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유리섬유 직물로 보강된 폴리프로필렌 복합체의 기계적 물성에 대한 가공 변수의 영향

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Effects of Process Parameters on the Mechanical Properties of Glass Fabric Reinforced Polypropylene Composites

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Abstract: The present study investigates the effects of process parameters such as fabric structure, fabric orientation and compatibilizer concentration on the mechanical properties of fabric reinforced polypropylene composites. Two types of glass fabric reinforced composites with two different stacking sequences were prepared by compressing molding. The composites were prepared with and without compatibilizer for the purpose of analyzing interfacial-bonding. The composites were investigated in terms of tensile, flexural and impact strength for evaluation of the effects of the process parameters. The factorial approach is used to design the experimental layout. The test results revealed that Type-2 fabric reinforced composite in $[0\text{-}90]_4$ orientation with 8 wt% compatibilizer possesses better mechanical properties. Tensile, flexural and impact strength were increased by 106%, 235% and 100%, respectively. Analysis of variance was used to identify the most significant factors which would affect the performance of composites. Morphological study explored the presence of strong interfacial-bonding between the materials.

Keywords: polypropylene, glass fabric, mechanical properties, morphological analysis, factorial design.

Introduction

Nowadays, advanced textile reinforced composites are broadly used in structural and automotive industries for load bearing applications in view of their good properties such as high specific strength and modulus, good dimensional stability, repair-ability, corrosion resistance and cost effectiveness.[1,2] Textile reinforced thermoplastic composites with these excellent features act as competitive materials for metals, alloys and thermoset counterparts.[2] The higher robustness feature of thermoplastic composites make them suitable for crash applications more than thermoset composites. However, improper fibre orientations,[3] poor wettability of matrix over fibre,[4-6] and inappropriate processing techniques are the vital aspects that control the usage and performance of thermoplastic composite materials. In order to motivate the extensive usage of thermoplastic composites for engineering applications, it is important to broaden the understanding of their mechanical behaviour. In-depth and extensive knowledge of processing and optimization of product performance has been developed for short/long fibre reinforced thermoplastic composites. But still the situation for continuous and woven fabric composites has to improve.

Okereke[3] studied the flexural response of glass fibre (GF) reinforced polypropylene (PP) composites with different ply orientations and noticed the influence of the plastic deformation of the matrix on the flexural behaviour of the composites. Ravikumar *et al.*[4] investigated the effect of maleated PP on the mechanical properties of corn fiber/PP composites using the Taguchi technique and found considerable improvement in the properties of composites. Greater improvement in flexural, impact and tensile strength was noticed by Liu *et al.*,[5] when maleic anhydride grafted polypropylene was added to the PP matrix. Sorrentino *et al.*[6] achieved mechanical per-

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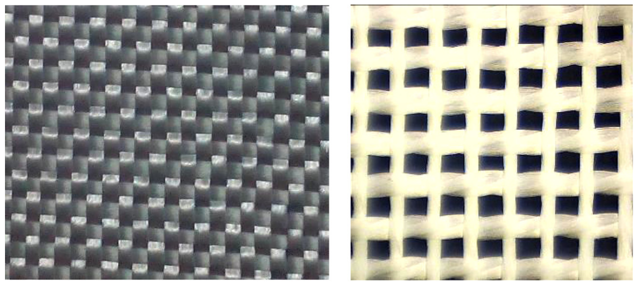
formance optimization through interfacial strength by adding maleated PP to the GF/PP composites.

