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Electromagnetic interference shielding properties of carbon nanotube buckypaper composites

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Abstract
Preformed carbon nanotube thin films (10–20 μm), or buckypapers (BPs), consist of dense and entangled nanotube networks, which demonstrate high electrical conductivity and provide potential lightweight electromagnetic interference (EMI) solutions for composite structures. Nanocomposite laminates consisting of various proportions of single-walled and multi-walled carbon nanotubes, having different conductivity, and with different stacking structures, were studied. Single-layer BP composites showed shielding effectiveness (SE) of 20–60 dB, depending on the BP conductivity within a 2–18 GHz frequency range. The effects on EMI SE performance of composite laminate structures made with BPs of different conductivity values and epoxy or polyethylene insulating layer stacking sequences were studied. The results were also compared against the predictions from a modified EMI SE model. The predicted trends of SE value and frequency dependence were consistent with the experimental results, revealing that adjusting the number of BP layers and appropriate arrangement of the BP conducting layers and insulators can increase the EMI SE from 45 dB to close to 100 dB owing to the utilization of the double-shielding effect.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Electromagnetic (EM) waves entering a shielding conductor attenuate exponentially. The depth at which the EM field decreases to $1/e$ of the incident value is called the skin depth (δ), and for highly conductive materials, such as metal, it is given by equation (1) [1]:

$$\delta = \frac{1}{\sqrt{(\pi f \sigma \mu)}} = \frac{1}{\alpha} \quad (1)$$

where σ (S m^{-1}) is the electrical conductivity, f (Hz) is the frequency, μ is the magnetic permeability, and α is the attenuation constant. At a given frequency, high conductivity and permeability are important for better shielding. Therefore, metal has been commonly used for electromagnetic interference (EMI) shielding due to its high conductivity. Carbon-based materials, such as carbon black, graphite, carbon fibers [1–4] and conducting polymers [5–7], are good candidates due to their relatively high conductivity and lighter weight, as well as their application to traditional fabrication processes.

Carbon nanotubes (CNTs) are a promising EMI shielding candidate [8–14] due to their lightweight, high conductivity, and exceptionally high mechanical properties. In the case of highly conductive single-walled carbon nanotube (SWCNT) film, 10 nm thickness gives more than 20 dB at 10 GHz and over 50 dB can be easily achieved over 10 μm thickness [9]. In the polymer composite, high aspect ratio of CNT filler has advantage compared to carbon nanofiber and carbon black in terms of conductivity due to their lower percolation limit. Remaining metal catalyst in the multi-walled carbon nanotube (MWCNT) [10] and higher CNT aspect ratio [12] make higher conductivity and this leads better shielding performance. For the lightweight EMI shielding performance and corrosion resistance, purified CNTs with less metal impurity are desired. Longer CNT is beneficial in terms of conductivity because of reduced intertube contact [15].

To maximize the electrical conductivity and mechanical properties, higher loading of CNTs in EMI shielding composites is preferred. However, most of CNT composite with polymer matrix has maximum loading around 40 wt% [10] and 10 wt% in the epoxy [16]. Higher loading will decrease the mechanical properties due to agglomeration. Buckypaper (BP) is a thin film of entangled CNTs. BPs can be infiltrated with resin and easily be incorporated into the structural composite and the conventional fiber-reinforced composite manufacturing processes [16]. We can achieve more than 50 wt% loading of nanotubes in BP composite without losing the mechanical properties [16]. This can provide a new technical approach toward realizing EMI shield/structural multifunctional composites. Unlike other CNT composites, BP composites can achieve a high concentration of CNTs and high conducting nanotube networks to further improve EMI shielding effectiveness (SE).

SE is the sum of the effectiveness of all attenuating mechanisms and EM losses measured in decibels (dB) and expressed by equation (2) [17]

$$\mathrm{SE} = 10 \log \frac{P_{\mathrm{in}}}{P_{\mathrm{out}}} = \mathrm{SE_A} + \mathrm{SE_R} + \mathrm{SE_M} \qquad (2)$$

where P_{in} and P_{out} are the power of incident and transmitted waves through a shielding material; $\mathrm{SE_A}$ and $\mathrm{SE_R}$ are the SE values from absorption and reflection, respectively, and the third term ($\mathrm{SE_M}$) is multiple reflections in the shielding. The primary mechanism for EMI shielding is usually reflection. The third term is generally neglected when $\mathrm{SE_A} > 15$ dB [17].

