

# Electromagnetic Shielding Potential of Graphene/Polyethylene Composites\*

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**Abstract**– Chemically reduced graphene oxide (RGO) was synthesized from graphite powder (G) by using improved Hummer method. Graphene oxide (GO) was chemically reduced by using ascorbic acid to RGO. Reduction protocol was illustrated briefly. Typically, SEM, TEM, Raman and XRD characterizations were carried out to confirm the successful complete conversion of graphite to RGO. RGO. Additionally, G and RGO polyethylene (PE) composites were prepared by using different mass loading of 10, 20, and 30%. Electromagnetic measurements (8-12 GHz) were done for G and RGO and their composites. It was observed that average reflection losses (RL) decrease and the average transmission loss (TL) increase as the percentage of G and RGO increased from 10% to 30% in the composite matrix. The higher shielding optional of RGO composites with respect to corresponding G composites was attributed to the higher conductivity of RGO with respect to G. The highest TL average value was -45 dB for optimum mass percentage 30% RGO. The EMI shielding potentials of 30% RGO /PE may have promising EMI applications.

**Keywords:** graphene, reduced graphene oxide, improved Hummer method, composites, electromagnetic shielding.

## I. INTRODUCTION (HEADING 1)

Electromagnetic shielding (EMI) materials can be classified into magnetic and dielectric materials, also it can be classified into three branches. The first one is metallic based materials, which contains metallic powders, magnetic metal oxides, and metal carbonyl [1-3]. The second is carbon based materials such as graphite and nanocarbons. Graphene and carbon nanotubes (CNTs) are the most interesting and promising nanocarbons for EMI applications [4-7]. Finally, conducting polymers such as polyaniline also have a potential applications and significance [8-10].

Nanostructured EMI materials have a great interest in the microwave absorption technology, because of the fascinating properties, like absorbing more microwave compared with the bulk counterparts due to having extremely small particle size and enormous surface area [11, 12]. Graphene is one of the most talk of nanostructured carbon materials due to its excellent properties, like high electrical conductivity and large surface area, good thermal and optical properties. Several methods are used for graphene preparation, such as liquid phase exfoliation [36-38], reduction of graphite oxide [39, 40], chemical vapor deposition [41-43] and organic synthesis [44,

45]. These different methods of production leads to a graphene with a different size, shape and chemical compositions [46, 47]. Graphene has a great interest in the application of microwave absorption, due to important features, like low density, high chemical stability, electrical conductivity and large specific surface area. [21, 22, 26-35]. The interesting physical and chemical properties of graphene make it an attractive filler for composite materials [21-23].

In this work GO was synthesized form graphite procures via improved Hummer method and then rescued with ascorbic acid to obtain RGO. EMI shielding potential of RGO/commercial polyethylene (PE) composites was examined and compared with corresponding graphite / PE composites.

## II. EXPERIMENTAL WORK

### A. Materials, Chemicals, and Synthesis Procedures

The chemicals used in the experimental work were in the reagent grade and obtained from the indicated sources, with some specified purities, as shown in table (1). No further purification was carried out.

Typically, 10 gm graphite powder as starting material are added to 1200 ml concentrated sulfuric acid  $H_2SO_4$  and 150 ml of concentrated phosphoric acid  $H_3PO_4$  in a 5 L beaker kept at room temperature 25-30 °C for 30 min. 60 gm of potassium permanganate  $KMnO_4$  gradually added to the mixture, stirring the mixture for 1 hour, then kept under room temperature for 24 hours. Cold distilled water is (1000 ml) are added to the mixture which kept at 5 °C in water path for 1 hour under continuous stirring. By adding distilled water (1500 ml) and hydrogen peroxide (30%, 300 ml) the reaction was ended. The product produced can be observed with a brown /yellowish color, which is the color that indicates the formation of graphite oxide GO. Distilled water was used to wash the product to remove any remnant byproducts in the solution by decantation. After repeating this process 5 times, the obtained GO suspension are reduced by using ascorbic acid. The reducing agent (ascorbic acid) was added to the 70 °C heated GO suspension under mechanical stir for 24 hours. Finally washing by distilled water to remove any remnant, and drying

in oven at 40 °C for 24 h to obtain the final product of reduced graphene oxide, with a black color.

