

# Comparison of MA-g-PP effectiveness through mechanical performance of functionalised graphene reinforced polypropylene

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## Abstract

This work aims in developing carboxyl functionalised graphene based PP nanocomposites by using melt mixing method to enhance the mechanical and thermal properties. Maleic anhydride grafted polypropylene was used as a compatibilizer to achieve better compatibility between the non-polar polymer and polar nanofiller. FTIR study confirms the presence of functional groups at corresponding absorption levels. TEM and SEM image shows the uniform distribution of COOH-Gr onto the PP matrix with the addition of MA-g-PP onto it. The tensile strength and young's modulus of PMG5 depicts better improvement of 62% and 20% compared to neat sample. The increase in storage modulus of 19.02% was obtained for PG and 43.48% for PMG samples. The reduction in  $\tan \delta$  peak confirms the minimum heat buildup and as a result, leads to better damping characteristics of the nanofiller incorporated PP matrix.

**Keywords:** *functionalised graphene, maleic anhydride grafted polypropylene, polypropylene, scanning electron microscopy, transmission electron microscopy.*

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## 1. Introduction

Polymer nanocomposites are the material wherein nanofillers are reinforced in a polymer matrix. Polypropylene (PP) is a thermoplastic material used in several applications like reusable containers of various types and stationery plastic parts, automotive components, laboratory equipments and polymer banknote<sup>[1]</sup>. PP is a breed of polymer which transforms its phase into liquid when it is heated and when freezing, it turns into a glassy state. Recently researches are being carried out with nanofillers such as exfoliated clay, alumina, silica, graphene, carbon nanofibers, carbon nanotubes, nanocrystalline metals and also nanoscale inorganic fillers or modified fibers<sup>[2]</sup>. Nanofillers enable better dispersal of the fillers into the matrix, outcome as enhancement of properties of base matrix<sup>[3,4]</sup>. Superior interaction of nanofillers with other particles and improvement in properties is because of high surface area of the nanofillers<sup>[1]</sup>. Another effect of the nanofillers is the change in glass transition temperature ( $T_g$ ). Both increase and decrease in  $T_g$  have been noticed depends on the interaction between the polymer and nanofiller. Some of the nanofillers used are carbon black, nano clay, carbon nanotubes, graphite oxide, graphene, copper nanoparticles

etc. Nanoparticles show improvement in properties at very low loadings. Thus it leads to significant reduction in weight for the same strength, performance and properties.

The present report emphasis on the use of graphene, a carbon based nanofiller. A distinct characteristics of carbon is the wide range of forms that it can assume when two or more atoms bond. Thus the carbon has attracted and continued in the direction to attract, a considerable research and development interest from researchers of all parts of the world<sup>[4,5]</sup>. The carbon usage in nanotechnology is quite promising research area and considerable funding was invested by the government to carbon nanotechnology research.

Pure PP without addition of any reinforcements is not suitable for potential applications. Some of the undesirable properties such as stiffness and rigidity may be enhanced by addition of suitable reinforcements. Though PP finds wide range of applications, its strength is not acceptable. The focus lies on enhancing its strength characteristics. Unfortunately, addition of reinforcements may not have superior adhesive

property. This research focuses at increasing the strength and adhesive characteristics of PP in order to extent its usage<sup>[5]</sup>.

PP nanocomposites have been synthesized with a variety of nanocomposite like modified calcium carbonate ( $\text{CaCO}_3$ ), nanoclay, layered silicate and multi walled carbon nanotubes (MWCNT). Derivatives of graphite have also been used as nanofillers. An effort has been made to use COOH functionalised graphene (COOH-Gr) as the nanofiller in turn to enhance the thermal property of the PP nanocomposite. Modification of graphene by adding carboxyl or hydroxyl groups on outer periphery of the graphene particles has found to be increases its interaction with polymer matrix<sup>[6-8]</sup>. Functional groups added may be carboxyl, amine or epoxy groups. COOH-Gr is highly polar and PP is non-polar, mismatch occurs during nanocomposite production. Maleic anhydride grafted polypropylene (MA-g-PP) is the compatibilizer used to further increase the interaction among the fillers and the matrix<sup>[9,10]</sup>. This is possible because of the polar to polar interaction between the compatibilizer and COOH-Gr. Polymer nanocomposite with graphite oxide derivative graphene material as fillers have made known dramatic improvements in its tensile strength, elastic modulus, thermal behavior and electrical conductivity of the nanocomposite. Enhancements at low loadings have been observed due to large interfacial area and high aspect ratio.