Absorption loss (penetration loss) of a material with a thickness of l is given by [17]

$$\mathrm{SE_A}(\mathrm{dB}) = 8.686\alpha l. \qquad (3)$$

Considering the typical conductivity of BP materials is in the range of 200–1000 S cm^{-1} at a thickness of 15 μm, the typical $\mathrm{SE_A}$ at 1 GHz is between 1.2 and 2.6 dB. Therefore, the multiple reflections (or correction term, $\mathrm{SE_M}$) in buckypaper composites cannot be neglected.

For this research, composite laminates were made with multiple layers of BP and employing different stacking sequences with two types of dielectric materials (EPON862 and polyethylene). The resulting structures were studied to determine how the multiple reflection contributions affected the overall EMI SE. In contrast to most of the previous EMI shielding measurements based on a coaxial techniques like ASTM D4935 [14, 18, 19], which covers up to 1.5 GHz with a small test panel, our research team used a relatively larger test panel size (6″ × 6″) and measured up to 18 GHz using modified MIL-STD-285 or IEEE-STD-299 standard using an antenna. The effects of the conductivity and laminate structures were revealed, and the modeling and experimental results were compared. Effective laminate structures of buckypaper composites for high EMI shielding effectiveness were identified and demonstrated.

2. Experiment

2.1. Materials

The nanotube materials used in the research were purified SWCNTs made by the HiPco process (Unidym Inc., TX) or chemical vapor deposition method (Thomas-Swan, UK). To reduce the cost, MWCNTs (Cnano, CA and Bayer, Germany) were mixed with SWCNTs for BP fabrication. Long-MWCNT BPs were purchased from Nanocomp and used as is without further filtration procedure.

Aqueous suspensions of nanotubes were prepared using a sonication with an aid of surfactant, Triton X-100. The suspensions are filtered through a 0.45 μm filter in order to develop randomly dispersed BP sheets [20]. SWCNT BPs 15 μm thick with an aerial density of 21.5 g m^{-2} were used because of their highly conductive properties, extreme lightweight and nanoscale porosity structure. In the case of mixed BPs, MWCNTs were mixed with SWCNTs at a weight ratio of 5:1 and dispersed using the same method as for the SWCNT suspension. The typical thickness of 20–25 μm was thicker than that of SWCNT BP, hence, the density was slightly lower. The thickness of long-MWCNT BPs from Nanocomp was greater than 40 μm, which is thicker than the other BP materials. In the case of SWCNT BPs, the typical room temperature conductivity was around 200 S cm^{-1}. Conductivity of the mixed BP of SWCNT and MWCNT was around 50 S cm^{-1}, irrespective of MWCNT manufacturers. The conductivity of a long-MWCNT BP was 400–1000 S cm^{-1} depending on the thickness variation.

To improve the conductivity further, we soaked the long-MWCNT BP in the $SOCl_2$ for 40 h as described previously [21]. After complete drying in the hood, its conductivity increased up to 6000 S cm^{-1} with reduced thickness of 20 μm due to dense packing. The conductivity of $SOCl_2$ doped nanotube decrease as time goes on [22] and the stabilized conductivity of doped long-MWCNT BP after heat treatment at 100 °C decreased down to 2000 S cm^{-1}.

2.2. Structure of BP composite laminates

To investigate the effects of lay-up structures of BP composites on the EMI SE, BPs were attached to polymethacrylimide (PMI) foam [23] with EPON862/CURE EPI W (Shell Chemicals) and a vacuum bagging process [16] was used to produce the samples. To develop lightweight flexible EMI shielding materials, BPs with low density polyethylene (PE) laminates were made by a vacuum bagging process, slightly above the melting temperature of PE (149 °C). In the BP composite layer, the CNT content was about 50 wt%.

