

Role of Compatibilizers on the Physicomechanical Performance of Tea Dust Polypropylene Composites

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ABSTRACT: Tea dust (TD) polypropylene (PP) biocomposites and glycidyl methacrylate (GMA)-grafted PP (GMA-g-PP) were manufactured using Brabender (PL-2000, Plasti-Corder, Germany) and hot press machine. The effects of fiber loading and the role of compatibilizer like maleic anhydride-grafted PP (MAH-g-PP) and GMA-g-PP on morphology and physicomechanical properties of the TD-reinforced PP biocomposites were investigated. On preliminary observation, it is found that the tensile strength of the biocomposites increased with increasing fiber content and presence of the compatibilizer at a certain limit. Scanning electron microscopic studies revealed that the dispersion of fiber in the ma-

trix became worse with increasing fiber content but improved with addition of compatibilizer. The improved mechanical properties of the biocomposites correspond to the presence of the compatibilizer, which is attributed to interactions between hydroxyl groups in the filler and epoxy groups in GMA-g-PP and carboxylic group in MAH-g-PP. Swelling properties and biodegradable behavior test of the composites have also been performed. © 2012 Wiley Periodicals, Inc. *J Appl Polym Sci* 000: 000–000, 2012

Key words: biofibers; polypropylene; compatibilization; composites; mechanical properties

INTRODUCTION

The continuous technological progress in the polymer field leads to growing interest in using natural fibers (NFs) as a reinforcement of polymer-based composites. It is mainly because of the advantages such as lower cost, renewability, acceptable specific properties, lower density, ease of preparation, lower energy requirements for processing, biodegradability, wide availability, and relative nonabrasiveness over traditional reinforcing fibers such as glass and carbon. However, some limitations in using NFs in composites are the lower allowable processing temperatures, incompatibility in between the hydrophilic NFs and hydrophobic polymers, and high moisture absorption of the fibers, resulting in the swelling of the manufactured composite.^{1–3}

Thermoplastics used in such composites consist of polyethylene (high and low density), polypropylene (PP), polyvinyl chloride, polystyrene, etc. However, kenaf, jute, sisal, coir, flax, banana, wood flour, rice hulls, newsprint, pulp, and cellulose fibers are the main NFs used as reinforcement.⁴ Most of the Asian

subcontinent countries, especially in Bangladesh, which is the largest delta country in the world, are able to grow tea plant that meet the demand sufficiently as required. The tea manufacturing industries always end up with lots of leftovers waste tea leaves or tea dust (TD) daily. Although a few amounts of waste tea leaves are used for fertilizer, fuel, and others, most are discarded. The tea leaves are rich in poly phenols, which are also referred as to tannins.⁵ Some tannin can actively react with formaldehyde.⁶ Protein and amino acid component of tea leaves can also react with formaldehyde as well.⁷ The plentiful phenolic extractive of tea leaves and their biological properties have long been noticed. Waste tea leaves have been used to prepare waste tealeaves particleboards.⁸ It is also reported that these boards possesses high resistance to decay-type fungi, and phenolic extractives from tealeaves could act as natural toxicants to termites.⁷ On encouragement of these report, we paid our attention on the waste tea leaves dust, i.e., TD to use as organic filler in polymer composites because of its lignocelluloses behaviors and toxicants to termites behavior in composites.

PP has been used in many applications but, in general, its use is limited by its lack of functional groups. Thus, functionalization reactions have been used to increase its interfacial interactions.^{9,10} The grafting process is one of the methods most frequently used to modify PP. Graft polymerization by

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hydrogen abstraction from tertiary carbon offers an effective approach to introducing some desirable properties into the polymer, thus expanding its applications without adversely affecting the backbone architecture.¹¹ Using benzoyl peroxide (BPO) as initiator and glycidyl methacrylate (GMA) as monomer, the copolymerization process occurs via a free-radical mechanism, through the scission of the double bond in the GMA.¹² GMA monomer possesses a dual functionality, containing both epoxy and acrylic groups, providing the design and performance versatility required for coating and resin applications. Besides, the acrylic and vinyl functionality (free-radical reactivity) allows copolymerization with a variety of other vinyl monomers in aqueous and nonaqueous systems. The resulting polymers feature a combination of epoxy functionality with an acrylic backbone. However, the epoxy group (functional reactivity) enables reactions with amines, carboxylic acids, anhydrides, and hydroxyl-containing polymers. It allows structural modification of the polymer backbone that can result in differentiated properties and higher performance. Furthermore, another compatibilizer, i.e., the maleic anhydride-grafted PP (MAH-g-PP) is a well known and widely applied graft copolymer used as a modifier to enhance the compatibility between fibers and PP.^{13–15} With the sufficient fiber source and with proper compounding conditions, the mechanical properties of composites are increased. Therefore, GMA-g-PP and MAH-g-PP have to be used in a wide range of applications as coating promoter, co-monomer in polymers with tailored properties or compatibilization agent for polymer blending and for polymer composite preparation. The aim of the research work is to manufacturer the TDPP biocomposites (TDPP) and to investigate the effect of different compatibilizer like GMA-g-PP and MAH-g-PP on the prepared TDPP biocomposites properties.

