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Research Article

Effects of Gamma Irradiation on the Dielectric Properties of Polypropylene-Based Composites

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Abstract

This work investigates the effect of gamma irradiation on the dielectric properties of polypropylene/α SiO_2 nanocomposites. The non-polar nature of polypropylene has allowed it to be used as an insulating material. At the same time, this property makes polypropylene a suitable matrix for nanoparticle-filled composites. In the study, the PP + α-SiO_2 composite was obtained by melt mixing at a temperature of 170–190 °C, and then samples were prepared from it by pressing, and the thickness of the prepared samples was 90 μm. The resulting composites were irradiated with gamma rays. Then, the effect of absorbed dose on the electrophysical parameters of composite samples after irradiation was studied. During the study, a comparative analysis of the electrophysical parameters of non-irradiated and irradiated samples was conducted. It was found that the resulting nanocomposites increase the absorption of electromagnetic waves. As the absorbed dose of radiation increases, the degree of crystallization of the material decreases, the crystalline structure gradually disintegrates, and at high doses it completely disappears. The value of ε increases significantly at the beginning and remains constant at subsequent values of dose increase, leading to a sharp increase in dielectric losses.

Keywords: polypropylene, polymer composites, agglomeration, electromagnetic, aerosil, dielectric permeability

1. Introduction

Nanotechnology enables the creation of new materials with unusual properties. Such materials are widely used in various fields of science and technology, biotechnology, the military industry, construction, and other areas [1], [2]. Polypropylene is a material widely used in household and technical applications. Its resistance to physical and chemical factors and non-polarity have led to its use as an insulating material. At the same time, the addition of nanoparticles makes polymers promising for use as one of their main ingredients.

Recently, extensive research has been devoted to the study of the effects of γ-irradiation on various types of polymers and polymer composites. Such research is aimed at obtaining new types of nanocomposites with elastic and antistatic, electroactive, physical-mechanical, electret, and radiation-resistant properties and expanding their application possibilities. When nanoparticles with high chemical activity are incorporated into a polymer, the physicochemical, electrical, optical, and other properties of the polymer change dramatically depending on the size, shape, agglomeration state, and volume fraction of the particles [3], [4].

In the preparation of electroactive composite materials as promising materials, researchers are also considering studying other effects (temperature, frequency of external electric field, various types of radiation) on the electrical, dielectric, magnetic, and other properties of composites [5], [6].

The electrical conductivity of polyaniline-polymethacrylic/SiO_2 nanocomposites synthesized by chemical oxidation increases with frequency, and the nanocomposites obtained by incorporating SiO_2 nanoparticles into the copolymer increase the absorption of electromagnetic waves [7], [8].

2. Materials and Methods

In the study, PP + α-SiO_2-based composite samples were prepared. During the preparation of composite samples, the particle size did not exceed 90 nm, while the thickness of the prepared samples was 90 μm. The α-SiO_2 content of the filler was taken as 5% and 10% (by volume). Sometimes α-SiO_2 nanoparticles are technically called aerosil. PP + 10% α-SiO_2 composites were obtained by melt mixing at a temperature of 170–190 °C, and then samples were prepared from them by pressing. The resulting composites were irradiated with gamma rays. Next, the effect of absorbed dose on the electrophysical parameters of the composite samples exposed to irradiation was studied. During the study, a comparative analysis of the electrophysical parameters of non-irradiated and irradiated samples was conducted.

3. Results and Discussion

Figure 1 shows the variation of dielectric permittivity (ε) as a function of D for composites with a volume fraction of pure PP and PP + α-SiO_2 filler of up to 5% and 10% before and after γ-irradiation. As can be seen from the figure, when the concentration of α-SiO_2 nanofiller increases, at room temperature (T = 293 K), ε = 2.2 (PP) increases to 3.75 (PP + 10% aerosil). When the radiation dose increases, ε increases very weakly in the PP + 10% aerosil composite sample. As can be seen from the graph, in samples with a filler volume fraction of 5% and 10%, the ε value increases significantly at the beginning and remains constant with subsequent dose increases. At the indicated exposure dose, the value of ε in the PP + 10% aerosil composite sample is 5.34, so a 1.42-fold increase was observed. Such dependence of ε on the absorbed radiation dose is explained by the formation of intra- and intermolecular radicals and their interaction with oxygen according to the reaction: $R^{\cdot} + O_2 \rightarrow ROO^{\cdot}$ [6], [7], [8].

