Investigation of Mechanical Properties of Low-Density Polyethylene with Copper Nanoparticles

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Abstract—**In this work, it is shown that the method of the in situ preparation of Cu/LLDPE by combining the formation of a composite and a nanodispersed phase in the viscous-flow state of a polymer makes it possible to achieve a uniform distribution of nanoparticles in the matrix and effectively regulate their mechanical and functional properties.**

The optimal concentration of Cu nanofiller was found to be 2-5%, allowing to achieve the best mechanical properties. Comparative analysis of the physical and mechanical properties of Cu/LLDPE nanocomposites obtained by various methods shows that the deformation and strength characteristics of the 3CuLLDPE nanocomposite obtained by the in situ method are improved in comparison with the properties of the 3CuLLDPE nanocomposite, prepared by ex situ method.

The relationship between the filler content and the modulus of elasticity/tensile strength has been determined. With an increase in the filler content, the elastic modulus increases by 10-20%, and the tensile strength decreases by 30%. Elongation at break for samples with nanofiller content up to 3 wt. % higher than unfilled polymer.

Keywords—**Mechanical properties, composites, nanoparticles, polymers.**

I. INTRODUCTION

POLYMER materials are widely used for matrix insulation of metallic and semiconducting nanoparticles. The development of modern technology requires the creation of new structural materials with high elastic-strength characteristics, and, on their basis, structures with more effective weight data. The creation of polymer composites based on nano-modified binders has been one of the priority areas of research in the field of composite materials manufacturing technologies for many years [1]-[12]. In such nanocomposites, the polymer matrix promotes the stabilization and organization of metal-containing nanoparticles, and the dielectric medium can affect their optical and electronic properties. In turn, there are numerous examples indicating a change in the mechanical, thermal, and other properties of polymers in the presence of nanoparticles [13]-[29].

A new approach in nanotechnology proposes the use of fillers at the nanometer scale, since nanofillers have a large surface area, which makes them chemically very reactive and helps them bind better to the matrix. Nanoparticles embedded in a polymer matrix are attracting more and more interest due to the unique mechanical, optical, electrical and magnetic properties exhibited by nanocomposites and significantly improve them [13]-[16]. These improvements depend on the volumetric content of the filler as well as the type and characteristics of the nanofillers [17]-[33]. Nanofillers of a metallic or inorganic type can be considered an excellent candidate for hardening polymer matrices [34]-[58].

The development of modern technology requires the creation of new structural materials with high elastic-strength characteristics, and, on their basis, structures with more effective weight data. The creation of polymer composites based on nano-modified binders has been one of the priority areas of research in the field of composite materials manufacturing technologies for many years [1]-[12].

Significant progress has been made in this area [13]-[21]. The development of composite materials that improve their operational limits is based on the reinforcement of two or more fibers into a single polymer matrix, which leads to an improved material system called hybrid composites with a wide variety of material properties [22]-[25].

When creating nanocomposites, the key tasks are the development of efficient, reliable, and affordable production technologies for mass production, which make it possible to obtain materials with stable characteristics. The hand lay technique, also called wet lay, is the simplest and most widely used process for producing flat reinforced composites. The process consists of laying layers of a polymer in successive layering using an epoxy matrix. Wet-laying is a molding process that combines layers of reinforced carbon fiber with epoxy to create a high-quality laminate. Before starting the installation process, you must prepare the appropriate form. This preparation consists of cleaning the table and applying a release agent to the surface. The manual laying process can be divided into four main steps: mold preparation, epoxy coating, laying and curing. Form preparation is one of the most important steps in the installation process. This process requires dry reinforcement layers and the application of a wet epoxy matrix. They are connected together - reinforcing material, impregnated with a matrix - epoxy resin.

Adding metal nanoparticles to polymers is a way to obtain materials that combine the properties of polymers (lightness, formability, elasticity), on the one hand, and, on the other, the properties of metals (conductive, optical, magnetic, catalytic properties, etc.) and thus, in in particular, to develop new materials with functional properties for biomedicine applications, protective packaging for products or special equipment [30]-[38].

The aim of this work is to obtain composite materials based on linear low-density polyethylene (LLDPE) and copper nanoparticles and to establish the effect of synthesis conditions on the physical, mechanical and functional properties of the nanocomposite material [59]-[76]. To achieve this goal, the following tasks were solved: the development of a method for obtaining composite materials based on LLDPE and Cu nanoparticles and the study of the effect of nanofiller and its content, technological modes of production on the physical and mechanical properties of nanocomposite materials.