The sandwich structures were designed to understand the effects of the thin buckypaper layer arrangement on EMI shielding performance and the shielding mechanisms. Since the thickness of BP is several tens of micrometers, BP/PE composites can be very thin and flexible, yet highly conductive. The total thickness of the EMI shielding sample (BP/PE composite) was kept at less than 2 mm, maintaining the lightweight and flexibility.

2.3. EMI shielding effectiveness tests

For EMI shielding effectiveness, the samples were tested by Parker-Chomerics, in accordance with modified IEEE-STD-299 standards, or Lockheed Martin Missiles and Fire Control in Orlando, in accordance with modified MIL-STD-285 standards. During the tests, the transmitter and receiver antennas were placed on either side of a 3.5″ diameter aperture (or 6″ side panel) in the shielded enclosure. Open reference measurements were taken through this opening. Each sample was placed at this location, and shielding effectiveness was estimated. The tests were performed at frequencies ranging from 1 GHz (or 4 GHz) to 18 GHz, with the reliability of the measurement at 3 dB.

3. Results and discussions

3.1. Multilayer EMI shielding

Generally, the EMI SE of composite materials can be expressed by the Simon formula [2]

$$\mathrm{SE(dB)} = 50 + 10\log_{10}\frac{\sigma}{f} + 1.7t\sqrt{f\sigma} \tag{4}$$

where σ is the volume conductivity (S cm^{-1}), t is the thickness of the sample (cm), and f is the measurement frequency (MHz). This ignored the multiple internal reflections. However, BP composites cannot exclude the multiple reflections because of smaller absorption loss and extra-large surface areas (~520 m^2 g^{-1}) [24] of the materials.

The absorption loss ($\mathrm{SE_A}$) of a multilayer structure (n-layer) is the sum of each layer [17] and can be written as

$$\mathrm{SE_A(dB)} = 8.686(\alpha_1 l_1 + \cdots + \alpha_n l_n) \tag{5}$$

where α is the attenuation constant and l is the thickness of each layer. The insulation layers barely impacted to the absorption loss. The $\mathrm{SE_A}$ is simply the sum of each conducting layer. However, the reflection loss ($\mathrm{SE_R}$) and multiple reflections induced the correction term ($\mathrm{SE_M}$) are not just the sum of each layer. $\mathrm{SE_R}$ was determined from the intrinsic impedance of each sheet material (η_i) and its ratio,

$$\mathrm{SE_R} = 20\log_{10}\left|\frac{(\eta_0+\eta_1)(\eta_1+\eta_2)\cdots(\eta_n+\eta_{n+1})}{2\eta_0\cdot 2\eta_1\cdots 2\eta_n}\right| \tag{6}$$

where $\eta = \sqrt{\frac{\mathrm{i}\omega\mu}{\sigma+\mathrm{i}\omega\varepsilon}}$ and can be written as $\eta = (1+\mathrm{i})\sqrt{\frac{\pi\mu f}{\sigma}}$ in the case of metal, and $\eta_0 = \sqrt{\mu_0/\varepsilon_0} \approx 377\ \Omega$ in the case of air where $\sigma \sim 0$.

In order to effectively utilize minimal thickness, high conductivity and a large surface area BP conducting layer was used and inserted insulating layers increase multiple reflections contribution. Since the inserted insulation layers possess a different conductivity and dielectric constant, the BP/insulating interface would lead to additional internal reflection loss. If multiple BP layers with inserted insulating layers were used, the reflection loss would be the sum of all new interfaces formed on the BP layers and insulating layers. The schematic structure of multiple BP layer composite was shown in figure 1(a). Figures 1(b) and (c) is the resulting SWCNT BP composite with PE after vacuum bagging process.