The effects of process parameters on the mechanical properties of kenaf/PP composites were analyzed by Sallih *et al.*[7] using the factorial approach. The optimization of process parameters for improving the flexural property of GF/PP composites was carried out using the Box-Behnken design.[8] Vamshi Krishna *et al.*[9] did evaluation of the mechanical properties of short GF/PP composites and observed the increase in the strength of the composite caused by an increase in fibre length. Lee *et al.*[10] investigated the effect of coupling agent on the flexural, tensile and impact properties of silica reinforced polypropylene composites and reported that the PP composite mixed with 5 wt% silane prepared using the dry method exhibited the highest mechanical properties. The morphological and mechanical properties of natural fibre reinforced[11-15] as well as synthetic fibre reinforced[16-19] PP composites were studied by the researchers. The mechanical properties of GF/PP composites under different fibre orientations[20] and various surface treatment methods[21,22] were examined and enhancement in the properties of composites was observed. Despite numerous publications on short/long fibre reinforced PP composites, reinforcement of woven fabric in PP composites has seen only a limited application.

In this study, the effects of process parameters on the tensile, flexural and impact properties of plain woven glass fibre reinforced thermoplastic (GFRTP) composites, were investigated. The optimum process parameters were obtained using the full factorial design of experiments (DOE) approach, to fabricate the thermoplastic composites with better performance characteristics. Analysis of variance (ANOVA) was used for identifying the most significant factors which would affect the performance of composites. Using a scanning electron microscope (SEM), the morphological study was conducted on the samples before and after the tensile testing, for the analysis of the interaction between the matrix and reinforcement.

Experimental

Materials. An isotactic film PP (0.5 mm) was chosen as the matrix material. Two types of E-glass fabrics (280 g/m^2) such as glass fibre cloth (Type-1) supplied by National traders, Chennai and glass fabric with holes (Type-2) supplied by Pyrotek India Pvt. Ltd., Chennai, as shown in Figure 1, were selected as the reinforcement material. Type-2 glass fabric was chosen in view of its better structural arrangement than type-1 glass fabric. The (3-aminopropyl) trimethoxy silane was selected as a silane coupling agent and was used for the glass fibre surface treatment. In order to improve the bonding between the silane treated hydrophilic GF surface and the hydrophobic PP matrix, the maleic anhydride grafted PP (MAPP) was used as a matrix compatibilizer. The properties of raw materials[23,24] are listed in Table 1.



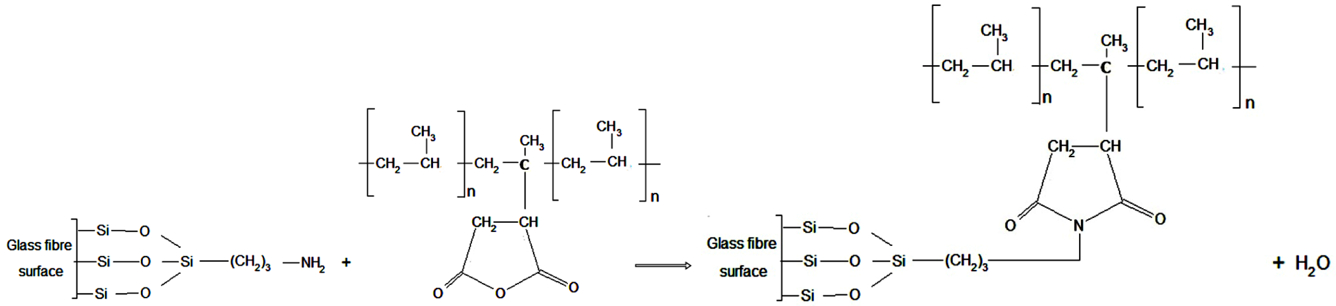
(a) (b)

Figure 1. Glass fabric structure: (a) Type-1; (b) Type-2.