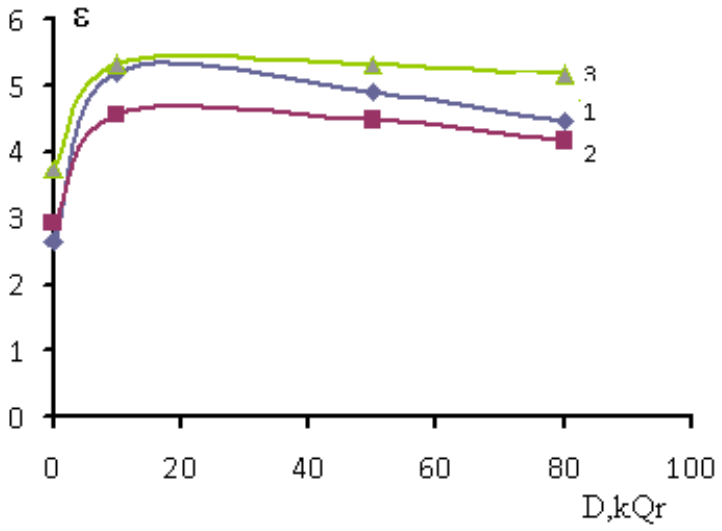


Figure 1. Dependence of the dielectric permittivity of composite samples with aerosil filler volume fraction φ = 5% and φ = 10% on the radiation dose ε = f(D): 1 – initial PP; 2 – PP + 5% α-SiO_2; 3 – PP + 10% α-SiO_2.

On the other hand, as the absorbed dose of radiation increases, the degree of crystallization of the material decreases, the crystalline structure gradually disintegrates, and at high doses it completely disappears. The main factor that determines the change in the properties of the polymer matrix and all composites based on it is the process of disruption of the order of the crystal structure.

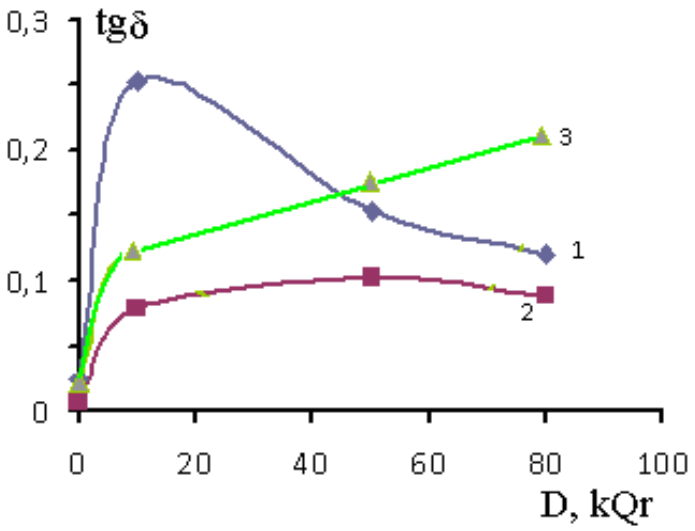


Figure 2. Dependence of the dielectric loss angle of composite samples with aerosil filler volume fraction φ = 5% and φ = 10% on the radiation dose tgδ = f(D): 1 – initial PP; 2 – PP + 5% α-SiO_2; 3 – PP + 10% α-SiO_2.