EXPERIMENTAL

Material

TD has been collected from Phulbari Tea Estate, Moulvibazar, Bangladesh. PP was used as the matrix. The PP grade (579S, SABIC) has an MFI of 47 g/10 min at 230°C/2.16 kg. Commercial compatibilizer maleic anhydride-grafted PP (MAH-g-PP) with a graft level of 1.4% and a melt flow rate of 75 g/10 min at 190°C, 2.16 kg was purchased from KOMETRA (SCONA TPPP 8112 CMB 6-4).

For preparation of GMA-grafted PP (GMA-g-PP) GMA, styrene (St), and BPO were purchased from Sigma-Aldrich. GMA-g-PP was functionalized in the molten state by free radical grafting. Melt grafting of PP with GMA was performed by charging PP

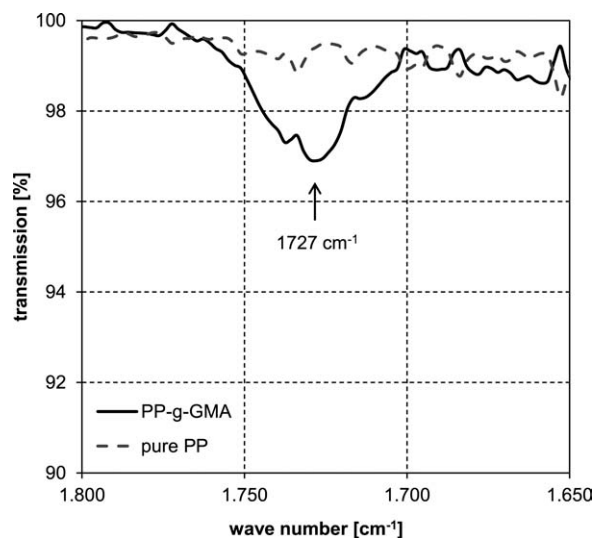


Figure 1 Excerpt of FTIR spectra of PP and GMA-g-PP.

pellets, GMA, and initiator (BPO) into a batch blender (Brabender PL-2000, Plasti-Corder, Brabender GmbH & Co. KG, Germany). The reaction temperature was set constant at 175°C. The grafting was performed for 10 min at a speed of 50 rpm. In the grafting formulation, the added concentration of GMA and styrene was 5 wt % and that of BPO was 1 wt %. After the designated reaction time, the molten grafted polymer was collected from the kneader. Modified PP was then purified by dissolution in hot xylene.

The resulting grafted polymer was dried in a vacuum oven for 24 h at 80°C. Presence of GMA in the purified polymer was proven by FTIR analysis. Figure 1 shows the characteristic peak of the ester bond of GMA at 1727 cm⁻¹. It is suggested that presence of styrene enhances the grafting efficiency of GMA and reduces radical induced β -scission of PP.

Composites preparation

The formulations of composites fabrication with different composition for optimization of filler content are listed in Tables I–III. The composition like PP, TD, MAH-g-PP, and GMA-g-PP were precisely weighed before mixing. Raw materials were well mixed by a Batch Blender at 285 rpm for 20 min. The compounding was performed by melt blending of the composition in a Brabender (PL-2000, Plasti-Corder, Germany) mixer with a rotating speed of 50 rpm at 180°C for 10 min.

The molten samples were transferred into a hot press machine to be pressed under 10 MPa for 5 min at 175°C into sheets of suitable thickness for making the specimens as per ASTM standard. Sheet size and thickness were dependent on the testing methods used in this study. The relative amount (wt %) of reinforcing materials and polymer matrix in composites are listed as mentioned.

