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Comparative study of the influence of carbon nanotubes and graphene nanoplatelets on dielectric and microwave properties of conductive rubber composites

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ABSTRACT

In this study the effect of carbon nanotubes (CNT) and graphene nanoplatelets (GNP) at various weight ratios on the dielectric (dielectric permittivity, dielectric loss angle tangent) and microwave (reflection coefficient, attenuation coefficient, shielding effectiveness) properties of conductive nanocomposites on the basis of natural rubber containing a constant amount of standard furnace carbon black has been investigated in the wide frequency range (1-12 GHz). Some additional investigations on the morphology and microstructure of the graphene particles, carbon nanotubes and furnace carbon black used have been carried out by transmission electron microscopy (TEM) and selected area electron diffraction (SAED). © 2012 Trade Science Inc. - INDIA

KEYWORDS

Rubber composites;
Carbon nanotubes;
Graphene nanoplatelets;
Dielectric and microwave
properties.

INTRODUCTION

Conductive polymer composites can be obtained by the combination of an insulating polymer matrix with electrically conductive fillers^[1-10]. Among these conductive fillers, carbon-based materials such as carbon black (CB)^[11-13], carbon nanotubes (CNTs)^[5,14-22], and graphene nanoplatelets (GNPs)^[2,3,7,23-26], have been studied extensively. In particular, significant property enhancements were reported, when carbon nanofillers

were incorporated into natural or synthetic elastomers^[27-31]. The reason for the interest in the above mentioned carbon nanostructures and their influence can be explained as follows: carbon is a versatile element. Carbon atoms are capable of bonding with other atoms in sp, sp², and sp³ hybridized structures, generating numerous stable molecules^[32]. Thus, carbon has a number of distinct molecular or crystalline forms termed allotropes or polymorphs, which include graphite, diamond, and the more recently discovered fullerenes and

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graphenes. It can also be shaped into different morphologies such as spherical, fibrous, and layered nanosized materials. Most of these carbon-based materials are electrical conductive and commercially available. These materials have different aspect ratios and specific surface areas. Figure 1 illustrates the typical carbon nanofillers with different geometries showing different surface area/volume relations^[33]. Some their specifications are shown in TABLE 1. Carbon black, composed of spherical particles with diameters from a few tens to hundreds of nanometers, has been one of the most widely used conductive fillers because of its predominant electrical property, low cost, and abundance. However, the low aspect ratio of CB nanoparticle structure leads to high percolation in the rubber nanocomposites (RNCs), typically ca. 20 wt% or higher for different rubber matrices^[34]. On the other hand, CNTs are known to have high aspect ratio (several hundred to thousand), outstanding electrical and mechanical properties with fibrous structure^[10,35,36]. Also, GNPs with layered structure have a high aspect ratio together with similar electrical and mechanical properties as those of CNTs^[37,38]. These fibrous and layered carbon nanomaterials are being widely used to reinforce polymers for improving electrical properties, thermal conductivity, and mechanical properties. It is obvious, that as CB, CNTs, and GNPs have different geometries, aspect ratios, and physical properties, their effects on the polymer nanocomposites properties should be different. Up to date, there has few reports to compare and analyze the properties of the polymer nanocomposites derived from their different structures and properties^[37,39]. There is also data about the influence of carbon black, especially carbon black with high electrical conductivity on electrical, dielectric and microwave properties of elastomeric composites in the frequency range from 1 to 12 GHz in the literature^[40-41]. Our previous studies^[42,43] were focused on the individual influence of CNT and GNP on these properties in that frequency range.

The price of CNTs and GNPs is still significantly higher than standard furnace carbon black. In this context, the aim of this study is to determine whether the addition of small quantities (1-5 phr) CNT and GNP to one standard significantly greater amount of active furnace carbon black (50 phr) can be used as a way to

modify and control, primarily to improve the dielectric (dielectric permittivity, dielectric loss) and microwave properties (coefficient of reflection, coefficient of attenuation, electromagnetic interference shielding effectiveness) of composites based on natural rubber in the high frequency range 1-12 GHz. Data for such a comparative study of those combinations of fillers that we mentioned was not found in the literature. A comparative study of the reinforcing effect of CNTs and GNPs in polyamide 12 based composites is published by S. Chatterjee et al^[44].