The results of the change in the dielectric loss angle tangent of the studied samples depending on the radiation dose (D) are given in Figure 2. A certain increase in ε up to the indicated values of the dose leads to a sharp increase in dielectric losses after the subsequent (up to 80–100 kGr) radiation dose (Figure 2). This may be due to the fact that at low doses, the rate of formation of spatial building bonds in the matrix prevails over the rates of destructive processes, while at high doses, D > 10 kGr, the rate of breaking of single bonds increases and the polymer matrix becomes brittle. In addition, studies of the sensitivity of nano-α-SiO_2 filler to γ and neutron radiation [9], [10] have shown that the effect of ionizing radiation leads to a decrease in dielectric loss tgδ. An interesting point, and at the same time contradictory to modern ideas, is that when nano-α-SiO_2 is irradiated with n-rays, the value of ε increases by up to 27 times. As can be seen from the figure, when the concentration of aerosil filler increases, tgδ = 0.0747 in pure PP at room temperature (T = 293 K) decreases from 0.0747 to 0.052 (PP + 5% α-SiO_2), and further decreases to 0.0163 (PP + 10% α-SiO_2).

Similar critical values are not obtained in the PP/nano-α-SiO_2 samples we studied. We believe that in the case of nanocomposites, SiO_2 nanoparticles can play the role of crystallization centers (Figure 3).

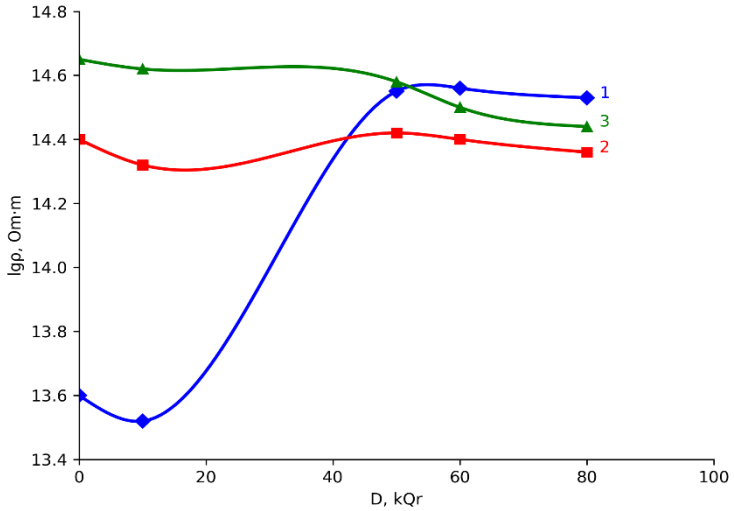
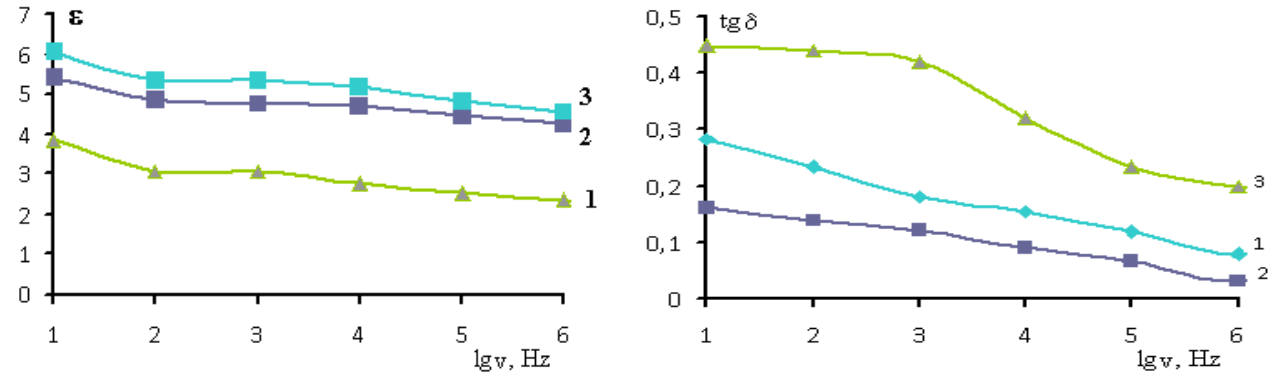


Figure 3. Dependence of the specific resistance of composite samples with aerosil filler volume fraction φ = 5% and φ = 10% on the radiation dose lgρ = f(D): 1 – initial PP; 2 – PP + 5% α-SiO_2; 3 – PP + 10% α-SiO_2.