As mentioned before, BP composites cannot neglect internal reflections in the shielding due to their small absorption loss, as compared to metal shielding structures. Therefore, multiple reflections induced correction terms must be utilized [17]

$$\mathrm{SE_M} = 20\log_{10}|[1-q_1\exp(-2\gamma_1\cdot l_1)]\cdots [1-q_n\exp(-2\gamma_n\cdot l_n)]| \tag{7}$$

$$q_n = \frac{(\eta_n-\eta_{n-1})[\eta_n - Z(l_n)]}{(\eta_n+\eta_{n-1})[\eta_n+Z(l_n)]}, \qquad \gamma = \alpha + \mathrm{i}\beta \tag{8}$$

where $Z(l_n)$ is the characteristic impedance and γ is the propagation constant with attenuation constant (α) and phase constant (β) (in the case of air or insulating material, $\gamma = \mathrm{i}\omega\sqrt{\mu\varepsilon}$ and in the case of metal, $\alpha = \beta$).

The estimate of the SE in this study is in the far-field limit [8, 17] with an incoming plane wave. This means that the distance from the source to the shielding barrier is sufficient compared to the wavelength.

Figure 2 shows the SE calculation of one layer of SWCNT BP (~15 μm thickness) composites based on its conductivity with and without multiple reflection induced correction terms ($\mathrm{SE_M}$). The solid lines are the sum of the SE, as shown in equations (5)–(7), which includes the correction term, and dashed lines from the conventional equation (equation (4)) that ignored the multiple internal reflection effect. In both cases, as the conductivity increased, the SE increased. However, the frequency dependence is different, especially at a low frequency range. This discrepancy lasts longer in the low conductivity sample. Since the absorption loss of the BP is small at a low frequency region of low conductive samples, the contribution of the correction term is significant.

3.2. Effect of conductivity

As shown in figure 2, conductivity is an important factor in determining the SE. Both the conventional equation and the model with internal reflection terms show SE improvement with higher conductivity. To determine the effects of conductivity in BP composites, BPs of different conductivity in the composites were studied. The characteristics of each BP were summarized in table 1.

Figure 3 shows the EMI SE comparison of the modeling and experimental results of a single BP layer/PE composite. DC conductivities of mixed BP/PE composites were less than 50 S cm^{-1} due to polymer infusion and SE of their laminates was less than 20 dB. Solid line is simulation based on σ = 50 S cm^{-1} and dashed line is the best fit of data with σ = 20 S cm^{-1} and this is the effective conductivity of BP/PE layer. Long-MWCNT BP with a conductivity of around

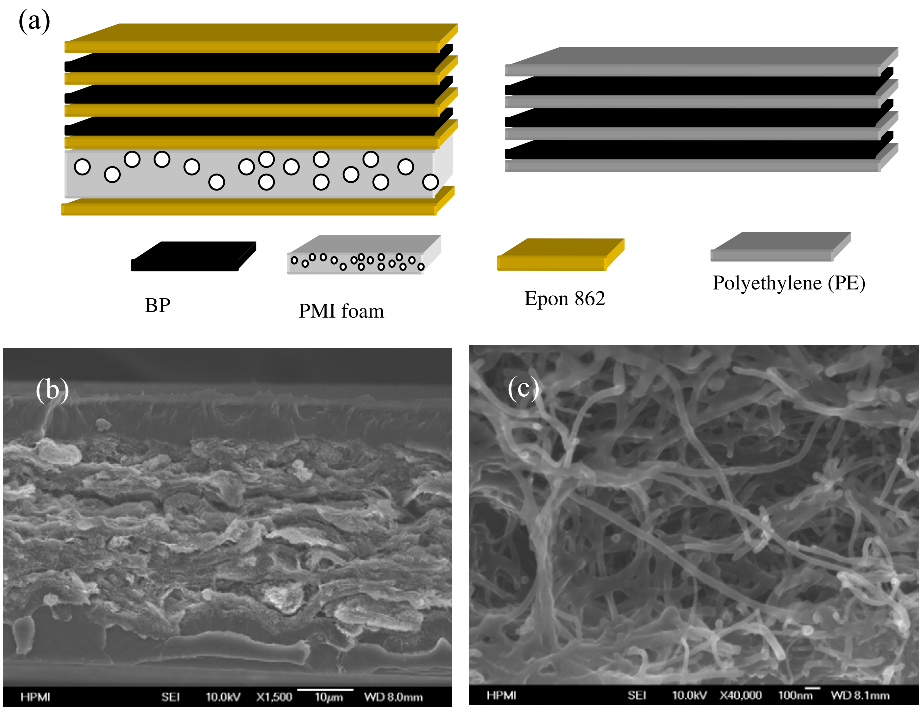

Figure 1. (a) Structures of buckypaper composite laminates with three SWCNT BP layers on the surface of the PMI foam and CNT BP layers with alternating PEs as separators. (b) Low magnification SEM images of the fracture surface of single BP layers with PE laminate. (c) High magnification SEM image shows PE infused to the BP, wrapped around nanotube networks and filling the voids.

