A Comparative Study of Multiwalled Carbon Nanotube Based Polystyrene and Toughened Polycarbonate Nanocomposites

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Submission: July 09, 2018; Published: July 17, 2018

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Abstract

The main objective of this article is to describe the effect of different length (aspect ratio) of carbon nanotubes (CNTs) on the electrical conductivity and electromagnetic shielding effectiveness of polystyrene/l-MWCNT and toughened polycarbonate/s-MWCNT composites. Long and short MWCNTs having aspect ratio of ~666 - 1333 and ~157 respectively were used for melt-mixed with polystyrene and toughened polycarbonate in a micro compounder. The uniform dispersion of MWCNT in matrix was confirmed by scanning electron microscopy. The realization shielding effectiveness value of -21dB respectively for PS/l-MWCNT composites: and -27dB for TPC/s-MWCNT composites at 10phr loading of MWCNTs, which show their potential use in making of mechanically strong and light weight EMI shield used for commercial application.

Keywords: Polycarbonate; Polystyrene; Multiwalled carbon nanotube; Electrical properties; EMI shielding effectiveness


Introduction

The broad developments of electronic systems and telecommunications has led to a novel type of pollution i.e. electromagnetic interference (EMI). EMI has appeared as a major problem, which not only affecting the proper working of electronic devices but as well as causing harmful effects to the health of human beings [1,2]. Generally, mobile phone, radar, radio transceivers, microwave oven, and various electronic devices are the main causes of EMI [3,4]. The long time exposure of EM waves have also been recognized as strong cancer-causing agent [5]. Therefore, appropriate shield is required to reduce the effect of EM waves. Generally, electrically conductive or magnetic filler are used as a EMI shielding material, because these materials have capability to interact with coupled electric and magnetic fields of the incident EM waves [6,7]. Metals and metal loaded composites have been widely used as EMI shielding materials, but these materials have disadvantages such as high density, corrosion prone, inconvenient processing etc. which limits their practical applicability. As compare to the metals, carbon nanomaterials have appeared as promising alternative conductive filler for production of EMI shield [6,8,9].

Nowadays, the use of carbon nanotubes (CNTs), having ultra-high modulus and strength, good thermal and electrical properties, as filler in polymer nanocomposites prepare material with lower filler loadings having improved electrical and EMI shielding properties[10-15]. As reported in literature, mechanical strength, electrical conductivity or thermal properties and EMI shielding effectiveness of the polymer nanocomposites are affected by different factors such as the aspect ratio, dispersion, processing methods, treatment methods, and loading of CNTs [16]. Li et al. [15] studied the conductivity and EMI SE of epoxy/SWCNT nanocomposites filled with SWCNTs having different aspect ratios. The maximum EMI shielding effectiveness has been reported for epoxy/SWCNT composites having 15wt% SWCNTs-long (SE ~49dB and 15-20dB obtained at 10MHz and in the 500MHz to 1.5GHz range respectively). In another study, Gupta et al. [17] reported EMI shielding properties (in Ku-band) of poly (trimethylene terephthalate) (PTT)/MWCNT nanocomposites. Electrical percolation of composites has been reported at 1wt% loading of MWCNT and SE of 36-42dB reported at 10wt% loading of MWCNT. Bai et al. [18] described the effect of nanotube aspect ratio on the electrical properties and mechanical strength of the
epoxy/MWCNT nanocomposites using three different length of MWCNTs (~1,10,50μm). It has been found that the short length MWCNTs (10μm) report good mechanical properties while long length MWCNTs (50μm) improved electrical properties of nanocomposites. Singh et al. [19] investigated the effect of CNTs having different aspect ratio on the electrical, mechanical and EMI shielding properties of epoxy/CNTs nanocomposites at low loading of CNT (0.5wt%). It has been observed that high aspect ratio CNTs filled nanocomposites show higher electrical (percolation threshold at 0.02wt% loading of l-MWCNT), mechanical (125MPa at 0.3wt% loading of l-MWCNT) and EMI shielding properties (highest SE ~16dB) in comparison to those filled with lower aspect ratio MWCNTs (percolation threshold at 0.11wt%, flexural strength ~113MPa at 0.3wt% and maximum shielding of ~11dB). Huang et al. [20] investigated the effect of heat treatment and CNTs with different aspect ratio on the EMI shielding effectiveness of epoxy/SWCNT nanocomposites (up to 15wt% SWCNTs). They reported that long SWCNTs based nanocomposites give high EMI SE (SE ~3 to 28dB) as compared to short annealed SWCNTs (SE ~21-23dB) and unannealed short SWCNTs (SE ~17-18dB). Al-Guo et al. [21] studied the effects of MWCNTs with high aspect ratio (313 and 474) on the electrical, mechanical, and thermal properties of PC/MWCNT nanocomposites. Above mentioned literature showed that most of studies either show EMI shielding effectiveness or mechanical properties of different types of MWCNTs based nanocomposites under higher loading of CNTs; or with both mechanical and EMI properties under lower loading of CNTs. Therefore, a comparative study of the preparation of higher loaded MWCNT nanocomposites and describing the effect of aspect ratio of MWCNTs on the mechanical, electrical and EMI shielding properties, of polymer nanocomposites is necessary.