TABLE I  
CHEMICAL USED IN PREPARATION

Chemicals	Purity	Source
Graphite	99.5%	NICE / India
Potassium Permanganate	98%	Alpha Chemicals/ India
Sulfuric Acid	98%	Alpha Chemicals/ India
Phosphoric Acid	85%	Alpha Chemicals/ India
Hydrogen Peroxide	35%	Alpha Chemicals/ India
Ascorbic Acid	98%	Alpha Chemicals/ India
Polyethylene	98%	Alpha Chemicals/ India

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### B. Instrumentations and Characterizations

Morphological and EDX characterizations of graphite precursor and the obtained RGO were carried out by scanning electron microscope (SEM, MIRA 3 XMU, TESCAN). Further, morphology of the RGO was investigated by High Resolution Transmission Electron Microscope (HRTEM) - model JEM-2100, JEOL, Japan. A small quantity of the RGO sample under investigation was dispersed in 10 ml ethanol then sonicated for 30 min. A few drops of the resulting suspension were placed on a copper grid, then photographed at acceleration rate of 200 KV. Moreover, Raman spectroscopic measurements are done, by using a dispersive Raman microscope (model Senterra, Bruker, Germany). A power of 10 mW used with a frequency range from 1000 to 4000 cm<sup>-1</sup> and resolution of 4 cm<sup>-1</sup>. Raman spectroscopy played an important role in the structural characterization of graphitic materials. X-ray powder diffractometer, Analytical XPERT PRO MPD, With ( $\lambda= 1.5418 \text{ \AA}$ ) with 40 kV rating, 40 mA, was employed to collect the XRD patterns of graphite precursor and obtained

RGO. The diffraction pattern covered the range of 10°-30° (2 $\theta$ ), with angular rang, 0.02° (2 $\theta$  step size) and 0.4 second scan step time.

TABLE II  
RGO AND GRAPHITE / POLYETHYLENE MATRIX

Sample	Graphite weight	RGO weight	PE weight
10% G	0.07 g	----	6.93 g
20% G	0.14 g	----	0.56 g
30% G	0.21 g	----	0.49 g
10% RGO	----	0.07 g	6.93 g
20% RGO	----	0.14 g	0.56 g
30% RGO	----	0.21 g	0.49 g



Fig. 1 RGO, PE before and after mixing

An Agilent E8363A PNA vector network analyzer figure (2) is used in order to achieve perfect electromagnetic measurements. The cross-section dimension of the waveguide (WR90) is 22.86 mm × 10.16 mm, with 7 mm length of sample holder. Reflection loss (RL) and transmission loss (TL) are measured in the frequency of the X-band (8.2–12.4 GHz). The waveguides filled with the prepared samples

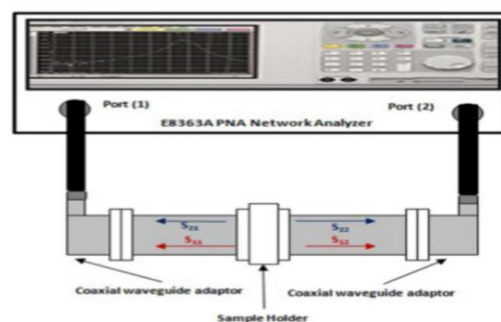


Fig. 2 EMI measuring setup

## III. RESULTS AND DISCUSSION

### A. Morphological Analysis & EDX Analysis

The microstructure of graphite and RGO are shown in Fig. 3. The SEM image of graphite shown in Fig. 3-a shows the flaky structures of graphite procures with thinness in the order of 2-5 $\mu\text{m}$  and lateral dimension in the order of 100-150  $\mu\text{m}$ . On the other hand, the SEM image of RGO shown in Fig. 3-b shows a shape of rankled thin layer than observed in the case of graphite. In addition, the SEM of RGO did not show any residual of graphite, which indicate the successive transformation of graphite to RGO. In order to obtain exact information about fine features, Transmission electron microscopy TEM was used. The dark areas shown in the TEM of RGO may indicate thick staging nanostructured layers of

