Application of multilayer carbon nanotubes for creation of coatings absorbing electromagnetic radiation

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Abstract. The article presents the use of carbon nanotubes as modifiers-materials for protection against electromagnetic radiation. The technique of obtaining nanomodified composites, as well as the method of characterization of carbon nanostructures is presented. Studies have shown that there is a linear dependence of the reflection coefficient on the frequency for composites modified multilayer carbon nanotubes based on Ni / 0.16 MgO catalyst. It was found that composites containing multilayer carbon nanotubes based on Ni / 0.16 MgO allow to achieve low values of the reflection coefficient and non-dependence of the reflection coefficient on the increase in frequency.

1. Introduction
The current state of scientific research in the field of radar absorbing coatings shows that an improvement in their characteristics can be achieved by creating new composite materials using nanoparticles [1]. In the past few years, much attention has been paid to functional materials with a hierarchical structure, since such an organization can allow the creation of new materials with improved operational characteristics [2].

Currently, there are hierarchical (multilayer) radar absorbing composite materials based on polymer matrices with good dielectric constant, but having a narrow range of operating frequencies. This disadvantage is due to the characteristics of the composite filler. Each filler or modifier has its own resonant frequency, at which the radio absorption will be most effective. The introduction of a large number of fillers to expand the operating range is impractical due to the increase in the cost of the resulting composite. To create a new type of composite material with reduced mass and size parameters, carbon nanostructured materials (fullerene, nanotubes, graphene, etc.) can be used as filler. This argument is partially confirmed in [1]. The development and creation of nanomodified materials is currently one of the relevant areas of scientific and applied research. Thanks to the modification, it is possible to create composite materials based on polymers that are resistant to mechanical stress [3], erosion wear [4], as well as materials with high electrical conductivity [5, 6].

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Obtaining a new type of composite material with reduced mass and size parameters can be achieved using carbon nanostructures, in particular multilayer carbon nanotubes (MWCNTs). The effect of introducing a filler modifier, which is a MWCNT, into the polymer matrix, will depend on their morphological features. At the active sites of the catalyst, nucleation and growth of MWCNTs occur. Thus, the catalyst affects the morphology and structure of MWCNTs.

The aim of the work is to obtain a nanomodified composite using MWNTs and to study the frequency dependence of the transmittance of electromagnetic radiation.

2. Methods and Materials

The Ni / MgO catalyst system obtained by thermal decomposition was chosen as a catalyst for the synthesis of MWCNTs. The catalyst preparation method consisted in dissolving the starting components and thermal decomposition of the preliminary catalyst. The dissolution temperature of the starting components should not exceed 60 °C, the thermal decomposition of the preliminary catalyst was carried out for 30 minutes at 500 °C. The resulting catalyst was used for the synthesis of MWCNTs using CVD. As a carbon-containing raw material, a propane-butane mixture was used.

To assess the morphology and structure of MWCNTs synthesized on the obtained catalysts, transmission (TEM) and scanning electron microscopy (SEM) were used (“Hitachi H-800” (Hitachi, Japan)).

Polyurethane (Silagerm's two-component polyurethane compound) acts as a polymer matrix in which a hierarchical structure is created. The technique for introducing multilayer carbon nanotubes into polyurethane involves the following steps:

1) Adding weighted in accordance with the calculation of the MWCNTs in component A; the addition was carried out in small portions with constant stirring for 5 minutes;

2) After dispersion of the MWCNTs, component B was added to component B so that the volume ratio of component A (excluding MWCNTs) and component B was ≈ 1/1; then the composition is stirred for 10 minutes;

3) Further, when the polyurethane composite hardens, it is exposed to a magnetic field from neodymium magnets with a magnetic induction of 2000 mT for 1 minute.

4) Polymerization of the nanocomposite in a vacuum cabinet at a temperature of 70 °C for 5 hours.

To modify the polyurethane, a wt. 4% concentration was used. To determine the effects of the interaction of microwave electromagnetic radiation with composite materials (transmission, reflection, absorption of electromagnetic radiation), a scalar network analyzer based on a oscillating frequency generator, a rectangular waveguide measuring channel. The measurements were carried out in four frequency bands: 8–12 GHz, 12–18 GHz, 18–26 GHz, 26–40 GHz. The studied samples of the composites were cut in the form of a parallelepiped with dimensions of 23 × 10 × 2 mm, 16 × 8 × 2 mm, 11 × 5.5 × 2 mm, 7.2 × 3.4 × 2 mm.

3. Results and Discussion

The morphology of MWCNTs synthesized on Ni / 0.16MgO is shown in Fig. 1(a). The structure of the synthesized MWCNTs is shown in Figure 1 (b).
The diameter of MWCNTs synthesized on a Ni / 0.16 MgO catalyst ~ 30 ÷ 60 nm.

Figure 2 shows optical micrographs of a modified polyurethane with 4% MWCNTs.

Figure 2. Optical micrographs of modified polyurethane with MWCNTs

From Figure 2 it follows that MWCNTs are fairly evenly distributed in the polymer matrix, which is the result of efficient distribution due to exposure to ultrasound.

The frequency dependence of the transmission of electromagnetic radiation by composite materials with MWNTs grown in the presence of a Ni / 0.16MgO catalyst is shown in Figure 3.
Figure 3. Frequency dependence of the transmittance of composite EMR materials with MWCNTs grown in the presence of a Ni / 0.16MgO catalyst.

For almost all samples of composite materials with MWCNTs grown on the Ni / 0.16MgO catalyst, similar frequency dependence and similar values of the EMR transmittance are observed — a decrease from 55% to 35% in the frequency range from 8 to 40 GHz. An exception is the sample with 4 wt.%, in which in the frequency range from 10 to 18 GHz there is a sharper decrease in the studied value in comparison with samples containing 2 and 6 wt.% MWCNTs.

The obtained results of the interaction of the nanomodified composite are related to the morphological structure of carbon nanotubes, namely their length, diameter, as well as the peculiarities of carbon nanotube interlacing in agglomerates. Changing the morphological features of carbon nanotubes can be used in the creation of layered coatings that effectively shield electromagnetic radiation.

4. Conclusion
The studies have shown that there is a linear dependence of the reflection coefficient on frequency for composites modified MWNTs Ni / 0.16MgO. For composites modified with MWNTs Ni / 0.16MgO, the reflection coefficient is practically independent of frequency and amounts to ~ 30–35% in the range from 8 to 40 GHz. The values of the transmission coefficients for the first and second systems monotonously decrease from 65% to 35% and from 55% to 35%, respectively, in the studied frequency range. Composites modified with MWNTs based on Ni / MgO 0.16 have a low value of the reflection coefficient. In this case, for these composites, there is an independence of the reflection coefficient from the increase in frequency.

Acknowledgments
This article is supported by the Program "5-100" of the Peoples’ Friendship University of Russia.

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