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Influence of Ultrasonic Treatment on Electromagnetic Characteristics of Composites Based on Multiwall Carbon Nanotubes at Wide Range of Frequencies (100 Hz – 258 GHz)

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In the present paper, we investigated microwave properties of polymer composites based on multiwall carbon nanotubes. The multiwall carbon nanotubes used in the composite were about 9.4 nm and 18.4 nm in diameter. The results show that the ultrasonic treatment modifies the dielectric properties of the composite. The dependence of the real and imaginary parts of the permittivity of sonication time is non linear. The results showed that composite material based on nanotubes with a diameter of 9.4 nm has a dispersive region of dielectric permittivity at the range about 10 GHz.

Keywords: MWCNT, Composites, Spectra of Complex Permittivity, Ultrasonic.

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1. INTRODUCTION

Polymer composite materials are used in widely industries and science. They have more suitable mechanical properties than ceramic composite.

The outstanding electrical, mechanical, and thermal properties of carbon nanotubes (CNTs) have made them among the most promising materials in a wide range of applications.

Carbon nanotubes draw particular interest for researchers due to the prospects of their application in various fields of engineering, microwave devices included [1-3]. There is a growing demand for light and radar absorbing materials in both commercial and military purposes [2].

Application of nanotubes as independent materials is difficult, they possess of volatility and low bulk density. For this reason, nanotubes are used as filler in the manufacture polymer composite, which can be used to solve problems of electromagnetic compatibility [3-5].

The purpose of this work is measuring permittivity spectra of composites based on urethane alkyd lacquer, with a concentration of multi-walled carbon nanotubes of 1 wt. % being sonicated before polymerization and measured on wide range of frequencies.

2. EXPERIMENTAL WORK

2.1 Experimental Samples

The filler of composites was multi-walled by carbon nanotubes (MWCNTs-2 and MWCNTs-3). Nanotubes were obtained by catalytic gas-phase deposition of ethylene in the presence of $FeCo/Al_2O_3$ catalyst in Institute of Catalysis SB RAS [6]. It is a lightweight, fluffy black powder containing individual nanotubes, tensions and aggregates of nanotubes, impurity metal particles which were encapsulated in nanotubes, the particles of the oxide supporter. The average diameter of nanotube is 9.4 nm, length > 15 μm, purity > 97.5 %.

The average MWCNTs-2 diameter is 9.4 nm, length > 15 μm, purity > 97.5 %. The average MWCNTs-3 diameter is 18.4 nm, length > 15 μm, purity > 90.0 %. MWCNTs-3 contain remainders of catalyst about 7.0 %.

Urethane alkyd lacquer was used for production of experimental samples. In the liquid state its viscosity is known to be small. This allows a filler to be moved easily.

For the production of mixture for experimental samples 1 wt.% of MWCNT was added to 99 wt. % of lacquer [5]. The mixture was placed in a glass beaker. The mixture was sonicated by the ultrasonic device "Alena" (Acoustic Processes & Devices lab Biysk Technological Institute, Russia). Probe was inserted into a glass beaker with mixture. The mixture was sonicated for 1, 2, 3, 4 and 5 minutes at 50 VA power. Mixtures were molded into a planar plate, which size is 70 mm × 20 mm × 2.0 mm. Process of polymerization was carried out for 48 hours at the room temperature.

From here abbreviations and notational conventions of samples will have the following meaning (Tables 1 and 2).

2.2 Measuring Equipment

Agilent's E4980A precision LCR meter was used to measure of resistance and capacitance of experimental samples at frequency range of 20 kHz-2 MHz. The bridge method capacitor is a system of two gold-plated plane parallel electrodes with a diameter of 1 cm. Sample located between this electrodes. A thin layer of petroleum jelly was used for the best ointment electrode with the sample.

The initial dielectric permittivity was calculated by formulas [7]

$$\varepsilon' = \frac{Cd}{\varepsilon_0 S}, \qquad \varepsilon'' = \frac{\varepsilon'}{2\pi f C R}, \tag{1}$$

where C is the capacitance, d is the thickness of sample, S is the plate area of the capacitor, $\varepsilon_0 = 8.85 \times 10^{-12}$ F/m, f is frequency, R is the resistance.

