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THE ROLE OF Cd + ZnS FILLER IN THE FORMATION OF RADIATION-RESISTANT DIELECTRIC PROPERTIES OF POLYPROPYLENE-BASED COMPOSITES

This work investigates the effect of γ-irradiation on the dielectric properties and electrical conductivity of polypropylene (PP) composites filled with CdS + ZnS at various volume fractions (10%, 30%, 50%). It was found that, for pure PP and composites with a low filler content (up to 30%), the permittivity and specific resistance remain stable at irradiation doses of up to 50 kGy. At a dose of 80 kGy, an increase in permittivity is observed in samples with 10–30% filler, which is associated with an increase in polarization and the formation of cross-links. In composites with a high CdS + ZnS content (50%), the permittivity first increases to 9.3, and then sharply decreases to 7.1 at 80 kGy, which indicates the destruction of the polymer matrix. The specific resistance at high filler content is significantly reduced by γ-irradiation, especially at 50% filler, which is associated with the formation of conductive channels.

Keywords: γ-irradiation, polypropylene (PP), filler, permittivity, resistivity, radiation resistance.

1. Introduction

Currently, electroactive polymeric materials are widely used in various fields of technology, microelectronics, electret microphones, dosimeters, nanotechnology, spacecraft, pressure sensors, air filters, *etc.* The scope of application of polymeric materials is constantly expanding, and their modification to create polymer composites combining the properties of different materials is of considerable interest [1]. Several methods are used to change the properties of polymers, one of which is the addition of dispersed fillers (20–25 μm) to the polymer volume. As a result, the composite material acquires new electret, electrophysical, strength, thermophysical and other proper-

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ties. In this case, additives change the supramolecular structure (SMS), which largely determines the mechanical and physicochemical properties of polymers and composites based on them. The nature of the influence of these additives and ionizing radiation on the supramolecular structure, physical, mechanical and electret properties of the polymer is due to the presence of amorphous and crystalline phases in it [2]. It is known that the nature of structure formation in polymers and composites based on them also depends on the properties of macromolecules and on the external conditions under which the formation of SMS occurs [3]. When fillers that act as artificial crystallization nuclei are introduced into polymers in small quantities, this leads to a change in the properties of the material. Depending on the type and properties of the filler, the rate of crystallization or hardening, the temperature of relaxation transitions, and the electrical, mechanical and other properties of the composite material change [4].

All of the above features indicate that by changing the type and dispersion of fillers, as well as the crystallization mode and γ-irradiation, it is possible to control the properties of polymer-based composite materials [5]. The introduction of fillers, including nanodispersed ones, into polymers is accompanied by the appearance of new artificial crystallization nuclei and stabilization centers (traps) for electric charges, which can contribute to an increase in the surface density of electret charges ($\sigma \sim 10^{-4}$ C/m^2) and their lifetime τ. The creation and application of new composite materials based on polyolefins and luminophore fillers are among the urgent tasks of radiation materials science [6]. Polypropylene (PP), as a radiation-sensitive polymer, was used as the main polymer matrix in this work [7]. Composites with different volume contents of the binary semiconductor filler cadmium sulfide and zinc sulfide ($CdS + ZnS$) were also prepared on its basis. After that, the electrophysical and luminescent properties of the obtained polymer/$CdS + ZnS$ composites and structural changes in these composites were studied [8].

2. Experimental Part

The samples for the study were prepared by hot pressing a mixture of polymer and filler powders in a heated hydraulic press. Two punches with heaters and smooth polished surfaces are fixed on the lower and upper nozzles of the press [1].

The procedure for preparing the composite samples, both the original PE and PP samples, and composites based on them, was as follows:

• mechanical mixing of a powdered phosphor additive with PE or PP powder in a porcelain mortar until a homogeneous mixture was obtained;

• the mixture, placed between two Al foils and a 100 μm thick gasket, is kept for some time at the melting temperature of the polymer under low pressure (∼1–5 MPa);

• at the same temperature, the pressure is slowly raised to 10–15 MPa, at which the sample is held for 5 minutes. The pressure is then released, and the sample is rapidly cooled in an ice-water mixture;

• the samples had a thickness of ∼100–120 μm and a diameter of ∼45 mm;

• to ensure reliable electrical contact during measurements of the electrophysical properties, electrodes made of ∼7 μm-thick aluminum foil were pressed onto both working surfaces of the composite samples.

For radiation studies, g-radiation sources of the isotope ^{60}Co, available at the Institute of Radiation Problems of the Ministry of Science and Education of Azerbaijan, were used. The main g-sources were isotope installations of the K-25 or MRX-g-25M type based on ^{60}Co with a half-life of 5.3 years and an emitter activity of 5×10^4 Ci at a temperature of 293 K [6].

