

Influence of polarization processes on the charge states and dielectric properties of polyethylene-based compositions with low-molecular additions PE+PbCrO₄ and PE+Cr

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There have been investigated charge states and dielectric properties of polyethylene –based compositions with low-molecular additions PE+PbCrO₄ and PE+Cr. New region of depolarization initiates to be manifested by electrets from polarized films. It is shown that observed effect is related to the repeated electron capture into deeper traps at the expense of internal local electric field. It is also established that the change of dielectric constant (ϵ), dielectric loss tangent ($\text{tg}\delta$) and specific resistance (ρ_v) is associated with the change of supra molecular structure of polymer matrix, structure and thickness of boundary layer and the change of interphase interactions of composition components depending on polarization condition.

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1. Introduction

Progress of electron-ion and polarization processes, space charges in polymer dielectrics can change substantially the process of electric breakdown progress of dielectrics. By introducing a major number of fillings the properties of polymer material are defined both by structural changes in polymer matrix and changes in subsurface layer of filling (1-4). The fillings have an influence on the crystallization or hardening rate, temperature of relaxation transitions, electric mechanical, thermo physical and other properties.

Good dielectric characteristics (dielectric constant ϵ , dielectric loss tangent $\text{tg}\delta$, logarithms of specific resistance $\lg\rho_v$) of polymers allow them as insulating materials in electronic instruments, condensers, generators to be used but also being included as used compounds. The nature of filling surface has a significant influence on $\text{tg}\delta$, $\lg\rho_v$, ϵ characteristics [8].

2. Experimental method

Production of composite sample has been carried out by method of hot forming at melting temperature of polymer and pressure 15MPa for 3min. with subsequent cooling under pressure to room temperature.

Polymer compositions have been subjected to electrothermopolarization at $T_i=353-413\text{K}$ and electric intensity $E_i=(3-12)\cdot 10^6$ V/m for 1 hour with subsequent cooling under the effect of electric field to room temperature.

Charge state and activation energy of charges have been studied by method of thermostimulated depolarization (TSD) [2].

To forecast stability of electret charges and investigation of their formation mechanism the method of thermostimulated depolarization is used. One of the method of thermoactivation spectroscopy is TSD method using to advantage for investigation of charge states and relaxation processes in dielectrics [7,8]. Activation energy of maximum has been calculated from curves by TSD method on temperature position of TSD current maxima:

$$E = k \frac{T_m(T_m - \Delta T)}{\Delta T}$$

From TSD area there has been calculated the magnitude of accumulated charge in sample polarization process.

Measurement of dielectric constant (ϵ) and dielectric loss tangent ($\text{tg}\delta$) is carried out by automatic bridge E8-4 on a frequency of 1kc within the temperatures (293-473)K. Logarithms of specific resistance of samples $\lg\rho_v$ have been measured by teraohmmeter E6-13A within T 293-473K by linear heating at a rate of 3 deg/min.

3. Discussion of obtained results

As a result of structurization on phase boundaries there have been formed deep traps about which TSD analysis data are indicative to the shift on temperature position of maxima (Fig. 1).

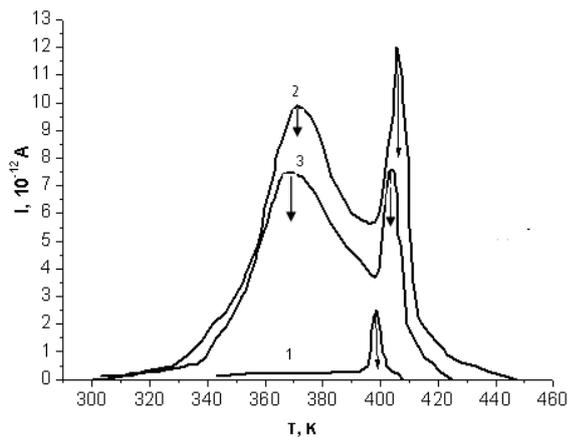


Fig. 1. TSD curves for PE+Cr composition previously undergone by electrothermopolarization at $T_i=373\text{K}$ for 1 hour: 1. $E_i=1\cdot 10^6\text{V/m}$; 2. $E_i=4\cdot 10^6\text{V/m}$; 3. $E_i=6\cdot 10^6\text{V/m}$.