After samples with a volume fraction of α-SiO_2 nanofiller φ = 5% and φ = 10% were irradiated with a γ-irradiation dose of D = 10 kGr, the ε = f(ν) and tgδ = f(ν) dependences of the dielectric properties of PP-initial,

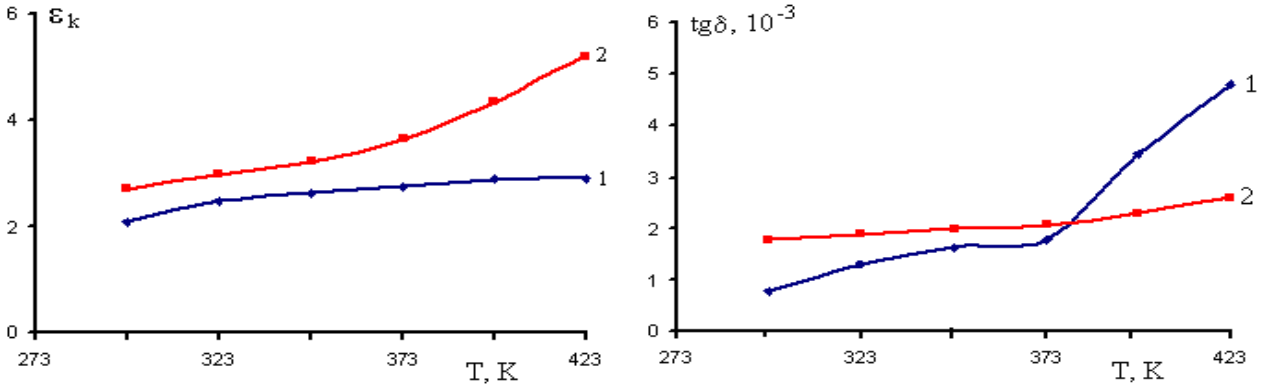
PP + 5% α-SiO_2, and PP + 10% α-SiO_2 composite samples after γ-irradiation were studied and compared in the frequency range of 25–10^6 Hz (Figure 4).



(a) **(b)**

Figure 4. Frequency dependences of dielectric permittivity **(a)** and dielectric loss angle **(b)** of polypropylene/α-SiO_2 nanocomposite samples ε = f(ν) (a), tgδ = f(ν) (b): 1 – D = 0; 2 – D = 10 kGr; 3 – D = 20 kGr.

Figure 5 shows the dependences of ε = f(T) and tgδ = f(T) of pure PP, PP + 10% aerosil, composites after 10 kGr γ-irradiation at room temperature (T = 293 K). The behavior of the initial PP ε = f(T) dependence does not change significantly.



(a) **(b)**

Figure 5. Temperature dependences of dielectric permittivity **(a)** and dielectric loss **(b)** of PP/α-SiO_2 nanocomposite samples after 10 kGr irradiation ε = f(T) (a), tgδ = f(T) (b). 1 – pure PP, 2 – PP + 10% α-SiO_2.

However, in samples with a volume fraction of aerosil filler φ = 10%, after irradiation with a γ-radiation dose of D = 10 kGr, its dielectric permittivity increases at the initial values of the frequency, but then decreases. At the same time, after irradiating samples with a volume fraction of aerosil filler φ = 10% with a γ-radiation dose D = 10 kGr, their dielectric losses decrease very rapidly (Figure 5). A characteristic feature of the dependences given in Figure 5 is that at higher temperatures, both the dielectric loss angle and the dielectric permittivity increase. Therefore, the addition of α-SiO_2 nanofiller reduces the structural homogeneity of the sample up to a dose value of D = 10 kGr.

The effect of γ-rays on polypropylene/α-SiO_2 nanocomposites has been poorly studied. In terms of structure, it can be expected that the nanoaerosil filler will change the surface molecular structure of PP. At the same time, the study of the role and mechanism of action of γ-irradiation should be considered as a guiding factor in modification processes. It is known that PP-based composites have a thermostabilizing effect at doses of D = (3–5) kGr [11], [12]. This is due to the fact that interchain crosslinking processes predominate at low

doses of γ-rays [11], [12]. At relatively high doses, the rate of destruction processes in PP increases, and the quality parameters of the composite decrease [13], [14].

