Studies on Microwave Shielding Materials Based on Ferrite- and Carbon Black-Filled EPDM Rubber in the X-Band Frequency

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ABSTRACT: The preparation and characterization of ferrite- and carbon black-filled ethylene-propylene-ethylidene norborene terpolymer rubber composites were studied for microwave applications. Both the insertion loss and return loss were measured in a high-frequency range (X-band) with a network analyzer. The results indicated a relatively low frequency dependence. The difference in the insertion losses measured for all samples, except for one containing 100 phr ferrite and 75 phr carbon black (B75), was ±3 dB. Sample B75 showed broadband absorption in the aforementioned frequency band. Furthermore, the incorporation of both ferrite and carbon black powder into the rubber matrix altered the electrical and microwave properties, that is, the insertion loss and return loss; this could be helpful in the design of suitable absorbing materials for microwave applications at high frequencies. © 2002 John Wiley & Sons, Inc. J Appl Polym Sci 83: 145–150, 2002

Key words: carbon black; electromagnetic shielding; ferrite; insertion loss; microwave; return loss

INTRODUCTION

Microwaves are electromagnetic waves with a frequency range in the electromagnetic spectrum of 300 MHz to 300 GHz. However, most applications of microwave technology make use of frequencies in the range of 1–40 GHz.[1] Polymeric materials are extensively used as passive and active elements in different technological applications, but they lack the ability to shield electromagnetic radiation.[2] The increasing number of electronic systems in vehicles, higher speeds, and the greater use of microwaves in advanced navigation and domestic appliances have resulted in a corresponding growth in electromagnetic interference (EMI). A solution for eliminating these types of problems is possible through the use of microwave shielding materials. Conductive fillers, including carbon, graphite, and magnetic materials such as ferrite, carbonyl iron, and cobalt–nickel alloys, are incorporated into the polymer matrix for the absorption of microwave radiation. Microwave absorbers are produced by the alteration of the dielectric and magnetic properties of the existing material to allow the absorption of microwave energy for broadband wave attenuation. The objective of this study was the development of microwave shielding composites based on ethylene-propylene-ethylidene norborene terpolymer (EPDM) because EPDM has good aging and

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Table I Formulation of Mixes

	Mix No.						
Ingredients	Gum	A	B	C	B25	B50	B75
EPDM	100	100	100	100	100	100	100
Zinc oxide	5	5	5	5	5	5	5
Stearic acid	2	2	2	2	2	2	2
2,2,4 Trimethyl 1,2 Dihydro Quinoline	1	1	1	1	1	1	1
Barium ferrite	—	80	100	120	100	100	100
High abrasion furnace	—	—	—	—	25	50	75
Mercaptobenzthiazole	1	1	1	1	1	1	1
Tetramethylthiuramdisulfide	1.5	1.5	1.5	1.5	1.5	1.5	1.5
Sulfur	1	1	1	1	1	1	1

weathering properties and excellent thermal, electrical, and dielectric properties.[3] This article deals with studies of ferrite- and carbon black-filled EPDM rubber composites with respect to their physical and microwave properties in a high-frequency range.

EXPERIMENTAL

Materials

The electrical properties of materials such as barium ferrite and high abrasion furnace (HAF) black are important for descriptions of the shielding and absorbing characteristics of composites. Barium ferrite, used as a magnetic filler, was supplied by Pacific Metals (Seoul, Korea); it had a specific gravity of 3.0 and a particle size of 0.9 μm. HAF carbon black, with a density of 1.8 g/cc, a particle diameter of 23 nm, and a Brunaur, Emmett and Teller (BET) surface area of 80 g/cc, was obtained from Philips Carbon Black (Durgapur, India). The base polymer used was EPDM rubber (BUNA AP541) with a density of 0.86, a viscosity of 70, and termonomer 5 ethylidine-2 norbornene (ENB); it was manufactured by Bayer AG (Leverkusen, West Germany). Other chemicals used in these formulations were commercial-grade.

Sample Preparation and Physical Testing

Barium ferrite and HAF were dispersed in an EPDM rubber matrix for various loadings. The mixture of the rubber and other ingredients was carried out with a two-roll mill according to the formulation given in Table I. After the elastomer was mixed with the filler, we had to cure the compound to study the cure characteristics, which were essential for getting an idea of the processability. The cure characteristics of the compounds were determined with a Monsanto rheometer (R.100). The compounds were molded at 160°C in agreement with their respective cure times. The cured samples were tested for their properties, including tensile strength, elongation at break, 100% modulus, and hardness, in accordance with ASTM specifications.

