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EFFECT OF γ-RADİATİON ON DİELECTRİC PROPERTİES OF HDPE- α-Al_2O_3 NANOCOMPOSİTES

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ABSTRACT

Dielectric properties of high-density polyethylene/ α-Al_2O_3 (HDPE+ α-Al_2O_3) composite films in the temperature range of 20°–110℃ and in the frequency range of 25–10^6 Hz before and after irradiation have been reported. HDPE matrix may result in changes to the dielectric properties, conductivity, and relaxation behavior. Therefore, investigating the electric modulus as a function of frequency and temperature in this composite system is essential for unraveling the underlying mechanisms and potential applications in electrical devices. The samples were irradiated by means of γ-rays from 0 up to 200 kGy. The experimental dielectric data have been analyzed with electric modulus formalism. The electric modulus representation shows well-defined relaxation peaks.

ВЛИЯНИЕ Г-ОБЛУЧЕНИЯ НА ДИЭЛЕКТРИЧЕСКИЕ СВОЙСТВА НАНОКОМПОЗИТОВ ПЭВП **+A-AL_2O_3**

АННОТАЦИЯ

Представлены диэлектрические свойства композитных пленок полиэтилена высокой плотности/α-Al_2O_3 (ПЭВП+ α-Al_2O_3) в интервале температур 20°–110°C и в диапазоне частот 25–10^6 Гц до и после облучения. Образцы облучались γ-лучами от 0 до 200 кГр. Матрица ПЭВП может привести к изменениям диэлектрических свойств, проводимости и релаксационного поведения. Следовательно, исследование электрического модуля как функции частоты и температуры в этой сложной системе имеет важное значение для раскрытия основных механизмов и потенциальных применений в электрических устройствах. Экспериментальные диэлектрические данные проанализированы формализмом электрического модуля. Представление электрического модуля показывает четко определенные пики релаксации.

***Ключевые слова*:** электрический модуль, диэлектрические свойства, ПЭВП, релаксация,

YSPE+A-AL_2O_3 NANOKOMPOZITLƏRININ DIELEKTRIK XASSƏLƏRINƏ Γ-ŞÜALANMANIN TƏSIRI

XÜLASƏ

Yüksək sıxlıqlı polietilen/α-Al_2O_3(YSPE+α-Al_2O_3) kompozitlərinin şüalanmadan əvvəl və sonra 20°–110°C temperatur diapazonunda və 25–10^6 Hs tezlik diapazonunda dielektrik xassələri məruzə edilmişdir. YSPE matrisi nümunənin dielektrik xassələrində, keçiriciliyində və relaksasiya davranışında dəyişikliklərə səbəb ola bilər. Buna görə də, bu mürəkkəb sistemdə elektrik modulunun tezlik və temperature asılılığını öyrənmək elektrik cihazlarında əsas mexanizmləri və potensial tətbiq sahələri aşkar etmək üçün vacibdir. Nümunələr 0-dan 200 kGy-ə qədər γ-şüaları vasitəsilə şüalandırılmışdır. Eksperimental dielektrik məlumatları elektrik modulu formalizmi ilə təhlil edilmişdir. Elektrik modulunun təsviri yaxşı müəyyən edilmiş relaksasiya piləri göstərir.

Açar sözlər: elektrik modulu, dielektrik xassələri, YSPE, relaksasiya

Introduction:

Polymer composite materials are widely used in microelectronics, electromagnetic compatibility, electromagnetic interference protection, and acoustoelectronics, as insulating systems for high-voltage applications, such as: cables, generators, motors, cast resin dry-type transformers [1]. The electrical and mechanical properties of polymers can be modified by adding inorganic nanofillers. Incorporation of nanosized particles improves the electrical and dielectric properties of polymers. Nanosized particles are more attractive because of the interesting properties that arise from the dimensions associated with the large surface area. The choice of the type and nature of micro- and nanoparticles used as fillers is determined by the given electrical, mechanical [2,4] and thermal [5,6] properties of the composite material. The introduction of nano-alumina oxide into the HDPE matrix may result in changes to the dielectric properties, conductivity, and relaxation behavior. Therefore, investigating the electric modulus as a function of frequency and temperature in this composite system is essential for unraveling the underlying mechanisms and potential applications in electrical devices. There are various methods to understand the dynamics of polymer composite materials. The method of dielectric spectroscopy is a good tool for studying the characteristics of materials. It should be noted that within the framework of the development of new polymer composite materials, it is necessary to have information about the dispersion of the real ε′ and imaginary ε″ parts of the complex dielectric conductivity, the temperature and frequency dependences of the tangent of the dielectric loss angle, and the basic laws of structural relaxation within the framework of the introduction of micro- and nanofillers into the polymer. The study of the electric modulus in polymer nanocomposites has gained significant attention due to its potential applications in various technological fields. In this context, the focus of our investigation is on the electric modulus of high-density polyethylene (HDPE) reinforced with nano-alumina oxide. Nano-alumina oxide (α-Al_2O_3), with its unique electrical and mechanical properties, holds promise for enhancing the electrical performance of polymers [7].