II. Obtaining polymer nanocomposites

In the case of Cu/LLDPE nanocomposites obtained by the in situ method, metal nanoparticles are formed during the thermal decomposition of the precursor at the moment of its mixing with the polymer melt during extrusion. The process of obtaining a nanocomposite material is carried out under conditions of coincidence of the temperature ranges of the decomposition of metal precursor with the temperature range of the presence of polymers in a viscous-fluid state. The technical result is to simplify the production technology of film composite materials by combining the production of a

composite material and a nanodispersed metal phase in one stage, as well as obtaining homogeneous (with uniformly distributed nanoparticles in a polymer matrix) film composite materials based on thermoplastic polymer matrices and copper nanoparticles with adjustable size and concentration of the dispersed phase.

The advantage of the proposed method is its technological simplicity, the availability of starting materials and thermoplastic polymers for large-scale production, the use of traditional technological cycles for processing polymeric materials (extrusion, mixing in micro-mixers, pressing) to obtain a film nanocomposite material.

Film samples of nanocomposite materials were obtained in two modes by casting using an injection molding machine (film thickness 1,85 mm) or hot pressing (film thickness 0,21 mm). The in situ approach was tested to obtain these nanocomposites. It was of interest to study how the technological conditions of the Cu/LLDPE production process affect their elastic and deformation properties.

Table 1 shows the main physical and mechanical characteristics of Cu/LLDPE nanocomposites obtained by the in situ method. The tensile curves of nanocomposite films are shown in Fig. 1.

Table I. Physical and mechanical properties of Cu/LLDPE nanocomposites

Sample	E, GPa	σ at rupture, MPa	εp, %
LLDPE	0,27±0,01	29,0±0,2	1015±6
0,5Cu LLDPE	0,29±0,01	22,3±1	1066±6
1Cu LLDPE	0,32±0,01	21,6±2,9	1049±118
3Cu LLDPE	0,30±0,02	22,2±2,2	1025±100
3,5Cu LLDPE	0,29±0,01	22,3±0,8	1024±27
5Cu LLDPE	0,33±0,01	20,8±2	1026±80
8Cu LLDPE	0,31±0,01	21,9±0,9	992±40
20Cu LLDPE	0,32±0,01	19,5±2,1	920±128

As evidenced by the obtained data, the general tendency of the influence of the content of nanofillers on the deformation-strength characteristics of materials is also preserved in the case of the studied Cu/LLDPE nanocomposites.

III. Physical and mechanical properties of polymer nanocomposites

Samples of the composite material were made on the basis of ED-20 epoxy resin with copper nanoparticles. The volumetric content of nanoparticles was up to 20%, the diameter of the fibers was 50 nm. Samples of three types were considered: 1) a quasi-homogeneous layered composite with a unidirectional reinforcement scheme, 2) a unidirectional composite with a wavelike layered structure, and 3) a pure matrix.

Mechanical tests were carried out on a universal electrical installation Instron 5969 (50kN) and Instron 5982 (100kN)

(UK) with Bluehill 3 software. Compression tests were carried out. Test speed was 1 mm/min.
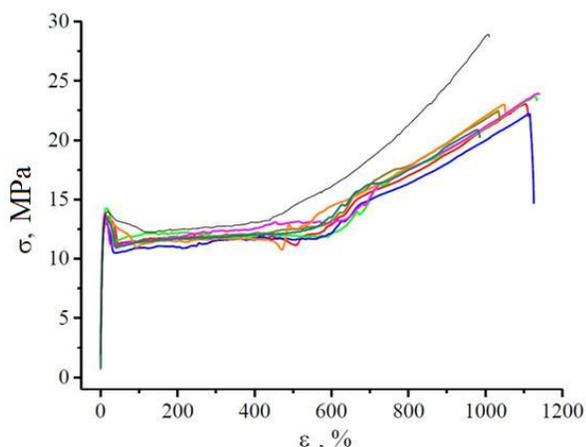

Fig. 1 Stress-Strain curves for initial LLDPE and Cu/LLDPE nanocomposites with different filler contents

The graph shows that with an increase in the filler content, the elastic modulus increases, and the tensile strength decreases (Fig. 2). Note that the elongation at break for samples with a nanofiller content up to 3 wt% is higher than that for an unfilled polymer, and then with a further increase in the nanofiller concentration, the elastic properties of nanocomposites deteriorate (Fig. 3, 4).
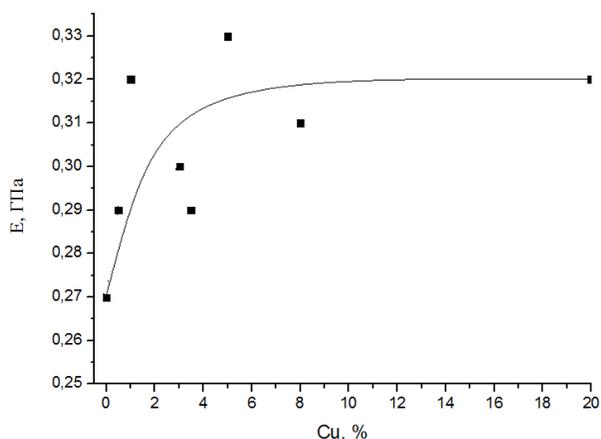

