Electromagnetic Interference Shielding Effectiveness of Ethylene Vinyl Acetate Based Conductive Composites Containing Carbon Fillers

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ABSTRACT: Conductive polymeric based composites were derived from ethylene vinyl acetate rubber filled with Vulcan XC-72, short carbon fiber (SCF), and their blends. The electromagnetic interference (EMI) shielding effectiveness (SE), return loss, and reflection coefficient were studied. The measurements of the SE of the composites were carried out in two different frequency ranges of 100-2000 MHz and 8-12 GHz (X band). It was observed that the SE of the composites was frequency dependent and it increased with increasing frequency. The increasing of filler loading also enhanced the SE of the composites. The 100% SCF filled composites showed a higher SE compared to that of the filler blend or purely carbon black filled composites. The correlation between the SE and bulk conductivity of various composites was also discussed. The compromise between EMI SE, electrical conductivity, and mechanical properties was obtained when the composites contained both types of filler like particulate carbon black and SCF. © 2001 John Wiley & Sons, Inc. J Appl Polym Sci 80: 1601–1608, 2001

Key words: composites materials; shielding effectiveness; return loss; reflection coefficient

INTRODUCTION

Any device or apparatus that transmits, distributes, processes, or otherwise utilizes any form of electrical energy be a source of electromagnetic interference (EMI), which is a conducted and/or radiated electromagnetic signal. This EMI may intentionally or unintentionally react with either electronic devices, causing a degradation of performance of other equipment, or systems that share the same environment. Furthermore, electronic equipment that produces electromagnetic

radiation must be isolated or shielded to prevent it from degrading the performance of surrounding equipment. The shielding of this radiated EMI has caused increased concern to materials scientists. Previously the problem of EMI was solved through isolation of the electronic setup in some metallic housing. Nowadays conductive polymeric housing has gained popularity due to its flexibility, light weight, corrosion resistance, and lower cost than metal. The polymeric housing must be made conductive by coating, metallizing, or compounding with a conductive filler in order to achieve the required shielding effectiveness (SE).¹ The shielding provided by different conductive composites depends on the conductivity of the composites and on the measuring frequency. De-

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Ingredients	EF	EVF	EVF	EVF	EVF	EV
EVA	100	100	100	100	100	100
DCP	1.5	1.5	1.5	1.5	1.5	1.5
TAC	1.5	1.5	1.5	1.5	1.5	1.5
SCF	00, 10, 20, 30, 40, 50	10	20	30	40	_
Vulcan XC-72		40	30	20	10	20, 30, 40, 50

Table I Formulation of Mixes

pending upon the SE at different frequency ranges, these composite materials are used in different microelectronics devices²⁻⁷ and microwave applications, such as absorbing materials.⁸⁻¹⁰ In fact, this kind of flexible conductive composites may also be used for the following applications: typical antenna systems; anechoic chambers to avoid interference due to unwanted electromagnetic fields during measurement; radar cross sections; computer housing; PCB shielding; and different types of pressure sensitive switches, connector gaskets, and floor heating elements, and so forth. A number of research- ers^{11-13} reported the result of their investigations on conductive polymeric composites as EMI shielding materials. The present article reports the results of EMI SE, return loss, and the reflection coefficient of composites based on an ethylene vinyl acetate (EVA) copolymer filled with conductive Vulcan XC-72 short carbon fiber (SCF), and their blends over frequency ranges of 100-2000 MHz and 8-12 GHz (X band). The aim of this study was to assess the EMI shielding capability of the composites in the different frequency regions.

EXPERIMENTAL

Materials

The EVA copolymer (Levaprene 450, 45% VA content) was supplied by Bayer. The Vulcan XC-72

Table IIPhysical Characteristics of VulcanXC-72Carbon Black

Nitrogen surface area (m ² /g)	180
DBP absorption number (mL/100 g)	178
Particle diameter (nm)	29
Electron microscopic surface area (m ² /g)	86
CTAB surface area (m^2/g)	86
Pore area (m ² /g)	94
Electrical resistivity (Ω cm)	0.015

 $\ensuremath{\mathsf{DBP}}$, dibutyl phthalate; CTAB, cetyl trimethyl ammonium bromide.

conductive carbon black was supplied by Cabot Carbon Ltd. The SCF (Indcarf 12K) was obtained from IPCL. Dicumyl peroxide (DCP) and triallyl cyanurate (TAC) were obtained from E. Merck Ltd. and Aldrich Chemical Ltd., respectively.

The materials used in the formulation (parts per hundred parts of rubber, phr) in this work are shown in Table I. The general physical properties of carbon black and SCF are given in Tables II and III, respectively. Other compounding ingredients used (such as DCP and TAC) were commercial grade. The carbon black and SCF of a critical length of $\sim 6 \text{ mm}$ (length/depth aspect ratio, L/D= 880) were mixed with the rubber in a Brabender plasticorder (PLE-330) under identical conditions of time, temperature, rotor speed, and sequence of mixing of all compounding ingredients. The optimum cure times at 170°C for these composites were determined from a Monsanto R-100S rheometer. The mixes were sheeted out in a laboratory size two-roll mixing mill. Then the mixes were cured at 170°C in an electrically heated press under an identical pressure of 5 MPa. These cured sheets were then kept at room temperature for 24 h for maturation before testing.