The irradiation of polymeric materials with ionizing radiation (gamma rays, X rays, accelerated electrons, ion beams) leads to the formation of very reactive intermediates products (excited states, ions and free radicals), which result in rearrangements and/or formation of new bonds. It is well known that irradiation enhance the electrical conductivity in insulating polymers. All materials have been found to break down at very high radiation doses, however, the range of doses under which a given polymer will maintain its desirable properties depends greatly on the chemical structure of the polymers. Indeed, below the destructive level of exposure, radiation treatment can impart many benefits and enhance properties of commercial value.

Therefore, in the present research, the frequency and temperature dependences of electrical modulus of the composites based on high-density polyethylene containing nanoparticles of α-Al_2O_3 before and after irradiation were experimentally studied to find the possibilities of controlled change in dielectric properties of composite materials. This article aims to provide a comprehensive analysis of the electric modulus in the HDPE/ α-Al_2O_3 nanocomposite system.

Experimental

As a polymer matrix powdered PE2NT11-285D high-density polyethylene (Russia, Kazan) with melting point of 130°C, and a density of 947g/m3 was selected. The choice of HDPE (particle sizes of no more than 300μm) as the matrix was mostly based on the high dielectric properties and process ability of the material. α-Al_2O_3 (Sky Spring Nanomaterials, Inc. Houston United States) with partical size d=40nm, dielectric permeability of ε≈10, and a density of 3,89g/cm3 was used as a filler.

Film samples of unfilled HDPE and composites based on it were prepared using a process flow sheet involving the following procedures:

– Mixing powders HDPE and α-Al_2O_3 to a visual uniform state in a porcelain mortar.

– Pressing a homogenous mixture of component powders in a hydraulic press with heated plates at a pressure of 15MPa with holding at a temperature of 130°C for 5 min, and obtaining samples of composites in the form of discs with a diameter of 20mm and a thickness of about 90-110nm.

–To ensure reliable electrical contact between the sample and the grounded stainless steel electrodes, pressing on the surface of the electrode made of a thin aluminum foil with a thickness of 7μm, followed by cooling in a water-ice mixture (quenching mode).

Thus, 0-3 composites containing 0÷10vol%α-Al_2O_3 in the HDPE matrix were prepared.

All concentrations given in this study are volumetric. The composite manufacturing mode allows one to obtain repeated electrophysical parameters for the bulk of the samples at the same concentration. Samples that had parameters different from the parameters of the main group (their number was small) were not taken into account in the analysis.

Dielectric spectroscopy analysis of the virgin and gamma irradiated specimens was carried out through a broadband dielectric impedance spectroscopy analyze for understanding the variation of dielectric constant (ε) and dielectric loss (tanδ) of the sample over a wide range frequency and temperature. The analysis of the behavior of the complex electric module was used to obtain additional information and to solve the usual difficulties associated with the influence of the nature of the electrodes, ohmic contact and the effects of space charge injection, so that the real and imaginary parts of the dielectric conductivity "hide" the relaxation in the frequency dependence of the imaginary parts of the dielectric conductivity. The complex electric modulus is defined by the equation M*.

$$M^* = \frac{1}{\varepsilon} = \frac{1}{\varepsilon' - j\varepsilon'} = \frac{\varepsilon'}{\varepsilon'^2 + \varepsilon''^2} + \frac{\varepsilon''}{\varepsilon'^2 + \varepsilon''^2} = M' + j$$

