THE DEPENDENCE OF DIELECTRIC PARAMETERS AND ELECTRIC CONDUCTION OF COMPOSITE ON THE BASE OF POLYETHYLENE AND IRON OXIDE ON RADIATION DOSE

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The radiation γ-influence at 0-500kGy doses on dielectric parameters (ε' and tgδ) and composite electric conduction (σ) on the base of high-density polyethylene (HDPE) and iron oxide (α-Fe_2O_3) radiated in air at room temperature is studied with the aim of modification of new class composite properties polymer –metal oxide. It is shown that ε'=f(D) and tgδ=f(D) dependences have extreme character at(10 and 20 %) α-Fe_2O_3 high concentrations. σ=f(D) dependence of investigated samples also has extreme character (besides composite HDPE+20%α-Fe_2O_3) and it is explained by accumulation of stabilized charge carriers in them.

Keywords: HDPE/α-Fe_2O_3, γ-radiation dose, dielectric constants, dielectric losses, electric conduction.
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INTRODUCTION

The modern state of development of cosmic, atomic and electronic industries requires the creation of new polymer materials and composites on their base for usage of multifunctional electron devices in the capacity of electro-active elements. In exploitation process these materials are subjected to influence of different external factors (mechanical load, thermal field, corrosive medium, humidity, ionizing radiation and etc), as a result of which their properties and structure change. In particular, the polymer composite materials (PCM) are successfully applied in the capacity of anti-damage nuclear reactor shielding, in power-supply system of artificial satellites and spaceships where there is always the nuclear radiation. The radiation-induced behavior of PCM properties is the important aspect, which should be taken into consideration at development of new PCM working in conditions of nuclear radiation. That's why new PCM should have high radiation resistance and minimal dependence of properties on change of environment parameter [1-4].

It is well known [5-6] that intermolecular linkage or destruction (chain breakage) of polymers at γ-radiation and this fact that which of these processes dominate depends on chemical structure, stability of formed free radicals and polymer aggregate state. The study of γ-radiation influence on PCM in particular, on their electrophysical properties still is of interest for many scientists and is important not only for fundamental investigation and for study of radiation process mechanism in these materials. The influence of γ-radiation is the topic of the many investigations [7-10].

The experimental investigation of γ-radiation influence on electrophysical properties of composite system HDPE/ α-Fe_2O_3 is the aim of the present paper.

EXPERIMENTAL PART

The powder-like high-density polyethylene by 20806-024 mark with average molecular mass 95000, crystallinity degree 52%, melting point 130˚C and density 958kg/m^3 is chosen in the capacity of dielectric (matrix). HDPE choice as matrix is caused mainly by well dielectric properties of material and its manufacturability.

For the obtaining of samples of unfulfilled HDPE and HDPE+α-Fe_2O_3 composites the following technological scheme is applied:

- the obtaining by way of sifting of α-Fe_2O_3 powders with particle dimensions not more 30μm;
- the drying during 24h. at T =100˚C and mixing in porcelain mortar of powder-like α-Fe_2O_3 with HDPE powder;
- the pressing of homogeneous mixture of component powders in hydraulic press with incalescent plates at pressure 15MPa with endurance at temperature 150˚C during 5min and obtaining of composite samples in the form of discs by diameter 20mm and thickness 140÷180μm. All concentrations given in the present paper are volume ones;
- the pressing of electrodes on both surfaces of samples from thin aluminum foil 7μm with further cooling in water-ice mixture (quenching mode) with aim of supply of reliable electric contact between sample and electrodes from stainless steel. All composites are subjected to temperature stabilization at 60˚C during 24h after which all samples are considered as suitable for investigations.

The previous radiation of samples by γ-rays are carried out in air on γ-source Co^{60} with dose rate D=3,3·10^3 Gy/h up to 500κGy. The dose rate in radiation point is defined with the help of ferro-sulfate dosimeter.

The investigations of dielectric characteristics (of real part of ε dielectric constant and tgδ dielectric loss tangent) are carried out by two-electrode system at 10^3Hz frequency, at 20˚C temperature and U=1V measuring voltage amplitude with application of specially screened and earthed heated measuring cell by "sandwich" type having the system of measuring and potential electrodes by diameters 10 and 15mm correspondingly. The samples are put into measuring cell with clamping electrodes from stainless steel. The sample temperature is controlled with the help of thermocouple cuprum-constantan. The measuring of C capacity and tgδ of samples are carried out with the help of wideband precision immittance measurer of E7-20 type and

measuring of R volume resistance with help of E6-13A teraohmmeter. Further, ε' value ($\varepsilon'=c\cdot d/\varepsilon_0\cdot S$, where S is plane sample square, d is film thickness) and electric conduction value σ ($\sigma = d/R\cdot S$, where R(Ом) is sample resistance) of measuring cell with sample is defined on base of measured values of given parameters and sample geometric dimensions by standard methods.