Methods

The EMI SE was measured by a scalar network analyzer (HP 8753 C/E, Hewlett Packard) couple with sweep oscillator (HP 8350B, Hewlett Packard), a power splitter, a detector, and a scalar network analyzer (HP 8753 C/E, Hewlett Pack-

Table III	Physical	Characteristics	of	Short
Carbon Fi	bers			

December	Deless such a 'taile
Precursor	Polyacrylonitrile
Average length (m)	$6 imes 10^{-3}$
Diameter (µm)	6.8
Density (kg/m)	$1.78 imes10^3$
Electrical resistivity (Ω cm)	$1.5 imes10^{-3}$
Aspect (L/D)	882



Figure 1 The shielding effectiveness as a function of frequency measured in the 100–2000 MHz range of EVA based composites filled with different concentrations of (a) carbon black, (b) SCF, and (c) their blends.

ard). All these instruments were connected to a test chamber. The sample holder in the test chamber was changed depending upon the measurement frequency range. The SE in the 100-2000MHz frequency range was measured using the coaxial cable line method. The SE in the 8-12GHz frequency range was measured using an Xband waveguide as a sample holder. Two different samples (1.8 and 3.5 mm thick) were used for both measurements. The EMI shielding measurement was carried out for each sample by continuously sweeping the frequency ranges of 100-2000 MHz and 8-12 GHz. In both measurement techniques the specimens were placed perpendicular to the waveguide axis. When the SE was found to be greater than 40 dB some noise was encountered; otherwise the result obtained was a smooth SE curve against the frequency as obtained from the instrument.

The SEM observations of the brittle fracture surface of SCF loaded composites were made using a Jeol scanning electron microscope (JSM 5800). The samples were subjected to brittle fracture in liquid nitrogen, and the fractured surfaces were sputter coated with gold within 24 h before testing. In this study the composites were identified by an alphanumeric system. The first letters represent the rubber used and the numbers represent the amount (phr) of carbon black in the composites; EF50 and EV50 represent EVA based composites containing 50 phr of SCF and 50 phr of Vulcan XC-72 conductive carbon black; EVF 10.40 represents the filler blend composites containing 10 phr of Vulcan XC-72 carbon black and 40 phr of SCF.

RESULTS AND DISCUSSION

The present study dealt with the effect of Vulcan XC-72 black, SCF, and their blends with different proportions embedded in EVA copolymer matrix on the electromagnetic characteristics and consequently on the shielding capability of the resulting composite materials. The variation of the SE over the frequency ranges of 100–2000 MHz and 8–12 GHz of the EVA based composites containing a particulate (Vulcan XC-72), fibers (SCF), and filler blend are presented in Figures 1(a–c) and 2(a–c). The SE of all composites appreciably increased with increasing frequency over 100–2000 MHz, but the change in SE over the X band (8–12 GHz) region was marginal. It is interesting to note that the rate of increase of the SE with



Figure 2 The variation of the shielding effectiveness over the X-band frequency range of EVA based composites containing different concentrations of (a) carbon black, (b) SCF, and (c) their blends.



Figure 3 The shielding effectiveness at 8 and 12 GHz as a function of carbon black and SCF loading.

filler loading was much faster in the fibrous filled composites than in the particulate filled composites in both frequency regions (Fig. 3). We found that a marginal increase in SE occurred with the variation SCF loading from 0 to 20 phr, but at higher loading at ~ 30 phr the system became more efficient in shielding and the SE increased appreciably. Again for a further increase of SCF loading above 30 phr the increase in the SE became marginal. This variation in the SE against SCF loading can be correlated with the change in conductivity of SCF-rubber composites against SCF loading. Initially with the incorporation of carbon black in the rubber matrixes the change in conductivity remained marginal up to the critical concentration (percolation limit); then the increase in conductivity was abrupt (i.e., the insulating rubber matrix became conductive at and above the critical concentration because of the formation of conductive networks). In addition, above this critical concentration the change in conductivity with filler loading was only marginal. The increase in filler loading increased the number of filler particles in the composite that were interacting with the incident radiation. But there was a significant difference in the efficiency of the fibrous and particulate filler in imparting SE to the composite. For example, 50 phr of SCF showed a higher SE than 50 phr of Vulcan XC-72 black. In the higher frequency region the difference was more prominent; even 20 phr of SCF gave comparable SE to that of 50 phr of Vulcan XC-72 black. The SE of the filler blend composite increased more than that of SCF and Vulcan



Figure 4 The variation of the shielding effectiveness with the filler blends at 8- and 12-GHz frequency.