In a low-conductivity system, the rapid increase in conductance at very low frequency is due to electrode polarization, and the effect of electrode polarization can completely mask the low-frequency relaxation. The "electrical modulus" formalism is used to study the dielectric relaxations to remove the electrode polarization effect and resolve the low-frequency relaxation. The electrical module is determined by the following formula

$$M' = \frac{\varepsilon'}{\varepsilon'^2 + \varepsilon''^2}$$

$$\frac{\varepsilon''}{\varepsilon'^2 + \varepsilon''^2}$$

The materials were examined using parallel plate capacitors in a two electrodes system in the equivalent circuit of a resistor and a capacitor connected in parallel in a frequency range of 25-10^6Hz at temperatures of 30-120°C and a measuring voltage amplitude of U=1V, using a special shielded and grounded heated "sandwich" measuring cell with a system of a measuring and potential electrode with a diameter of 14 and 20mm, respectively. Temperature measurements were conducted at a frequency of 1kHz. The samples were placed in the measuring cell with pressure exerting stainless steel electrodes. The temperature of sample was controlled intelligent digital temperature controller using a CH-B702 type (China). The centering of the electrodes was provided by a special mandrel in a heated chamber. The distance between the electrodes was determined by the thickness of the test samples. The measurements of capacitance C, dielectric loss tangent tanδ were conducted in a direction perpendicular to the plane of compression of the samples using an E7-20 broadband precision immittance meter. The accuracy of measurement is within 2-5%.

Studies of the effect of filler concentration and of gamma irradiation of HDPE and composites based on it before and after exposure to these factors. The irradiation of nanocomposites were carried out at room temperature by γ– exposure at a dose rate of 5,65 kGy in an irradiator MRX-γ-30 provided with (^{60}Co) sourse. HDPE film and composite samples were irradiated to absorbed doses D=50 and 200kGy.

Discussion

Modulus spectra are frequently used for characterization of nanodielectric because these spectra are independent of electrode polarization effect, electrode material and the adsorbed impurities, and only give the bulk response of the dielectric material [8]. Fig 1. shows the real and imaginary parts of the electric modulus respectively obtained through equation (2) and (3) as a function of frequency for different volume percentage (up to 10%) of α-Al_2O_3 in different doses (0 kGy, 50 kGy,200 kGy). Measurements were made in the 25-10^6Hz frequency range, at room temperature for examined systems is presented. It can be seen that Fig1a the value of M′ increase with frequency up to 2*10^5Hz (for pure HDPE from 0.31 to 0.34, for HDPE+ 3%α-Al_2O_3 composites from 0.30 to 0.32, for HDPE+ 10%α-Al_2O_3 from 0.27 to 0.30), then deacrease to the end of frequency scale. As expected, the real part of electric modulus of HDPE and HDPE+ α-Al_2O_3 nanocomposites decrease with increase α-Al_2O_3 content: for 3% M′=0.32 and 10% M′=0.33. But value of M′ for 1% and 5% nanocomposites higher than pure HDPE(for 1% M′=0.33, for 5% M′=0.34)

After 50 kGy exposure dose value of M′ decrease than 0 kGy irradiation for all tested composite system. The reduction in the values of M′ with increasing dose results from the increase in the mobility of the polymer segment and charge carriers [8]. It is clearly seen that the real part of electric modulus increases slowly up to 10^3Hz, then increases high rate in comparison previous frequencies up to 2*10^5Hz and obtaion maximum value then decrease end to the frequency scale. The value of M′ decreases with an increase in volume content α-Al_2O_3 in the base mixture as a result of increase of the real part of complex dieletric permittivity and nature of all curves are similar. For all the studied composite samples there is a transition from low values to high ones, which implies the relaxation process. The value of M′ for pure HDPE decreases 1.76 times (from 0.300 to 0.176) after 50 kGy irradiation. The maximum value of M′ for pure HDPE is 0.2.

