural features.

It should be noted that the dendritic resistance of crystallizing polymers is significantly influenced by the chemical nature and concentration of the introduced additives, dispersion, surface area and shape of the particles, the presence and nature of functional groups on the surface of the additives, and the physical and electrophysical nature of the additive particles [21,22]. The observed increase in the induction period of dendrite formation in LDPE with the introduction of the proposed phthalic acid (PhAc) additives in the optimal amount (0.05wt%) can be explained by their structuring characteristics, which help to slow down the process of local heating near the tip in a strong electric field and the occurrence of initial defects due to thermal decomposition of the polymer [23-25]. Moreover, the increase in dendritic resistance of LDPE + 0.05 wt% PhAc should contribute to the improvement of the dielectric properties of the polymer composition, the experimental results of which are presented in the table.

Table 1. Influence of additives on the dendritic resistance of LDPE

Composition	U_d kV	Λ - dendrite at 9 kV	$tg\delta$ at ($\nu = 10^6$ Hs)	ε at ($\nu = 10^6$ Hs)	ρ_u, Om · m
LDPE	6.8	16.5	$4.5 \cdot 10^{-4}$	2.3	$1 \cdot 10^{5}$
LDPE+0.05 wt %PhAc	9.5	13	$3 \cdot 10^{-4}$	2.1	$1 \cdot 10^{17}$
LDPE+0.1wt%PhAc	7	16	$4 \cdot 10^{-4}$	2.2	$1 \cdot 10^{11}$

The table shows the influence of organic additives on the dendritic resistance and dielectric properties of low-density polyethene. Indeed, when modifying the properties of polymers by changing their supramolecular structure (by introducing artificial nucleating agents or other methods), it is important to assess the stability of the supramolecular structure through various mechanical, thermal, ionizing, and other external influences.

Furthermore, the study of the stability of the supramolecular structure showed that after several repeated melting, the size of the structural elements in the LDPE samples containing nucleating agents remains practically unchanged, while in the control samples, they increase.

CONCLUSIONS

The optimal composition of the low-molecular-weight organic additive phthalic acid was determined, and its influence on the process of dendrite formation in low-density polyethene (LDPE) was investigated. It was found that composites with 0.05% by mass of phthalic acid significantly increase the dendritic resistance of LDPE, as expected. The effect of mechanical stretching on the development of dendrites in polymer dielectrics was studied. It was shown that the rate of dendrite growth increases with increasing mechanical tensile stress.

The observed increase in the induction period of dendrite formation in LDPE with the proposed additive in the optimal amount can be explained by their structuring characteristics. This contributes to slowing down the process of local heating near the tip in a strong electric field and the occurrence of initial defects due to the thermal decomposition of the polymer.

It should be noted that we are conducting comprehensive studies on the structural properties of LDPE and its modifications using electron scanning microscopy, X-ray diffraction, and IR spectroscopy. We are also investigating the effects of gamma radiation doses and UV irradiation on the electrophysical ($\varepsilon, tg\delta, \rho_V$) and mechanical properties of LDPE and its modifications. Additionally, we are studying the influence of the small mentioned additive on the rate and mechanism of polyethene crystallization. The results of these studies will be published in prestigious scientific journals.

ORCID

Sh.A. Zeynalov, https://orcid.org/0000-0002-0040-6057

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ВПЛИВ ФТАЛЕВОЇ КИСЛОТИ НА ПРОЦЕС РОЗВИТКУ ДЕНДРИТІВ ПОЛІЕТИЛЕНУ НИЗЬКОЇ ГУСТИНИ ПРИ ЕЛЕКТРИЧНОМУ ПРОБОЇ

Ш.А. Зейналов[a], Х.Н. Везіров[b], Ф.Ш. Керімов[a], С.І. Сафарова[a], К.Й. Гульмамедов[a], А.С. Алекперов[c,d]
[a]*Азербайджанський технічний університет, Баку, AZ-1073, Азербайджан*
[b]*Інститут фізики Міністерства науки і освіти Азербайджанської Республіки, Баку, AZ-1143, Азербайджан*
[c]*Азербайджанський державний педагогічний університет, Баку, AZ-1000, Азербайджан*
[d]*Західно-Каспійський університет, Баку AZ-1001, Азербайджан*

У представленій роботі представлені результати дослідження впливу невеликих кількостей добавок фталевої кислоти на утворення дендритів у поліетилені низької щільності (ПЕНЩ). На підставі отриманих результатів показано, що дендритна стійкість ПЕНЩ, як і очікувалося, зростає при введенні 0,05 мас.% фталевої кислоти. Встановлене підвищення дендритостійкості ПЕНЩ із введенням фталевої кислоти в першу чергу можна пояснити зменшенням неоднорідностей у вигляді повітряних пор внаслідок прискореного структуроутворення та виникнення більш однорідної надмолекулярної структури. Виявлено, що підвищення опору дендритів корелює з поліпшенням діелектричних характеристик композиції. Досліджено вплив механічного навантаження на розвиток дендритів у полімерних діелектриках. У результаті дослідження росту дендритів у зразках ПЕНЩ та його оптимального складу, підданих однобічному розтягуванню, встановлено, що під дією механічних розтягуючих напружень змінюється форма поверхні, що обмежує деревоподібні пагони, ця поверхня сплющується в напрямок розтягування. Було показано, що швидкість росту дендритів збільшується зі збільшенням механічних сил розтягування.

Ключові слова: *LDPE; дендрити; напруга; супрамолекулярна структура; фталева кислота*