The main objective of this paper to describe the effect of aspect ratio of MWCNTs on the electrical and EMI shielding properties of MWCNT based polystyrene (PS) and toughened polycarbonate (TPC) composites containing up to 10phr MWCNTs. The nanocomposites have been prepared by melt compounding of PS and TPC with MWCNTs in a twin screw micro-compounder. Two types of MWCNTs viz. long (l-MWCNT) having diameter ~7.5nm & length ~5-10μm (aspect ratio ~666-1333) and short (s-MWCNTs) having diameter 9.5nm & length ~1.5μm (aspect ratio ~157) were used for the fabrication of nanocomposites. The dispersion of MWCNTs in matrix is investigated by scanning electron microscopy. The EMI shielding of nanocomposites was measured in X-band (frequency range of 8.2-12.4GHz). The morphology, electrical conductivity and EMI shielding effectiveness of composites have been interrelated with distribution density and aspect ratio of CNTs, under both lower and higher loading.

Materials and Methods

Materials

Polystyrene (SC206) was purchased from Supreme Petrochem limited. Polycarbonate (PC) Lexan 143, procured from Sabic Innovative Plastic. Ethylene methyl acrylate (EMA) copolymer (Elvaloy® AC 1330 from DuPont) having 70% by weight ethylene and 30% by weight methyl acrylate are used in this investigation for toughening of PC [22,23]. Here, 5wt% EMA containing PC (TPC) is used for study. The long length MWCNTs (l-MWCNTs) were produced by CVD method using pongamia oil as a carbon source and ferrocene as catalyst and preparation process is mentioned elsewhere [24]. The diameter of l-MWCNT
was in the range of 50-200nm and length is in the range 5-10µm. The short length MWCNTs (s-MWCNTs), grade NC7000 with 90% carbon purity (length ~1.5µm and diameter ~9.5nm) were obtained from Nanocyl, Belgium.