Table 1. Summary of BP properties.

	Nanotube type	Typical BP conductivity (S cm^{-1})	Thickness (μm)
SWCNT BP	HiPco SWCNT	200	10–15
Mixed BP 1	SWCNT, MWCNT (large diameter)	50	20–25
Mixed BP 2	SWCNT, MWCNT (small diameter)	50	20–25
Long-MWCNT BP	Long-MWCNT	400–1000	20–60
$SOCl_2$ doped long-MWCNT BP	Long-MWCNT	6000	20–25

1000 S cm^{-1} resulted[4] in 50 dB throughout the frequency range up to 18 GHz and σ = 600 S cm^{-1} gives the best fit. To improve the conductivity, long-MWCNT BP was treated with $SOCl_2$ as described previously. Heat treatment during the lamination will decrease the conductivity. However, this doped long-MWCNT BP composite sample demonstrated the most enhanced EMI SE, over 70 dB at 12 GHz, with only one sheet of BP. And the estimated conductivity from the fitting is around 1500 S cm^{-1}. Even though the estimated effective conductivity of BP layer after polymer infusion is reduced, the SE increases proportional to the conductivity of original BP. Therefore, higher conductivity of BP is an important factor toward achieving high EMI shielding performance.

Several factors need to be considered for comparing theoretical estimates and the experimental results of the EMI SE. In the case of doped long-MWCNT BP/PE, the SE varies more than other sample. This may be related to the sample properties like irregular doping or non-uniform thickness after doping. In an actual measurement of SE, proper grounding is important [1]. A larger gap between the BP layers or BP layer to ground may also work as a leak and will reduce the SE. Additionally, constant conductivity was assumed at all of the frequency ranges, but the conductivity and dielectric constant can differ, especially at high frequency ranges, as mentioned in the reference [9]. Therefore, for a more rigorous estimate, frequency dependent conductivity and dielectric constant of BP materials must be considered. Attenuation constant (α) determined in equation (1), was determined with the assumption of the large loss tangent ($\tan\Delta = \varepsilon_i/\varepsilon_r \gg 1$). At a low frequency of below 1 GHz, $\tan\Delta \gg 1$ and BP is a good conductor. However, at a high frequency range of over 10 GHz, it is close to approximately 1 [9]. Generally, the attenuation constant is given as [5, 8].

$$\alpha = \frac{2\pi}{\lambda_0}\sqrt{\frac{\varepsilon_r(\sqrt{1+\tan^2\Delta}\mp 1)}{2}} \tag{9}$$

where λ_0 is the wavelength, ε_r is the real part of the complex relative permittivity, and $\mp$ signs are applied for positive and

[4] After vacuum bagging and hot-press process, the thickness of long-MWCNT BP was reduced down to 20–25 μm based on SEM measurement. Pressed long-MWCNT BP has similar thickness of 20 μm and it shows conductivity of 1000 S cm^{-1}.

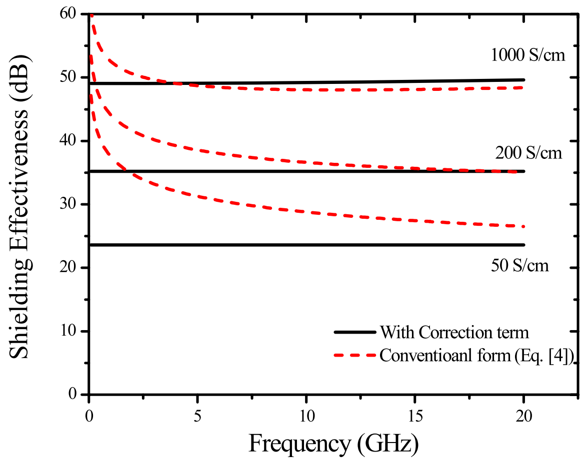

Figure 2. Theoretical calculation of SE in SWCNT BP ($\sim$15 μm thick) without multiple internal reflection correction terms (dashed line) and with correction terms (solid line) at different BP bulk conductivities.