TABLE I
Relative Amount (wt %) Filler and Polymer Matrix in Composites to Optimize the Filler (TD) Content in the Composites

Filler (%)	Polymer matrix (%)	Composites
None	PP: 100	PP
TD: 10	PP: 90	TD10
TD: 20	PP: 80	TD20
TD: 30	PP: 70	TD30
TD: 40	PP: 60	TD40
TD: 50	PP: 50	TD50

Mechanical and morphological characterization

Testing bars were conditioned at 23°C, 50% humidity before characterization (ASTM D 618). Tensile properties were tested by use of a Zwick 1446 (Zwick GmbH, Germany) testing machine with a load cell capacity of 10 kN at a testing speed of 50 mm/min (ISO 527). At least five specimens were measured for each sample, and mean values and standard deviations were calculated.

Scanning electron microscopy (SEM) was done on an S-520 (Hitachi, Japan) under 20 kV accelerating voltage to analyze the fracture morphology of the biocomposites. The test specimens were fractured within liquid nitrogen, and the fracture surface was coated with gold before characterization.

Swelling properties and outdoor degradation test

The water absorption capacity of different composites determines the water-swelling behavior of the composites. The different composites were allowed to swell for 2–30 h in water at ambient temperature and in steam (at 100°C) at atmospheric pressure. The swelling of composites was determined by the weight difference of the composite before and after treatment with water and steam.

To assess the degradability of different composites, they were placed on a roof, on ground, and indoors for 120 days. The samples were fixed to a wooden board which was then placed at 45°C facing south on the roof of a two-story building. The

TABLE II
Relative Amount (wt %) of GMA-g-PP Compatibilizer and Polymer Matrix in Composites to Optimize the Amount of GMA-g-PP Content in the Composites

Compatibilizer GMA-g-PP (%)	Filler TD (%)	Polymer matrix PP (%)	Composites
0.5	40	59.5	TDG0.5
1.0	40	59.0	TDG1
1.5	40	58.5	TDG1.5
2	40	58	TDG2
2.5	40	57.5	TDG2.5
3	40	57	TDG3
4	40	56	TDG4

TABLE III
Relative Amount (wt %) of MAH-g-PP Compatibilizer and Polymer Matrix in Composites to Optimize the Amount of MAH-g-PP Content in the Composites

Compatibilizer MAH-g-PP (%)	Filler TD (%)	Polymer matrix PP (%)	Composites
0.5	40	59.5	TDM0.5
1.0	40	59.0	TDM1
1.5	40	58.5	TDM1.5
2	40	58	TDM2
2.5	40	57.5	TDM2.5
3	40	57	TDM3
4	40	56	TDM4

temperature and the humidity during testing period were 20 to 35°C and 50 to 90% rel. H, respectively. The samples were also placed on ground where the temperature and humidity were almost the same as on the roof. The composite samples were also stored indoors as control experiments.

RESULTS AND DISCUSSION

Effect of TD content

The effect of TD content (10, 20, 30, 40, and 50 wt %) on the mechanical properties of the resulting composites has been studied and the variation of tensile strength (TS) and elongation at break (Eb%) and tensile modulus (TM) with TD fiber content in the composite matrix is represented in Figures 2–4, respectively. As the TD content increases, the stress is more evenly distributed and the TS (Fig. 2) of the composites increases up to 40% TD content; after that the composite exhibits a decreased value over the fiber content. At lower levels of TD fiber content, the composites show low TS because of poor fiber

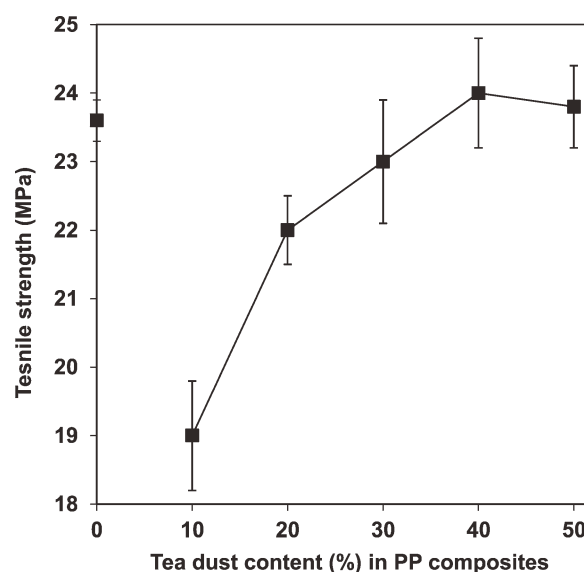


Figure 2 TS (MPa) of the TDPP composites.

