

Tribological Properties of Polymer Composite with Impregnated Quasicrystal Nanoparticles

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Abstract—In this work, a study is carried out on the introduction of quasicrystal particles into a thermoplastic polymer and it is shown that this leads to changes in the structure of polyethylene. The introduction of quasicrystal particles into a thermoplastic polymer leads to changes in the structure of polyethylene: the degree of crystallinity decreases from 42% (PE) to 27% (10AlCuFe/PE) with increasing concentration of the filler, the ratio of bands corresponding to amorphous and crystalline regions in the IR spectra changes, which indicates on the amorphization of the PE structure.

The specimens have improved wear resistance (the wear rate decreased by 96% compared to the original PE), but the friction coefficient remained practically unchanged. It is shown that the addition of quasicrystal nanoparticles in a small amount (up to 10 wt.%) leads to an increase in hardness, but does not have a noticeable effect on the surface roughness. The results obtained indicate that quasicrystals can serve as effective fillers for promising polymeric materials in products for aerospace, instrument making, and other industries.

Keywords—Composites, nanoparticles, tribological properties, polyethylene.

I. INTRODUCTION

EPOXY resins are commonly used as polymer matrices in high performance composites due to their good heat resistance, durability, electrical, chemical and mechanical properties. However, its range of applications is limited due to its fragility and poor toughness. One of the ways to overcome this problem is to change the properties of epoxy matrices by

introducing a rigid filler into the polymer matrix [1]-[12]. 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]-[43].

Modern composites have not only a wide range of physical and mechanical properties, but are also capable of directionally changing them, for example, increasing fracture toughness, regulating rigidity, strength, and other properties. These possibilities are expanded when fibers of different nature and geometry are used in composites, i.e., when creating hybrid composites. In addition, these materials are characterized by the appearance of a synergistic effect (coordinated joint action of several factors in one direction).

The main purpose of the polymer binder is to bind the filler together, to ensure the joint operation of all monofilaments (or particles, if a dispersed filler is used), to ensure the solidity of the material and the transfer (distribution) of stresses. The properties of the binder almost completely depend on: heat and heat resistance, resistance to the action of various working media (water, steam, fuel, oils, etc.), impact strength, impact strength, resistance to prolonged exposure to alternating loads, creep, stress relaxation.

After curing (for thermosetting materials) or hardening (for thermoplastic materials), the binder turns into a matrix. The

matrix is a continuous phase, the layer thickness of which can vary from 1 to 1000 μm [19].

In the “ideal” case, the binder should have the following properties: the deformation properties of the matrix should be no lower than that of the filler $\epsilon_m > \epsilon_n$; the binder should have a relatively high modulus of elasticity ($E > 2000$ MPa); the binder should have good adhesion to the filler ($\tau_{sd} > 20$ MPa).

The matrix and filler must necessarily have good compatibility, however, must not dissolve in each other. Based on the factors of matrix selection: molding technology; type of production; geometric features of the resulting product; technological and operational properties in this study will be studied linear low density polyethylene.

The properties of the interface or interfacial zone, first of all, the adhesive interaction between the fiber and the matrix, determine the level of properties of composites and their retention during operation. Local stresses in the composite reach their maximum values just near or directly at the interface, where material destruction usually begins. The interface must have certain properties to ensure efficient transfer of the mechanical load from the matrix to the fiber. The adhesion bond at the interface should not be destroyed under the action of thermal and shrinkage stresses arising from the difference in the temperature coefficients of linear expansion of the matrix and fiber or as a result of chemical shrinkage of the binder during its curing.

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 [44]-[71].

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.

The prospects for using quasicrystals are associated with coatings and films, steel hardening, and the preparation of composites with dispersed quasicrystalline fillers. The introduction of a quasicrystalline Al-Cu-Fe alloy powder into various polymers significantly increases their wear resistance at a lower abrasiveness in comparison with other fillers (plastic metals or ceramics) [1-12]. Quasicrystals (QC) are solids. As a rule, these are intermetallic phases characterized by perfect long-range order in the absence of translational symmetry and in the presence of rotational symmetry with axes of the 5th, 8th, 10th, or 12th orders, which are forbidden in crystalline materials.

Stable QC phases are present in phase equilibrium diagrams and can be obtained by sufficiently slow cooling from the liquid. Quasicrystals of the Al — Pd — Mn system were obtained in the form of large perfect single crystals, on which, in particular, extensive studies of the regularities of their deformation were carried out.

Quasicrystals are extremely fragile. They have few dislocations, and those that are present have low mobility. As metals bend, creating and moving dislocations, the almost complete absence of movement of dislocations causes brittleness. On the positive side, the difficulty of dislocation movement makes quasicrystals extremely solid. They are highly resistant to deformation. This makes them excellent candidates for high strength surface coatings [13]-[21].