Methods

Fabrication of PS/MWCNT and TPC/MWCNT nanocomposites

<table>
<thead>
<tr>
<th>Sample Designation*</th>
<th>Polystyrene (wt%)</th>
<th>Toughened Polycarbonate (wt%)</th>
<th>MWCNT Loading (phr)</th>
</tr>
</thead>
<tbody>
<tr>
<td>l-CNT/PS-0</td>
<td>100</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>l-CNT/PS-1</td>
<td>100</td>
<td>-</td>
<td>1</td>
</tr>
<tr>
<td>l-CNT/PS-2</td>
<td>100</td>
<td>-</td>
<td>2</td>
</tr>
<tr>
<td>l-CNT/PS-5</td>
<td>100</td>
<td>-</td>
<td>5</td>
</tr>
<tr>
<td>l-CNT/PS-10</td>
<td>100</td>
<td>-</td>
<td>10</td>
</tr>
<tr>
<td>s-CNT/TPC-0</td>
<td>-</td>
<td>100</td>
<td>-</td>
</tr>
<tr>
<td>s-CNT/TPC-1</td>
<td>-</td>
<td>100</td>
<td>1</td>
</tr>
<tr>
<td>s-CNT/TPC-2</td>
<td>-</td>
<td>100</td>
<td>2</td>
</tr>
<tr>
<td>s-CNT/TPC-5</td>
<td>-</td>
<td>100</td>
<td>5</td>
</tr>
<tr>
<td>s-CNT/TPC-10</td>
<td>-</td>
<td>100</td>
<td>10</td>
</tr>
</tbody>
</table>

*where PS-polystyrene, TPC- toughened polycarbonate, l-CNT- long multiwalled carbon nanotube, s-CNT- small multiwalled carbon nanotube, Numerical value- MWCNT content.

Characterization of PS/MWCNT and TPC/MWCNT nanocomposites

PS and TPC nanocomposites were characterized by morphological, electrical, EMI shielding thermal and spectroscopy study. Surface morphology of these nanocomposites were inspected using scanning electron microscope (SEM Zeiss EVO 50 at magnification of 30,000) and transmission electron microscope (TEM Zeiss 200kV). The cryofractured surface of composites were used for SEM imaging. The electrical conductivity (EC) of the nanocomposites at room temperature was measured by two point contact method using a Keithley 224 programmable current source. Electromagnetic interference shielding of composites were recorded on Agilent E8362B Vector Network Analyzer (VNA) in 8.2-12.4GHz frequency range (X-band).

Results and Discussion

Morphological characterization

SEM images of l-MWCNTs and s-MWCNT based PS and TPU nanocomposites are presented in Figure 2a - 2h respectively. MWCNTs are homogenously dispersed in the matrix, even at 10phr loading as observed from the images. Thus, melt blending using micro-compounder is an effective technique for homogenous dispersion of MWCNTs in matrix. From SEM images, it is also observed that, at any specific MWCNT loading, the distribution density of s-MWCNT in TPC matrix is higher compared to l-MWCNTs in PS matrix. These s-MWCNTs have more surface area in comparison to l-MWCNTs for interfacial interactions with TPC matrix, which are necessary for good mechanical strength and EMI shielding. On other side, l-MWCNTs with higher aspect ratio are expected to provide good stress transfer properties, low percolation threshold and long-range charge transport in PS matrix. Therefore, depending on the aspect ratio of MWCNTs, their distribution and loading inside matrix, significant mechanical, electrical and EMI shielding
properties are expected.

**Electrical conductivity (EC)**

The EC of PS/l-MWCNT and TPC/s-MWCNT nanocomposites with respect to MWCNT loading is presented in Figure 3. The EC of nanocomposites based on l- and s-MWCNT increased with increasing MWCNT loading and shows a drastic increase (~eight and seven orders of magnitude respectively) below 1phr MWCNT content, which show the formation of 3D conducting networks. Such a low percolation threshold value is due to the uniform dispersion of MWCNTs in matrix. It is reported in literature that percolation threshold is basically dependent on the different factors like the aspect ratio, intrinsic conductivity and dispersion of the conductive nanofiller [25-27]. Therefore, the exact value of threshold of l- and s-MWCNTs, has been projected by plotting the EC as a function of the MWCNT loading and performing data fitting using the scaling law [28-].

$$\sigma = \sigma_0 (\rho - \rho_0)^t \quad (1)$$

where $\sigma$, $\sigma_0$, $\rho$, $\rho_0$ and $t$ represents the EC of the nanocomposites the intrinsic conductivity of the nanofiller, volume fraction of nanofiller, the volume fraction at the percolation threshold and the critical exponent related to the system dimensionality respectively.