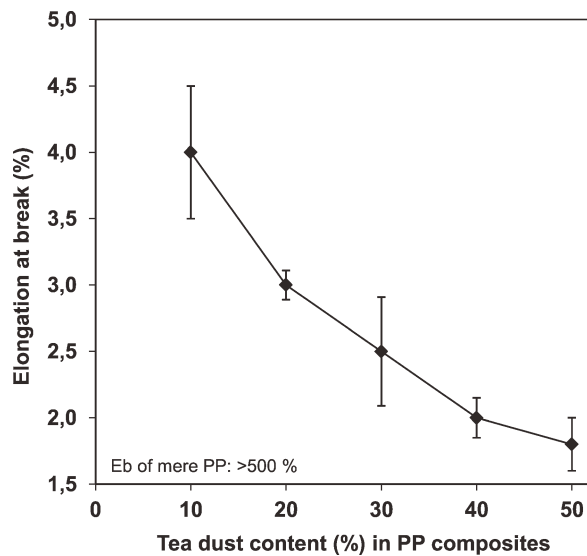


Figure 3 Eb (%) of the TDPP composites.

population and low load transfer capacity to one another. As a result, stress gets accumulated at certain points of composites and highly localized strains occur in the matrix. At intermediate level of TD loading (40%), the population of the fibers is just right for maximum orientation and the fibers actively participate in stress transfer. High levels of TD content showed that the increased population of fibers leads to agglomeration and stress transfer gets blocked and the resulting composite property is again found to decrease.^{16,17} Figures 3 and 4 show that with the increase of the TD content to the polymer composites, the Eb decreases and the TM increases. From the above interpretation, it may be concluded that according to the TS value, the optimized content of TD is 40% with 60% PP polymer matrix, which showed a considerable mechanical

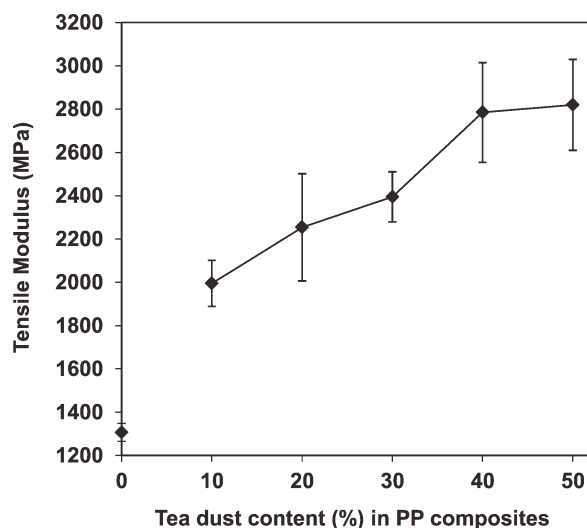


Figure 4 TM of the TDPP composites.

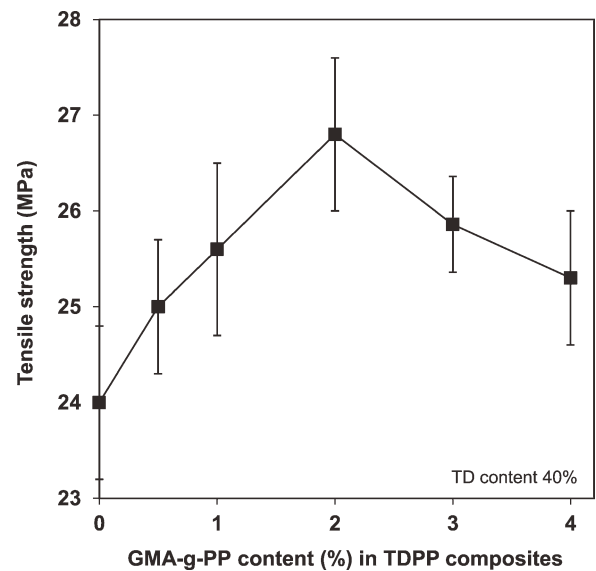


Figure 5 Effect of GMA-g-PP on TS of the TDPP composites.

performance with TS = 24 MPa, Eb = 2%, and TM = 2785 MPa.