This work aims to prepare PP/functionalised graphene nanocomposite and compare the reinforcing result of COOH-Gr on the mechanical and thermal behavior of PP nanocomposite. This study also explains the morphology of prepared nanocomposite and to review the distribution of nanofillers in polymer matrix.

## 2. Materials and methods

### 2.1 Materials

Commercial grade of PP was provided by Reliance Industries Limited, Mumbai, India. MA-g-PP with

8-10 wt.-% maleic anhydride content was obtained from SIGMA-ALDRICH and the chemical formula for the same is  $(\text{C}_3\text{H}_6)_m(\text{C}_7\text{H}_8\text{O}_3)_n$ . COOH-functionalised graphene with carboxyl content of 20% was supplied by United Nanotech Innovations Pvt. Ltd., Bangalore.

### 2.2 Methods

#### 2.2.1 Melt mixing

The preparation of polymeric nanocomposite was carried out using melt mixing method<sup>[9,11]</sup>. The mixing operations were done at 160°C for a time period of 15 minutes. The speed is constantly maintained 50 rpm for throughout the mixing process. Initially the PP granules were feed in between the two rolls and allowed to mix for 5 minutes, Compatibilizer (MA-g-PP) is added to semisolid state PP content and mixed together for 3 more minutes. Functionalized graphene was poured in between the rolls once after the polymer gets completely melted. The whole blend is mixed for further 7 minutes to attain homogeneous mixing.

#### 2.2.2 Mixing formulation

The mixing formulation for preparing the PP nanocomposite is shown in Table 1. "PP" denotes pure polypropylene, "PM" denotes PP with MA-g-PP, "PG" denotes PP with COOH-Gr and "PMG" denotes PP with COOH-Gr and MA-g-PP.

#### 2.2.3 Compression molding

Compression molding is one of the plastic forming process in which a plastic material is placed directly in metal mold and kept in between two hot plates. Polymers softened by the applied heat and pressure to get the shape of the mold. The temperature of 160 °C and pressure of 0.8 MPa are maintained upto the molding polymer gets cured. Tensile and DMA tests specimens were made from compression molding process. The DMA specimens dimension were 6.4 cm length x 1.27 cm width x 0.32 cm thick.

**Table 1.** Samples mixing formulation.

Sl. No.	Sample code	PP (wt. - %)	MA-g-PP (wt.-%)	COOH - Gr (wt.-%)
1.	PP	100	-	-
2.	PM1	97.5	2.5	-
3.	PM2	95	5	-
4.	PM3	92.5	7.5	-
5.	PM4	90	10	-
6.	PM5	87.5	12.5	-
7.	PM6	85	15	-
8.	PG1	100	-	0.5
9.	PG2	100	-	1
10.	PG3	100	-	1.5
11.	PG4	100	-	2
12.	PG5	100	-	2.5
13.	PG6	100	-	3
14.	PMG1	97.5	2.5	0.5
15.	PMG2	95	5	1
16.	PMG3	92.5	7.5	1.5
17.	PMG4	90	10	2
18.	PMG5	87.5	12.5	2.5
19.	PMG6	85	15	3

### 2.2.4 Fourier Transform Infrared Spectroscopy (FTIR)

The Fourier transform infrared spectroscopy (FTIR) spectrum of graphene and modified graphene has been documented using a NEXUS 870 FTIR (Thermo Nicolet) equipment. The samples were prepared by making pallets with KBr for characterization<sup>[12,13]</sup>. The signal resolution was  $2\text{ cm}^{-1}$  and a minimum of 64 scans were within the range of  $4000\text{--}500\text{ cm}^{-1}$ .

### 2.2.5 Transmission electron microscopy (TEM)

Transmission electron microscopy (TEM) imaging was carried out using JEM-2100 (JEOL JEM Co.) electron microscope. Dispersion morphology of composite samples are microtomed as very thin slices of  $80\text{ nm}$  approximately by means of ultra microtome with a diamond knife and the temperature of samples were retained at  $-80\text{ }^{\circ}\text{C}$  using liquid nitrogen.

