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SYNTHESIS OF METAL-CARBON NANOCOMPOSITES BASED ON THE HIGH PRESSURE POLYETHYLENE PROPERTIES

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ABSTRACT

Metal-carbon nano composites are of a great interest as new generation materials with a set of valuable practical properties. The effect of cobalt nanoparticles and multilayer carbon nanotubes stabilized in a polyethylene matrix on the physic mechanical and thermal properties of these nano composites has been studied. An improvement was detected in the strength and deformation parameters, as well as in the thermal-oxidative stability of the obtained nano composites. This can be attributed to the effects of structural and chemical stabilization of the polymer matrix. Small amounts of these nano fillers introduced into the polymer, play the key role as structure-forming agents-artificial nuclei of crystallization, which contributes to the formation of a small-spherolite structure in the polymer, characterized by improved physical, mechanical and thermal properties in the resulting nano composite.

Keywords: Cobalt-containing, Nanoparticles, Multilayer carbon nanotube, Metal-carbon, Polyethylene matrix, High pressure polyethylene.

INTRODUCTION

For recent years, there has been considerable interest in composite materials based on polymer matrices and nano sized metal particles, which is related to a widespread application from catalysis to nanotechnology in information technology. The unique properties and improved characteristics of nano materials are related to their size, surface structure, and interfacial interaction. Metal-carbon nano composites are of great interest as new-generation materials with a set of valuable practical properties.

The use of metal nanoparticles of variable valence (copper, cobalt, nickel, etc.) incorporated in polymers allows to obtain fundamentally new materials, that can be widely used in radio and optoelectronics as magnetic, electrically conductive and optical medium (Gubin et al., 2005; Pomogajlo et al., 2018; Yurkov et al., 2014). Polymer nano composites can be obtained by the in-situ method, i.e. by polymerizing the monomer in the presence of a nano filler previously dispersed in the reaction medium (Antipov, et al., 2003). The method of introducing a nano filler into a polymer melt is more preferable. This method is most convenient for application in modern industry. It makes possible to obtain nano composite polymers for a wide range of manufacturers, which makes this method promising and cost effective (Savinova et al., 2011; Kurbanova et al., 2020). Modification of polyethylene by generating various composite materials can significantly expand the scope of its application. Filled polyethylene takes one of the first places among filled thermoplastics. Currently, more and more attention is paid to the development of composites with nano scale fillers. Such composite materials own higher performance than composite materials with micro- and macro- fillers. Even, the introduction of a small amount of nano sized filler into polyethylene can significantly increase the physical properties, improve the barrier properties, increase the thermal stability, electrical conductivity, etc (Antipov, et al., 2003; Yurkov et al., 2014; Kurbanova et al., 2020).

Carbon nano fillers provide ample opportunities for regulating the properties of substances without significant changes in their composition due to the manifestation of size effects that affect the electronic, thermal, mechanical, electrical and other properties of the filler and affect the properties of materials (Rakov, 2013). Most often, polymer composite materials with CNTs used as filler are of interest from the point of view of their electrically conductive properties, but no less important are their physical, mechanical and operational properties (wear, oil, petrol resistance, etc.). It is also of great interest to increase the heat and temperature resistance of products made from such nano composites, their gas and air tightness, which is important in the production of pipes, containers and other similar products (Mordkovich et al., 2009). The presented work is devoted to preparation and study of the properties of nano composites based on the high-pressure polyethylene using metal-containing nanoparticles stabilized by a polymer matrix and multi-walled carbon nanotubes used as Nano filler.

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MATERIALS AND METHODS

Experimental part

High-pressure Poly Ethylene grade 15803-020 (PE), as nano fillers (NPh) was used cobalt-containing nanoparticles stabilized by a polymer matrix of high-pressure polyethylene, obtained by a mechanochemical method (NPhCoO) (Mordkovich et al., 2009) and multi-walled carbon nanotubes MWCNT (CVD-179, purified from resins and Fe, hydrocarbon feedstock-cyclohexane). Nano composite polymer materials are obtained from mixing PE with a cobalt-containing nano filler and MWCNT on laboratory rollers at a temperature of 130-135°C for 15 minutes. For mechanical tests, the resulting mixtures were pressed in the form of 1 mm thick plates at 170°C and a pressure of 10 MPa for 10 minutes. The physicomechanical parameters of the obtained compositions were determined by using an RMI- 250 device. The thermal stability of the researched nano composite samples was studied on a Q-1500D derivatograph (MOM, Hungary). The tests were carried out in an air atmosphere in a dynamic mode when the sample was heated to 5 degrees • min^{-1} from 20 to 500°C, a sample of 100 mg, the sensitivity of the channels DTA-250 mkV, TG-100, DTG-1 mV.

RESULTS

Nano composite polymeric materials based on PE with cobalt-containing nano filler and MWCNT have been acquired. The physicomechanical, thermo physical and thermal properties of the obtained nano composites have been investigated. The data are presented in Table 1.