At the moment, there is a wide range of methods for the production of quasicrystalline materials, such as crystallization from melt, spontaneous crystallization methods, gas spraying, mechanical fusion, electrodeposition, physical vapor deposition, gas evaporation, laser or electron beam melting and electron irradiation, low temperature annealing of amorphous phases or high-temperature heat treatment of crystalline intermetallic phases [22]-[31]. However, the most common methods for the manufacture of Al — Cu — Fe quasicrystals are those that are accompanied by melting processes - crystallization from the melt, gas sputtering, and mechanical alloying.

The use of Al-Cu-Fe QC as a filler can significantly improve the mechanical, tribological and thermal properties of composites with a polymer matrix. Polymers filled with Al-Cu-Fe QC show improved wear resistance and mechanical properties due to tough core with low aspect ratio. In addition, the QC powder in the polymer matrix practically does not cause abrasion of the steel counter-surface materials during wear tests [32-37]. Among the most interesting examples of Al-Cu-Fe QC-filled polymers are composites based on epoxy resin, ultra-high molecular weight polyethylene (UHMWPE), polyamides, polytetrafluoroethylene, ethylene vinyl acetate, and others.

II. PREPARATION AND STUDY OF THE PROPERTIES OF NANOCOMPOSITES WITH QUASICRYSTALS

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.

The matrix can be a thermosetting polymer - epoxy resin, which has already found many applications: from structural composites to adhesives and surface coatings. Epoxy resins already have a number of unique qualities among polymers: no shrinkage during curing, high adhesion to various substrates, good dielectric and other valuable properties [36]-[44]. Nanocomposites using thermoplastic polymers are well known and studied to improve mechanical, electrical, thermal and insulating properties.

Currently, there are a huge variety of approaches to the production of metal-polymer nanocomposites, but the most technologically advanced are processes for the production and processing of polymers, methods of introducing metal nanoparticles (ex situ and / or in situ) into a polymer melt, methods of polymerization dispersion or joint formation of a dispersed phase and a polymer matrix in one reactor.

Methods for the synthesis of nanocomposite materials (or nanocomposites) are divided into two main large approaches: in situ and ex situ. The creation of nanocomposites by the in situ method makes it possible to simultaneously (in one stage) obtain both a matrix and nanoparticles, obtaining a nanocomposite at the output. The advantage of the method is that it prevents particle agglomeration while maintaining a good spatial distribution in the polymer matrix. The main disadvantage of the method is that all the products of the synthesis of nanoparticles remain in the nanocomposite, which can deteriorate the quality and purity of the obtained material.

When using the ex situ method, each stage of nanocomposite creation is brought into a separate process: from the synthesis of nanoparticles to the preparation of a nanocomposite. This method is more energy and labor intensive compared to in situ, and requires much more time. Also, special attention should be paid to the dispersion of the nanocomponent in the matrix, since during long-term storage

nanoparticles are collected into larger ones and they must be dispersed by various methods, for example, by ultrasonic treatment [1]-[8]. The ex situ synthesis method is more suitable for large scale industrial applications than the in situ method.

The ex situ method, despite all its drawbacks, is used more often than the more technologically advanced in situ method. First of all, this is due to the simplicity of the approach based on the fact that in such reactions the synthesis of nanoparticles is not complicated by additional reactions with the polymer matrix, the resulting nanocomposite will not contain by-products associated with the formation of nanoparticles in the polymer matrix, etc.

Linear low density polyethylene (LLDPE) was used as a polymer matrix, and nanoparticles of quasicrystalline alloy Al₆₅Cu₂₂Fe₁₃. The ex situ method was used for preparation of the nanocomposites – the nanoparticles were introduced into the polymer matrix by the stirring and distributed by volume. The composition, morphology and microstructure of the obtained nanocomposite materials were investigated by XRF methods (Table 1).

Table 1. XRF data for the 10Al₆₅Cu₂₂Fe₁₃/LLDPE nanocomposite.

2 θ /deg	d/Å	I imp/s	Irel (%)	Phase
21,70	4,095	1261	100	LLDPE
24,10	3,693	639	51	LLDPE
26,30	3,389	56	4	LLDPE
36,50	2,462	67	5	LLDPE
43,05	2,101	405	32	Al ₆₅ Cu ₂₂ Fe ₁₃
45,35	2,000	379	30	Al ₆₅ Cu ₂₂ Fe ₁₃
53,15	1,723	51	4	LLDPE
55,00	1,670	63	5	LLDPE
64,10	1,453	59	5	LLDPE
77,05	1,238	119	9	Al ₆₅ Cu ₂₂ Fe ₁₃

It was found that during dispersion, filler nanoparticles are evenly fairly distributed in the volume of the matrix, and conglomerates are observed. This is confirmed by broadened peaks in X-ray diffraction patterns corresponding to the nanofiller phase (Fig. 1). Appearances other lines are not observed, which indicates the progress of the process of dispersion of the filler in the polymer matrix without phase transformations.