Fig. 2 Dependences of the elastic modulus of the Cu/LLDPE nanocomposite on the filler content
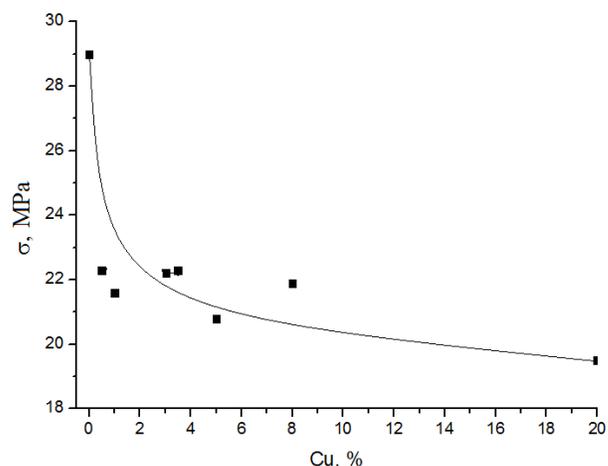

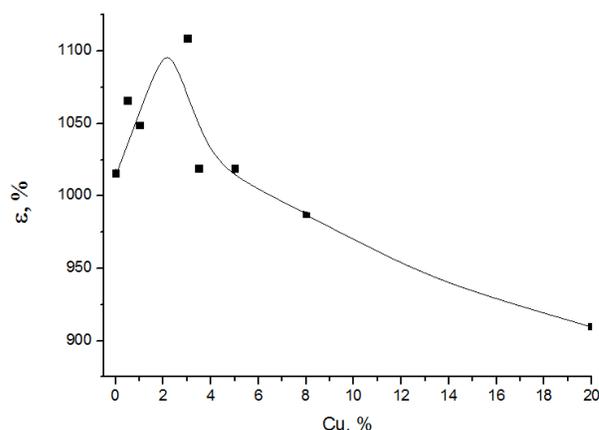Fig. 3 Dependences of the tensile strength of the Cu/LLDPE nanocomposite on the filler content

Fig. 4 dependence of elongation at break of Cu/LLDPE samples on nanofiller content

Comparative analysis of the physical and mechanical properties of Cu/LLDPE nanocomposites obtained by various methods shows that the deformation and strength characteristics of the 3CuLLDPE nanocomposite obtained by the in situ method are improved in comparison with the properties of the 3CuLLDPE nanocomposite, prepared by ex situ method. This opens new prospective for tailored fabrication of polymer composite materials for subsequent practical application as functional and construction materials.

IV. DISCUSSION AND THEORETICAL ISSUES

To identify the elastic properties of a monolayer from the known values of the elastic moduli of composite samples with different reinforcement schemes, we will use the classical model of layered composites.

We use the model of spherical inclusions to model the properties of the filled matrix, assuming that the reinforcing particles of titanium dioxide are spheres. We assume that the particles are absolutely solid and do not collapse (the upper estimate). Bulk content 0,6%. We use the Digimat - MF module, the averaging method of Mori - Tanaka. Strength criterion - according to the maximum principal stresses acting

in the matrix.

If the initial volumetric content of inclusions is set to 3%, the model predicts that the properties of the matrix will not change, since there are too few inclusions. The effect of the interfacial layer must be taken into account. For this, we will carry out a calculation with the setting of the effective volumetric content (volumetric content of the filler + volumetric content of the interfacial layer, under the assumption that their properties are equal). Let us select the effective volumetric content that allows us to describe the obtained experimental data in relation to the elastic modulus and ultimate strength.

The conducted studies allowed to investigate residual deformations in panels with an asymmetric reinforcement scheme based on the obtained analytical solution, as well as numerical modeling. Comparison of the results of analytical and numerical solutions with the obtained experimental data confirms the reliability and validity of the developed mathematical models and research methods for effective thermomechanical characteristics and residual stress-strain state of panels made of layered nanomodified materials.

An analytical calculation to determine the effective elastic properties of a monolayer will be carried out in the Digimat-MF module using the Mori-Tanaka averaging method. Separately, we will also evaluate the effect of the filler on the elastic modulus of the matrix, considering the material containing only nanoparticles as inclusions. Based on these calculations, we will determine the "effective" volumetric content of inclusions, taking into account the influence of interphase layers formed around the inclusions. The influence of these layers, as will be shown below, cannot be neglected, since in this case underestimated values of the elastic characteristics of the matrix will be obtained. Therefore, in fact, knowing the modulus of elasticity of the nanomodified matrix from experiments, the content of the filler will be selected such that the calculation and experiment will coincide. The found value of the effective volumetric content of inclusions is further used in analytical and numerical calculations of the properties of a monolayer.