RGO and on the other hand the high transparency areas indicates much thinner films [13, 14]. Also, the TEM result of RGO indicating the thin layer profile of graphene without any contamination on the surface or residual buck graphite. Further, the EDX comparative analysis between graphite and RGO that shown in table 3. EDX analysis of G indicates a 95.72% of carbon atomic ratio and with a 4.04% of oxygen ratio, where the high carbon / oxygen ratio refers to the pure graphite chemical structure, with low functional groups or oxidizing agents [200]. Where RGO shows a very similar trend confirming the successive reduction process. Fig. 4 shows the SEM images of neat PE matrix, 10% RGO/PE, and 10% graphite/PE. As shown in Fig. 4-b, which represents 10% graphite/PE sample, it is clear to distinguish the graphite flakes within the PE matrix. On the other hand, the SEM of 10% RGO/PE sample shown in Fig. 4-c indicate the better distribution of RGO in PE matrix rather than in case of graphite in PE matrix. The relative larger surface area of RGO and thinner thickness of RGO with respect of graphite may lead to better homogeneity of RGO distribution in PE matrix rather than graphite.

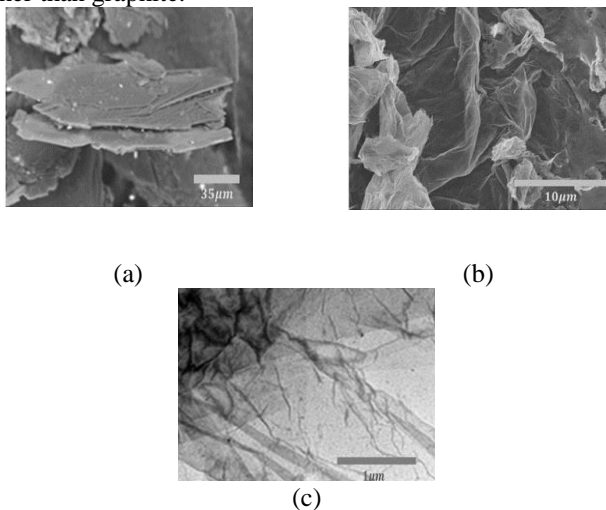


Fig. 3 EMI measuring setup

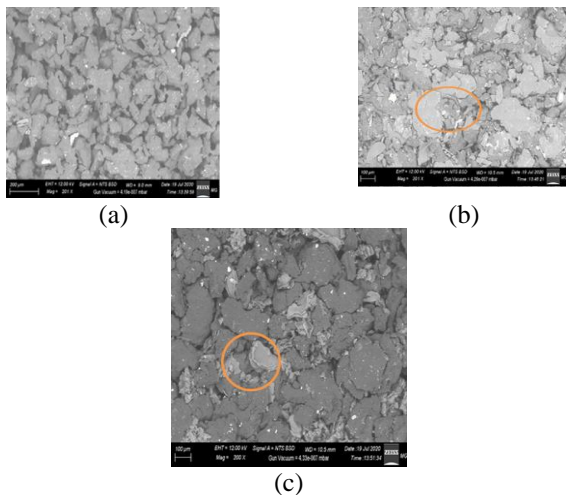


Fig. 4 (a) SEM of PE matrix, (b) 10% graphite / PE composite and (c) 10% graphene / PE composite

TABLE II  
EDX ANALYSIS OF G AND RGO

Sample	C%	O%
G	95.72	4.04
RGO	94.3	5.7

### B. Raman Spectroscopy

The Raman analysis of RGO produced by improved Hummer method, with respect to pure graphite powder is shown in figure (5). The G peak for reduced graphene oxide RGO is shifted to lower frequency with a value of 1575  $\text{cm}^{-1}$  with respect to pure graphite of 1580  $\text{cm}^{-1}$ . The recovery of the hexagonal shape of carbon atoms with defects is the main reason of lowering the value of G peak. The broad of the G peak for RGO is implemented to the presence of oxidizing functional groups in carbon lattice. The high value of the D peak in RGO with respect to G-NICE refers to defects in RGO due to the chemical reduction of GO using ascorbic acid which leads to the high ID/IG intensity ratio. While the high decrease in the 2D peak of RGO with respect to G-NICE, that indicates the chemical reduction process to produce RGO [15]. The relative peak intensity values of both pure graphite G-NICE, and RGO produced by improved Hummer method are shown table (4).