Table 1. Properties of Raw Materials

Raw materials	Weight fraction (wt%)	Density (g/cm^3)	Tensile strength (MPa)	Tensile modulus (GPa)
Polypropylene	60	0.905	33	1.34
E-Glass fibre	40	2.55	2400	73

Hot Compression Moulding. In this study, the GFRTP composite laminates were prepared in a hot press through compression molding using the film stacking technique.[25] PP film and 4 layers of amino-silane treated glass woven fabric were stacked, one over the other for 2.2 mm thickness and the MAPP powder was distributed uniformly in between them in all the layers. The weight fraction of GF (W_f) was 40 wt% and that of matrix and compatibilizer (W_m) was 60 wt%. The stacked materials (300×300 mm size) were placed in a hot press and then heated above the melting temperature of the matrix. The forming pressure and forming temperature used for the laminates were 90 bar and 190 ºC respectively. At the forming temperature, the molten MAPP copolymer was physically absorbed into the PP matrix and chemically attached to the organo-functional amine group of silane treated GF surface as in equation 1. The bridged oxygen in the maleic anhydride condensed with the hydrogen in the amine group of silane treated glass fibre and released the water molecule. This chemical reaction subsequently developed the strong interfacial covalent (carbon-nitrogen) bonding between the constituent materials. The GFRTP composite laminate was removed from



Equation 1.

the hot press, after the room temperature cooling.

Factorial Design. Factorial design is a systematic method for determining the optimum parameters that would affect the performance of the materials. In order to analyze three factors which are studied at two levels, a total of $2^3 = 8$ runs are required. Therefore, a full factorial experimental design was considered for the investigation of the effects of process parameters such as fabric structure, fabric orientation and compatibilizer concentration on the mechanical properties of GFRTP composites. The process parameters and their levels are presented in Table 2. The GFRTP composite laminates were prepared in the hot press according to the full factorial design as in Table 3.

Table 2. Process Parameters and Their Levels

Process parameters	Level 1	Level 2
Fabric structure	Type-1	Type-2
Fabric orientation	$(0\text{-}90°)_4$[a]	$(0\text{-}90°/\pm45°)_s$[b]
Compatibilizer concentration (wt%)	0	8

[a]4-4 layers of (0-90°) glass fabric. [b]s-symmetric layers of glass fabric.

Table 3. Experimental Layout Using Full Factorial Design

Exp. runs	Sample Id	Fabric structure	Fabric orientation	Compatibilizer concentration (wt%)
1	GFRTP-1	Type-1	$(0\text{-}90°)_4$	0
2	GFRTP-2	Type-1	$(0\text{-}90°)_4$	8
3	GFRTP-3	Type-1	$(0\text{-}90°/\pm45°)_s$	0
4	GFRTP-4	Type-1	$(0\text{-}90°/\pm45°)_s$	8
5	GFRTP-5	Type-2	$(0\text{-}90°)_4$	0
6	GFRTP-6	Type-2	$(0\text{-}90°)_4$	8
7	GFRTP-7	Type-2	$(0\text{-}90°/\pm45°)_s$	0
8	GFRTP-8	Type-2	$(0\text{-}90°/\pm45°)_s$	8

Characterization Testing. The composite laminates thus prepared were subjected to mechanical characterization testing, and each test was repeated thrice to minimize the errors. The tensile test was conducted as per ASTM D 638 Type I standard with the specimen size of 165 mm length, 19 mm width, and 2.2 mm thick. The test was performed to assess the tensile behavior of composite materials under uniaxial loading condition and was carried out at room temperature with a crosshead velocity of 2 mm/min using 50 kN universal testing machine (UTM). During the tests, an electronic load cell was used for measuring the load and a linear variable differential transducer was used for measuring the displacement.

The flexural test was conducted as per ASTM D790-03 standard with the specimen size of 127 mm length, 12.7 mm width, and 2.2 mm thick. The test was performed to get an understanding of the bending behavior of composite materials under three-point bending condition and was carried out at room temperature with a crosshead velocity of 2 mm/min using 50 kN UTM. Flexural strength was calculated from the peak load of the specimen.[3,8]

Impact test was conducted as per ASTM D256 standard with the specimen size of 64 mm length, 12.7 mm width, and 2.2 mm thick. The Charpy "V" notch impact test was performed using an impact testing machine to find the impact behavior of composite materials.