### 2.2.6 Mechanical properties

Dumb-bell shaped samples were made from compression molding process. The investigations were done under ambient condition with a universal testing machine (Hounsfield H10KS), the specimens were prepared for ASTM D 412-06 and ASTM D 624-00(2007) standards. Tensile strength and Modulus of elasticity were measured at ambient temperature. The primary length of the specimens was  $40\text{ mm}$  and  $50\text{ mm/min}$  speed was maintained for jaw separation.

### 2.2.7 Dynamic Mechanical Analysis (DMA)

The TA Instruments DMA 2980 model instrument is used to characterize the materials mechanical behavior at different temperatures. The dynamic properties of the samples with dimensions  $35\text{ mm}$  length,  $12\text{ mm}$  width and  $3\text{ mm}$  thick was studied at constant frequency mode with  $1\text{ Hz}$  frequency<sup>[14]</sup>. The samples were heated at a rate of  $3^{\circ}\text{C/min}$  to the temperature range of  $30\text{ }^{\circ}\text{C}$  to  $130\text{ }^{\circ}\text{C}$ .

### 2.2.8 Scanning electron microscopy (SEM)

The tensile fractured face of the specimens were imaged with scanning electron microscope (Model: JSM-5800 of JEOL Co., Acceleration voltage:  $20\text{ kV}$ , Coating type: gold) at  $1000$  times zooming<sup>[15]</sup>. The surface morphology of fabricated specimens can also be studied using scanning electron microscopy<sup>[1,5,16]</sup>.

## 3. Results and discussions

### 3.1 FTIR

The existence of functional groups O-H and C-O are confirmed from FTIR (Figure 1) results. This is arrived by the identification of peaks at  $1276$ ,  $1710$  and  $3432\text{ cm}^{-1}$  which match up to C-O stretching, C=O stretching and O-H stretching correspondingly. Hence the sample was confirmed to be COOH modified graphene<sup>[3,13,17]</sup>. The covalent bonds of molecules are like springs that are stiff but stretch and bend. At normal temperatures these bonds vibrate in accordance with their electronic states.

The stretching takes place during the absorption of infrared rays when these bonds experience a transition between the energy levels induced by photons of appropriate energy.

Each functional group has corresponding absorption levels which are identified from the FTIR results.

### 3.2 TEM

TEM image of  $2\text{ wt.-%}$  of functionalised graphene dispersed in PP matrix is revealed in Figure 2. The agglomeration of COOH-Gr in the matrix is observed in Figure 2. However, Figure 3 illustrates the TEM image of polymer matrix with  $2\text{ wt.-%}$  of functionalised graphene and the addition of  $10\text{ wt.-%}$  compatibilizer onto it. It proves the successful exfoliation of single layer graphene which gets uniformly distributed in the polymer matrix<sup>[9,18]</sup>, this is because of the addition of compatibilizer which improved the bonding between matrix and the filler. However, Figure 3 reveals that addition of compatibilizer shows better dispersion of nanoparticles compared to Figure 2.

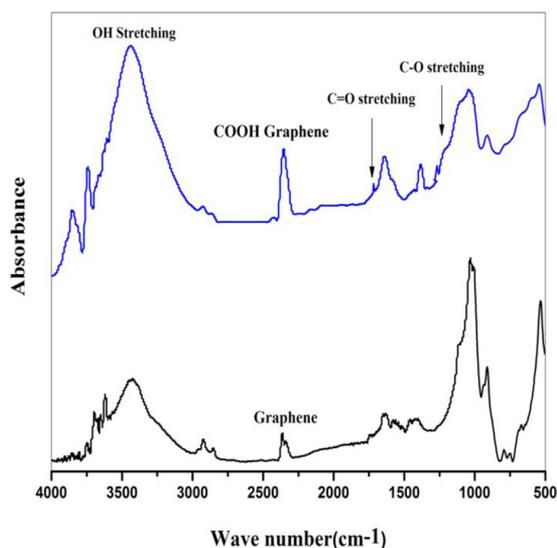
### 3.3 Mechanical properties

Mechanical properties were studied for pure and functionalised graphene filled PP matrix is shown in Figure 4. For the uncompatibilized system (PG1 to PG4) the tensile strength gradually enhances with increase in nanofiller content. The tensile strength values of the samples PG1, PG2, PG3 and PG4 increased by  $18\%$ ,  $22\%$ ,  $36\%$  and  $47\%$  respectively when compared to pure PP.