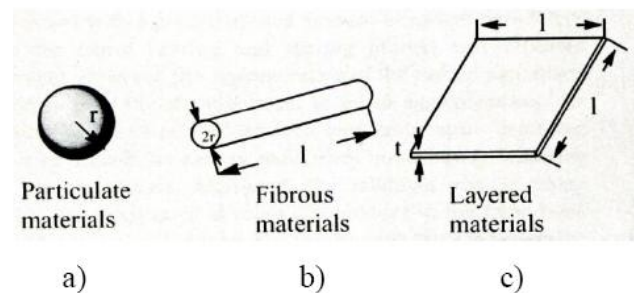


Figure 1 : Carbon nanofillers with different geometric characteristics: a) carbon black; b) carbon nanotubes; c) graphene nanoplatelets

TABLE 1 : Characteristics of different carbon nanofillers^[33]

Material	Density, g/cm ³	Specific area, m ² /g	Length, μm	Diameter or thickness, nm	Aspect ratio
CB (furnace)	1,8	100	-	20	1
CNTs	2,1	250	>1	6-20	120-200
GNPs	2,2	150	<50	<100	500

EXPERIMENTAL

Characterization of the carbon nanofillers used

Multiwalled carbon nanotubes as produced by Hayzen Engineering Co., Ankara, Turkey were used in our investigation. The material's purity is more than 95%, density – 150 kg/m³. Carbon nanotubes are with average diameter about 15 nm and length 1-10 microns (Figure 2). The pattern taken in an electron diffraction regime (in the right angle) shows that they have a polycrystal structure typical for CNT.

Graphene as produced by Hayzen Engineering Co., Ankara, Turkey was used in our investigation (Figure

3). Graphene nanoplatelets (GNP) have a “platelet” morphology, meaning they have a very thin but wide aspect (“sheet”-like structure). Aspect ratios for this material can range into the thousands. Each particle consists of several sheets of graphene with an overall thickness of 50 nm and average plate diameter 40 microns. The pattern taken in SAED regime shows a considerable number of spot-like reflections typical for single-crystal structures (Figure 3 - in the right angle).

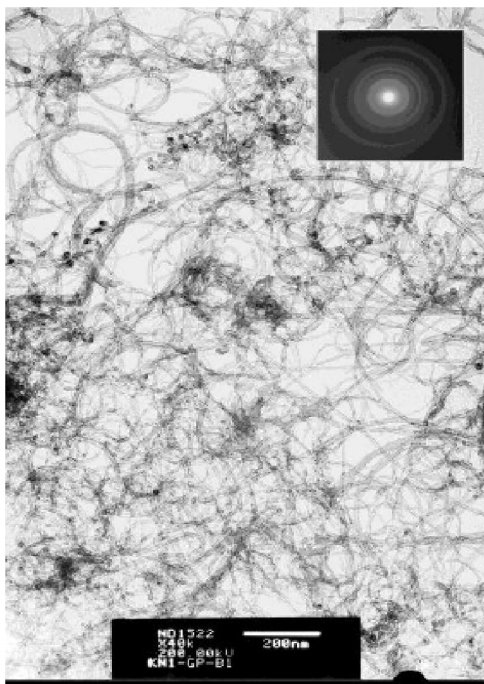


Figure 2 : TEM micrographs of CNT in transmission regime and selected area electron diffraction regime (in the right angle)

The specific structure features of the carbon black used (Corax N 220, produced by Evonik) are shown in Figure 4.

It may be seen that the primary particle size of the carbon black used is about 20 nm, but their capability to form secondary aggregates and agglomerates is considerable. SAED indicates for absence of crystal structures.