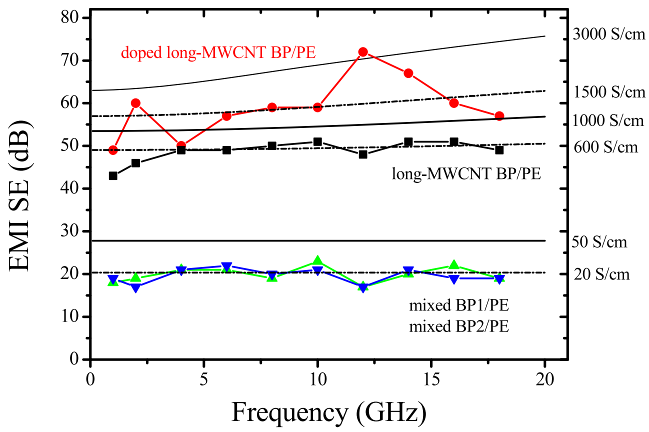

Figure 3. EMI SE of single-layer BP/PE composites with low conducting mixed BP and high conducting long-MWCNT nanotube BP from an IEEE-STD-299 measurement. Conductivity is the main factor in improving the EMI SE. Solid lines are based on the conductivity of original BP with 50 S cm^{-1}, 1000 S cm^{-1}, 3000 S cm^{-1} with 25 μm thickness and dashed lines are the best fit to the data with 20, 600, and 1500 S cm^{-1} from the bottom with thickness of 25 μm.

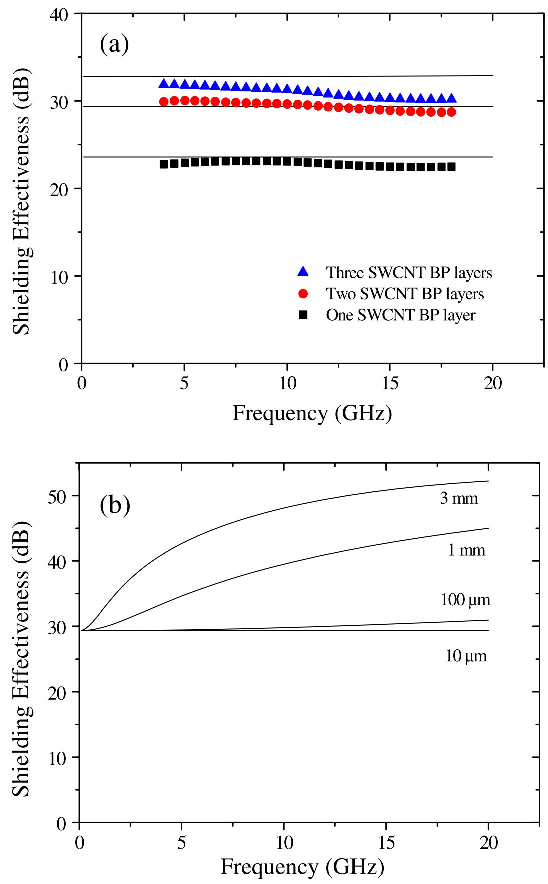

Figure 4. (a) EMI SE of multiple-layer SWCNT BP composites on the surface of PMI foam. The lines are the theoretical calculation of multiple SWCNT BP layers having different total thicknesses of 15 (one layer of BP), 30 (two layers of BP), and 45 μm (three layers of BP) with σ = 50 S cm^{-1}. (b) Modeled EMI SE of two separated BP layers with different air gap (ε_0) distances ($=d$). The BP layer was considered as a good conductor with $\tan\Delta \gg 1$, and σ_{BP} = 50 S cm^{-1}.

negative ε_r, respectively. Therefore, both the conductivity and ε_r should be considered together in the EMI SE of the BP samples at a high frequency range of over 10 GHz, which requires further study.