Microwave Absorption Measurements

The mechanism of EMI shielding effectiveness (SE) is shown in Figure 1. The microwave absorber is an electromagnetic shielding material that attenuates radiated electromagnetic energy. When an electromagnetic wave impinges on the surface of a material, the wave undergoes a combination of reflection, absorption, and transmission. The EMI shielding theory is explained elsewhere in the literature.[4,5] SE is defined as the insertion loss due to shielding materials and is expressed as

$$SE(db) = 10 \log_{10} (Ps/Po)$$

where P_s is the electromagnetic powder transmitted through the sample and P_o is the electromag-

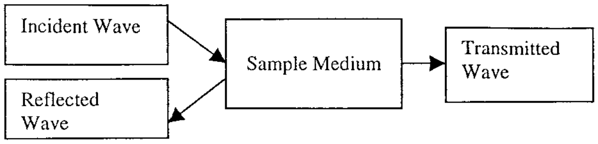
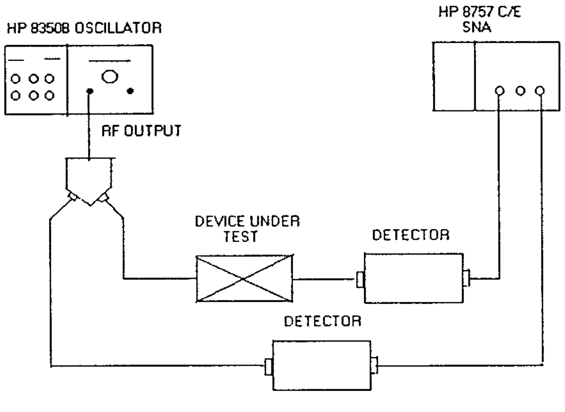
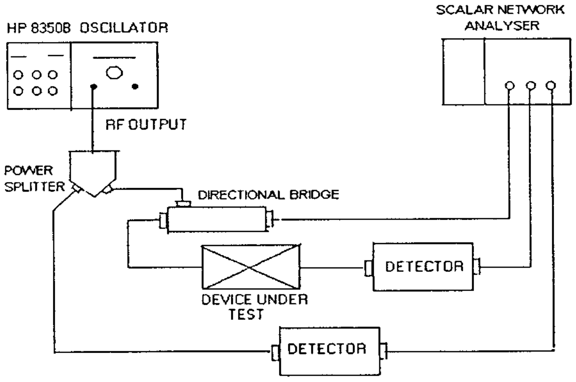


Figure 1 Specular behavior of waves at the interface and bulk of the material.



a) INSERTION LOSS



b) RETURN LOSS

Figure 2 Schematic diagram of the experimental setup for the measurement of (a) the insertion loss and (b) the return loss.

netic power incident to the sample. The electromagnetic wave interacts with the material via impurities, air insulation, pores. and voids.[6] The insertion loss is a measure of the total power loss due to attenuation and reflection and is defined as the ratio of incident energy to transmitted energy. It is function of a material's properties and thickness. The return loss is the portion of incident energy reflected off the surface and is defined as the ratio of reflected energy to incident energy. A schematic diagram of the experimental setup for the measurement of the insertion loss in plane wave radiation is shown in Figure 2(a,b).

The network analyzer was calibrated with a standard one-port calibration procedure. The sweep oscillator arrangement was capable of generating a wide frequency range. In this experiment, an 8–12-GHz frequency range was used. The specimen was prepared from a sheet according to the internal dimensions of the X-band rectangular wave-guide holder, with a cross section of 2.286 cm × 1.143 cm, and was placed perpendicular to the wave-guide axis for measurement. The sample thickness was 4 mm, which was arbitrarily chosen because 2 mm and 6 mm samples present difficulties in their placement in sample holders. To obtain all the parameters within the measurement limit, we used 4-mm-thick samples for the measurements.

RESULTS AND DISCUSSION

The cure characteristics of the compounds containing various loadings of barium ferrite and carbon black (HAF) are shown in Table II. The addition of barium ferrite reduces the scorch safety considerably; in addition, the maximum torque increases with the addition of both ferrite and carbon black. Table III summarizes results obtained after the evaluation of various physical properties of barium ferrite and carbon black compounds at 160°C.

Neither the 100% modulus nor the tensile strength changes appreciably with the addition of ferrite powder. However, the addition of carbon black shows a marked influence on the physical properties. This is due to the enhancement of the polymer–filler interaction, which results in im-

Table II Cure Parameters of the Compounds Containing Barium Ferrite and Carbon Black

	Mix No.						
Cure Parameter	Gum	A	B	C	B25	B50	B75
Minimum torque (N m)	1.36	1.58	1.92	1.81	1.36	2.49	3.16
Maximum torque (N m)	10.40	10.74	10.96	11.75	11.30	12.66	15.93
Scorch time (min)	3.25	2.13	1.95	1.75	1.75	1.50	1.38
Optimum cure time (min)	11.00	12.00	11.75	10.75	9.00	9.25	10.00