Effect of GMA-g-PP as a compatibilizer for TDPP composites

Effect of different percentage (0.5, 1, 2, 3, and 4) of GMA-g-PP compatibilizer was studied using the optimized composite system (40% TD content with PP matrix). The TS and TM values of the composites are represented in the Figures 5 and 6, respectively. It is observed that there is increase in TS (Fig. 5) with increase in compatibilizer dose up to 2% which might be expected as reported earlier.¹⁸ However, a decrease in modulus is noticed in the Figure 5 when

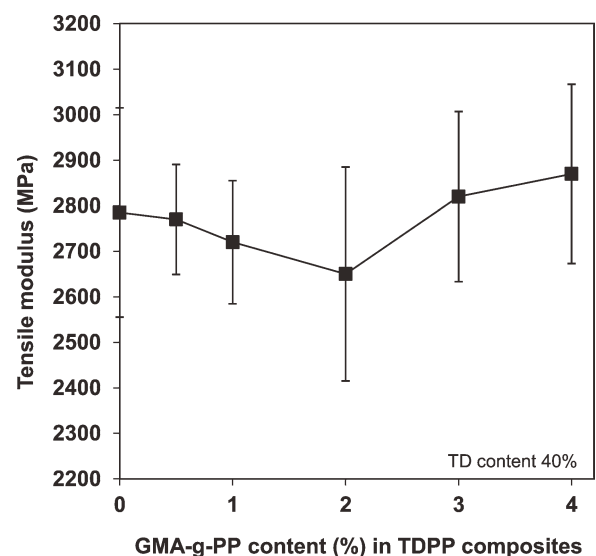


Figure 6 Effect of GMA-g-PP on TM (MPa) of the TDPP composites.

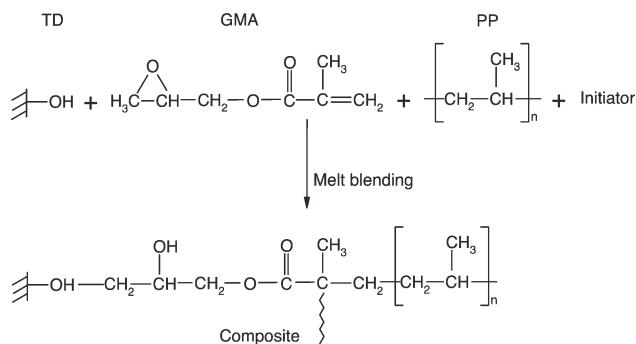


Figure 7 Schematic representation of the reaction among TD, GMA, and PP.

compatibilizer dose increases from 0.5 to 2%. The reason might be attributed to the fact that migration of too much compatibilizers around the fibers causing self-entanglement among compatibilizers rather than the polymer matrix resulting slippage.¹⁸ The effect of GMA-g-PP as compatibilizer on TDPP composites may be attributed to the presence of epoxy functions for better bonding with the TD and its homogeneity with the PP matrix. From the observation (Figs. 5 and 6), TDG2 composite sample which prepared with 2% GMA-g-PP shows remarkable tensile behavior. Figure 7 represents the probable reaction among hydroxyl group of TD and epoxy group of GMA-g-PP with polymer matrix.^{19–25} Therefore, significant improvement in mechanical properties of the GMA-g-PP-treated TDPP composites is supposed as a result of compatibilization, which is represented in the Figure 7.

Effect of MAH-g-PP compatibilizer on TDPP composites

Different percentage of MAH-g-PP (0.5, 1, 2, 3, and 4) as compatibilizer was added to study its influence

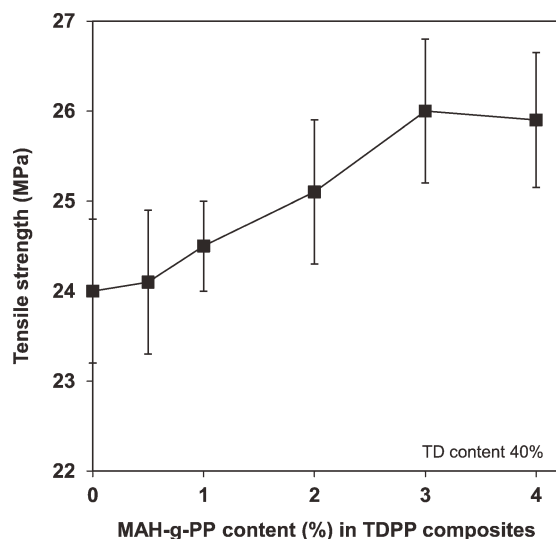


Figure 8 Effect of MAH-g-PP on TS of TDPP composites.