From TSD curves it is seen that according to E_i the area of TSD curves from which there has been calculated the number of polarization charges changes. It is shown that the main region of depolarization is the temperature range from 355 up to 410 (curves 2,3). At first the number of charges Q increases then it decreases gradually. New depolarization region initiates to be manifested by electrets from polarization films at T 406-410K (curves 2,3). Observed effect of charge transfer from low-temperature traps (T 355-385K) into high-temperature ones within T 406-410K can be related to repeated election capture into deeper traps at the expense of internal electric field. Besides TSD peak intensity and hence the number of stabilized charges for polarized samples at $E_i=4\cdot 10^6\text{V/m}$ is higher than at $E_i=6\cdot 10^6\text{V/m}$. In Table 1 there has been presented activation energy of maximum and the number of charges depending on electrothermopolarization intensity.

Table 1.

| E_i , 10^6 V/m | 0 | 4 | | 6 | |
|---|------|--------------------------------------|--------------------------------------|--------------------------------------|-------------------------------------|
| E_a , eV | 2,6 | $T_{\text{max}}=370\text{K}$ 0,27 | $T_{\text{max}}=405\text{K}$ 1,25 | $T_{\text{max}}=370\text{K}$ 0,24 | $T_{\text{max}}=405\text{K}$ 4,6 |
| Q , 10^{-10} Kl | 1,89 | 60,5 | 23 | 46 | 15 |
| σ , 10^{-6} Kl/m ² | 0,27 | 8,64 | 3,29 | 6,53 | 2,12 |

In Fig. 2 there have been shown TSD curves taken for PE+PbCrO₄ compositions before and after undergone by electrothermopolarization at $T_i=373\text{K}$ for 1 hour.

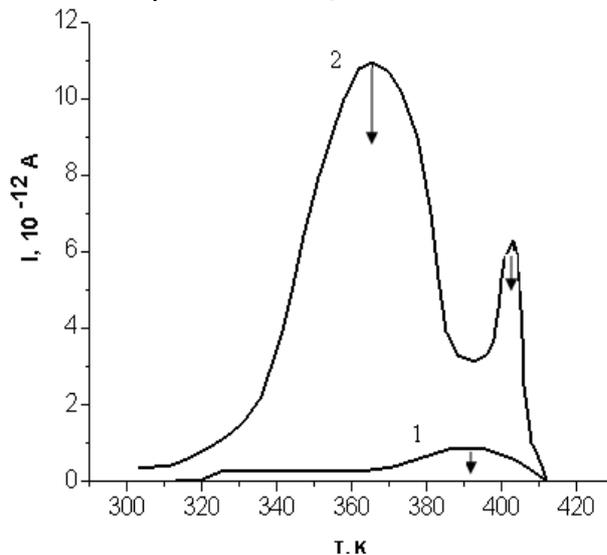


Fig. 2. TSD curves for PE+PbCrO₄ composition previously undergone by electrothermopolarization at $T=373\text{K}$ for 1 hour: 1. $E_i=0$; 2. $E_i=6\cdot 10^6\text{V/m}$.

From TSD current curves the detailed information about density, relaxation time and activation energy of charges being responsible for electret state has been obtained. It is seen from TSD curves that according to E_i the area of TSD curves from which there has been calculated the number of polarization charges changes.

In current spectra of TSD from PE+PbCrO₄ up to electrothermopolarization maximum at $T_{393\text{K}}$ is observed (curve 1) and two maxima at T 365-402K after electrothermopolarization (curve 2). New region of depolarization initiates to be manifested by electrets from polarization films at T 406-410K (curve 2). In Table 2 activation energy of maximum and the number of charges according to the intensity of electrothermopolarization has been given.

There has been also investigated dependence of dielectric constant ϵ , dielectric loss tangent $\text{tg}\delta$, logarithms of specific resistance $\text{lg}\rho_v$ of PE+PbCrO₄ and PE+Cr compositions on temperature after polarization.

Table 2.

| E_i , 10^6 V/m | 0 | 6 | |
|---|------|--------------------------------------|-------------------------------------|
| E_a , eV | 0,52 | $T_{\text{max}}=365\text{K}$ 0,25 | $T_{\text{max}}=405\text{K}$ 1,3 |
| Q , 10^{-10} Kl | 5,41 | 56 | 10,1 |
| σ , 10^{-6} Kl/m ² | 0,77 | 8,12 | 1,4 |

From Fig. 3 it is seen that in dependence $\text{tg}\delta(T)$ for PE+Cr composition the most pronounced maximum of losses at T 375K is available. For PE+PbCrO₄ two maxima of losses at T 305K and 340K are available.

