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Electrical Destruction Processes in Polypropylene + Nanoclay Nanocomposites after Exposure to an Electric Field

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The effect of the D_{k1}-brand nanoclay (NC) additive on the electrical durability (τ) and electrical conductivity (κ) of polypropylene (PP) is investigated. Depending on the amount of NC, the electrical strength (E) decreases and κ increases. To investigate the molecular processes, which take place during electrical destruction, the electrophysical properties are compared with the changes occurring in the structures of pure PP and nanocomposite by pre-exposure to an electric field (ageing). C=C groups formed as a result of the rupture of interatomic chemical bonds in macromolecules after ageing lead to a decrease in E in both PP and nanocomposite, and the rate of decrease depending on ageing time (t) is lower in nanocomposite compared to PP.

Досліджено вплив добавки наноглини (НГ) марки D_{k1} на електричний строк служби (τ) й електропровідність (κ) поліпропілену (ПП). Залежно від кількости НГ, електрична довговічність (E) зменшується, а κ збільшується. Для дослідження молекулярних процесів, які відбуваються під час електричного зруйнування, електрофізичні властивості порівняно зі змінами, що відбуваються у структурах чистого ПП та нанокомпозиту шляхом попереднього впливу електричного поля (старіння). Групи C=C, що утворюються в результаті розриву міжатомових хемічних зв'язків у макромолекулах після старіння, приводять до зменшення E як у ПП, так і в нанокомпозиті, причому швидкість зменшення, залежно від часу старіння (t), у нанокомпозиту нижча порівняно з ПП.

Key words: polypropylene, nanoclay, electrical strength, conductivity, optical density.

Ключові слова: поліпропілен, наноглина, електрична довговічність, елек-

тропровідність, оптична густина.

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1. INTRODUCTION

It is not reasonable to resynthesize polymers and develop their production when there is a need for materials with new complex properties. This is a very long, expensive, difficult and not always successful way out of the situation. One of the promising solutions to this problem is to change the physical modification of existing polymers or add substances of a different nature and structure to give them new complex properties and make them more durable. Therefore, it is no coincidence that in recent years the main focus of polymer physics and chemistry has been on the production and use of composite materials [1].

In studies [2–8], the role of nanoclay (NC) in the mechanical and electrical destruction of polyolefin-based NC nanocomposites was investigated. Changes in physical properties under the influence of external factors have shown that in polymers and nanocomposites, destruction begins with the rupture of interatomic chemical bonds that make up macromolecules. The mechanism of destruction can be explained by comparing such changes in structure with physical properties and by studying the molecular processes occurring under the influence of external factors.

Polypropylene (PP) is a plastic material characterized by high mechanical and electrical strength, high chemical resistance, low moisture and gas permeability. It is easy to process, can mix well with organic and inorganic additives. Operating temperature is of 120–140°C.

It is extensively used in the production of plastic pipes for clean and waste water drainage, in the cable industry as an insulating material, in the manufacture of household items.

Whatever the quality of the raw material, it will inevitably decompose under the influence of external factors during operation.

Due to the fact that polymer products, which have expired, are difficult to reuse (recycle), they cause environmental pollution. Products improved in quality by various methods wear out and fail over time.

The study of destruction of polymeric materials under the influence of external factors is of both scientific and practical importance.

By studying the causes of destruction, critical information such as changes in the properties of polymeric substances and the increase in service life during operation can be obtained.

2. EXPERIMENTAL

D_{k1} brand NC containing Na^+-montmorillonite was used as an inorganic additive included in PP as a composite material. NC is the common name of clay minerals with a layered structure of about 1–3 nm thickness and of 50–150 nm length. Montmorillonite is a grey or white highly dispersed layered silicate. Its chemical structure is $NaO_3(Al,Mg)_2Si_4O_{16}(OH)_2 \cdot nH_2O$.