Irradiation with γ-rays during the tests was carried out using remote control of the radiation source in special concrete chambers providing biological shielding. Samples of polyethylene- and polypropylene-based composites containing the binary compound ($CdS + ZnS$) were irradiated with γ-rays in sealed ampoules at a temperature of 77 K, i.e. in liquid nitrogen, up to an absorbed dose of 10 kGy. The dose was estimated with a ferrosulfate dosimeter. Dosimetry was carried out under the same conditions as the irradiation [9].

3. Experimental Results and Discussion

As can be seen from Fig. 1, the value of ε remained practically constant at absorbed radiation doses of D = 30, 50, and 80 kGy in a sample of pure PP (without additives) (curve *1*, $\varepsilon = 2.25$).

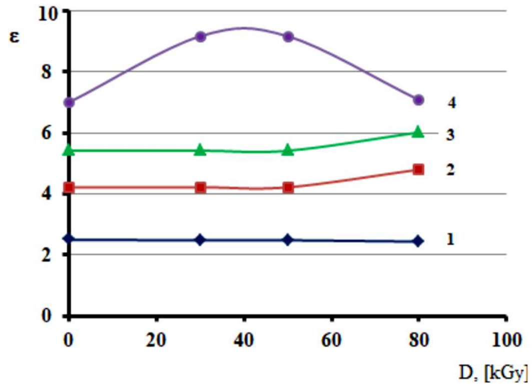

Fig. 1. Dependence of the real part of the permittivity of PP/CdS + ZnS composite samples on the γ-irradiation dose, $\varepsilon = f(D)$: PP (*1*); CdS + ZnS – 10% (*2*); CdS + ZnS – 30% (*3*); CdS + ZnS – 50% (*4*)

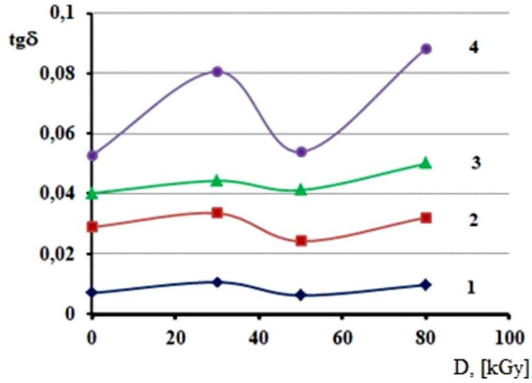

Fig. 2. Dependence of the dielectric loss tangent ($\tan\delta$) of PP/CdS + ZnS composite samples on the γ-irradiation dose, $\tan\delta = f(D)$. PP (*1*); CdS + ZnS – 10% (*2*); CdS + ZnS – 30% (*3*); CdS + ZnS – 50% (*4*)

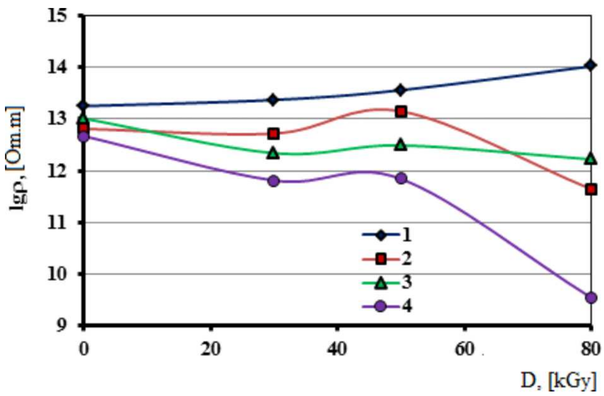

Fig. 3. Dependence of the specific resistance of PP and PP/CdS + ZnS composites with different filler contents on the γ-irradiation dose: PP (*1*); 10%CdS + ZnS (*2*); 30%CdS + ZnS (*3*); 50%CdS + ZnS (*4*)

In the composite sample with a filler volume fraction of $\Phi = 10\%$ and 30%, the initial value was $\varepsilon = 4.15 \div 5.4$, which is associated with both polarization and conductivity. At absorbed radiation doses of 30 and 50 kGy, the ε' value remains virtually constant. At an absorbed radiation dose of $D = 80$ kGy, a significant increase in the ε value is observed ($\varepsilon = 4.8 \div 6$). This increase may be associated with the formation of cross-links between molecules, as well as with an increase in polarization and conductivity. In the sample with a filler volume fraction of $\Phi = 50\%$, the permittivity initially (before irradiation) was $\varepsilon = 6.9$. In this sample ($\Phi = 50\%$), the permittivity reached $\varepsilon = 9.3$ at irradiation doses of $D = 30$ and 50 kGy. At an absorbed dose of $D = 80$ kGy, it decreased to $\varepsilon = 7.1$. This decrease may be associated with the destruction of bonds in the main chain and side groups of the polymer, depending on the filler volume fraction ($\Phi = 50\%$) [1].

