

Structure and Physical Properties of Solid Fullerene-polymer Systems

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With usage of different physical methods (X-ray scattering, mechanical tests, and dielectric spectroscopy) the structure and physical properties of fullerene-containing systems on the basis of amorphous (PMMÑ and PS of the laboratory synthesis) and semicrystalline (PTFE with trade mark F-4) polymers were studied. The concentration C of \tilde{N}_{60} and fullerene soot (FS) in polymer matrix were varied within $C=0-10$ mass% interval. In all investigations were used the films prepared from polymer solutions or melts with additives of C_{60} or FS. The main results of investigations are the following.

Mechanical properties. Changes of mechanical properties (tensile strength σ and breaking elongation ϵ) are different and ambiguous for amorphous and semicrystalline fullerene-containing polymers. With increase of fullerene \tilde{N}_{60} and FS concentration in PMMÑ and PS the tendency of σ and ϵ increasing was observed. In the case of F-4 we observed a monotonous increase of σ and constancy of ϵ in $C=0-3\%$ interval, and a sharp decrease at $\tilde{N}>3\%$. These changes apparently are connected with a competition of the degradation and cross-linking processes in the polymer matrix under the fullerene action.

Structure. On WAXS of films from atactic PMMÑ and PS two amorphous halos were observed. With increase of \tilde{N}_{60} concentration some transformations of WAXS take place: on the background of amorphous halo the tendency of reflection development from aggregates of \tilde{N}_{60} was observed. The order in these aggregates is lower than crystalline but higher than amorphous one. A well-developed micro-heterogeneity of the structure is typical for amorphous fullerene-containing polymers. In comparison with original samples it manifests in the sharp increase of diffuse scattering intensity on SAXS diffractograms with an increasing of \tilde{N}_{60} concentration in polymer matrix.

In the case of F-4 modified by fullerene soot in accordance of WAXS patterns take place the polymorphous transformation of crystalline lattice of F-4 and an increase of its crystal sizes. Fullerene-containing samples of F-4 also possess the well-developed micro-heterogeneity: with increase of the FS content on SAXS patterns can be observed an increase of diffuse scattering, whereas on WAXS can be seen the most strong reflection from the crystalline lattice of graphite having in fullerene soot.

Thermal properties of crystalline lattice and amorphous phase of initial and soot-filled F-4 in the temperature interval $20-330-20\tilde{N}$ were investigated. It was established the influences of FS on thermal coefficients of linear extension of crystal lattice $\alpha_{\tilde{n}}$ and amorphous phase $\alpha_{\tilde{a}}$ are similar, however the rate of temperature growth for $\alpha_{\tilde{n}}$ is more than for $\alpha_{\tilde{a}}$ one and this difference increases with increase of FS concentration. The analysis of the obtained results shows that defects of crystal lattice caused by the degradation of polymer chains are located in a-axis crystallographic direction.

Dielectric properties. Dielectric properties of fullerene-containing PMMÑ were studied. It was established that with increase of \tilde{N}_{60} concentration the dielectric permeability ϵ at the frequency 1 kHz is reduced almost twice. In the interval $20-180\tilde{N}$ the value of ϵ monotonously increases with a growth of the temperature. On dependence $\text{tg}\delta(\tilde{O})$ at the frequency 1 kHz for initial sample the maximum of dielectric losses was observed at $80\tilde{N}$; this maximum is due by dipole-segmental mobility of polymer kinetic units. For samples with $C=1-5\%$ of C_{60} this maximum was also observed but its position was displaced on $10-12\tilde{N}$ in the direction of higher temperature and the maximum intensity decreases. For the sample with $\tilde{N}=10\%$ two maxima were observed on $\text{tg}\delta(\tilde{O})$ curves, at 80°C and 108°C . Apparently, the first maximum corresponds to the dipole-segmental relaxation of PMMÑ molecules, and the second is due by the same relaxation of the PMMÑ molecules with attached to them of \tilde{N}_{60} molecules. Thus the introduction of fullerene \tilde{N}_{60} and FS into the amorphous and semicrystalline polymer matrices is accompanied by changes in their structure and physical properties.