Natural rubber (SMR 10) was used as a rubber matrix in our investigations. Other ingredients such as zinc oxide (ZnO), stearic acid (SA), N-(1,3-dimethylbutyl)-N'-phenyl-p-phenylenediamine (Vulkanox 4020, produced by Lanxess), dibenzothiazyl disulphide, MBTS (Vulkacit DM, produced by

Lanxess), and sulphur (S) were commercial grades and used without further purification.



Figure 3 : TEM micrographs of GNP in transmission regime and selected area electron diffraction regime (in the right angle)

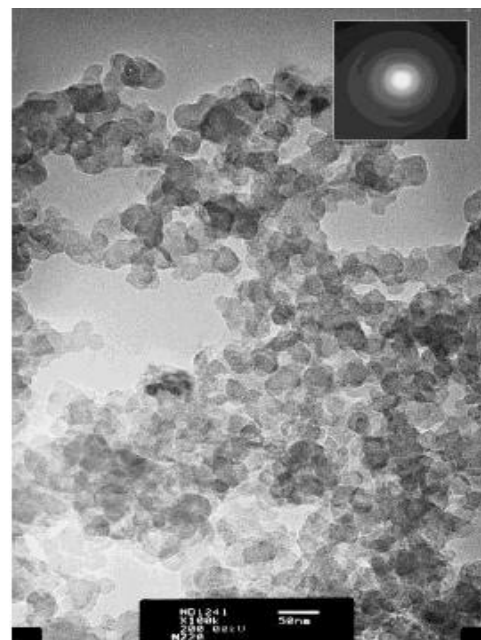


Figure 4 : TEM micrographs of furnace carbon black Corax N220 particles together with SAED (in the right angle)

Preparation and vulcanization of rubber compounds

TABLE 2 summarizes the formulation characteristics of the rubber compounds (in phr) used for the investigations.

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TABLE 2 : Composition of the rubber compounds studied

	NR 1	NR 2	NR 3	NR 4
Natural Rubber	100	100	100	100
Foaming agent	8	8	8	8
Stearic acid	1	1	1	1
ZnO	4	4	4	4
Processing oil	10	10	10	10
Carbon black N220	50	50	50	50
CNT/ GNP	0	1	3	5
MBTS	2	2	2	2
TMTD	1	1	1	1
IPPD 4020	1	1	1	1
Sulphur	2	2	2	2

The rubber compounds were prepared on an open two-roll laboratory mill (L/D 320×360 and friction 1.27) by incorporating pre-characterized CB, CNTs and GNPs into a natural rubber matrix at various loadings (TABLE 2). The speed of the slow roll was 25 min⁻¹. The experiments were repeated for verifying the statistical significance. The ready compounds in the form of sheets stayed 24 hours prior to their vulcanization.

The optimal vulcanization time was determined by the vulcanization isotherms, taken on an oscillating disc vulcameter MDR 2000 (Alpha Technologies) at 150°C according to ISO 3417:2002.

These composites were evaluated for their dielectric (dielectric permittivity, dielectric loss angle tangent) and microwave (reflection coefficient, attenuation coefficient, shielding effectiveness) properties in the 1-12 GHz frequency range.

MEASUREMENTS

Microwave properties

Reflection and attenuation

Measurements of reflection and attenuation were carried out using the measurement of output (adopted) power P_a in the output of a measuring line without losses, where samples of materials may be included. Because of the wide frequency measurement a coaxial line was used. Samples of the materials were shaped into discs with an external diameter $D=20.6\text{mm}$, equal to the outer diameter of the coaxial line and thickness of $\Delta \approx 2\text{mm}$. The internal diameter depended on the

relative dielectric permittivity of the material.

The sample reflected a part of the incident electromagnetic wave with power P_{in} . The rest of the wave with power P_p penetrated the material, so that the attenuation L depended on the coefficient of reflection $|\Gamma|$. Its module was determined by a reflect meter.