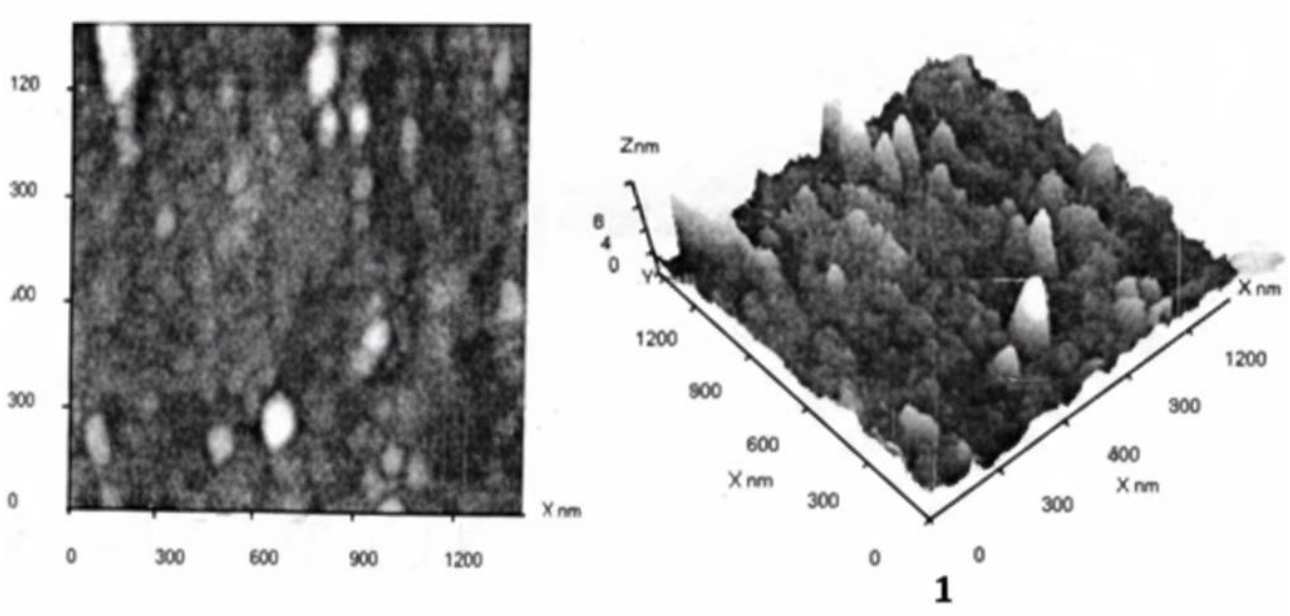


Figure 6. AFM image of a PP/α-SiO_2 (95:5 vol.%) sample irradiated with γ-quanta in air with a dose of D = 5 kGr.

Figure 6 shows the AFM image of PP/α-SiO_2 (95:5%) nanocomposite samples. The surface morphology appears uniform. In addition, the filler is evenly distributed throughout the matrix [13], [14]. Studies show that when the amount of amorphous α-SiO_2 in polypropylene exceeds 10% (by volume), the heterogeneity of nanocomposite samples increases, affecting other structural parameters in the form of agglomerates in the nanoparticles.

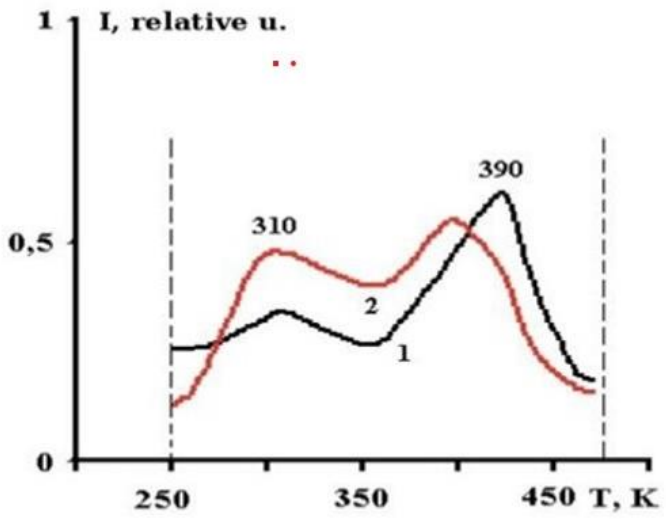


Figure 7. RTL spectra of PP + 5% α-SiO_2 nanocomposite samples: 1 – D = 0; 2 – D = 50 kGr.