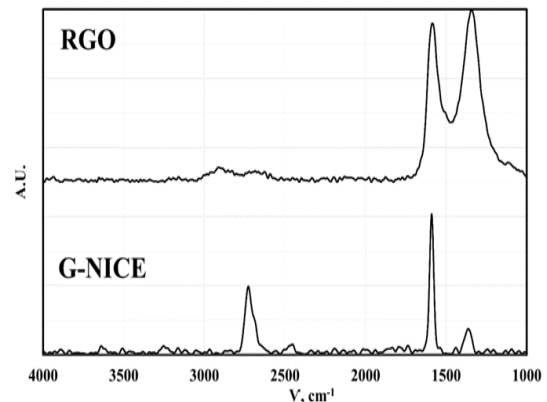


Fig. 5 Raman analysis for graphite and reduced graphene oxide

### C. XRD Analysis

The XRD of the RGO prepared by the improved Hummer method RGO, with respect to the G-NICE can be shown in figure (6). The 002 peak of G-NICE can be observed at  $2\theta$  value of  $26.5^\circ$ , which is a sharp peak with high intensity indicating the interlayer spacing and crystal structure of pure graphite. The observation of a low intensity peak of RGO at  $2\theta$  value of  $26.1^\circ$ , with a broad shape, indicates the reduction process of graphite oxide GO and the removal of oxidizing functional groups. The conclusion from the previous results that RGO is suitable to use as graphene-based composites. The XRD analysis indicates reduction process of GO and removal of functional groups.

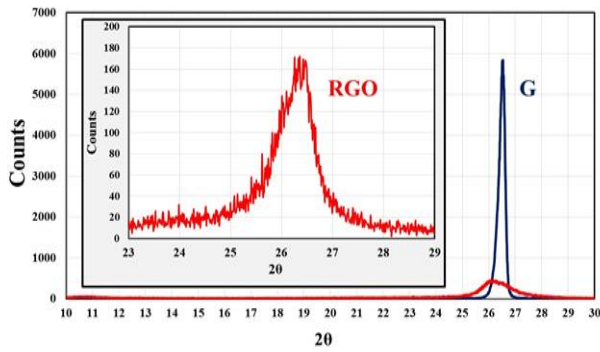


Fig. 6 the XRD analysis of RGO with respect to G-NICE

#### D. Electromagnetic measurements

Fig. 7 shows the RL and TL profiles obtained by 0.7g of G and RGO within frequency band 8-12GHz. It was found that the average RL values achieved by G and RGO was  $-2.9 \pm 0.4$  dB and  $-1.8 \pm 0.6$ , respectively. On the other hand, the average TL values achieved by G and RGO was  $-30.8 \pm 0.7$  and  $-59 \pm 0.8$  dB, respectively. It was obvious to report that RGO achieved almost double the TL of G. This results may be due to the high conductivity and hence imaginary part of the permittivity  $\epsilon''$  of RGO with respect to G. Also, the observed very low values of RL of G and RGO indication the relatively lower impedance values of G and RGO with respect to air leading to decreasing matching between the incident wave interface [16-18].

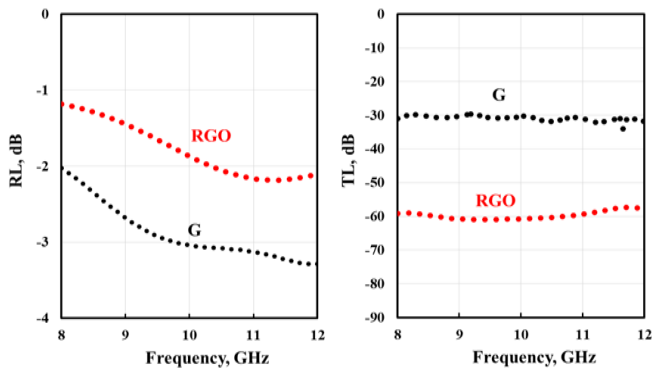


Fig. 7 Reflection losses and Transmission losses by 0.7g of G and RGO

Fig. 8 and 9 shows TL and RL in dB of 0.7 gm of G and RGO composites with PE with different loading percentages over a frequency range of X-band (8-12) GHz. It was obvious to report that as the percentage of G and RGO increased from 10 to 30% lead to increasing TL and reducing RL. According to the obtained results, as the G percentage increased from 10 to 30% the RL decreased from  $-4.6 \pm 0.8$  to  $-2.5 \pm 0.3$  dB, respectively. Also, TL of increased from  $-8.4 \pm 2.1$  to  $-20.4 \pm 2.6$  dB, respectively. On the other hand, as the RGO percentage increased from 10 to 30% the RL decreased from  $-4.3 \pm 0.4$  to  $-2.4 \pm 0.5$  dB, respectively.