Thus the attenuation was determined by

$$L = 10 \log \frac{P_a}{P_p}, \text{dB} \quad (1)$$

where

$$P_p = P_{in} \cdot (1 - |\Gamma|^2) \quad (2)$$

The following scheme presents the equipment used for testing both parameters (Figure 5)

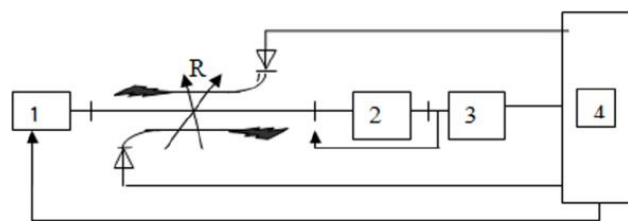


Figure 5 : Scheme of the equipment for measuring the microwave properties

- 1 - a set of generators for the whole range: HP686A and G4 - 79 to 82;
 - 2 - Coaxial section of the deck E2M Orion, with samples of material;
 - 3 - Power meter HP432A;
 - 4 - Scalar reflectance meter HP416A;
- R - Reflect meter, including:
- two directional couplers Narda 4222.16
 - two crystal detectors Narda 4503-N.

Shielding effectiveness (S. E.)

This parameter is defined as the sum of the reflection losses R , dB and attenuation L , dB in the material. It can be directly measured or calculated from the measured reflectance and attenuation in the material. In the first case, as measured: incident power on the sample P_{in} and adopted after the sample P_a (Figure 1), S.E. is determined by

$$\text{S.E.} = 10 \log \frac{P_0}{P_a}, \text{dB} \quad (3)$$

In the second, if known reflection and absorption in the material, S.E. is determined, by a definition, as

$$S.E. = R, dB + L, dB \tag{4}$$

where R, dB is the attenuation due to the reflection of power at the interfaces.

In the present work the shielding effectiveness was determined by equation (4).

Dielectric properties

Complex permittivity

The determination of complex permittivity was carried out by the resonance method, based on the cavity perturbation technique.

Having measured the resonance frequency of the empty cavity resonator f_r , a measurement of the shift in resonance frequency f_ϵ was carried out in the presence of the sample material. Then the dielectric constant ϵ_r was calculated from the shift in resonance frequency, cavity and the sample cross sections S_r and S_ϵ , respectively

$$\epsilon_r = 1 + \frac{S_r}{2S_\epsilon} \cdot \frac{f_r - f_\epsilon}{f_r} \tag{5}$$

The sample was in the form of a disc with a diameter of 10 mm and about 2 mm thickness. It was placed at the maximum electric field location of the cavity. Because the thickness of the sample was not equal to the height of the resonator, a dielectric occurred with an equivalent permittivity ϵ_e at the place of its inclusion. The parameter was determined by (5) and instead be saved. Then was determined by

$$\epsilon_r \approx \epsilon_e (k + 1) - k, (\Delta \ll 1) \tag{6}$$

where $k = l/\Delta$ and l is the distance from the disc to the top of the resonator.

Loss factor $\tan \delta$

The loss factor $\tan \delta$ was calculated from the quality factor of the cavity with Q_e and without sample Q_r

$$\tan \delta = \frac{1}{4\epsilon_r} \frac{S_r}{S_\epsilon} \left(\frac{1}{Q_e} - \frac{1}{Q_r} \right) \tag{7}$$

The measurement setup used several cavity resonators for the whole range, generators for the whole range, frequency meter and oscilloscope.

The following scheme presents the set used for measuring the dielectric properties (Figure 6):

1 - Generators for the whole range: HP686A and G4 -

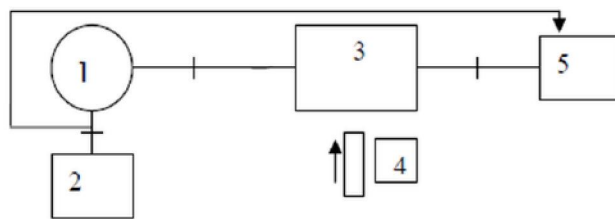


Figure 6 : Scheme of the equipment for measuring the dielectric properties

- 79 to 82;
- 2 - Frequency meters: H 532A; FS – 54;
- 3 - Cavity resonator;
- 4 - Sample;
- 5 - Oscilloscope EO 213.