As can be seen from Fig. 2, at absorbed doses of 30 and 80 kGy after γ-irradiation, an increase in the $\tan\delta$ value is observed compared to the unirradiated sample. It should also be noted that the observed changes (increase or decrease) in the electrophysical properties of polymer composite samples, namely permittivity (ε), dielectric loss tangent ($\tan\delta$) and electrical conductivity (σ) depending on the absorbed radiation dose indicate that the effect of radiation is specific to each material, including polymer composite materials (PCMs), which is a heterogeneous system. This behavior is governed by the fact that each material absorbs radiation differently depending on its chemical structure and properties, as well as its mechanical and physical properties [10].

The values of specific resistance of composites with a low filler content (10%) remain nearly unchanged after irradiation up to 50 kGy (Fig. 3). It is known from the literature that at low filler volume fractions, composites are more resistant to radiation. If the filler volume fraction exceeds a certain value, then irradiation worsens the physical properties of the composite [11]. We believe that at both low radiation doses (10 kGy) and low CdS + ZnS filler contents (up to 30%), the electrical conductivity of the composites remains practically unchanged. The obtained results may contribute to the future development of polymer composites resistant to γ-radiation. From the dose dependence it is evident that a further increase in the filler volume fraction to 50% CdS + ZnS leads to a sharp decrease in specific resistance [12].

4. Conclusion

The conducted research demonstrated that the electrophysical, dielectric, and conductivity properties of polypropylene (PP)-based composites containing CdS + ZnS fillers are strongly influenced by both the filler concentration and the absorbed γ-irradiation dose. The introduction of a nanoscale CdS + ZnS luminophore into the polymer matrix leads to the formation of artificial crystallization nuclei, modifications in the supramolecular structure, and enhancement of dipolar, interfacial, and volume polarization mechanisms. As a result, the initial permittivity (ε) and dielectric loss tangent ($\tan\delta$) values increase significantly compared to pure PP. The study showed that at low filler contents (10–30%), γ-irradiation up to 50 kGy does not lead to substantial changes in the electrophysical characteristics of the composites. This is associated with the stabilizing role of CdS + ZnS particles in the polymer matrix, which form additional traps for charge carriers and thus improve structural stability under irradiation. However, at an absorbed dose of 80 kGy, an increase in ε and $\tan\delta$ was observed, which is attributed to radiation-induced crosslinking, enhanced charge mobility, and intensified polarization processes. At higher filler concentrations (50%), the behavior of the composite changes considerably. Although ε initially increases at doses of 30–50 kGy, a noticeable decrease is observed at 80 kGy. This decrease is associated with intensified radiation-induced chain scission, oxidative degradation, and destruction of intermolecular bonds within the polymer matrix under conditions of high filler content. Therefore, high concentrations of CdS + ZnS weaken the radiation resistance of the polymer and lead to nonlinear variations in its dielectric properties. The analysis of electrical resistivity revealed that composites with 10–30% filler maintain their specific resistance almost unchanged up to 50 kGy. This confirms that low-filler composites possess greater radiation stability. In contrast, samples with 50% CdS + ZnS exhibit a sharp decrease in resistivity with increasing irradiation dose. This behavior is linked to the formation of conductive percolation pathways resulting from the close packing of semiconductor particles, which becomes more pronounced under γ-irradiation.

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РОЛЬ НАПОВНЮВАЧА CdS + ZnS
У ФОРМУВАННІ РАДІАЦІЙНО СТІЙКИХ
ДІЕЛЕКТРИЧНИХ ВЛАСТИВОСТЕЙ
КОМПОЗИТІВ НА ОСНОВІ ПОЛІПРОПІЛЕНУ

У роботі досліджується вплив γ-опромінення на діелектричні властивості та електропровідність поліпропіленових (PP) композитів, наповнених CdS + ZnS у різних об'ємних концентраціях (10%, 30%, 50%). Встановлено, що для чистого PP і композитів з низьким вмістом наповнювача (до 30%) діелектрична проникність і питомий опір залишаються стабільними для доз опромінення до 50 кГр. У випадку дози 80 кГр спостерігається збільшення діелектричної проникності у зразках з 10–30% наповнювача, що пов'язано зі збільшенням поляризації та утворенням зшивок. У композитах з високим вмістом CdS + ZnS (50%) діелектрична проникність спочатку зростає (до 9,3), а потім різко зменшується, коли доза становить 80 кГр (до 7,1), що свідчить про руйнування полімерної матриці. Питомий опір за високого вмісту наповнювача значно знижується при γ-опроміненні, особливо коли концентрація наповнювача становить 50%, що пов'язано з утворенням провідних каналів.

Ключові слова: γ-опромінення, поліпропілен (ПП), наповнювач, діелектрична проникність, питомий опір, радіаційна стійкість.