Table III Physical Properties of Barium Ferrite- and Carbon-Black-Filled Compounds

	Mix No.						
Property	Gum	A	B	C	B25	B50	B75
Specific gravity	0.84	1.35	1.47	1.56	1.54	1.60	1.72
Hardness, shore A	38	39	41	47	62	71	78
Tensile strength (MPa)	1.53	2.32	2.10	1.88	8.56	12.44	13.43
Elongation (%)	149.35	268.25	246.64	205.04	456.02	402.80	275.89
Modulus							
100%	1.22	1.49	1.46	1.48	3.56	6.13	5.46
200%	—	1.84	1.84	1.83	5.42	9.02	10.18
Resistivity Ohm cm	$>10^{15}$	0.25×10^{15}	7.84×10^{15}	9.8×10^{15}	4.9×10^{11}	$<9.8 \times 10^{7}$	$<9.8 \times 10^{7}$
Aging (70 h at 100°C)							
Hardness change (pts)	45 (+7)	47 (+8)	46 (+5)	51 (+4)	62	73 (+2)	81 (+3)
Tensile strength change (MPa %)	1.55 (+1)	2.35 (+1)	2.23 (+6)	2.33 (+23)	7.11 (−16)	11.99 (−4)	12.81 (−5)
Elongation change (%)	−8	−4	−5	−9	−26	−17	−17

proved reinforcement, leading to improvements in the physical properties of the compounds.

The volume resistivities of all the mixes, measured with a Hewlett–Packard (United States) model 16008A resistivity cell and a Hewlett–Packard model 4329A high-resistance meter, are reported in Table III. The results for all ferrite-loaded polymers show high resistivity values due to the high resistivity nature of ferrite. In general, the addition of ferrite powder to the polymer matrix enhances the SE without increasing conductivity.[7,8] However, the addition of carbon black to ferrite-loaded mixes (B25, B50, and B75) causes a marginal change in the conductivity that is attributed to the formation of a conductive network.

X-band microwave absorption spectra for the insertion loss and return loss were obtained for ferrite-loaded rubber and ferrite/carbon black-loaded rubber samples 4 mm thick. Variations in the insertion loss for ferrite-loaded rubbers as a function of frequency are shown in Figure 3, in which the insertion loss is shown to be relatively frequency-dependent. Variations in the insertion loss are observed over the entire X-band.

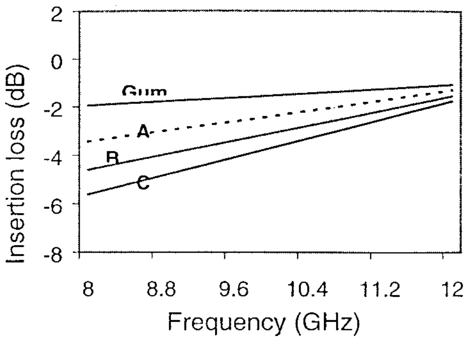


Figure 3 Effect of barium ferrite on the insertion loss.

The insertion loss increases with an increasing loading of the magnetic filler. This may be due to a high interaction of impurities (magnetic powders) with electromagnetic waves when the loading of the magnetic ferrite powder is increased. Attenuation in the material is achieved through dielectric conductive loss, magnetic loss, or both. The improvement is more pronounced in the lower side of the X-band frequency.

Figure 4 shows the effect of both ferrite- and carbon black-filled EPDM on the insertion loss. The addition of carbon black increases insertion loss and shows maximum broadband absorption over the entire X-band for sample B75 (Table I). This can be explained on the basis of electromagnetic waves, which become more attenuated as the conductivity is increased.

The return loss is given by the difference between the incident and reflected signal levels. Variations in the return loss with frequency (8–12 GHz) for mixes with ferrite contents of 80, 100, and 120 phr are shown in Figure 5. The return loss value decreases with the addition of ferrite loading. The minimum value of the return

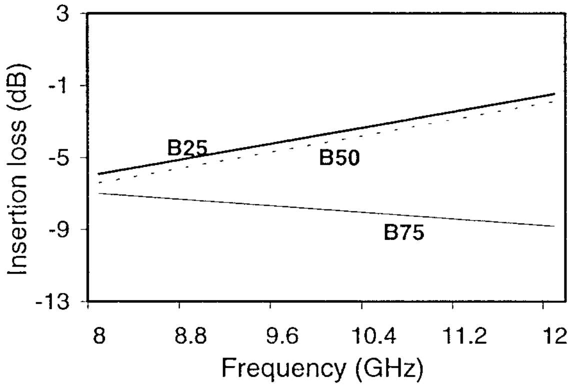


Figure 4 Effect of barium ferrite/HAF on the insertion loss.