RESULTS AND DISCUSSION

Dielectric properties

Complex relative dielectric permittivity. Real part

The dependences of the real part of complex dielectric permittivity (the so-called relative dielectric permittivity ϵ_r) on the frequency and amount of the second filler are shown in Figures 7-8. The figure shows that there are common trends in these dependences, namely:

- with the increase of frequency and amount of the

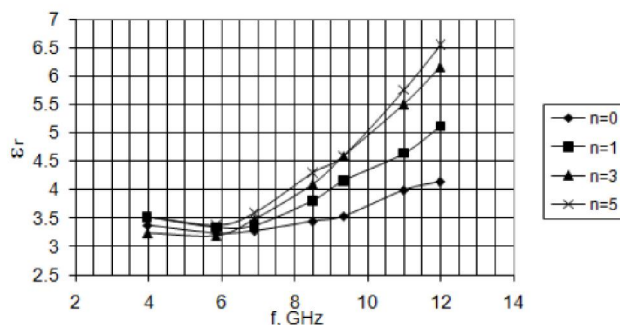


Figure 7 : Frequency dependence of relative dielectric permittivity ϵ_r at constant first filler amount (CB-50 phr) and various second filler amount (n-phr of carbon nanotubes)

second filler (CNT or GNP) at constant amount of carbon black (50 phr), the values of relative dielectric permittivity increase.

- variation is hardly observed in the range 1-7 GHz in both types of composites depending on frequency whereas in the range 7-12 GHz there is a variation, with the composites containing CNT the influence the frequency and amount of the second

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filler is significantly more pronounced compared to composites containing GNP, whose dependencies remain grouped. It is noteworthy, that the overall values of the composites containing CNT (between 3.2 and 6.5) are much higher than those of composites containing GNP (between 3.2 and 3.8).

The polarization mechanism operating in the giga-

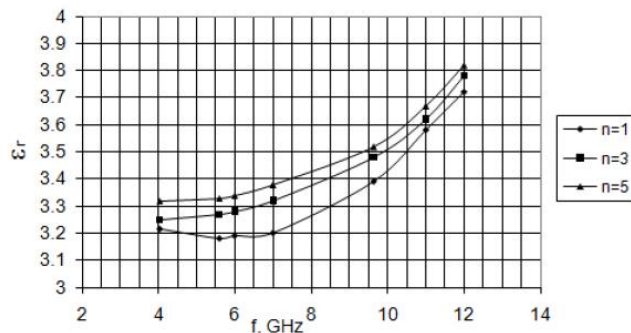


Figure 8 : Frequency dependence of relative dielectric permittivity ϵ_r at constant first filler amount (CB-50 phr) and various second filler amount (n-phr of graphene nanoplatelets)

hertz frequency is purely electronic or orientational with relaxation times smaller than the time period of the applied signals. Interfacial polarization, which is the basic reason for the dispersion in dielectric permittivity at radio frequency (RF) regime, has no role to play in microwave frequencies as it does not produce dispersion in ϵ_r because of its much smaller relaxation time. But ϵ_r was found to increase with the increase of phr of carbon nanotubes and graphene nanoplatelets in the composite as it is evident from Figures 7 and 8. This phenomenon of increase in dielectric permittivity with the increase in second filler concentration can be attributed to the enhancement of electrical conductivity of the composites, but the differences can be attributed first of all to the differences in the resistivity/conductivity and the structure features of CNT and GNP.

Complex relative dielectric permittivity. Imaginary part (Dielectric loss)

The dependencies of the imaginary part of the complex dielectric permittivity (dielectric losses) on the amount of the second filler and the frequency at constant amount of the first filler are shown in Figures 9-10. It can be seen from the figures that with the increase of frequency, the dielectric losses decrease, but by increasing the amount of the second filler the losses

increase.