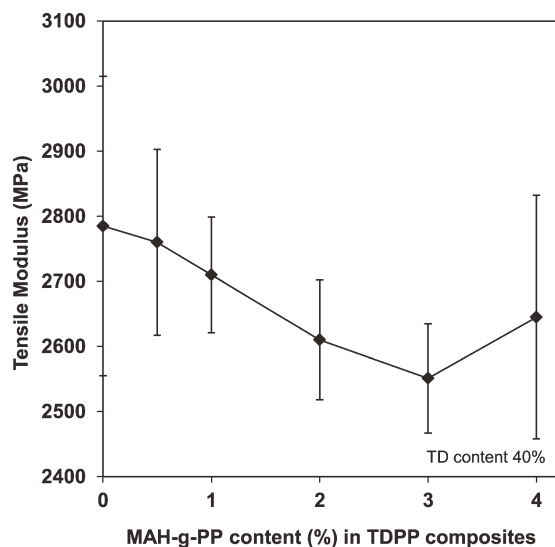


Figure 9 Effect of MAH-g-PP on TM (MPa) of TDPP composites.

on TDPP composites in the optimum system. Figures 8 and 9 show the effect of MAH-g-PP on TS and TM of the composites, respectively. For the TDPP composites containing 40% TD, the TS increase gradually with an increase in the compatibilizer MAH-g-PP from 0.5 to 3%. However, in this case, maximum TS have produced at 3% MAH-g-PP, and the TS value up to 4% MAH-g-PP remains nearly the same. The TM of the MAH-g-PP-treated composites can be observed in the Figure 9, which shows the similar trend as the Figure 5 for the effect of GMA-g-PP on TDPP composite. This result support that MAH-g-PP produces good adhesion between TD and polymer matrix, which indicate that there is some kind of interfacial contact between TD and PP in the presence of MAH-g-PP. So far, the role of MAH-g-PP as compatibilizer in NFs composites has also been reported in recent years^{13–15,26–28} that corroborate the present observation.

Swelling properties of the composites

Figure 10 shows the results of absorption of steam (solid line) and water (broken line) in the different composites, e.g., without compatibilizer (TD40) and with compatibilizer (TDM3 and TDG2). The absorption of water by different composites is largely dependent on the availability of free ---OH groups on the surface of the reinforcing fiber. The difference in absorption of water among the without and with compatibilizer-treated composites are due to blocking of ---OH in which the detailed mechanism has already been established. Absorption of water is much less in comparison with absorption of steam, because water cannot penetrate the resin matrix as much as steam can. The polymer matrix PP is

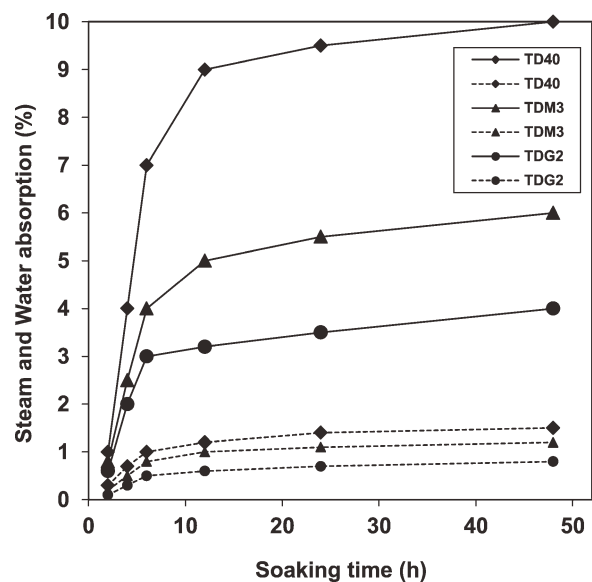


Figure 10 Steam (solid line) and water (broken line) absorption of the composites.

hydrophobic in nature and the fibers, containing —OH groups, are prone to moisture absorption by formation of hydrogen bonding between water molecules and cellulose. By the virtue of the physico-chemical interaction of the fiber and polymer matrix during compression and well fabrication of the composites, the fibers become nonpolar in nature, form the bond with polymer matrix, and result in the reduction in penetration of steam/water into the cell wall. The hydrophilic NFs have normally low compatibility with the hydrophobic polymer. Influence of compatibilizer in the composites leads to improvement in the compatibility between polymer matrix and fiber that draws them nearer to each



Figure 11 SEM micrograph of TD.

