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Carbon black/silicone rubber blends as absorbing materials to reduce Electro Magnetic Interferences (EMI)

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Summary

In this work the microwave absorbing properties of carbon black/silicone rubber blends were investigated in the frequency range 0.2÷6.0 GHz, varying the carbon black contents. In particular, the permittivity of the samples was measured by a network analyzer and then the experimental data were fitted as function of microwave frequency and carbon black content, $\varepsilon^* = \varepsilon^*(f, C)$.
The reflection loss for a layer of blend attached on a metal plate, which is related to the shielding effectiveness, was simulated as function of frequencies and of layer thickness for some values of carbon black content, and the simulation results were summarized in contour plots. These plots can be used to select thickness and carbon black content necessary to achieve the desired shielding effectiveness for a given frequency.

1. Introduction

The use of electronic devices is experiencing an exponential growth in all the fields of human life, and most of them (personal computers, communication, medical and analytic devices, a lot of domestic appliances) work in the microwave frequency range. This growth gives rise to an increase of Electro Magnetic Interference (EMI) so that it is mandatory to develop systems to protect electronic devices from external interferences.
To this aim electromagnetic shielding are used to confine electromagnetic energy within the bounds of a specific region and/or to prevent the propagation of such energy into a designated locale. Depending on the interference due to any of forms of electromagnetic energy (radiated, static or time-varying electromagnetic force field), a variety of materials have been developed and used in the fabrication of shields. In particular, shields structures could be realized by using conductive and/or absorbing materials such as metallic conductor, polymeric composites with metallic and non metallic conducting inclusions, etc. These materials must have specific requisites: first of all shielding effectiveness, lightness, good mechanical properties, good processability, low cost [1].

Rubbers are among the best materials candidate to realize absorbing layers, because they're light, easy-to-form and they usually have interesting mechanical properties, but they're not e.m. wave absorbing media. The desired absorbing properties could be induced by adding conductive fillers (carbon black, graphite, ferrite, ionic salts) in a rubber matrix.

Estimation of conductive filled-rubbers absorbing capability requires a deep knowledge of their dielectric behavior, in term of relative permittivity (ε^*) and permeability (μ^*). To our knowledge, it is not a widely investigated field and some studies are briefly summarized in the following. Annadurai *et al.* [2] examined systems made of ferrite- and carbon black-filled EPDM (Ethylene-Propylene-Diene-Monomer) rubber; Kim *et al.* [3] studied the ferrite-silicone rubber composite; Kwon *et al.* [4] considered the carbon black-silicone rubber blends. All of them found interesting potentiality in adopting the investigated materials as shields for protection from EMI. The properties of conductive filled-rubbers and polymers, useful to realize shielding materials, have been studied also by Paul and Thomas [5], who analyzed carbon black- and glass fiber-LDPE (Low Density Poly-Ethylene); by Achour *et al.* [6], who tested the applicability of mixture laws to permittivity of carbon black-epoxy resin composites; by Moon *et al.* [7], who considered the epoxy-dielectrics-carbon black composites as phantom materials to simulate human tissues, with aim to provide model materials for analysis of electromagnetic wave interaction with human body; and by Choi *et al.* [8], who analyzed the $SrTiO_3$-epoxy systems, and found that the microwave attenuation maxima shift toward lower frequencies increasing filler content and layer thickness.

Aims of this work are to investigate the microwave behavior of carbon black-silicone rubber blends, and to select the design parameters (the carbon black content and the thickness) which give rise to the best shield effectiveness (the shield which ensures the maximum attenuation).

2. Experimental

Blends were prepared mixing a silicone rubber (a commercial one, pure silicone with acetic acid as curing agent) with carbon black. In particular, the two blend components were weighted, well homogenized and placed in sample holders. The blends were cured at room temperature and humidity for about 100 hours.

The blend samples dielectric spectroscopy was performed using a network analyzer (HP8753ES, *Agilent Technologies*), equipped with a coaxial probe system (HP85070B, *Agilent Technologies*). The instrument was calibrated using distilled water at room temperature, open and short circuit loads. The measurements were performed by pressing the open end of the coaxial probe against the samples. Dielectric constant and loss factor were then measured in the frequency range 0.2÷6.0 GHz.

3. Results and discussions

3.1 Filled rubbers dielectric behaviour

Interactions between materials and electromagnetic fields can be expressed by materials permittivity and permeability. These properties are reported in terms of relative complex numbers: $\varepsilon^* = \varepsilon' - j\varepsilon''$ and $\mu^* = \mu' - j\mu''$. The real part of the permittivity, ε', named *dielectric constant*, measures how much energy from an external electric field is stored in the material. The imaginary part, ε'', named *loss factor*, accounts for the loss energy dissipative mechanisms in the materials. For the carbon black/silicone rubber blends a reasonable assumption for $\mu^* = 1 - j0$ ($\mu' = 1$ and $\mu'' = 0$) was made [4], while the real and the imaginary part of the permittivity were measured.