Overall, the values of the composites containing

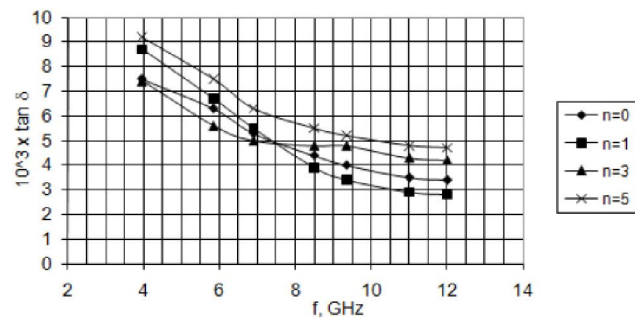


Figure 9 : Frequency dependence of dielectric loss at constant first filler amount (50 phr) and various second filler amount (n-phr of carbon nanotubes)

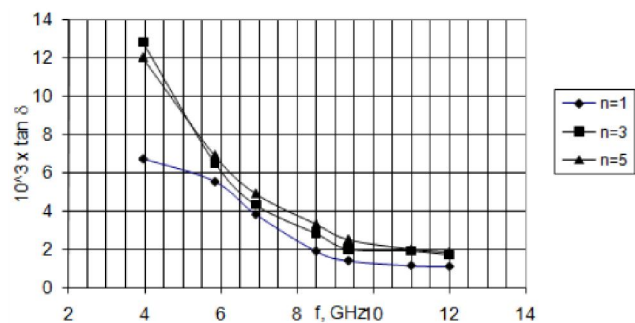


Figure 10 : Frequency dependence of dielectric loss at constant first filler amount (50 phr) and various second filler amount (n-phr of graphene nanoplatelets)

CNT are lower ($3 \cdot 10^{-3}$ - $9 \cdot 10^{-3}$) than those of composites containing GNP ($1.7 \cdot 10^{-3}$ - $13 \cdot 10^{-3}$). There are, however, some differences in the nature of variation of these dependences – with composites containing CNT the decrease of dielectric losses is smooth throughout the entire frequency range studied and the influence of the amount of filler is not so pronounced.

Microwave properties

Coefficient of reflection

The dependencies of the coefficients of reflection of electromagnetic waves on the frequency and amount of the second filler are shown in Figures 11-12.

It can be seen from the graphs that with the increase of frequency and amount of the second filler, the reflection coefficients also increase. The nature of this increase, however, is different. With composites containing CNTs, the influence of the amount of the second filler is stronger whereas the influence of frequency - less strong compared to composites containing GNP, where the effects are exactly the opposite. Thus, at lower

frequencies, composites with carbon nanotubes have higher coefficients of reflection, but at higher frequencies - lower than the composites containing equal amounts of GNP at constant amount of the second filler. The highest values of the coefficients of reflection for composites containing CNT are about 0.7, while those of composites containing GNP - about 0.8.

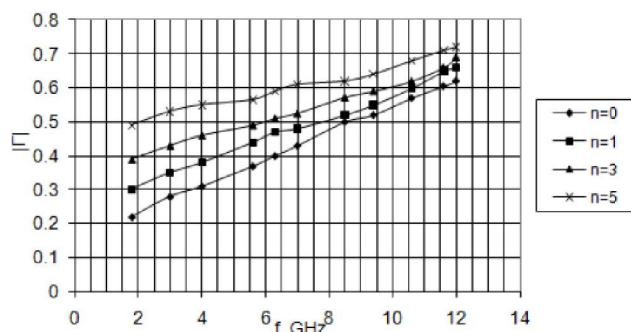


Figure 11 : Frequency dependence of coefficients of reflection at constant first filler amount (50 phr) and various second filler amount (n-phr of carbon nanotubes)

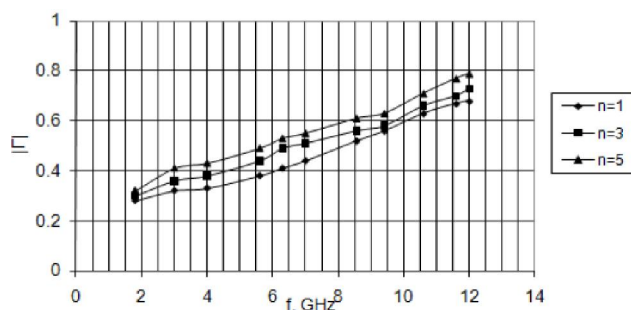


Figure 12 : Frequency dependence of coefficients of reflection at constant first filler amount (50 phr) and various second filler amount (n-phr of graphene nanoplatelets)

Attenuation coefficient

The dependencies of the coefficients of attenuation on the amount of the second filler and frequency are shown in Figures 13-14.