3.3. Effect of shielding composite structure

Figure 4(a) is the SE of multiple SWCNT BP layers attached to the PMI foam surface using the vacuum bagging process, as shown in figure 1(a). By increasing the number of BP layers, the SE improved from 22 dB to more than 30 dB. However, the SE increase was not proportional to the number of layers, and the increment of EMI attenuation was sharply reduced with the increase of the number of BP layers. The shielding performance of multiple-layer structures did not linearly increase due to the multiple reflection induced correction term. In the figure, the line is the SE calculation of SWCNT BP possessing different total thicknesses (d = 15, 30 and 45 μm) representing one, two, and three BPs with σ_{BP} = 50 S cm^{-1}.[5] They showed more or less the same SE value all over the frequency range. Therefore, the effectiveness of continuously adding shielding materials on the surface of the composites was limited toward achieving high SE because of the lack of reflection and multiple reflection terms.

Conversely, figure 4(b) shows the theoretical SE estimation of two separated BP layers with different air gap distances (ε_0). In the case of two BP layers with a small gap distance (10 μm), the SE was more or less the same as the multiple directly attached BP structures in figure 4(a). With the assumption of the BP with $\tan\Delta \gg 1$ and σ = 50 S cm^{-1}, the SE increased with a gap distance at a higher frequency range. This means that the insulating material thickness between the conducting BP layers is important to improve

[5] Resin infused to the BP layer and reduced the intertube contact. The conductivity of BP/PE composite is less than that of original BP and we used 50 S cm^{-1} instead of 200 S cm^{-1} as briefly discussed in section 3.2.

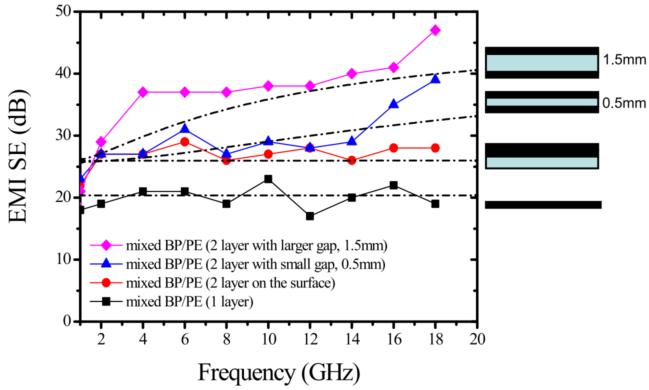

Figure 5. EMI SE of a single BP layer and a double BP layer with different gap distances from PE based on mixed BP1 ($t = 25$ μm). While increasing the gap distance, the EMI SE increased because of multiple reflection induced correction terms, as described in figure 4(b). A schematic illustration of the composite structure is shown on the right. Dashed lines are estimated SE values based on the simulation with BP/PE layer conductivity of 20 S cm^{-1}.

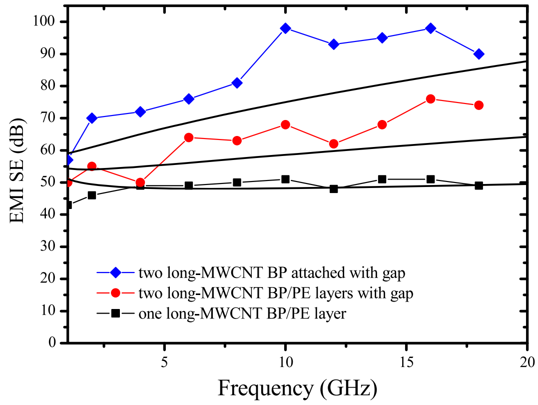

Figure 6. Effects of polymer impregnation on the EMI SE. Solid lines are based on the conventional form since the absorption loss is over 15 dB over 5 GHz. Without polymer impregnation, the effective conductivity of BP is higher and leads to better EMI SE.