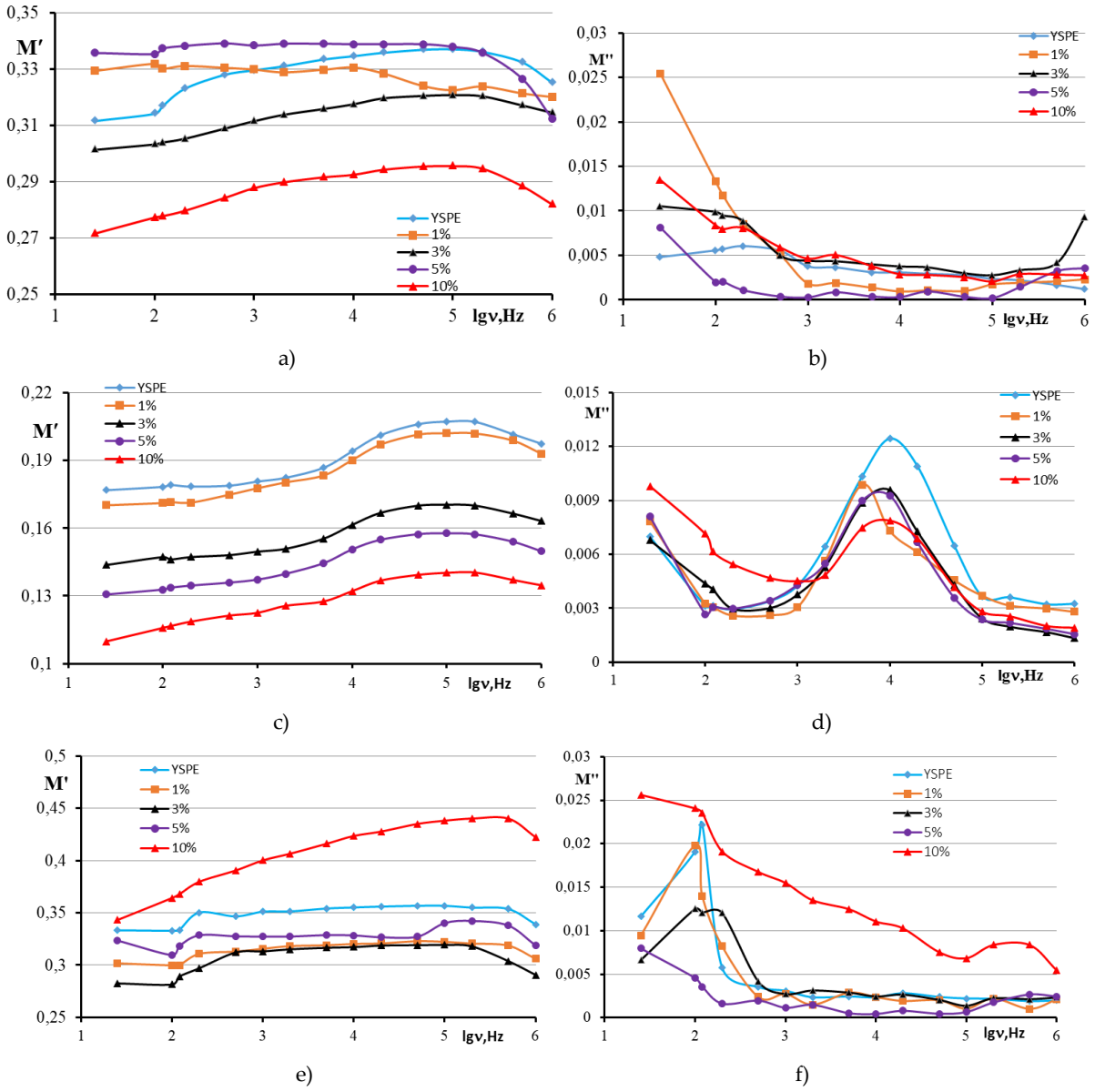

Fig.1. Frequency dependence of real and imaginary parts of electric modulus of HDPE+ x%α-Al_2O_3 at different radiation doses:a,b-0kGy;c,d-50kGy; e,f-200kGy.

At 200 kGy exposure dose value of the real parts of electric modulus for HDPE and HDPE+ 10%α-Al_2O_3 are close to each other, 0.33 and 0.34 respectively. The nature of curve remains the same for HDPE+ 10%α-Al_2O_3 but peak shifts to high frequency (10^5Hz) after irradiation which can be attributed to the release of more trapped charge carriers[9]. The M′of other samples show a plateau in intermediate frequency ($5*10^2$ -$2*10^5$Hz) range and gets maximum value at 10^5Hz then decrease to the end of the frequency range.

The frequency dependence of the imaginary parts of electric modulus M″ in alternating fields for pure HDPE and HDPE+ x% α-Al_2O_3 composite containing different amount of filler at D=0 kGy are given in Fig1b. It is found that M″ value firstly decrease with increase of frequency within (25-10^3Hz) frequency range, then remain constant intermediate frequency range 10^3 - 10^5Hz for all examined system, for 3% composites the value of M″increases from 0.003 to 0.009 (3times).