It is obvious that with the increase of the amount of the second filler and frequency the coefficients of attenuation also increase. Composites containing CNTs, have significantly higher attenuation than composites containing GNP under the same other conditions. This is particularly obvious when the amount of the second filler is higher (e.g. at 5 phr, the attenuation coefficient of composites containing GNP is within 22-39 dB/cm, depending on the frequency, and for composites containing CNTs –it is in the range of 42-54

dB/cm). With the last type of composites all the effects are more pronounced.

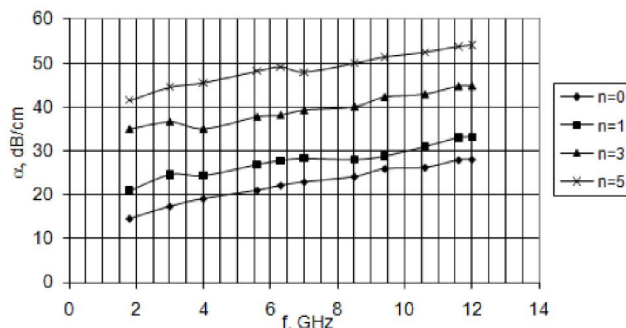


Figure 13 : Frequency dependence of coefficients of attenuation at constant first filler amount (50 phr) and various second filler amount (n-phr of carbon nanotubes)

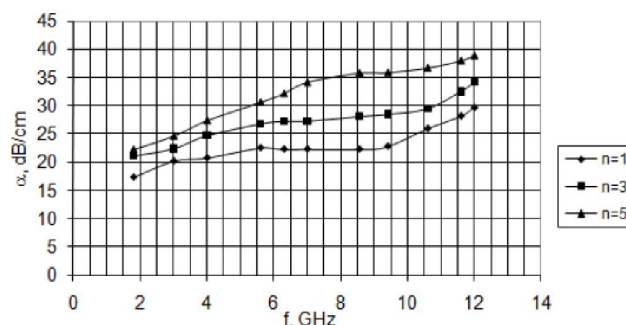


Figure 14 : Frequency dependence of coefficients of attenuation at constant first filler amount (50 phr) and various second filler amount (n-phr of graphene nanoplatelets)

Shielding effectiveness

The dependencies of the shielding effectiveness on the amount of the second filler and the frequency are shown in Figures 15-16. The decrease in the values of this characteristic depending on frequency is quite obvious since the growth of the coefficients of attenuation is apparently not high enough to make up for the increase in reflection coefficients. These two coefficients according to equation (4) are crucial to the formation of the value of the shielding effectiveness.

In most cases, with composites containing nanotubes, this parameter has better values than with composites containing graphene structures, mainly due to the lower reflection and higher attenuation of the first, especially at higher frequencies. The composite containing 5 phr CNTs is characterized with the best values of this parameter which remain within the 11-12 dB for the entire frequency range studied.