Numerical calculations will be carried out using the Digimat-FE module. The size of a cubic representative fragment was set by the system automatically. The effective elastic properties were calculated by determining the ratio of the volume-averaged representative fragment of the stress level to a given value of homogeneous deformations. In this case, the boundary conditions and the geometry of the fragment itself are periodic. The calculations were carried out using the finite element method.

Nanoparticles, even with a very low volumetric content (less than 1%), are contained in such a fragment in a very large amount, and it is impossible to model their effect at this scale level. For example, a cubic fragment of a 1 μm matrix contains more than thousand nanoparticles for a given volumetric content. Therefore, in particular, the nano-modified binder is white, while the usual binder is yellow. To model such

materials, it is necessary to resort to multiscale approaches and to carry out a consistent determination of effective properties at various scale levels. This task is greatly simplified if the properties of the nanomodified matrix are known from experiments. In particular, it is known that its Young's modulus is 2.5 GPa. The missing characteristic is Poisson's ratio, which can be approximately taken unchanged, or estimated on the basis of analytical calculations using the found value of the "effective" volumetric content of the filler, which was done. Further, it suffices to numerically solve the averaging problem on a representative fragment containing only nanoparticles.

To obtain an analytical assessment of the properties of the nanomodified sample, the "effective" volumetric content of nanoparticles in the matrix was preliminarily determined, at which the calculated value of the effective Young's modulus of the nanomodified matrix coincides with the known experimental value (2,5 GPa). It was found that if we do not take into account interfacial effects, then the calculation predicts the effective Young's modulus equal to 2,04 GPa, with an initial value of 2 GPa. That is, with such a low content (1%) of even very hard inclusions, they should not have a significant effect on the properties of the material. The experimentally found increase in the matrix modulus can be explained by the influence of hardened and rigid interphase zones formed around the inclusions. For an approximate assessment of their influence, the concept of "effective" volumetric content of inclusions is introduced. It is assumed that the properties of the interphase zones and inclusions are the same, and the calculation should use the value of the "effective" volumetric content of inclusions, which is the sum of their real volumetric content and the content of interfacial zones. This value was approximately 10% for the considered composite. In this case, the predicted Young's modulus of the modified matrix is 2,5 GPa, Poisson's ratio is 0,39, and the thermal expansion coefficient is $5,76 \cdot 10^{-5}$.

Based on the results of experimental studies, the effective characteristics of a monolayer made of a composite material based on both conventional and nanomodified matrices were determined, and a solution to the corresponding inverse problem was obtained. It was found that the addition of nanoparticles within the experimentally found range of 2-5% leads to a slight increase in the longitudinal elastic modulus and shear modulus of the monolayer, as it can be seen from experimental data. In this case, there is an almost twofold decrease in the elastic modulus in the transverse direction and a decrease to zero Poisson's ratio. The reliability of the developed numerical models is confirmed by a good correlation between the results of both numerical and analytical solutions and the obtained experimental data on the study of the thermomechanical characteristics of nanomodified materials.

Currently, a large number of studies have been carried out to obtain and study the strength of composite materials. A large amount of experimental data dedicated to the synthesis

and exploration of heterophase composites were obtained. However, well-known studies were carried out mainly for the case of quasi-static loading of materials. The effects of strain rate influence on the strength of composites with bent fibers have been studied to a much lesser extent, although they have also been considered. Analytical and numerical models are proposed to take into account the effects of curved stacking and the effect of fiber curvature. Some applied models, in particular, are used in standard software packages for finite element modeling. The buckling effects of fibers, even in the case of ideal straight-line packing, leads to a decrease in the compressive strength in relation to the ultimate tensile strength of the fiber composite by more than 1,5 times.

V. Conclusion

As a result of the studies carried out, it was shown that the in situ method of obtaining CuLLDLP nanocomposites makes it possible to obtain highly dispersed nanoparticles and achieve their more uniform distribution in the polymer matrix, which is reflected, as demonstrated, in improved deformation and strength characteristics of the materials obtained. The optimal concentration of Cu nanofiller was found to be 2-5%, allowing to achieve the best mechanical properties. The relationship between the filler content and the modulus of elasticity/tensile strength has been determined. With an increase in the filler content, the elastic modulus increases by 10-20%, and the tensile strength decreases by 30%. Elongation at break for samples with nanofiller content up to 3 wt. % higher than unfilled polymer.

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