At D=50 kGy exposure dose spectra of M″ exbihit electric modulus relaxation peak around 10^4Hz which is attributed to PE local chain motion [10]. It can be seen from fig1d, the value of M″ decreases at 25-10^3 Hz frequency range, then increase up to 5*10^3 Hz and 10^4Hz, respectively, with the increase frequency value of M″ almost same for all examined system. The relaxation times can be calculated from the relation, $\tau_x = 1/2\pi f_{max}$, for HDPE $\tau_x = 2 \cdot 10^{-4}$s, for 1% $\tau_x = 3 \cdot 10^{-4}$s, for 3,5,10% $\tau_x = 1.6 \cdot 10^{-5}$s. There observed an increase in the value of M″ of composite system. Similar observasions were made for PVDF/$CaCu_3TiO_{12}$ nanocrystal composite[11]. It should be noted that, with increasing irradiation dose up to 50kGy the value of M'' increase 1.4 times measured at 25Hz frequence than 0 kGy, for HDPE M' increases from about 0.007 to 0.005.

From Fig.1f. it can be noted that after 200kGy irradiation peaks shifts to low frequency range in a 0, 1 and 3% composites. There is not peaks 5% and 10% composites. For 0, 1% specimens relaxation time is $\tau_x = 8 \cdot 10^{-4}$s. Also value of M″ increases than 50 kGy irradiation dose in stuied samples. The value of M'' for 10% composites decreases from 0.25 to 0.005 and peak observed at 5*10^6Hz.