In relation to the investigation of the influence of the

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second filler (CNTs or GNP) on the dielectric and microwave properties of composites containing 50 phr standard furnace carbon black, the following further explanation of the above experimental results can be added: the observed differences are mostly due to differences in the geometry and microstructure of CNT and GNP, as well as to differences in their electrical conductivity, which leads to different percolation threshold of the studied composites. It was found out that the percolation threshold for composites filled with the investigated fillers, but individually and not in combination, is at 3, 12 and 15 phr respectively for CNTs, GNPs and CB^[33]. It is possible because CNTs have fiber-like geometry giving lower percolation threshold than GNPs, having sheet-like geometry. According to Balberg^[45] the average interparticle distance can be different in the carbon nanofillers with different aspect ratios. The more spherical-like structure, the larger the interparticle distance will be formed. For the elongated particles, their composites have a very narrow distribution of the distances due to the entangled particles structures^[45] and the volume resistivity of the composites decreases monotonically with increasing the aspect ratio. The interparticle distance, composites microstructure and the volume resistivity have a strong influence on the microwave properties^[46]. As from TABLE 1, the CNTs and GNPs have much higher aspect ratio than that of the CB nanoparticles. The differences in the step-like increase of the dielectric permittivity of the composites, containing CNTs and GNPs toward high frequencies is induced by the differences in their dielectric relaxation which suggests that the established percolation network structure is not stable and easily affected by the external frequency disturbances. The effects of composites containing CNTs are more pronounced than those containing GNPs, respectively, the improvement of the properties of the first is better pronounced. S. Chatterjee et al^[44] came to a similar conclusion comparing the influence of CNTs and GNPs on the mechanical and electrical properties of composites based on polyamide 12. The authors ascribe the observed effects to the better dispersion and distribution of CNTs in the polymer matrix. However, our investigation using TEM did not ascertain such differences (probably due to the significant amount of carbon black in the composites), which makes us believe that the effects we observed are due

to differences in geometry, microstructure and the properties of CNTs and GNPs and the related differences in the structure and characteristics of the composites themselves.

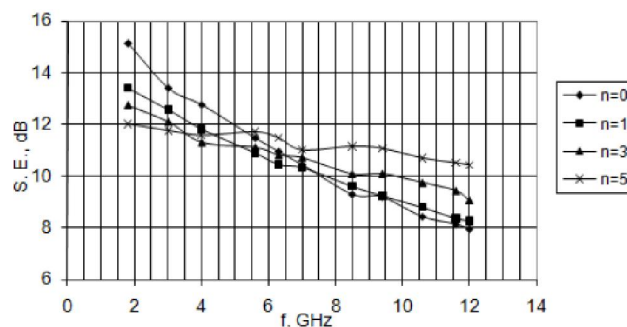


Figure 15 : Frequency dependence of electromagnetic interference shielding effectiveness at constant first filler amount (50 phr) and various second filler amount (n-phr of carbon nanotubes)

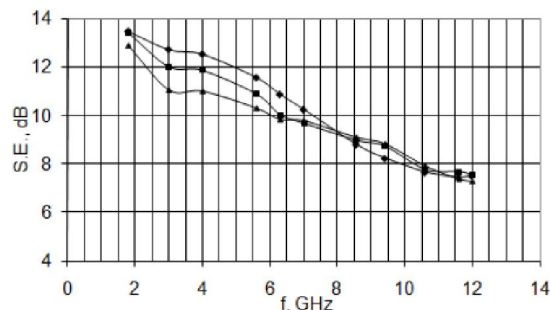


Figure 16 : Frequency dependence of electromagnetic interference shielding effectiveness at constant first filler amount (50 phr) and various second filler amount (n-phr of graphene nanoplatelets)

CONCLUSIONS

1. The influence of CNTs and GNPs on the dielectric and microwave properties of the composites on the basis of natural rubber containing a constant amount of standard furnace carbon black has been studied.
2. It has been found out that adding CNTs and GNPs in small quantities to the furnace carbon black has the effect of modifying the properties of the composites and in most cases improvement is observed.
3. It has been found out that CNTs and GNPs influence quantitatively the investigated properties in different ways and the effects on CNTs are more pronounced.
4. The observed differences are due mainly to differ-

ences in geometry and microstructure of CNTs and GNPs (fiber-like and sheet-like, respectively), as well as to the differences in their electric properties.

5. Adding small amounts (1-5 phr) of CNTs and GNPs to a standard constant amount of active furnace carbon black can be used as a way to modify and control, primarily to improve the dielectric (dielectric permittivity, dielectric loss) and microwave properties (coefficient of reflection, coefficient of attenuation, electromagnetic interference shielding effectiveness) of composites based on natural rubber in the high frequency range 1-12 GHz.

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