RESULTS AND THEIR DISCUSSION

The changes of real component of complex dielectric constant ε' of HDPE and composites with filler concentrations α-Fe_2O_3 up to 20 % before and after influence of γ-radiation are shown on fig.1. From the figure it is seen that ε' increases from 2,3 for HDPE up to 3,8 for HDPE+20% α-Fe_2O_3 composite with α-Fe_2O_3 concentration increase and it slowly increases with absorption dose increase. ε' firstly insignificantly increases for samples with volume contents 10 and 20 % α - Fe_2O_3, achieves maximum at dose D=300kGy and further decreases. ε' value in maximum is 4,8, i.e. it increases approximately in 1,26 times (≈26%), in given region of exposure radiation dose for HDPE+20%α-Fe_2O_3 composite. Such character of ε' dependence on absorbed radiation dose is explained by formation of end and intra-chain peroxide radicals and their destruction by reaction:

$$R^{\cdot} + O_2 \rightleftarrows RO^{\cdot}_2$$

By other hand [11], the material crystallinity degree decreases with the increase of absorbed radiation dose, crystal structure gradually destroys and totally disappears at high doses. Especially this process of crystal structure disorder and also the increase of dipole element number (C=0 group, dipole moment $9{,}35\cdot10^{-30}$ C·m) leading to ε' increase are main factors defining the change of HDPE polymer matrix properties and composites as a whole on its base. It is necessary to note that dipole element number increase leading to formation of wide series of polar elements appearing on boundary particle-polymer matrix takes place at composite obtaining too (as a result of partial matrix oxidation and stabilization of α - Fe_2O_3 particles in it). The results of dielectric loss tangent change of investigated samples are given on fig.2. It is seen that introduction of α-Fe_2O_3 up to 20% into HDPE leads to dielectric loss increase from 0,008 up to 0,1 with respect to pure HDPE. From the figure it is also seen that tgδ composite values in given region of absorption dose increase with D increase, pass through the maximum at 300 and 100kGy and later decrease. tgδ increase observed in initial period is connected with increase of oxide groups (formed mainly because of solved oxygen), unsaturation and amorphism degree of polyethylene matrix. In HDPE+5%α-Fe_2O_3 case tgδ with D increase insignificantly decreasing achieves the minimum at 100kGy in interval (100÷300)кGy practically remains constant and further also insignificantly increases. For pure HDPE tgδ value with D increase practically doesn't change up to absorbed dose 500kGy. The typical curve of function σ = f(D) are shown on fig.3. σ value extreme change with radiation dose is observed practically in all cases. The electric conduction on constant current of sample composites with 5 and 10% volume content of filler α-Fe_2O_3 correspondingly is subjected to the biggest change.

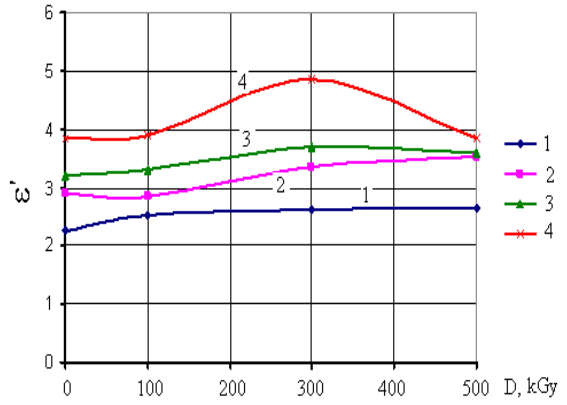

Fig.1. The dependences of dielectric constant (ε′) on HDPE+αFe_2O_3 composite radiation dose: 1 is pure HDPE, 2 is HDPE+5%α-Fe_2O_3 ,3 is HDPE+10% α-Fe_2O_3, 4 is HDPE+20% α-Fe_2O_3.