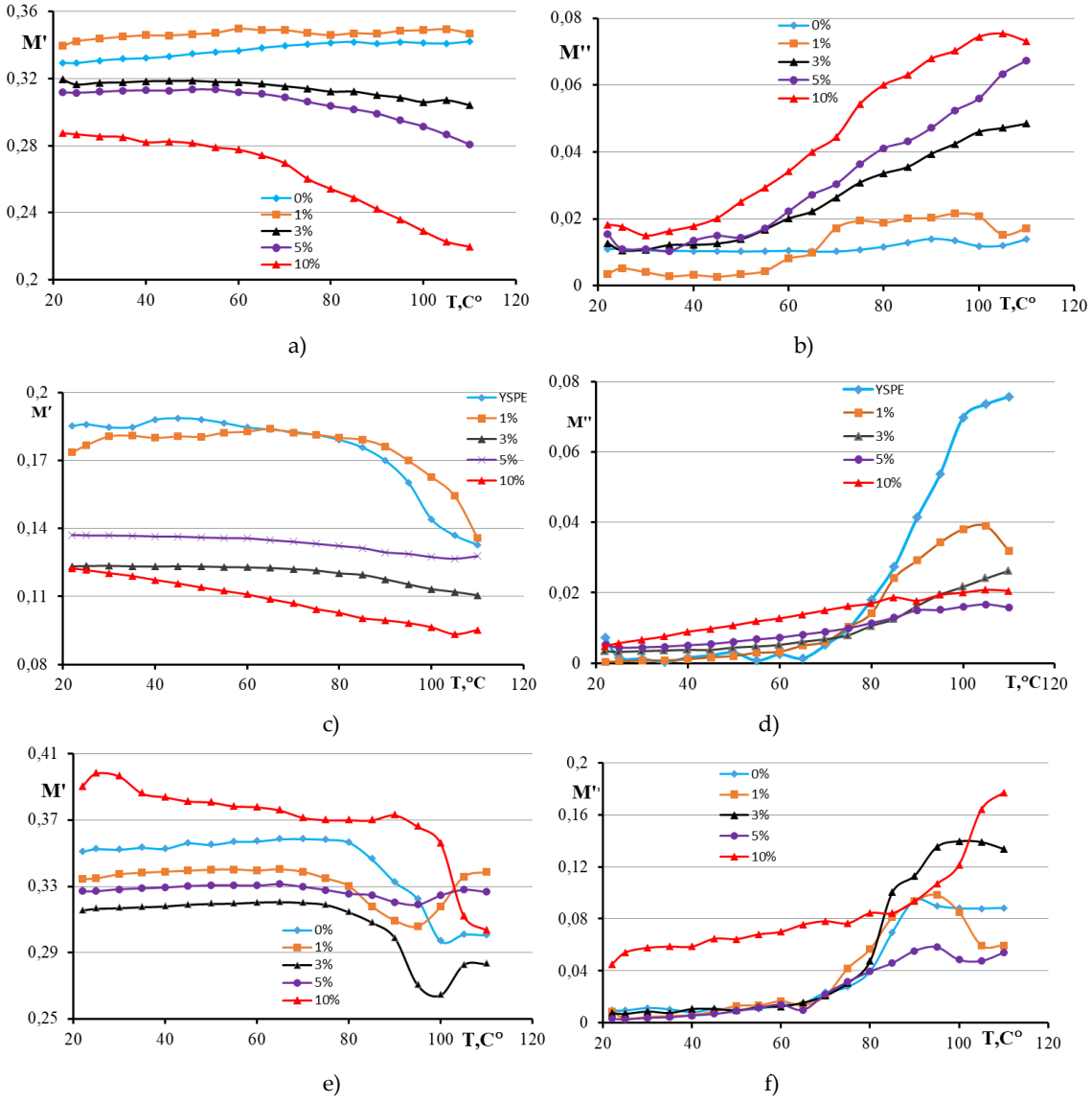

Fig.2. Temperature dependence of real and imaginary parts of electric modulus of HDPE+ x%α-Al_2O_3 at different radiation doses:a,b-0kGy;c,d-50kGy; e,f-200kGy.

Fig. 2 shows the variation of real (M') and imaginary (M'') parts of the electric modulus as a function of temperature for different volume concentrations of α-Al_2O_3 in different doses at costant frequency 10^3Hz . Measurements were made in the temperature range of 20°C-110°C. It can be seen from Figure 1a, the value of M' for pure HDPE and HDPE+1% α-Al_2O_3 composite almost does not change depending on the temperature, the value of M' decreases with increasing temperature at higher temperatures (starting from 60°C), at D=0kGy exposure dose, as the filler content increases in the in the HDPE matrix, at 10% composite the rate of decrease was greater than other samples. The increase of α-Al_2O_3 content results in lower values of M', implying that the real part of dielectric permittivity increases ceramic filler. The value of M'' increases as the filler content increase from 3% to 10% volume percentage in HDPE (Fig.2b.) which is a characteristic of Maxwell-Wagner-Sillar (MWS) relaxation.

In Fig.2c, d M' and M'' as a function of temperature is presented for all the examined systems, after 50kGy γ-irradiation. The value of M' decreases than 0 kGy irradiated samples. It can be seen one relaxation M'' as a function of temperature process, located in the high temperature range are clearly recorded, for pure HDPE, and HDPE +1% α-Al_2O_3 specimens is formed in the temperature range where transition occurs. Thus, values are very close to the glass transition temperature of the pure HDPE. In consequence it is reasonable to suggest the relatively slow dielectric relaxation process corresponds to glass/rubber transition.

Fig.2e, f shows functions M'(T) and M''(T) for HDPE and HDPE+ x% α-Al_2O_3 after γ-irradiation at dose 200kGy. As seen in Fig.2e the value of M' higher than 50kGy irradiated samples. The imaginary part of electric modulus of 0;1 and 3% nanocomposites show plateau in low temperature range (T<80°C), while a broad peak emerges in temperature range of 80°-100°C. Amplitude of peak decreases for 5% composite. Also, value of M'' increases from 0.009 to 0.045 for 10% composite system than pure HDPE and there is not peak in HDPE+ 10% α-Al_2O_3 this temperature range (80°-100°C). It is well known (Aliev, Kh.S., at al., 2018) that α- relaxation process is associated with glass to rubber transition. If sufficient thermal energy is provided to the polymer, then large parts of amorphous macromolecular chains can relax simultaneously in a cooperative motion. The whole process is characterized by glass transition temperature (T_g), which is considered as the temperature where transition occurs. In polymer matrix composite systems, glass transition is related to the chemical structure of the polymer chains and in many cases to the type of the applied filler. As a rule of thumb, glass transition temperature is taken as the temperature at which the α- relaxation loss peak is recorded in the dielectric spectrum at constant frequency.

Conclusion:

The dielectric properties of nanocomposites "nonpolar HDPE/ α-Al_2O_3" were studied within the frequency range of 25–10^6 Hz. It was shown that the dielectric parameters of composites significantly depend on both the α-Al_2O_3 concentration in the polymer and the frequency of the electric field. The appearance of peaks at higher temperature reveals that there may be the MWS-relaxation peak.

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