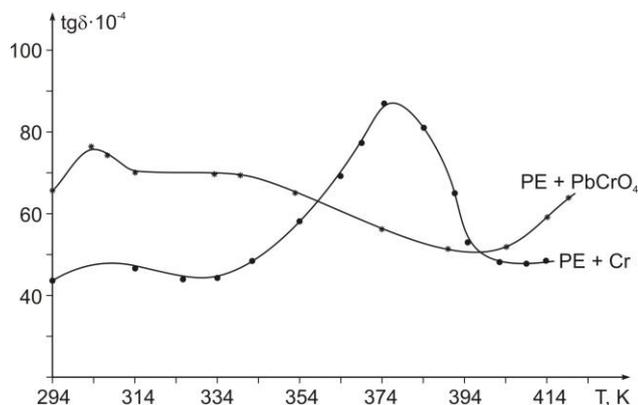


Fig. 3. Temperature dependences of dielectric loss tangent $\text{tg}\delta$ of PE+PbCrO₄ and PE+Cr compositions after polarization.

From T 396K there has been kept increasing dielectric loss tangent $\text{tg}\delta$ that can be related to as the increase of sample conductance as the structural changes in polymer.

In Fig. 4 there have been presented dependences of dielectric constant ϵ of PE+PbCrO₄ and PE+Cr compositions on temperatures under the action of which the polarization is carried out. It is seen from the figure that some increase of ϵ values with the growth of temperature appears to be due to the process of additional crystallization of polymer.

In Fig. 5 dependences of specific resistance logarithm $\lg\rho_v$ of PE+PbCrO₄ and PE+Cr on temperature dependence after polarization have been given. It is seen that at certain temperatures the value of ρ_v increases, i.e. composition samples take posistor effect. For PE+Cr composition at T=324K, for PE+PbCrO₄ at T=350K values of ρ_v are maximum. Increase of $\lg\rho_v$ indicates the decrease of space charges in compositions.

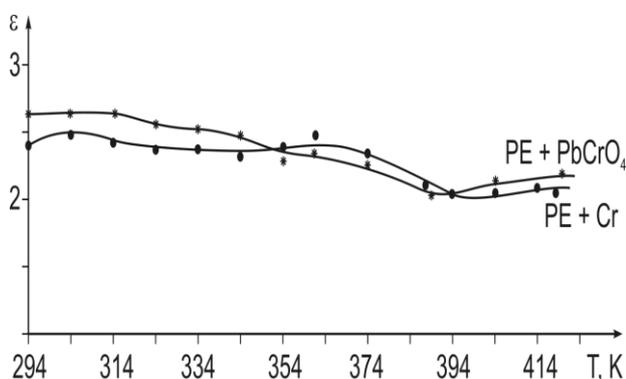


Fig. 4. Temperature dependences of dielectric constant ϵ of PE + PbCrO₄ and PE + Cr compositions after polarization.

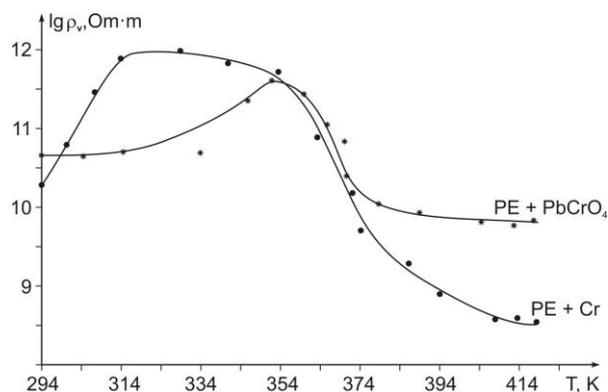


Fig. 5. Temperature dependences of specific resistance logarithms $\lg\rho_v$ of PE+PbCrO₄ and PE+Cr compositions after polarization.

4. Conclusion

Analysis of experimental results indicates that the component composition has a substantial influence on the charge state. The new region of depolarization initiates to be manifested by the electrets from polarized films. Given effect can be related to the repeated electron capture into deeper traps at the expense of internal electric field. Besides TSD peak intensity, and hence the number of stabilized charges for polarized samples depends on polarization condition. Analogously the change of supramolecular structure of polymer matrix, thickness of boundary layer and interphase interaction of composition components depending on polarization condition lead to the change of electret and dielectric properties of ϵ , $\text{tg}\delta$ and $\lg\rho_v$ of compositions with low-molecular additions.

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