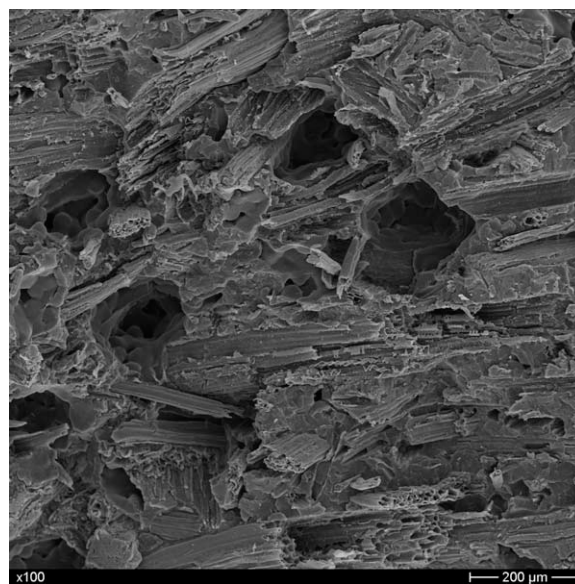


Figure 12 SEM micrograph of the TD40 composite.

other, and, because of that, the penetration of steam and/or water becomes somewhat restricted.^{29,30}

SEM analysis of the composites

Examination of the fracture surfaces of the composites by SEM gave information about how a compatibilizer affects the interphase of the composites. Scanning electron micrographs for tea dust (TD) and different composite samples are shown in Figures 11–14, which represent the microstructure of TD and the composites TD40, TDM3, and TDG2, respectively. The effect of compatibilizer on the microstructure of TDM3 and TDG2 composites is clearly seen in the SEM photographs. In Figure 12 of the composite

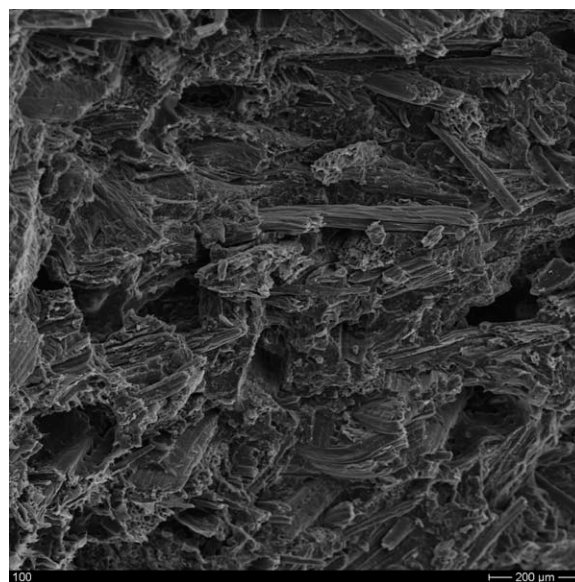


Figure 13 SEM micrograph of the TDM3 composite.

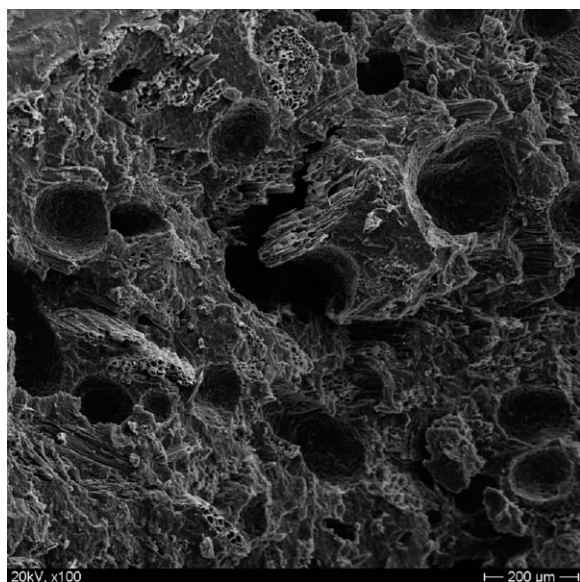


Figure 14 SEM micrograph of the TDG2 composite.