SE due to the multiple shielding effect. Similarly, stacking of the MWCNT/PMMA coated layers with the gap [13] and laminating CNT layer [14] show better SE than a mixed composite. These improvements can be ascribed to multiple reflections.

Figure 5 shows the EMI SE of the mixed BP/PE composites with different stacking or lay-up structures. One layer BP/PE composite shows the lowest SE at around 20 dB all over the frequency range. Two layers of BP/PE composites on the surface of the PE substrate show minimal improvement (a 5–7 dB increase). These results were consistent with the results of figure 4(a). By adding 0.5 and 1.5 mm PE layers between the BP/PE layers, as shown in the right side of figure 5, the SE further improved. As expected in the theoretical estimation of figure 4(b), a larger gap between the conducting layers provides significantly better shielding performance with the same amount of BP especially at higher frequency range over 10 GHz. The SE difference between small gap and large gap distance is negligible at low frequency, but the SE increase faster in the case of larger gap as expected in figure 4(b). Dashed lines are estimated results based on BP/PE layer conductivity of 20 S cm^{-1} that is from the best fit from the data in figure 3(a). Theoretical estimation and experimental data are following similar frequency dependence. A slight deviation between them might be originated from the conductivity and gap variation. Since the absorption contribution of two BP/PE layers is around 5 dB at 20 GHz, most of the difference at higher frequency is coming from the correction term (SE_M). Therefore, the stacking conducting BP layer with proper insulation gap is important for higher EMI SE.

In the previous composite preparation procedure, EPON862 resin or PE was infused into the BP layer, which increased the intertube contact resistance of BP. Hence, the effective conductivity of BP/PE layer reduced around 40–60% compared to that of original BP and this will decrease the EMI SE in the composite. To maximize the shielding performance, we stacked two long-MWCNT BP layers that show higher conductivity than others. Instead of impregnation with a polymer, the BP layer was attached to the surface of the PE substrate to maintain its original conductivity. Figure 6 shows the comparison between the long-MWCNT BP layer with and without PE impregnation and compared to the SE of one layer long-MWCNT BP/PE that is shown in figure 3. The estimated conductivity of one long-MWCNT BP/PE layer is around 600 S cm^{-1}, and absorption loss range from 3.3 to 13 dB from 1 to 18 GHz. Therefore, two highly conducting long-MWCNT BP layers give absorption loss of 6.7–25.9 dB and simplified form in equation (4) can be used especially at high frequency range over 5 GHz. The two solid lines at the bottom are based on the conventional form with one and two long-MWCNT BP/PE layer with effective conductivity of 600 S cm^{-1} ($t = 25$ and 50 μm respectively see footnote 4). The top line is for two long-MWCNT BP with conductivity of 500 S cm^{-1} ($t = 100$ μm). With insertion of PE gap (1.5 mm), SE data show better than the theoretical estimation without it. Therefore, insertion of insulation gap between conducting layer and reduce the polymer infusion in the BP layer will be helpful to get a high EMI SE with the same amount of BP.

4. Conclusion

In this study, large EMI shielding test panels of over 6″ × 6″ with SWCNT, SWCNT/MWCNT mixed BPs and long-MWCNT BP were fabricated with PMI foam and a PE substrate. The SE performance of the EMI shielding was measured in accordance with MIL-STD-285 and IEEE-STD-299 standards. The SE measures the sum of absorption, reflection, and multiple reflections in the shield materials. For metal-based shielding materials, the third term from multiple internal reflections is usually ignored in modeling, but this cannot be ignored in the BP composites due to small absorption loss and more multiple reflection contributions due to large surface areas and internal structures. The SE of BP composites mainly depended on the conductivity and thickness of the BP layers. SWCNTs, or long-MWCNT BP, provide better EMI shielding due to their high electrical conductivity. However, increasing the number of BP layers by adding on to the composite surface showed some limitations toward realizing high EMI shielding performance due to a lack of multiple reflections. Therefore, in addition to the improvements in electrical conductivity, proper stacking or lay-up of the BP and insulation layers in the laminates is also important in achieving higher EMI attenuation. Finally, achieving up to 100 dB SE with only two highly conducting BPs with a designed insulating gap was demonstrated.

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