It can be seen from the obtained radiothermoluminescent curves that the intensities of the high-temperature radiation peaks at 310 K and 300 ± 2 K were redistributed by the effect of γ-radiation, that is, the first peak shifted to lower temperatures by 5–80 ^{0}C. Both facts clearly demonstrate the possible structural changes as a result of irradiation and the role of α-SiO_2 nanofiller as a nanofiller in these changes. Microscopic studies show that the α-SiO_2 filler prevents the formation of large spherulites in the polymer Spherulite and radial crystallites. The crystallization process ("secondary" crystallization) in PP does not increase the size of spherulites in the material, but rather their number. These structural changes allow for the radiation-induced tracks to be evenly distributed in the polymer and, as a result, allow for the prediction of radiation modification processes.

4. Conclusion

Recently, extensive attention has been paid to the study of the effect of γ-irradiation on various types of polymers and polymer composites such as PP/nano-α-SiO_2. In the study, PP + α-SiO_2-based composite samples were prepared. During the preparation of composite samples, the particle sizes did not exceed 90 nm. The α-SiO_2 content of the filler was taken as 5% and 10% (by volume). PP + α-SiO_2 composites were obtained by melt mixing at a temperature of 170–190 °C, and then samples were prepared from them by pressing. The resulting composites were irradiated with gamma rays. Then, the effect of absorbed dose on the electrophysical parameters of composite samples after irradiation was studied. During the study, a comparative analysis of the electrophysical parameters of non-irradiated and irradiated samples was conducted.

The variation of dielectric permittivity (ε) as a function of D was studied for composites with pure PP and PP + α-SiO_2 filler volume fractions of up to 5% and 10% before and after γ-irradiation. It was found that as the concentration of α-SiO_2 nanofiller increases, ε = 2.2 (PP) increases to 3.75 (PP + 10% aerosil) at room temperature (T = 293 K). As can be seen from the graph, in samples with a filler volume fraction of 5% and 10%, the ε value increases significantly at the beginning and remains constant with subsequent dose increases. Such dependence of ε on the absorbed dose of radiation is explained by the formation of intra- and inter-chain radicals and their destruction by the reaction: $R^{\cdot} + O_2 \rightarrow ROO^{\cdot}$.

The tgδ = f(ν) dependences of PP + 10% α-SiO_2 composite samples before and after γ-irradiation show that the dielectric losses of the irradiated composite change little. However, after 10–50 kGr of radiation, although the value of tgδ increases sharply at the initial frequency, stabilization is observed later. After 50 kGr of radiation, the value of tgδ decreases sharply. It has been observed that the nanocomposites obtained by incorporating SiO_2 nanoparticles increase the absorption of electromagnetic waves.

At the same time, the dielectric losses of samples with aerosil filler volume fractions of φ = 5% and φ = 10% decrease very rapidly after being irradiated with a γ-irradiation dose of D = 10 kGr. At higher temperatures, both the dielectric loss angle and dielectric permittivity increase. Therefore, up to a dose value of D = 10 kGr, the α-SiO_2 nanofiller reduces the homogeneity (in terms of structure) of the sample. The reason for the changes is that radiation-destruction or crosslinking processes, as a result of the influence of radiation, change the mobility of various structural units (segments, lamellae, crystallites, etc.) in the polymer.

On the other hand, as the absorbed dose of radiation increases, the degree of crystallization of the material decreases, the crystalline structure gradually disintegrates, and at high doses it completely disappears. The main factor determining the change in the properties of the polymer matrix and all composites based on it is the process of disruption of the order of the crystal structure.

Author Contributions

Both authors reviewed, edited, and approved the manuscript.

Conflict of Interest

The authors declare no conflicts of interest.

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Abbreviations

Silicon Dioxide (SiO_2), Atomic Force Microscopy (AFM).

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