The maximum increase in tensile strength was achieved for PG4 ( $29.4\text{ MPa}$  for  $2.0\text{ wt.-%}$  of graphene). Further loading of COOH-Gr in the PP matrix (PG5 and PG6) shows drop in tensile strength compared to PG4. The increase in addition of graphene may lead to agglomeration of nanoparticles in the PP matrix<sup>[3,19]</sup>. Hence the tensile strength starts to decrease.

The blend PM1, PM2, PM3, PM4, PM5 and PM6 shows a slow gradual improvement on tensile strength when compared with pure polymer.

The compatibilized system shows considerable enhancement in tensile strength compared to uncompatibilized and pure systems. The percentage enhancement in tensile strength for the samples PMG1, PMG2, PMG3, PMG4 and PMG5 compared



**Figure 1.** FTIR spectrum of graphene and COOH modified graphene.

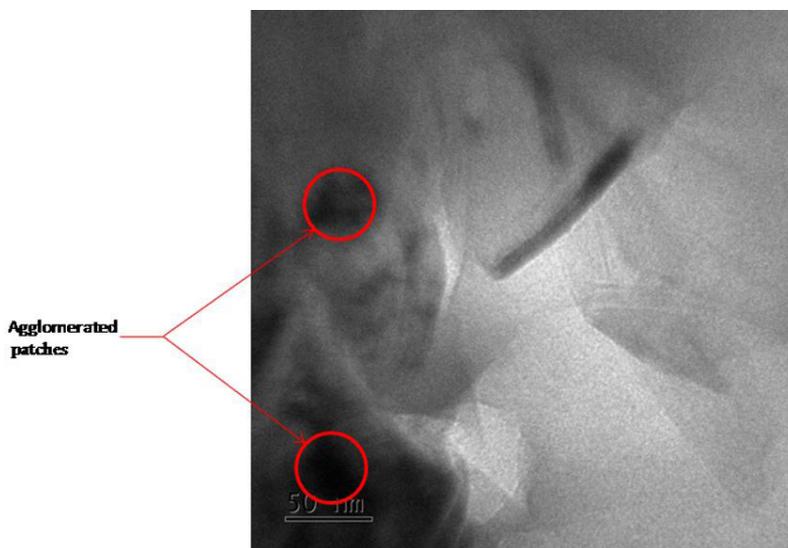


Figure 2. TEM image of PP+2 wt. % of COOH-Gr.

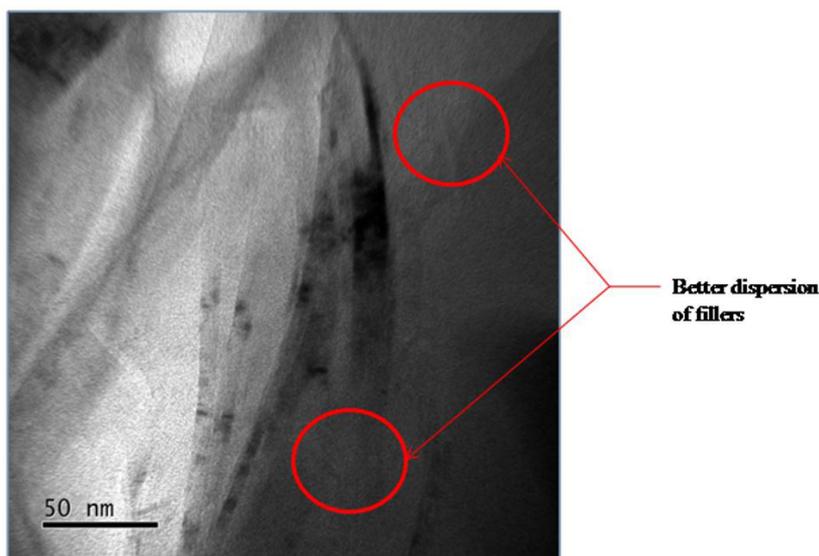


Figure 3. TEM image of compatibilized PP + 2 wt.-% of modified graphene.