Electrical resistivity may be calculated by the for-

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mula shown below

$$\rho = \frac{RS}{d}. \quad (2)$$

The cavity perturbation techniques were used for the evaluation of dielectric and magnetic properties of the material in S-band of microwave frequencies. In this technique, a cavity was designed with a very small slot at the centre of the broad/short wall of rectangular waveguide in order to insert the sample material.

The device consists of vector network analyzer, Agilent Technologies E8363B, and different volume multimode rectangular cavities, that cover the frequency range 3-13 GHz [1].

The frequency dependence of the dielectric constant composite film in the terahertz frequency range were measured using the Mach-Zehnder interferometer, working backward wave oscillator in the range of 115-258 GHz [2].

Measurements were made at the room temperature of 24 ± 1 C.

2.3 Results and Discussion

The electrical resistivity, calculated by formula (2), is presented in Tables 1 and 2. They suggest that electrical resistivity decrease with frequency increasing. This indicates that there is a sample conductivity, which has greater effect at higher frequencies. Increase processing time result in the appearance of the maximum electrical resistivity of samples with MWCNTs-3 at a frequency of 100 kHz under three minute ultrasonic treatment, but samples with MWCNTs-2 don't show the same pattern. Different results electrical resistivity can be explain by the composition of MWCNTs, as was mentioned above, MWCNTs-3 contain remainders of catalyst more than MWCNTs-2.

The results of measurement of complex permittivity are presented in the Fig. 1-4.

Figures 1 and 3 illustrate the frequency dependences of the dielectric constant MWCNTs-3 and MWCNTs-2 have monotone character at wide range of frequencies. This indicates that the electronic and ionic

Table 1 – Abbreviations and notational conventions of samples with MWCNTs-3

Sample No MWCNTs-3	*T*, time treatment (min)	PV, MOm/m	
		100 kHz	1MHz
A1	0	0.21	0.12
A2	1	0.25	0.14
A3	2	2.58	0.58
A4	3	5.12	1.09
A5	4	1.71	2.62
A6	5	1.53	2.21

Table 2 – Abbreviations and notational conventions of samples with MWCNTs-2

Sample No MWCNTs-2	*T*, time treatment (min)	PV, MOm/m	
		100 kHz	100 kHz
B1	0	1.34	0.61
B2	1	0.01	0.01
B3	2	1.30	1.82
B4	3	6.50	1.20
B5	4	5.90	0.80
B6	5	9.17	1.20

polarizations in this material are exist. Increasing imaginary permittivity of samples at the frequencies 10 GHz and above indicates that there is orientation polarization. At that, frequency dependence is stronger for the MWCNTs-2 (see figure 1 and 3). At present time, we can't precisely define the value of dispersive region, because there is no technical opportunity to measure this value at frequencies from 14 GHz to 120 GHz.

Dielectric permittivity (DP) samples without of ultrasonic treatment have different values. The permittivity of sample A1 is less than the permittivity of the sample B1. It indicates about different characteristics of original filler, namely, MWCNTs with varied diameter have various conductivities.

From the results can be seen that the ultrasonic treatment change value of dielectric permittivity of composites it should be noted that this dependence is nonlinear both real and imaginary part of dielectric permittivity. Notice, that for different types of carbon nanotubes these changes are unfolding in different ways.

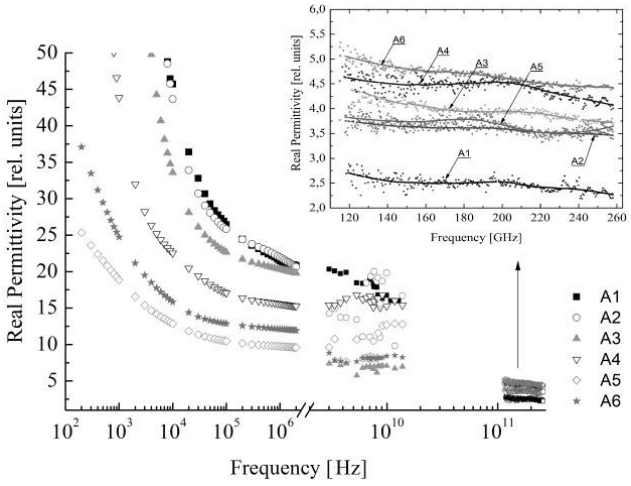

Fig. 1 – Real permittivity of composites based on MWCNT-3 for different time of ultrasonic treatment