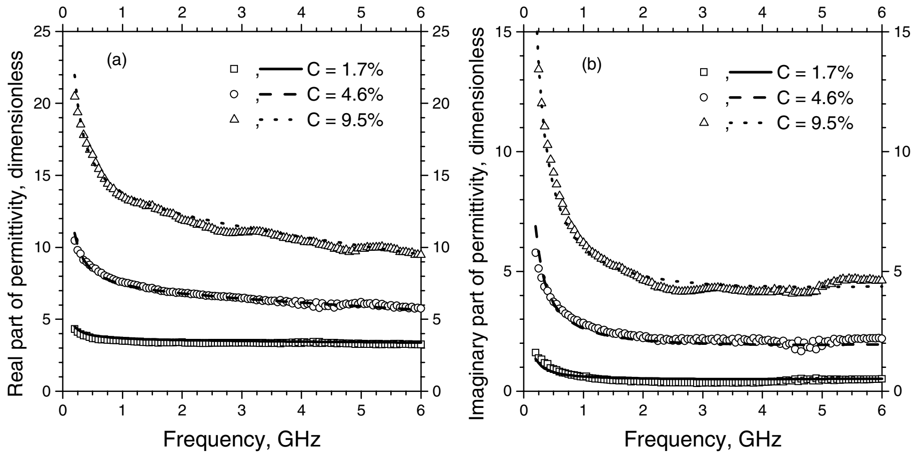

Figure 1. Complex permittivity vs. frequency, for samples of several carbon black contents: (a) Dielectric constant **ε'**, (b) dielectric loss **ε''**.

Figure 1 reports ε' and ε'' for some samples investigated in this work, as function of frequency (open symbols). Data curves are reported with the parameter C = carbon black content (w/w %), chosen below 10% to avoid conductive behavior due to percolation phenomena [4].

The real part and the imaginary part of permittivity show similar behavior: they slightly decrease with frequencies, and increase with carbon black content, both of the effects being larger at lower frequencies.
The dependence of loss factor upon frequency is usually described by a sum of two terms: one, $\sigma_0/2\pi f$, accounting for conductive effects and one, ε''_R, accounting for relaxation effects [6]:

$$\varepsilon''(f) = \frac{\sigma_0}{2\pi f} + \varepsilon''_R(f) \qquad (\mathbf{1})$$

In the carbon black content range investigated, the conductivity term plays a secondary role, since the silicone rubber is not conductive and the carbon black itself, even if it is conductive and not dielectric, when encapsulated (dispersed at low concentration) in a dielectric matrix does not show a conductive behavior [7]. The main term affecting the loss factor is therefore the relaxation one. The loss factor in blends of a conductive filler dispersed in a dielectric matrix is due to a phenomenon known as Maxwell-Wagner polarization (the polarization that occurs at the interfaces between the silicon rubber -dielectric- and the carbon black -conductive filler-). When the material is subjected to an electric field, dipoles can be induced at the interfaces, and the loss factor frequency dependence can be explained by the relaxation of that dipoles.

However, there is not a simple way to model the relaxation effects, i.e. the term ε”$_R$(*f*). Furthermore, as already noted above, the dielectric constant, ε’, dependence upon frequency is similar to the loss factor one, ε”. Following this reasoning, and on the basis of the mathematical structure of equation 1, we decided to describe the frequency effects on both the complex permittivity parts by the following fitting equation:

$$\varepsilon(f) = a + bf + cf^{-1} \qquad (\mathbf{2})$$

where ε(*f*) can be both ε’(*f*) and ε”(*f*), and {*a*, *b*, *c*} are fitting parameters. This correspond to model the relaxation effects by assuming a linear dependence upon frequency: ε”$_R$ = *a* + *bf*. It is not supported by any physical reasoning, but it is simple and, at will be seen in the following, it works well with our data.
A preliminary fitting session, in which each data curve has been described by equation 2, gave rise to a set of parameters {*a*, *b*, *c*} for each curve. These parameters have been plotted versus the carbon black content, and we found that they vary linearly with carbon black content (this result is not reported here). This suggested us to adopt a linear function of carbon black content for each parameter. Thus, equation 2 was modified accordingly:

$$\varepsilon(f, C) = (a_1C + a_2) + (b_1C + b_2)f + (c_1C + c_2)f^{-1} \qquad (\mathbf{3})$$