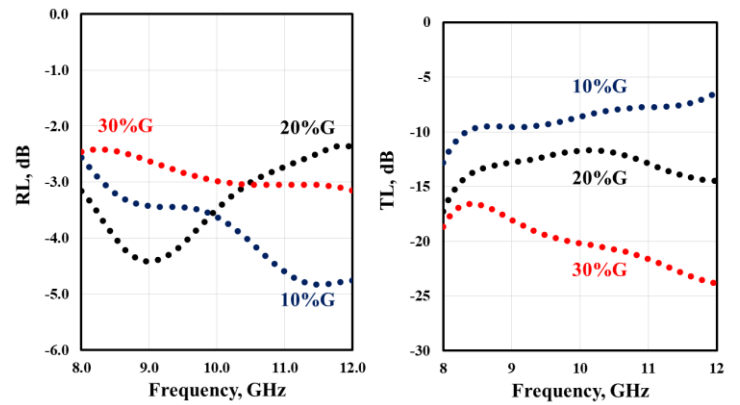


Fig. 8 Reflection losses and Transmission losses by 0.7g of 10, 20 and 30% G /PE composite

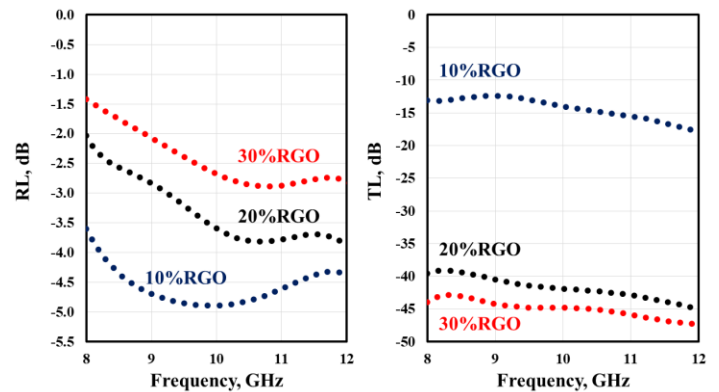


Fig. 9 Reflection losses and Transmission losses by 0.7g of 10, 20 and 30% of RGO-Polyethylene

Also, TL of increased from  $-14.7 \pm 1.9$  to  $-45.3 \pm 1.6$  dB, respectively. This phenomena is observed due to increasing composite conductivity as the concentration of conductive filler increased. Interestingly, RGO/ PE composites with PE achieved higher TL losses than the corresponding G/ PE composites. These results confirm the higher conductivity of RGO/PE than the corresponding G/ PE composites. Table 6 summarized all the obtained EMI shielding results. It was obvious that 30% RGO/PE composite has a promising EMI potential

TABLE VI  
Mass loading percentage impact on RL and TL

	10%		20%		30%	
	RL	TL	RL	TL	RL	TL
G	$-4.6 \pm 0.8$	$-8.4 \pm 2.1$	$-3.5 \pm 0.7$	$-13.2 \pm 1.6$	$-2.5 \pm 0.3$	$-20.4 \pm 2.6$
RGO	$-4.3 \pm 0.4$	$-14.7 \pm 1.9$	$-3.3 \pm 0.6$	$-42.1 \pm 2.1$	$-2.4 \pm 0.5$	$-45.3 \pm 1.6$

#### IV. CONCLUSIONS

RGO was prepared successfully with high purity by improved Hummer method followed by chemical reduction. RGO/PE composites was prepared successfully and compared with the corresponding G/PE composites. Instrumentations and

Characterizations using XRD, SEM, TEM, EDX and Raman spectroscopy was carried for G, RGO and their composites confirming the successive preparations.

It was proved that the average RL values and TL values changes with the change in mass loading percentage of G and RGO utilized in PE composite, as we discussed the average RL values was decreased and TL values was increased with the increase in mass loading percentage from 10 % to 30 %. It was found that 10% RGO-PE composite has the highest average RL value (-5 dB) and 30% RGO-PE composite has the highest average TL value (-45 dB). The EMI potential of 30% RGO/PE composite may have a promising EMI shielding applications.

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