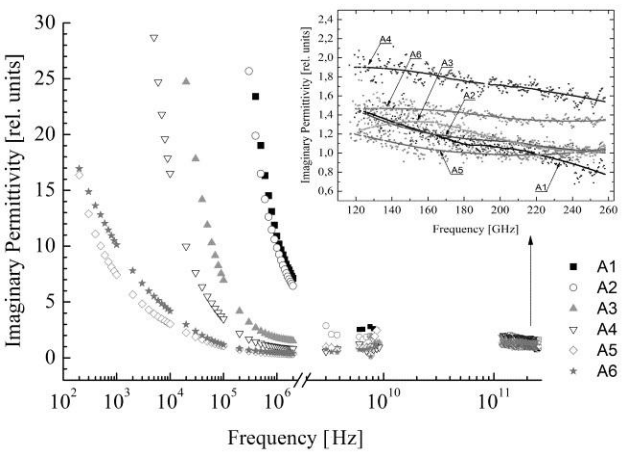

Fig. 2 – Imaginary permittivity of composites based on MWCNT-3 for different time of ultrasonic treatment

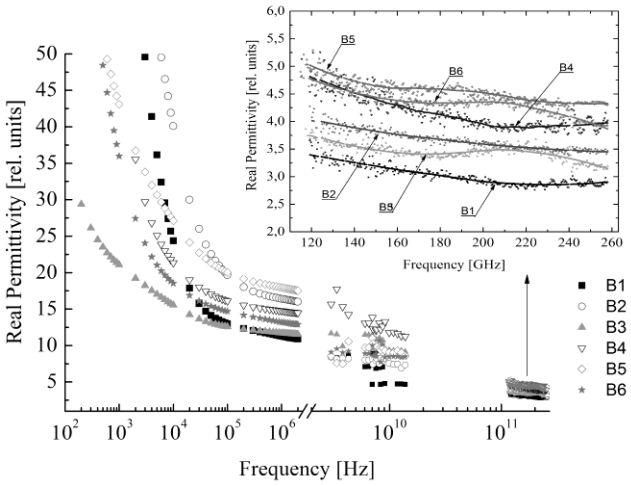

Fig. 3 – Real permittivity of composites based on MWCNT-2 for different time of ultrasonic treatment

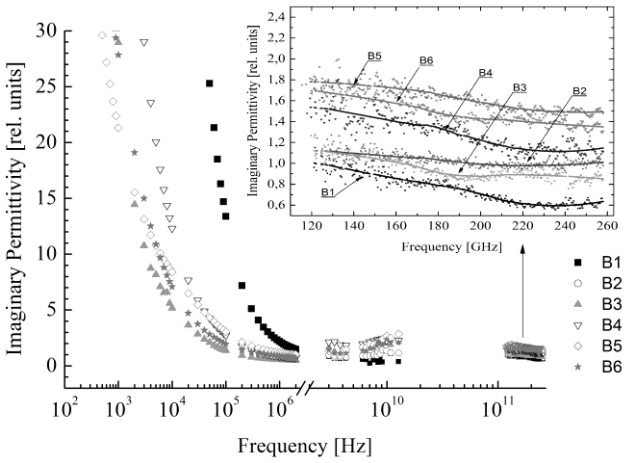

Fig. 4 – Imaginary permittivity of composites based on MWCNT-3 for different time of ultrasonic treatment

Real permittivity of samples A1-A6 changes with increasing processing time of ultrasonic treatment in a way which give the same measures of DP on 1 and 4 minute at wide range of frequencies. Processing time about 5 minute gives maximum value of real permittivity at all frequency range.

At the same time imaginary permittivity changes in another way. Samples A1-A6 have maximum values of imaginary permittivity under three minute ultrasonic treatments.

Real permittivity of samples B1-B6 with the increasing of processing time ultrasonic treatment have a nonlinear dependence – growth has been accompanied by recession, then there is value growth again.

Lake samples A1-A6, samples B1-B6 also have processing time gives maximum value of imaginary permittivity on 4 minute.

The destruction of MWCNTs agglomerates is a reason of this fact.

3. CONCLUSION

In this work, we have presented electromagnetic characteristics of polymer composites based on multiwall carbon nanotubes. It is observed that with the increase of time ultrasonic treatment, the permittivity is grows, but there is non-monotone increase. Increase processing time of ultrasonic treatment leads to increase electrical resistivity of composites with MWCNTs-2.

The growth of treatment time leads to emergence of the orientation polarization samples.

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