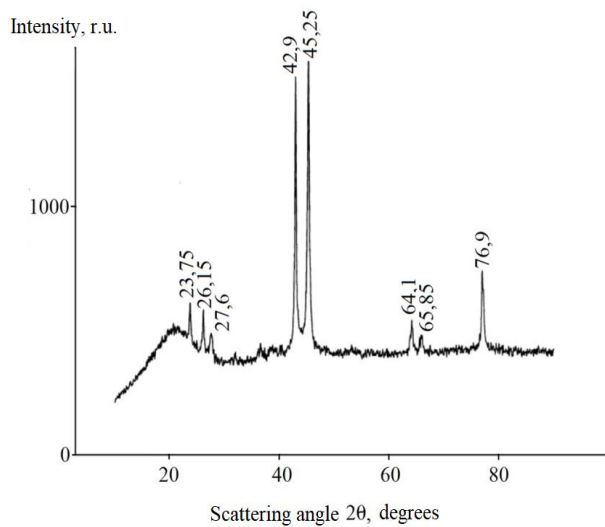


Fig. 1 x-ray diffraction pattern of Al₆₅Cu₂₂Fe₁₃/LLDPE nanocomposite.

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).

Ensuring high wear resistance is associated with the prevention of wear, a decrease in the rates of initial and steady-state wear. Wear resistance refers to the properties of a material to resist wear under certain friction conditions estimated by the inverse of the wear rate or wear rate, therefore, the wear rate can be used to judge the durability of the part. Table 2 shows the values of the wear rate and the coefficient of friction of the obtained composites depending on the amount of filler content Al₆₅Cu₂₂Fe₁₃.

Table 2. The value of the coefficient of friction and the rate of wear of the composite depending on the content of the quasicrystalline alloy.

Material	Wear rate, I, 10 ⁻⁶ kg/h	Friction coefficient, f
LLDPE	40	0,2
LLDPE + Al ₆₅ Cu ₂₂ Fe ₁₃ 0,1%	20-25	0,2
LLDPE + Al ₆₅ Cu ₂₂ Fe ₁₃ 1%	1,5-1,7	0,2
LLDPE + Al ₆₅ Cu ₂₂ Fe ₁₃ 5%	0,3-0,6	0,3
LLDPE + Al ₆₅ Cu ₂₂ Fe ₁₃ 10%	0,8-1,2	0,18-0,22

As a result of the studies carried out, it was found that the friction coefficients of the obtained composites did not differ

significantly and their values were in the range $f = 0.18-0.30$. When studying the wear rate (Fig. 2), data were obtained that indicate that with an increase in the content of the quasicrystalline alloy in the polymer matrix, the wear rate decreases, therefore, this contributes to an increase in wear resistance.

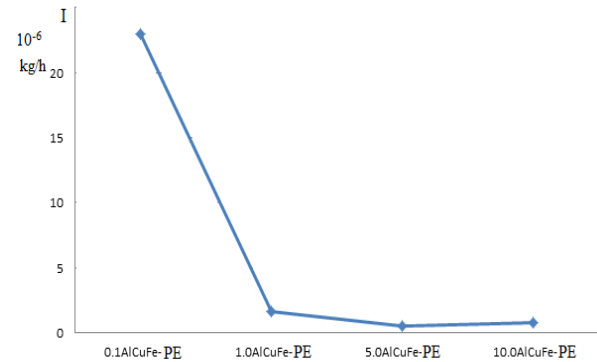


Fig. 2 The value of the wear rate of the composite depending on the content of the quasicrystalline alloy.

One of the most important characteristics of metals and alloys that affect the wear rate is hardness: with increasing hardness, the wear resistance of the material increases. In addition, the wear resistance of the material also depends on the coefficient of friction of the alloy. Since the friction coefficients of the studied samples differ insignificantly, the priority role in the formation of high wear resistance of the obtained composite should be given to the high value of the hardness of the quasicrystalline alloy.

III. CONCLUSION

The introduction of quasicrystal particles into a thermoplastic polymer leads to changes in the structure of polyethylene: the degree of crystallinity decreases from 42% (PE) to 27% (10AlCuFe/PE) with increasing concentration of the filler, the ratio of bands corresponding to amorphous and crystalline regions in the IR spectra changes, which indicates on the amorphization of the PE structure. The specimens have improved wear resistance (the wear rate decreased by 96% compared to the original PE), but the friction coefficient remained practically unchanged. From the data obtained it is seen that the addition of quasicrystal nanoparticles in a small amount (up to 10 wt.%) leads to an increase in hardness, but does not have a noticeable effect on the surface roughness. This means that the improved mechanical properties in this case are achieved. The results obtained indicate that quasicrystals can serve as effective fillers for promising polymeric materials in products for aerospace, instrument making, and other industries.

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