Equation 3 was adopted to describe the dependence of both ε’ and ε” from frequency, *f*, and carbon black content, *C*. Even if equation 3 is a semi-empirical one, being the dependence of ε from carbon black content linear, at a given frequency, it is in agreement with the mixing rule due to Lichtenecker and Rother, reported by [6], when their model parameter is unity. The final fitting session, which was performed applying the non-linear method of Marquart-Levemberg to all the set of experimental data (see Figure 1), gave rise to six parameters for dielectric constant and six for loss factor. The parameter values are summarized in Table 1, and the model predictions are reported in Figure 1 as curves. The agreement between data and the semi-empirical model is good, even if the model is not able to describe some secondary effects as the scattering observed around the frequency of 4.5 GHz for all the samples. It is worth noticing that the proposed model has a theoretical basis, but it was strongly modified and its fitting nature causes it to be predictive only if applied in the same range of frequency and carbon black content investigated.

Table 1. Parameters value to describe ε’(*f*, *C*) and ε”(*f*, *C*) by equation 3

Parameters value to describe ε’(*f*, *C*).					
a_1	0.2233	b_1	1.1366	c_1	-0.6460
a_2	-0.1837	b_2	1.5835	c_2	0.8183
Parameters value to describe ε”(*f*, *C*).					
a_1	0.3063	b_1	0.3722	C_1	0.1179
a_2	-0.3328	b_2	-0.2199	C_2	-0.0723

3.2 Shield design criteria

An EMI shield can be can be made up by an absorbing layer attached on a metallic plate. The impedance of this layer, normalized by the impedance of the free space, can be calculated as [3]:

$$Z = \sqrt{\frac{\mu^*}{\varepsilon^*}} \tanh\left(j \frac{2\pi}{c} \sqrt{\mu^* \varepsilon^*} fd \right) \quad \textbf{(4)}$$

where all the symbols are already known, except the speed of light, *c*, and the layer thickness, *d*. The shielding effectiveness is usually expressed by the attenuation experienced by the wave interacting with the shield itself. In turn, they are usually related by the reflection loss, that is a measure of the amount of e.m. wave reflected, and it can be expressed in dB by [3]:

$$RL = -20 \log\left(\left| \frac{Z-1}{Z+1} \right| \right) \quad \textbf{(5)}$$

Thus, the lower the reflection loss, the better the shielding effectiveness, since a lesser amount of incident wave is reflected. Therefore, the shield design specification is the minimum value of *RL* attainable, in the frequency range of interest. Once the designer has decided to work with carbon black/silicone rubber, the design variables are the layer thickness, *d*, and the carbon black content, *C*.

Figure 2 reports contour plots of the reflection loss as function of frequency (in the range investigated, 0.2÷6.0 GHz) and of layer thickness (from 0 to 20 mm), simulated for four different carbon black content (2.5, 5.0, 7.5 and 10.0% w/w). Calculations were made by equations 4 and 5, using the complex permittivity model proposed above (equation 3 with parameters from Table 1), which was tuned on the basis of the experimental measures performed in this work.

The simulation results compare well with the results of Kwon *et al.* [4] and of Choi *et al.* [8] who found a shift in the frequency where the reflection loss is minimum increasing the layer thickness. Furthermore, from inspection of Figure 2 it is evident that, for a given frequency the increase in layer thickness does not guarantee a better shield behavior (a low level of reflection loss).

The absolute value of reflection loss increases with carbon black content, but the lowest values of *RL* were found for carbon black content equal to 7.5% (the dark spots located, for example, around f = 5GHz and d = 5 mm, C = 7.5%, where $RL = -30$). However, the windows of operating conditions useful to obtain such a low reflection loss are very narrow.

Results of our investigation can be summarized as the following guidelines, useful for the design of a e.m. wave shield made of carbon black/silicone rubber:

1. carbon black content less than 5.0% are not useful,
2. carbon black content very high are unnecessary,
3. if the shield has to protect from EMI at a single frequency, the optimal thickness can be read from Figure 2: lower frequencies will require higher thickness,

If the shield has to work in a range of frequencies, the thickness that guarantee the optimum protection (the minimum in *RL*) can be selected from Figure 2, taking an appropriate average, but the layer will work not very well at the ends of frequency range of interest. For example, for C = 7.5%, to absorb the EMI in the range 2÷5 GHz, since the optimal thicknesses are 5 mm at 5 GHz and 12 mm at 2 GHz, the layer could be realized of thickness 8.5 mm. This thickness, however, will produce only a *RL* limited to −5 for frequency less than 2.4 and greater than 4.6 GHz.