Table 1. Physical and mechanical properties of the synthesized nano composites

Compositions (wt %)	**The tensile strength**	**Elongation at break (ε_p), %**	**Vicat softening point, ºC**
PE	11.39	400	130
PE/NPhCoO (100/0.5)	10.94	450	134
PE/NPhCoO (100/1.0)	12.45	500	138
PE/NPhCoO (100/2.0)	10.79	440	133
PE/MWCNT (100/0.01)	9.56	215	137
PE/MWCNT(100/0.05)	10.05	220	140
PE/MWCNT (100/0.1)	9.17	210	135
PE/NPhCoO/MWCNT (100/0.5)	11.83	680	145

As can be seen from the data in Table 1, the introduction of 0.5-1.0 mass% of NPhCoO into the composition of the composition leads to an increase in the strength index from 11.39 to 12.45 MPa and the deformation value at break of the composite from 400 to 500%. An increase in the concentration of NPhCoO more than 1.0 wt% leads to a decrease in the strength of the composite (10.79 MPa) and the magnitude of deformation upon fracture of the composite (440%), which is probably due to the aggregation of nanoparticles, which leads to the formation of micro-defects in the bulk of the polymer matrix. Investigation of the Vicat heat resistance of the obtained compositions showed that the introduction of the nano filler NPhCoO (1.0 wt.%) into the PE composition leads to an increase in the heat resistance index from 130 to 138°C. Investigation of the Vicat heat resistance of the obtained compositions showed that the introduction of the nano filler NPhCoO (1.0 wt.%) into the PE composition leads to an increase in the heat resistance index from 130 to 138°C. An increase or decrease in the amount of NPhCoO leads to a decrease in the heat resistance index, which is probably related to the micro defect of the obtained composite. An increase or decrease in the amount of NPhCoO leads to a decrease in the heat resistance index, which is probably related to the micro defect of the obtained composite.

A necessary condition for obtaining the best properties of carbon nano materials in a polymer composite is to achieve the maximum degree of dispersion of the filler and its optimal orientation in the polymer matrix (Mordkovich et al., 2009). As can be seen from the data in Table 1, the introduction of 0.01-0.1 wt.% MWCNTs into the composition leads to a significant decrease in physical and mechanical parameters: σp from 11.39 to 10.05 MPa, εp from 400 to 220%, but the Wick heat resistance increases by 10°C. To overcome the low affinity of MCNTs for the polymer, it is necessary to use low molecular weight compounds or polymers with functional groups (Mordkovich et al., 2009). We have used PE as a functionalized polymer, modified with cobalt oxide nanoparticles. The combined use of both NPhCoO and MWCNTs in the PE composition increases the ultimate strength at break to 11.83 MPa, relative elongation to 680%, and Wick heat resistance to 145°C.

The thermal stability of the studied samples based on PE, containing separately or together NPs of copper oxide and MWCNTs, was estimated from the value of the activation energy (E_a) of the decomposition of thermal oxidative destruction, calculated by the method of double logarithm using the TG curve according to the method (Mordkovich et al., 2009; Kurenkova, 1990), at the temperature of 10% (T_{10}), 20% (T_{20}) and 50% (T_{50}) decay of the samples under study, as well as their half-life-τ1/2. The data, achieved as a result of derivatograph studies, are shown in Table 2.

Table 2. Thermal properties of the studied nano composite samples

Compositions (wt.%)	**T_{10} °C**	**T_{20} °C**	**T_{50} °C**	**$\tau_{1/2}$, min**	**E_a, кJ/mol**
PE	325	345	380	63.2	191.45
PE/NPhCoO (100/0.5)	335	355	390	78.3	221.48
PE/NPhCoO (100/1.0)	340	360	400	79.8	229.56
PE/NPhCoO (100/2.0)	330	350	385	76.4	219.74
PE/MWCNT (100/0.01)	355	375	405	79.1	248.17
PE/MWCNT(100/0.05)	360	380	410	81.2	256,22

It was shown, the insertion of copper oxide NPs containing NPh into the PE composition contributes an increase in the sample half-life ($T_5$0) from 380 to 400°C, the half-life ($\tau_{1/2}$) increases from 63.2 to 79.8 min., the activation energy (E_a) of the decomposition for thermal oxidative destruction of the obtained nano composites increases from 191.45 to 229.56 kJ/mol. The introduction of only MWCNTs into PE leads to an increase in T_{50} from 380 to 410°C, $\tau_{1/2}$ from 63.2 to 81.2 min., E_a from 191.45 to 256.22 kJ/mol. The combined use of both NPhCoO and MWCNTs in PE promotes an increase in T_{50} to 420°C, $\tau_{1/2}$ to 83.1, E_a to 291.38 kJ/mol.

DISCUSSION

Thermal analysis studies have shown that the joint insertion of NPs of cobalt oxide and MWCNTs into the composition of the composition improves the thermal-oxidative stability of the obtained nano composites. Numerous experimental data on the mechanical, strength, relaxation, and other properties of polymer-polymer and polymer-filler mixtures are explained within the framework of the concept of the presence of an interphase layer (Kurenkova, 1990).The properties of polymer composites are significantly influenced by the supra molecular structure of the polymer (the size of spherulites, the degree of crystallinity, the presence of C=O groups and various branches, etc.) and the interfacial interaction at the interface (Pomogajilo, 2002). The metal-containing nanoparticles and MWCNTs used in this work, located in the interphase layer of the structural elements of PE, contribute to the formation of a composition of heterogeneous nucleation centers in the melt, which, in the process of stepwise cooling of the nano composite, contribute to an increase in crystallization centers, leading in general to an improvement in the crystallization process and the formation of a relatively small-spherulite structure.

CONCLUSIONS

1. A separate and joint effects of cobalt oxide nanoparticles stabilized by the high-pressure polyethylene matrix, obtained *via* the mechanochemical method and MWCNTs on the properties of metal-carbon composites based on high-pressure polyethylene have been studied.
2. An improvement in the strength, deformation parameters, and also thermal-oxidative stability of the obtained nano composites was revealed, which is apparently associated with the synergistic effect of the interaction of cobalt-containing nanoparticles with MWCNTs.
3. The results obtained indicate that small amounts of nano fillers introduced into the polymer obviously play the role of structure-forming agents-artificial nuclei of crystallization, which contributes to the appearance in the polymer a fine-spherolite structure imparted the improved physical, mechanical and thermal properties to the obtained nano composites.

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