Thin samples of 50–80 μm were obtained by hot pressing at 15 MPa, 440 K, 10 min after mechanical mixing of different percentage concentrations of NC (1.0%, 2.0%, 3.0%, 4.0%, 6.0%, 8.0%, 10.0%) with PP powder. As both components are in powder form, a homogeneous mixture is obtained. The unchanged density of nanocomposites in low percentages ensures the lightness of fabricated products that is very important from a practical point of view.

According to experiments conducted by Giannelis [9], the production of nanocomposites is carried out in three stages (Fig. 1).

At the first stage (*a*), the tactoid is formed: polymer chains cover the clay agglomerates from the outside. At the second stage (*b*), the chain segments enter the gaps between the clay layers, causing the layers to separate by 2–3 nm. At the third stage (*c*), the clay layers begin to diverge relative to each other, an uneven arrangement of the layers begins, and the layers are completely separated. The uniform distribution of NC in the matrix indicates that the structure of the nanocomposites is flawless.

During the experiment, it is necessary to ensure the uniformity of the electric field and exclude the influence of extraneous discharges on the breakdown voltage value. The electrodes consist of a system with a diameter of 30 mm and 2 mm with rounded edges,

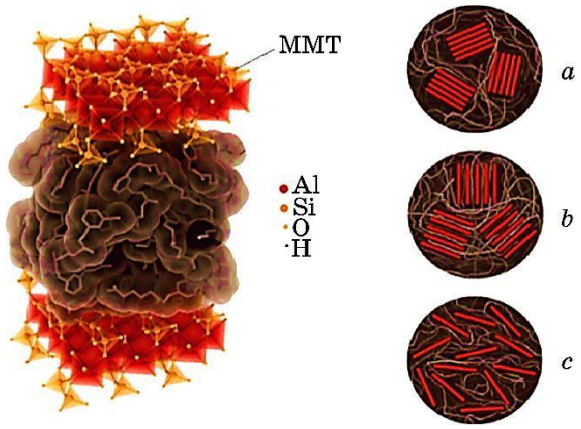

Fig. 1. Three types of 'polymer-layered silicate' composites.

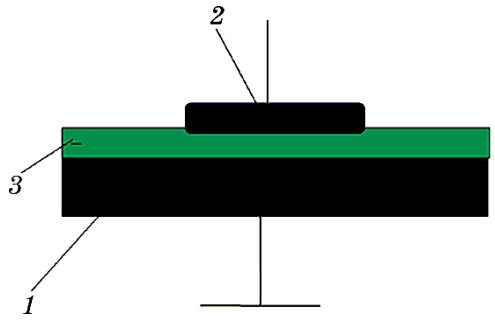

Fig. 2. Test cell: *1*—grounding electrode; *2*—high-voltage electrode; *3*—studied sample.

made of stainless steel. Time τ is measured by applying a voltage to the sample between the electrodes that is less than the breakdown voltage (electrical durability τ is the time that passes from the moment the voltage is applied to the sample until the moment of breakdown [10]). The χ values of the samples were measured at room temperature in a special measuring rod consisting of a system of electrodes. Ageing of PP and composite in an electric field was carried out in a test cell schematically shown in Fig. 2.

To study the changes occurring in the structure of nanocomposites after electric field exposure, the optical density (D) of the C=C group in a double bond corresponding to the frequency of 1640 cm^{-1} was calculated by infrared (IR) spectroscopy at a frequency of 400–2500 cm^{-1} and compared with E. PP and PP + 4.0% NC nanocomposite samples were chosen as the object of study.

3. RESULTS AND DISCUSSION

The dependence of the durability of PP-based nanocomposites with the addition of NC on E is shown in Fig. 3. It is seen that, for each sample, τ decreases as E increases. Using this graph, a decrease is observed when establishing the dependence of E on the amount of NC at the value of $\log\tau = 0$ ($\tau = 1$ sec) (Fig. 4), that is, the electrical strength of PP decreases depending on the amount of NC. The decrease in electrical strength may be caused by the increase in electrical conductivity of nanocomposites. The experimental results of the effect of NC on χ of PP are demonstrated in Fig. 4. Depending on the amount of NC, E decreases and χ increases.