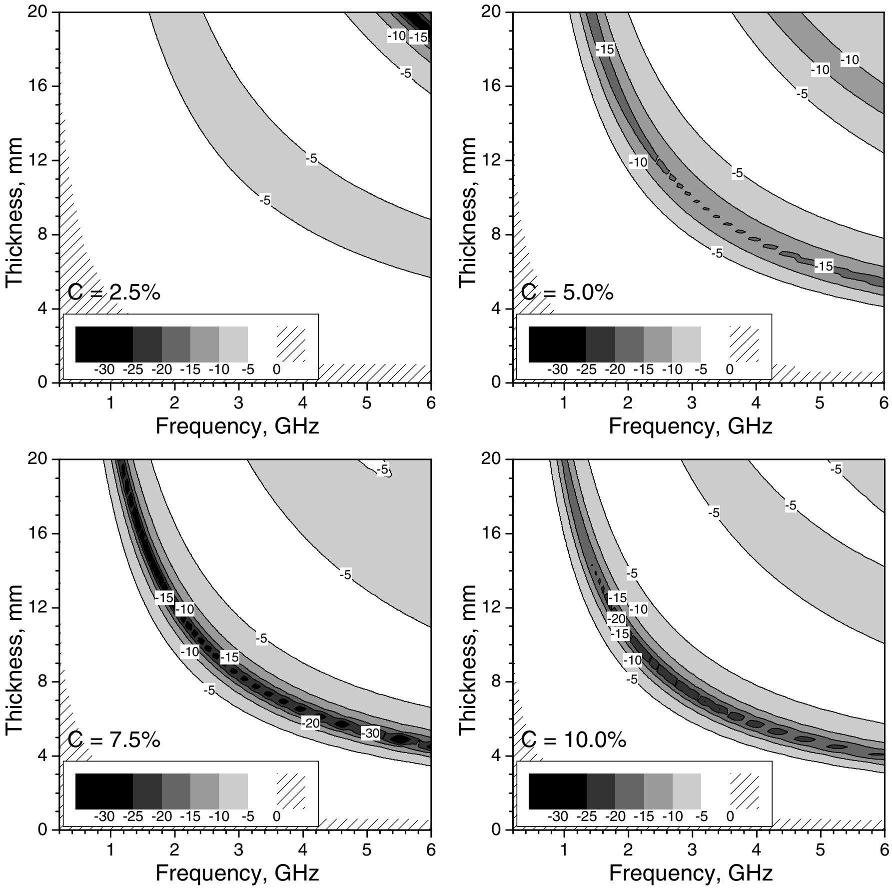

Figure 2. Contour plots of reflection loss as function of microwave frequency and layer thickness, for four different carbon black contents (2.5, 5.0, 7.5 and 10.0% w/w).

4. Conclusions

The permittivity of carbon black/silicone rubber blends were measured using a network analyzer in the frequency range 0.2÷6.0 GHz, varying the carbon black content up to about 10%.

The data were fitted by a suitable semi-empirical model and this model was than adopted as the basis for the calculation of reflection loss realizable by a microwave absorbing layer attached to a metal plate (a shield) made of carbon black/silicone rubber blends, with different carbon black content and different thickness, estimating the normalized input impedance of the layer.

Due to their microwave absorbing properties, the carbon black/silicone rubber blends could be adopted as shielding materials in the microwave frequencies. A predictive tool to design the optimal thickness and carbon black content of the shield, in order to realize the largest absorption of the electromagnetic wave (the minimum in reflection loss), has been pointed out in this work.

References

1. Neelakanta P.S., Handbook of electromagnetic materials, CRC Press, Boca Raton, (1995)
2. Annadurai P., Mallick A.K., Tripathy D.K., J. Appl. Polym. Sci. 2002; 83(1): 145-150
3. Kim S.S., Jo S.B., Gueon K.I., Choi K.K., Kim J.M., Churn K.S., IEEE Trans. on Magn. 1991; 27(6): 5462-5464
4. Kwon S.K., Ahn J.M., Kim G.H., Chun C.H., Hwang J.S., Lee J.H., Polym. Eng. Sci. 2002; 42(11): 2165-2171
5. Paul A., Thomas S., J. Appl. Polym. Sci. 1997: 63(2): 247-266
6. Achour M.E., El Malhi M., Miane J.L., Carmona F., Lahjomri F., J. Appl. Polym. Sci.; 1999: 73(6): 969-973
7. Moon K.S., Choi H.D., Lee A.K., Cho K.Y., Yoon H.G., Suh K.S., J. Appl. Polym. Sci.; 2000; 77(6): 1294-1302
8. Choi H.D., Shim H.W., Cho K.Y., Lee H.J., Park C.S., Yoon H.G., J. Appl. Polym. Sci.; 1999; 72(1): 75-83