XC-72 black filled composites, and the SE decreased when the SCF was partly replaced by carbon black. For example, 20 phr of SCF and 30 phr Vulcan XC-72 blend composites showed higher SE than 30 phr SCF or 50 phr Vulcan XC-72 black filled composites. Incorporation of carbon fiber in the rubber matrix imparted higher conductivity compared to Vulcan XC-72 black with the same filler loading. The main purpose of partial replacement of the SCF with conductive particulate black was to enhance some of the reinforcement of the rubber matrix because, unlike carbon black, SCF does not impart high reinforcement to a rubber matrix.

It is well established that the SE of a conductive composite is related to its conductivity. The SE depends not only on the conductivity but also on the reflection and absorption coefficient of the dispersed filler. The variation of the SE at 8 and 12 GHz with filler blends is presented in Figure 4. We found that replacing the carbon black with SCF increased the SE. The distribution of filler in the matrix determined the void space between the filler aggregates. The closely packed arrangement of filler particles in the matrix gave higher SE. The fibrous filler may be considered to be a rigid, long aggregate of carbon black that gives rise to a more continuous network chain formation compared to particulate filler in the insulating polymer matrix. The SCF filled composites therefore impart higher conductivity and higher SE. The SCF were found to be randomly distributed in the composites containing only carbon fiber, as well



Figure 5 The variation of the return loss versus the frequency in the X-band frequency range of EVA based composites containing different concentrations of (a) carbon black, (b) SCF, and (c) their blends.



Figure 6 The variation of the return loss at 8 and 12 GHz as a function of carbon black and SCF blends.

as the blend of carbon fiber and carbon black. Because of the brittle nature of carbon, severe fiber breakdown of fibers occurred during mixing. The average aspect ratio (L/D) of the carbon fiber in the composites was found to be reduced by 95–98%, depending on the fiber loading and filler blend ratio.¹⁴ In filler blend composites, black aggregate can bridge the gap between two short carbon fibers. However, this efficiency of plugging the void space of the conducting mesh can be increased when carbon black is added to the ma-



Figure 7 The variation of the reflection coefficient versus the frequency in the X-band frequency range of EVA based composites containing carbon black, SCF, and their blends.







Figure 8 The shielding effectiveness at 100 and 2000 MHz as a function of the conductivity of EVA based composites filled with (a) carbon black and SCF and (b) the filler blend.

trix. Hence, the SE of the composites containing filler blend was higher.

The void in the conductive composites also affected the absorption, return loss, and reflection coefficient because of their internal reflection. Figure 5(a-c) shows the return loss as a function of frequency (8–12 GHz) at varying concentrations of SCF, Vulcan XC-72, and their blends. Figure 6 shows the variation of the return loss as a function of the filler blend at 8 and 12 GHz. It is apparent that the replacement of the Vulcan XC-72 by the SCF decreased the return loss at any particular frequency, but the change was



Figure 9 The shielding effectiveness at 8 and 12 GHz as a function of the conductivity of EVA based composites filled with (a) carbon black and SCF and (b) the filler blend.

marginal because the fiber length decreased during mixing in the presence of carbon black. Consequently, the reflection coefficient of these composites (Fig. 7) increased with the increase in SCF concentration in the filler blend matrix. The SE is the sum of the absorption loss and return loss (SE = A + R). This indicates that the loss due to the absorption increases with SCF concentration.

Although it is well established that the SE of a conductive composite is related to its conductivity, no quantification of this relationship via some function that is universally applicable to all materials at all frequencies has yet been reached.



a

b







Figure 10 An SEM micrograph of the fractured surface of EVA based composites containing (a) 50 phr SCF, (b) a filler blend of 10 phr Vulcan XC-72 and 40 phr SCF, and (c) a filler blend of 40 phr Vulcan XC-72 and 10 phr SCF at 200% magnification.

Figures 8(a,b) and 9(a,b) depict the relation between the SE and conductivity of the composites filled with Vulcan XC-72, SCF, and their blends at 100–2000 MHz and 8–12 GHz, respectively. Conductivity is a consequence of conductive network formation in the insulating rubber matrix. The network formations in the fiber filled composites are shown in a scanning electron micrograph (Fig. 10). This conductive network or conductive mesh is uniformly distributed in the matrix and interacts with electromagnetic waves. This conductive mesh is responsible for the SE.

CONCLUSION

- 1. Electrically conductive SCF filled composites are more effective for EMI shielding than conductive carbon black filled ones. SCF filled composites exhibit higher SE at comparatively lower filler concentration.
- 2. The SE increases with increasing fiber and carbon black concentrations. In the frequency range of 100–2000 MHz the variation of the SE with frequency is not systematic, but in the 8–12 GHz frequency range the SE increases slowly with the increase in frequency. The composites with high fiber concentration show maximum SE and the least return loss, which indicates that the loss due to the absorption increases.
- 3. The composites containing SCF and conductive particulate filler blend show higher SE than the composites filled with conductive particulate or SCF only. The addition of a small amount of particulate filler to the

fiber filled composites enhances the EMI shielding, as well as the mechanical properties, of the composites, which can be utilized as an effective shielding material for some electronic devices and microwave applications.

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