The increase in κ in nanocomposites depends on the type of intracrystalline bonds of the clay minerals, which we use as additives. Covalently bonded dielectric minerals (quartz, mica, feldspar, *etc.*) have a very high resistivity 10^{12}–10^{15} Ohm·m. Semiconductor miner-

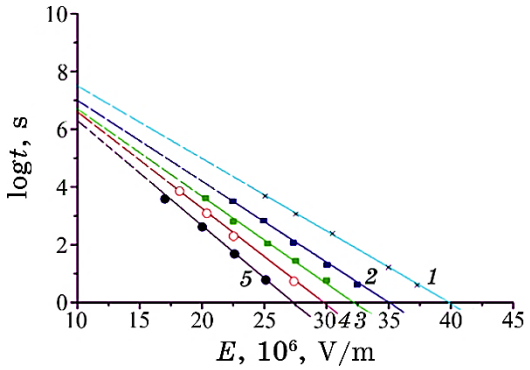

Fig. 3. Dependence of the electrical durability (logτ) of PP + NC nanocomposites on the electric field strength (E): *1*—PP; *2*—PP + 2.0% NC; *3*—PP + 4.0% NC; *4*—PP + 6.0% NC; *5*—PP + 8.0% NC. T = 298 K.

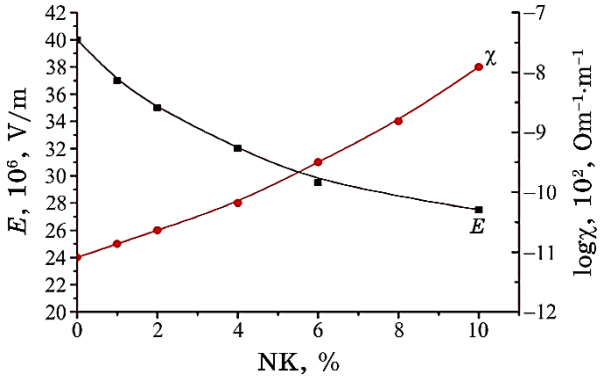

Fig. 4. Dependence of electrical strength and electrical conductivity of PP on the amount of NC.

als (carbonate, sulphate, halide, *etc*.) known mainly for ionic bonds are characterized by high resistivity of 10^4–10^8 Ohm·m. Finally, the resistivity $\rho < 10^4$ Om·m of clay minerals (hydromica, montmorillonite, kaolinite, *etc*.) with ion–covalent bonds is quite low [11]. It would be unreasonable to expect an increase in E and a decrease in κ in nanocomposites obtained with D_{k1}-grade NC, which we used as an additive (Fig. 4), since D_{k1} belongs to the class of minerals of medium permeability ($\chi_{NC} \cong 10^{-4}$ Om^{-1}·m^{-1}, $\chi_{PP} \cong 10^{-12}$–10^{-13} Om^{-1}·m^{-1}).

To study the influence of the electric field, the samples were aged for different hours (t = 5, 10, 20, 30, 40, 50 hours) in a field of strength $E = 1.2 \cdot 10^7$ V/m. From the log$\tau = f(E)$ dependence, it can be seen that E for both PP and PP + 4.0% NC nanocomposite decreased after 50 hours (Fig. 5).

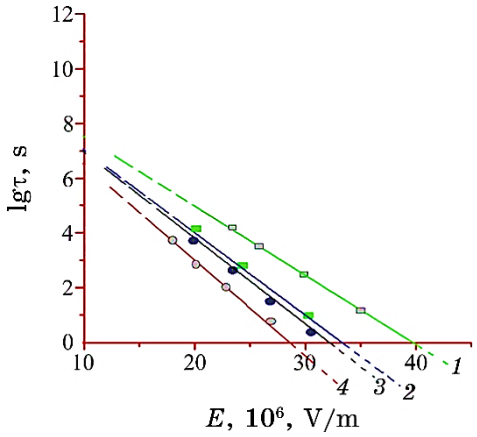

Fig. 5. $\log\tau = f(E)$ dependence for PP and PP + 4.0% NC nanocomposite: *1*, *2*—PP; *3*, *4*—PP + 4.0% NC ($T = 298$ K, $E = 1.2 \cdot 10^7$ V/m; *1*, *3*—$t = 0$; *2*, *4*—$t = 50$ hours).