The linear regression data fitting (Figure 4 b & 4c) gives $\rho_0 = 0.83$phr & $\beta = 1.54$ for PS/l-MWCNT and $\rho_0 = 1.07$ wt.% & $\beta = 1.09$ for TPC/s-MWCNT nanocomposites. Considering the good dispersion of both l- and s-MWCNTs, the low percolation threshold for l-MWCNTs can be attributed to its high aspect ratio as compared to s-MWCNTs. It can also be seen that at any loading level, EC of PS/l-MWCNT nanocomposite is higher than respective TPC/s-MWCNT nanocomposite. This can again be attributed to the higher aspect ratio of l-MWCNTs responsible for long range charge transport in nanocomposites. As the EMI shielding is also related to the EC, the observed EC trend of nanocomposites recommend that, at comparative loading, EMI shielding effectiveness of PS/l-MWCNT nanocomposites should be higher than TPC/s-MWCNT nanocomposites.

**Electromagnetic interference shielding (EMI SE) of nanocomposites**

The EMI SE is the capacity of a material to attenuate incident electromagnetic waves. The EMI shielding is a direct result of the absorption of the wave as it passes through the shield's thickness, the reflection of the wave from the front face of the shield and multiple reflections of the waves at various interfaces. The presence of charge carriers in material helps in electromagnetic wave reflection via reflection mechanism electromagnetic wave penetrate through the material and get attenuated via the absorption. Absorption loss is more important for the magnetic field of electromagnetic wave than the electric field. Therefore, the electric field of electromagnetic wave is mostly reflected at the interface. The total SE of a material can be expressed in logarithmic power ratio as [32]

$$SE(db) = (SER + SEA + SEM) = 10 \log \frac{p_t}{p_i} \quad (2)$$

Where SEA, SER and SEM are the SE due to absorption, reflection and multiple reflections respectively and $pi$ is the power of incident wave and $Pt$ is transmitted EM wave.
Figure 4a & 4b shows the total SE of PS/l-MWCNT and TPC/s-MWCNT nanocomposites respectively. It can be observed that neat matrix (PS and TPC) sample gives negligible attenuation. However, incorporation of MWCNTs causes enhancement in SE for both PS/l-MWCNT and TPC/s-MWCNT nanocomposites obtaining SE value of approximately 21dB and 27dB at 10phr loading of MWCNT, respectively as given in Table 2 & 3. This can be ascribed to the formation of conducting networks throughout the insulating PS and TPC matrix. The EMI shielding results also shown that at lower loading (up to 2phr MWCNT loading), SE of PS/l-MWCNT nanocomposites is higher compared to s-MWCNT based nanocomposites, but at higher loadings, s-MWCNT based composites show higher EMI SE. Accordingly, PS/l-MWCNT-1 and TPC/s-MWCNT-2 show approximate similar shielding effectiveness (~7.8dB). These results indicate the effectiveness of l-MWCNTs at lower loading and s-MWCNTs at higher loading.

Figure 4: Variation in total shielding effectiveness with frequency of the (a) PS/l-MWCNT composites & (b) TPC/s-MWCNT composites; variation of SEA & SER as a function of MWCNTs content for (c) PS/l-MWCNT composites and (d) TPC/s-MWCNT composites.

The total SE have two components i.e. reflection and absorption (SER and SEA). Both SER and SEA increases with MWCNT loading can be observed from Figure 4c & 4d. The SEA increases at much faster rate as compare to SER in both the cases with increase in MWCNT loading. A brief investigation shows that at any given loading l-MWCNTs filled nanocomposites show superior SER value as compare to s-MWCNTs filled nanocomposites. However, absorption trend shown that below 5phr loading of CNT, PS/l-MWCNT nanocomposites show superior SEA values whereas at higher loadings, SEA of TPC/s-