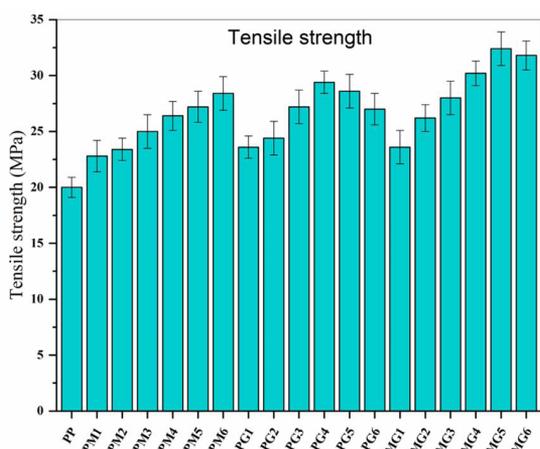


Figure 4. Tensile strength of PP, PG and PMG.

to pure PP are 18%, 31%, 40%, 51% and 62% respectively. The maximum tensile strength of 32.4 MPa was achieved for the sample PMG5. The same shows 62% enhancement in tensile strength match up to pure PP. The presence of compatibilizer enhances the dispersal of graphene in the base PP matrix<sup>[9,10,20]</sup>. However with increase in filler content beyond 5 wt.-% for PMG5, the tensile strength starts to decrease.

For both uncompatibilized and compatibilized system the elastic modulus gradually increases with increase in addition of graphene in the PP matrix. The Young's modulus for blend PM1, PM2, PM3, PM4 and PM5 increases upto 6% compared to pure polymer, later on decreases for PM6. For the uncompatibilized system 5%, 7%, 8%, and 10% increase of modulus was obtained for the samples PG1, PG2, PG3 and PG4 respectively. From Figure 5, on further adding of COOH-Gr (2.5 wt.-% and 3 wt.-%) decreases the elastic modulus of PG5 and PG6. Similarly for the compatibilized system shows 10%, 12%, 13%, 18% and

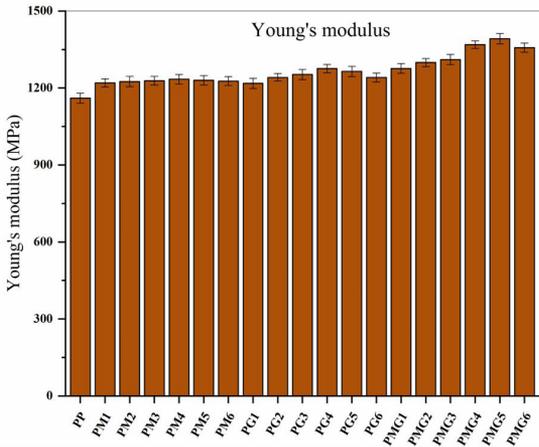


Figure 5. Young's modulus of PP, PG and PMG.

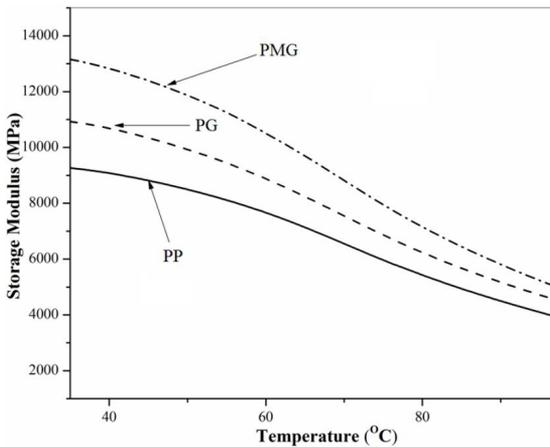


Figure 6. Storage modulus for PP, PG and PMG.

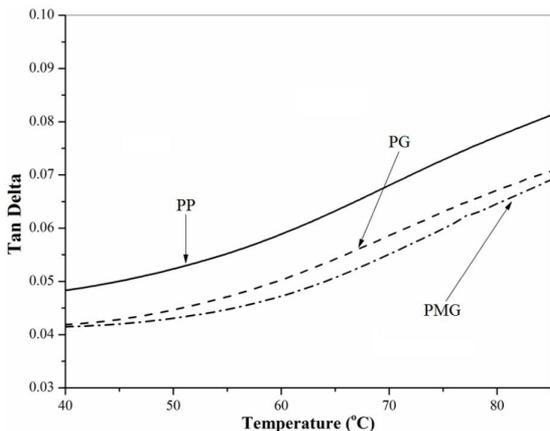


Figure 7. Tan  $\delta$  peak for PP, PG and PMG.