TD40, there are some void spaces around the TD fibers and some spaces where TD fibers have been pulled out. These may be due to the poor adhesion between TD fibers and polymers matrix in the absence of a compatibilizer. However, relatively less void spaces and a lower proportion of pulled out TD fibers are found in Figures 13 and 14, which represent the microstructure of the composites prepared by using the compatibilizers. These indicate that there is some kind of interfacial contact between TD fibers and PP in the presence of MAH-g-PP and GMA-g-PP. It means that the compatibilizer produces good adhesion between TD fiber and polymer matrix.

Outdoor degradation of the composites

To perform outdoor degradation of the different composites, samples were placed on roof and ground. The TSs of the samples were measured periodically and the results of the loss of TS are tabulated in the Table IV. The loss of TS is found to

increase with increase of storage time on roof.³¹ Without compatibilizer, the composite (TD40) was found to lose its strength faster than compatibilizer-treated composites (TDG2 and TDM3). TDG2 composite shows the lowest degradation.

In the case degradation during ground storage, maximum loss of TS for TD40 composite is 42%, whereas that for TDM3 and TDG2 is about 20% and 14%, respectively. Composites samples were placed indoor at 25°C and 30% relative humidity to study the indoor degradation. In that case, TD40 showed about 20% TS loss after 120 d and TS loss of the compatibilizer-treated composites TDM3 and TDG2 is about 10% and 8%, respectively at 120 days.

CONCLUSION

The results of this study showed that melt blending of the composition in a kneader and hot press molding process could be used to manufacture TDPP composites. The study demonstrates that the prepared composite with 40% content of TD exhibits best mechanical properties within the tested mixtures. A series of TDPP composites have been prepared using different amount of (0.5–4%) GMA-g-PP and (0.5–4%) MAH-g-PP as compatibilizers (whereas purchased MAH-g-PP is based on a different PP). In this work, optimum compatibilization and therefore best mechanical properties within the mixtures with 40 wt % TD were observed for 2% GMA-g-PP (composite TDG2). Compatibilization with GMA-g-PP showed 12% enhancement in TS and a minimum in absorption of steam and water at ambient temperature. Furthermore, the TDG2 composite showed the lowest level of degradation in comparison with MAH-grafted samples and the composite without compatibilizer. This implies better compatibilization properties for GMA-g-PP, which might be explained by a higher graft level. The phenomenon of decreased outdoor degradation might be due to better physical bonding of PP and TD. Therefore, TD may be washed out slower. Nevertheless, the mechanism of the compatibilizing agent in the TD/PP composite is not fully understood. Hence

TABLE IV
Loss of TS (%TS) of the Composites during Outdoor Degradation

Degradation time (day)	On roof			On ground			Indoor		
	Loss of TS (%)			Loss of TS (%)			Loss of TS (%)		
	TD40	TDM3	TDG2	TD40	TDM3	TDG2	TD40	TDM3	TDG2
10	2 ± 0.8	1.2 ± 0.7	1 ± 0.5	5 ± 0.9	2 ± 0.8	1.5 ± 0.6	1.5 ± 0.4	0.5 ± 0.3	0.3 ± 0.2
20	5 ± 0.8	2.5 ± 0.7	2 ± 0.5	9 ± 0.9	3.5 ± 0.8	3 ± 0.6	3 ± 0.4	1 ± 0.3	0.8 ± 0.2
60	10 ± 0.8	4 ± 0.7	3 ± 0.5	14 ± 0.9	6 ± 0.8	4 ± 0.6	7 ± 0.4	3 ± 0.3	2 ± 0.2
80	17 ± 0.7	6 ± 0.6	5 ± 0.4	21 ± 0.8	10 ± 0.7	6 ± 0.5	10 ± 0.3	5 ± 0.2	3 ± 0.09
100	25 ± 0.7	9 ± 0.5	7 ± 0.3	30 ± 0.8	15 ± 0.7	8 ± 0.5	14 ± 0.3	7 ± 0.2	5 ± 0.09
120	35 ± 0.7	15 ± 0.5	10 ± 0.3	42 ± 0.8	20 ± 0.7	14 ± 0.5	20 ± 0.3	10 ± 0.2	8 ± 0.08

additional research demands the mechanism of the activities of the compatibilizing agent in the TDPP composite fabrication.

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