### Table 2: Result of EC and EMI shielding properties of PS/l-MWCNT nanocomposites.

<table>
<thead>
<tr>
<th>Sample Designation</th>
<th>EC of PS/l-MWCNT (S/cm)</th>
<th>SER (dB) at 10.5GHz</th>
<th>SEA (dB) at 10.5GHz</th>
<th>Total SE (dB) at 10.5GHz</th>
</tr>
</thead>
<tbody>
<tr>
<td>l-CNT/PS-0</td>
<td>3.3 x 10^-16</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>l-CNT/PS-1</td>
<td>1.2 x 10^-8</td>
<td>-2.5</td>
<td>-5.3</td>
<td>-7.8</td>
</tr>
<tr>
<td>l-CNT/PS-2</td>
<td>2.3 x 10^-3</td>
<td>-3</td>
<td>-9.34</td>
<td>-12.36</td>
</tr>
<tr>
<td>l-CNT/PS-5</td>
<td>0.1</td>
<td>-3.8</td>
<td>-16.2</td>
<td>-20</td>
</tr>
<tr>
<td>l-CNT/PS-10</td>
<td>1.3</td>
<td>-4.5</td>
<td>-18.2</td>
<td>-21.2</td>
</tr>
</tbody>
</table>

### Table 3: Result of EC and EMI shielding properties of TPC/s-MWCNT nanocomposites.

<table>
<thead>
<tr>
<th>Sample Designation</th>
<th>EC of TPC/s-MWCNT (S/cm)</th>
<th>SER (dB) at 10.5GHz</th>
<th>SEA (dB) at 10.5GHz</th>
<th>Total SE (dB) at 10.5GHz</th>
</tr>
</thead>
<tbody>
<tr>
<td>s-CNT/TPC-0</td>
<td>1.23 x 10^-16</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>s-CNT/TPC-1</td>
<td>3.07 x 10^-9</td>
<td>-1.9</td>
<td>-2.5</td>
<td>-4.46</td>
</tr>
<tr>
<td>s-CNT/TPC-2</td>
<td>3.18 x 10^-6</td>
<td>-2.2</td>
<td>-5.7</td>
<td>-7.9</td>
</tr>
<tr>
<td>s-CNT/TPC-5</td>
<td>8.77 x 10^-4</td>
<td>-3.1</td>
<td>-17.8</td>
<td>-19.9</td>
</tr>
<tr>
<td>s-CNT/TPC-10</td>
<td>1.56 x 10^-3</td>
<td>-4.2</td>
<td>-25.7</td>
<td>-27.9</td>
</tr>
</tbody>
</table>
MWCNT nanocomposites dominate. This can be attributed to the fact that at lower loadings, the interfacial polarization effects are not too dominant, and conductivity shows the important role. Consequently, the TPC/s-MWCNT nanocomposites with a lower conductivity compare to PS/l-MWCNT nanocomposites show lesser attenuation. However, at higher loadings, better input impedance matching in case of TPC/s-MWCNT nanocomposites allows more incident waves to enter inside the shield that can be effectively dominated by absorption mechanism.

Conclusion

PS/MWCNT and TPC/MWCNT nanocomposites having up to 10phr loading of different aspect ratio MWCNTs (i.e. long and short MWCNTs respectively) have been prepared by melt mixing method. These nanocomposites show improved electrical conductivity and low electrical percolation threshold (i.e. 0.83 and 0.91phr for PS/l-MWCNT and TPC/s-MWCNT composites respectively) which is the sign of uniform dispersion of MWCNTs in the matrix. Further, the good electrical conductivity, processing induced morphology and difference in aspect ratio are responsible for observed maximum attenuation of -21dB and -27dB for PS/l-MWCNT and TPC/s-MWCNT nanocomposites respectively. It was also detected that PS/l-MWCNT nanocomposites show better SE at lower loading (up to 5phr) whereas TPC/s-MWCNT nanocomposites give better SE at higher loading. The aspect ratio of MWCNTs have been interconnected with the observed trends of electrical conductivity and EMI SE. These nanocomposites with good electrical conductivity along with high EMI shielding are considered as potential aspirant for making commercially feasible EMI shields.

References


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DOI: 10.19080/AJOP.2018.01.555561