20% increase of modulus for the samples PMG1, PMG2, PMG3, PMG4 and PMG5 respectively. On further addition of COOH-Gr (3 wt.-%) decreases the modulus of PMG6. At last, this overall increase of mechanical properties is due to homogeneity in mixing and better dispersion of functionalised fillers in matrix.

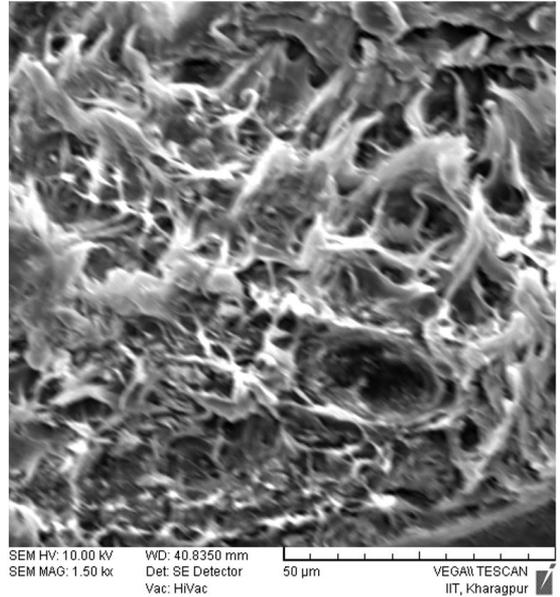


Figure 8. SEM image of fracture surface with 2.5 wt.-% COOH-Gr + MA-g-PP + PP.

### 3.4 DMA studies

Figure 6 characterizes the temperature dependence of storage modulus ( $E'$ ) of nanocomposite. The nanocomposite containing modified graphene showed a development in storage modulus on compared with PP and PG. An increase of 19.02% was observed for PP and graphene nanocomposite compared to pure PP.

Several studies<sup>[21,22]</sup> have confirmed that inclusion of graphene (particles, sheets) can boost the thermal behavior of the polymer to different degree. Some studies<sup>[23,24]</sup> exposed that incorporation of functionalized graphene (particles, sheets) accelerates the thermal degradation of polymer in inert condition. The enhancement in storage modulus is additional evidence for the compatibilized nanocomposites. The compatibilized COOH-Gr system showed an increase of 43.48% in storage modulus compared to PP.

With increase in filler loading the peak intensity of tan  $\delta$  consecutively decreases. This may be due to better reinforcing effectiveness of the filler with the PP matrix. Figure 7 signifies the tan  $\delta$  of PP, PG and PMG. The reduction in tan  $\delta$  peak confirms the minimum heat buildup and as a result, leads to better damping characteristics of the nanofiller incorporated PP matrix.

### 3.5 Morphological study

The polar nature of COOH-Gr will not be compatible upon mixing in bulk non-polar PP matrix. In order to avoid the compatibility mismatch, a polar compatibilizer MA-g-PP derived from base PP is used as a medium to attain effective reinforcement of COOH-Gr in base matrix.

However, PMG5 sample shows better tensile strength and Young's modulus. So, the SEM study of PMG5 sample has also been done to add more proof of the obtained results. SEM image (Figure 8) shows a fibrillated morphological texture, which proves the enhancement in compatibility between polar COOH-Gr and non-polar PP matrix by introducing MA-g-PP as compatibilizer<sup>[18,20]</sup>.

## 4. Conclusion

In summary, PP nanocomposite was prepared by incorporating COOH functionalised graphene to investigate the enrichment of mechanical and thermal stability of PP. FTIR results confirm the presence of functional groups in modified graphene by corresponding absorption levels. TEM image justifies the successful exfoliation of single layer graphene, uniformly distributed in PP matrix. Significant enhancement in the mechanical properties and dynamic mechanical properties was achieved for compatibilized PP nanocomposites compared to uncompatibilized and pure system. The surface morphology shows a fibrillated morphological texture, which proves the enhancement in compatibility between polar COOH-Gr and non-polar PP matrix by introducing MA-g-PP as compatibilizer. As of the results, it confirms that addition of nanofiller reinforcement into the polymer matrix, compatibility match between the fillers and base matrix plays a role in varying the properties of the polymer.

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