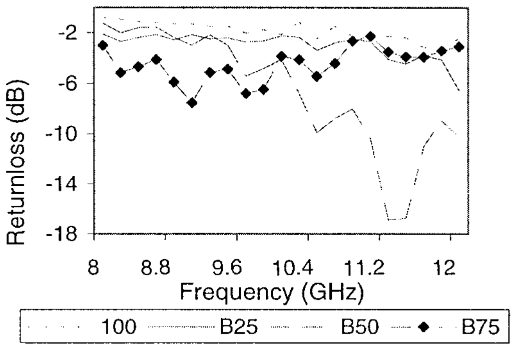


Figure 6 Effect of barium ferrite/HAF on the return loss.

loss is observed for a 120 phr ferrite-filled composite.

The increase in the insertion loss is associated with a decrease in the return loss, and this is in agreement with the behavior of most polymer conductive composites.[9] This can be explained on the basis of a change in the electrical conductivity of the composite. Ferrite-filled composites show little variation in their resistivity values on loading, but the incorporation of various loadings of carbon black into ferrite-filled mixes increases the conductivity, which increases the value of the return loss. The return losses for a composite with 100 phr ferrite and for composites with 25, 50, and 75 phr carbon black are shown in Figure 6.

The return loss increases with the carbon black loading, and a sample of mix B50 exhibits relaxation peaks in the X-band frequency range. This behavior may be the result of a phase transition from an insulator to a conducting composite around this concentration. The given composites seem to be characterized by a critical concentration of carbon black. At this concentration, a drastic change in the electrical resistivity with a corresponding change in the behavior of its electromagnetic characteristics is observed. Any further increase in the filler content will only improve the resulting electromagnetic characteristics. Because of the limited resolution of the high-resistance meter used for the resistivity measurements, the exact values for samples B50 and B75 are not reported in Table III. The variations in the return losses for all the mixes (Table I) are given in Figure 7 for frequencies of 8 and 12 GHz.

The minimum return loss is observed for all mixes at lower frequencies. Therefore, both the insertion loss and return loss can be controlled with the type of ferrite, the type of carbon black, and the loadings. The results obtained in this study reveal that the polymer is an insulator, and it can be considered transparent to high frequencies. When a filler is added to a matrix, the electrical behavior of the matrix may change according to the type and concentration of the filler.[10]

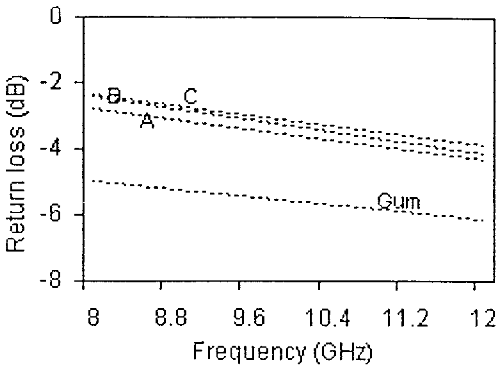


Figure 5 Effect of barium ferrite on the return loss.

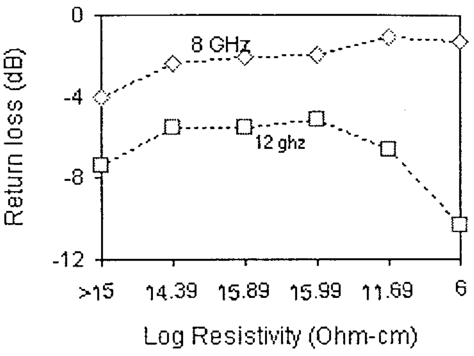


Figure 7 Effect of frequency and resistivity on the return loss.

CONCLUSIONS

The results of our study on ferrite- and carbon black-filled EPDM rubber in a high-frequency range are as follows:

1. The insertion loss shows a relatively small frequency dependence, and a composite with 100 phr ferrite and 75 phr carbon black (B75) exhibits broadband absorption characteristics.
2. At all frequencies of the incident radiation, ferrite-filled rubber shows an increased value of the insertion loss with loading without much of a change in conductivity. For example, maximum absorptions of −5.18 and −6.05 dB at 8 GHz were found for ferrite-loaded composites filled with 100 and 120 phr ferrite, respectively.
3. The addition of carbon black with a ferrite-loaded rubber matrix reduces resistivity. This is due to a marginal change in the conductivity, which is attributed to the formation of a conductive network.
4. The return loss decreases with the addition of ferrite loading, but there is an increase with carbon black loading.
5. Ferrite-filled composites have inferior mechanical properties in comparison with both ferrite- and carbon black-filled composites, but the presence of both ferrite- and carbon black-filled composites improves the physical properties of aging and SE.
6. Thus, ferrite- and carbon black-filled EPDM rubber composites are suitable as EMI shielding materials in the radar and television industries.

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