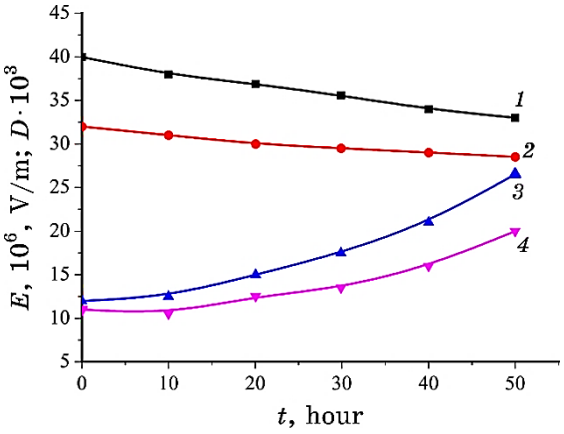

Fig. 6. Dependence of electrical strength and optical density on ageing time: *1*, *2*—*E*; *3*, *4*—*D*; *1*, *3*—PP; *2*, *4*—PP + 4.0% NC.

If we plot the dependence of E on t at $\log\tau = 0$, we will see that the reduction in PP is 10%, and 17.5% in a nanocomposite (Fig. 6). The changes occurring in the structure depending on t were considered to explain the decrease and the difference in decrease.

The IR spectroscopy method has been used to explain the molecular mechanism of destruction at the atomic and molecular level by studying the molecular groups formed as a result of the rupture of interatomic bonds in macromolecules. It is possible to follow the progress of the destruction process by comparing the results ob-

tained in terms of structure and strength properties.

From the absorption spectra obtained under the influence of external factors, it is evident that molecular groups formed at different *wavelength* frequencies (hydroxyl C–OH, carbonyl C=O, methyl CH_3, double bonds C=C corresponding to stretching vibrations, *etc*.) cause changes in electrophysical properties. Starting from $t = 10$ hours, an increase in the intensity of the peak (band) belonging to the C=C group, corresponding to the *wavelength* frequency of 1640 cm^{-1}, is observed, both in PP and in the nanocomposite. The concentration of newly formed groups depends on the intensity of the absorption band [11, 12].

An increase in the concentration of the double bond group C=C formed due to a radical in a macromolecule after the action of an electric field depending on t is the result of shortening of the length of polymer chains and decreasing in molecular weight [12]. If a macromolecule splits into several parts due to the formation of a C=C group, the mobility of the molecules increases and causes a decrease in E depending on t (Fig. 6).

The optical density of the C=C group calculated from the spectra is shown in Fig. 6. The dependence of $D = f(t)$ for both samples shows that the electric field is destructive. In PP, D increases faster than in the nanocomposite, whereas E decreases faster. NC inhibits its destruction depending on the time of exposure to the electric field and suppresses the growth of the C=C group. The destructive processes are faster in pure PP, in the nanocomposite PP + 4.0% NC this process slows down.

4. CONCLUSION

Depending on the amount of NC, the electrical strength of PP decreases and the electrical conductivity increases. It has been noticed that there is a correlation between the electrical strength and optical density of the samples studied by us after exposure to an electric field. Analysis of the spectra obtained by IR spectroscopy has showed that C=C molecular groups are formed as a result of the rupture of interatomic chemical bonds in macromolecules after exposure to an electric field, and the number of these groups increases in both PP and nanocomposite depending on the exposure time.

NC reduces the speed of destructive processes caused by the impact of an electric field.

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