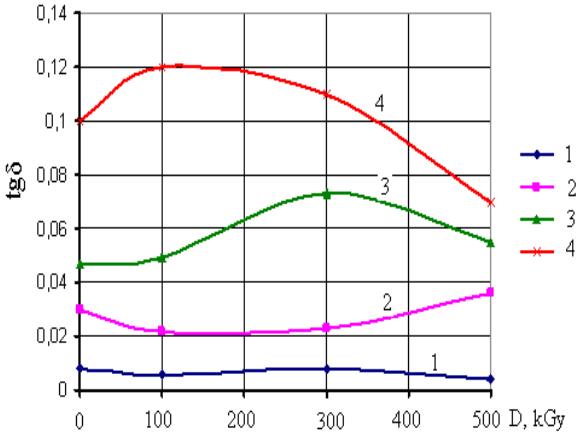

Fig.2. The dependences of dielectric loss (tgδ) on HDPE+αFe_2O_3 composite radiation dose: 1 is pure HDPE, 2 is HDPE+5%α-Fe_2O_3 ,3 is HDPE+10% α-Fe_2O_3, 4 is HDPE+20% α-Fe_2O_3.

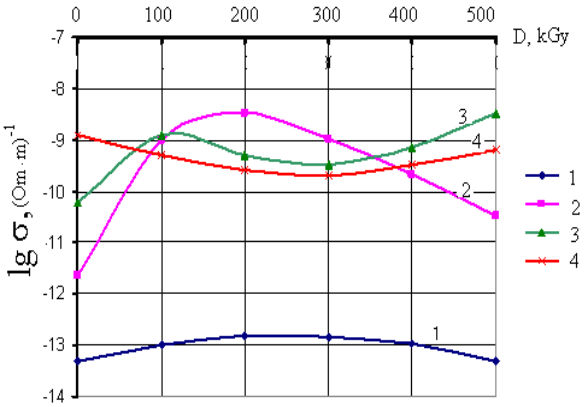

Fig.3. The electric conduction dependences (σ) on HDPE+αFe_2O_3 composite radiation dose: 1 is pure HDPE, 2 is HDPE+5%α-Fe_2O_3 ,3 is HDPE+10% α-Fe_2O_3, 4 is HDPE+20% α-Fe_2O_3.

The electric conduction of these samples with D increase achieves the maximum at D=200 and D=100kGy correspondingly. The further increase of absorption dose leads to σ decrease of HDPE+5% α-Fe_2O_3 composites and in case of HDPE+10%α-Fe_2O_3 composite the electric conduction decreasing achieves minimum at 300kGy and later increases again. The electric conduction of pure HDPE and HDPE+20% α-Fe_2O_3 composites in given dose interval practically remains constant. It is seen that HDPE filling up to 20%α-Fe_2O_3 leads to electric conduction increase from $4{,}44\cdot10^{-14}$ $(Om\cdot m)^{-1}$ up to $3{,}5\cdot10^{-10}(Om\cdot m)^{-1}$. Note that as a whole the absorbed dose weakly influences on dose dependences of investigated samples (besides HDPE+5%α-Fe_2O_3). One can suppose that some increase of electric conduction observing at γ-radiation is caused by ionization. The ionization produces the charges able to motion (free electrons and positive ions) which can take part into conduction process. In [12] it is seen that at γ-radiation of HDPE the accumulation of volume electric charge correlates with formation and accumulation of peroxide radicals, which are electron acceptors and define the whole process of charge accumulation. By authors' opinion [13] the dose effects are the direct consequence of free radical formation and their recombination between each other with formation of character postradiational changes in polymer structure. Moreover, the some ovservable increase of electric conduction of investigated samples one can connect with linkage processes of polymer chains leading to three-dimensional structure formation and further its decrease one can connect with oxide destruction processes.

The authors [14] confirm that electric conduction of polyethylene matrix is caused by charge carriers which appear at γ-radiation and not by charges injected from electrodes. The concentration of trapped electrons increases with radiation dose increase: the more radiation doses the bigger number of charge carriers.

The radiation dose increase leads to decrease of relation of crystal phase to amorphous one and by this it causes the decrease of phase interface square. As a result of this the concentration density of deep traps on phase interface increases.

Thus, the dose effects, i.e. the influence of absorbed dose of γ-radiation on electro-physical properties of previous radiation is caused not by spectrum change of molecular motions, but accumulation in radiated material of stabilized charge carriers and less radical or molecular radiolysis products which act as trap centers.

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