The measured tensile properties, flexural properties and impact strength are presented in Table 4. The design of experiment software MINITAB-17 is used to analyze the characterization testing data.

Results and Discussion

Optimum Level of Parameters. The average strength (tensile, flexural and impact) values of each levels of the factors were calculated using the facts presented in Table 4 and are

Table 4. Mechanical Properties of GFRTP Composites

Sample Id	Tensile properties			Flexural properties		Impact strength (J)
	Maximum load (kN)	Tensile strength (MPa)	Tensile modulus (GPa)	Maximum load (N)	Flexural strength (MPa)	
GFRTP-1	2.42	94	1.12	35	43	2.37
GFRTP-2	2.79	102	1.01	62	77	3.37
GFRTP-3	1.98	68	0.85	30	37	2.12
GFRTP-4	1.80	75	0.89	55	68	3.01
GFRTP-5	3.01	116	1.36	30	37	3.37
GFRTP-6	3.83	140	1.50	100	124	4.23
GFRTP-7	2.12	81	0.99	40	50	3.03
GFRTP-8	2.33	89	1.20	60	74	3.85

presented in Tables 5-7. The optimum level of factors as well as the ranking of the factors are presented in Tables 5-7. The response tables (Tables 5-7) helped in the identification of the optimum performance characteristics through the process parameters with the highest tensile strength, flexural strength and impact strength.

The main effects plots of tensile, flexural and impact strength are illustrated in Figure 2(a-c), respectively. Based on the highest strength values, the optimum level of factors was found as (fabric structure)$_2$ (fabric orientation)$_1$ and (compatibilizer concentration)$_2$, i.e., Type-2 fabric structure, [0-90]$_4$ fabric orientation, and 8 wt% compatibilizer concentration for all the test conditions.

Table 5. Response Table for Tensile Strength

Level	Fabric structure	Fabric orientation	Compatibilizer concentration
Level 1	84.75	**113.00**	89.75
Level 2	**106.5**	78.25	**101.5**
Delta	21.75	34.75	11.75
Rank	2	1	3

Table 6. Response Table for Flexural Strength

Level	Fabric structure	Fabric orientation	Compatibilizer concentration
Level 1	56.25	**70.25**	41.75
Level 2	**71.25**	57.25	**85.75**
Delta	15	13	44
Rank	2	3	1

Table 7. Response Table for Impact Strength

Level	Fabric structure	Fabric orientation	Compatibilizer concentration
Level 1	2.72	**3.34**	2.72
Level 2	**3.62**	3.00	**3.62**
Delta	0.90	0.34	0.90
Rank	1	3	2

Analysis of Variance. The purpose of ANOVA is to identify the most significant factors which would affect the performance characteristics.[26] The F-test was carried out for understanding the significance of each process parameters. The factor with high F-value has a great significance in the effect of the response of the process. The effects of process parameters such as fabric structure, fabric orientation, and compatibilizer concentration on the tensile strength, flexural strength and impact strength were analyzed using MINITAB-17 software. The ANOVA details for tensile strength, flexural strength and impact strength are presented in Tables 8-10.

ANOVA was conducted at 5% significance level for a study of the contribution of the parameters. A P-values for each independent parameter in the model are shown in the ANOVA tables (Tables 8-10). When the P-value is less than 0.05, the parameter could be considered statistically highly significant. The percentage contribution of each parameter to the total variation is highlighted in Tables 8-10 indicating the degree of influence of each parameter on the mechanical properties. The analysis revealed the fabric orientation (62.34%) as a dominant parameter for tensile strength, followed by fabric structure (24.42%) and compatibilizer concentration (7.14%). However, the compatibilizer concentration (64.74%) is a dominant parameter for flexural strength, followed by fabric structure (7.53%) and fabric orientation (5.65%). On the other hand, fabric structure (47.17%) is a dominant parameter for the impact strength, followed by compatibilizer concentration

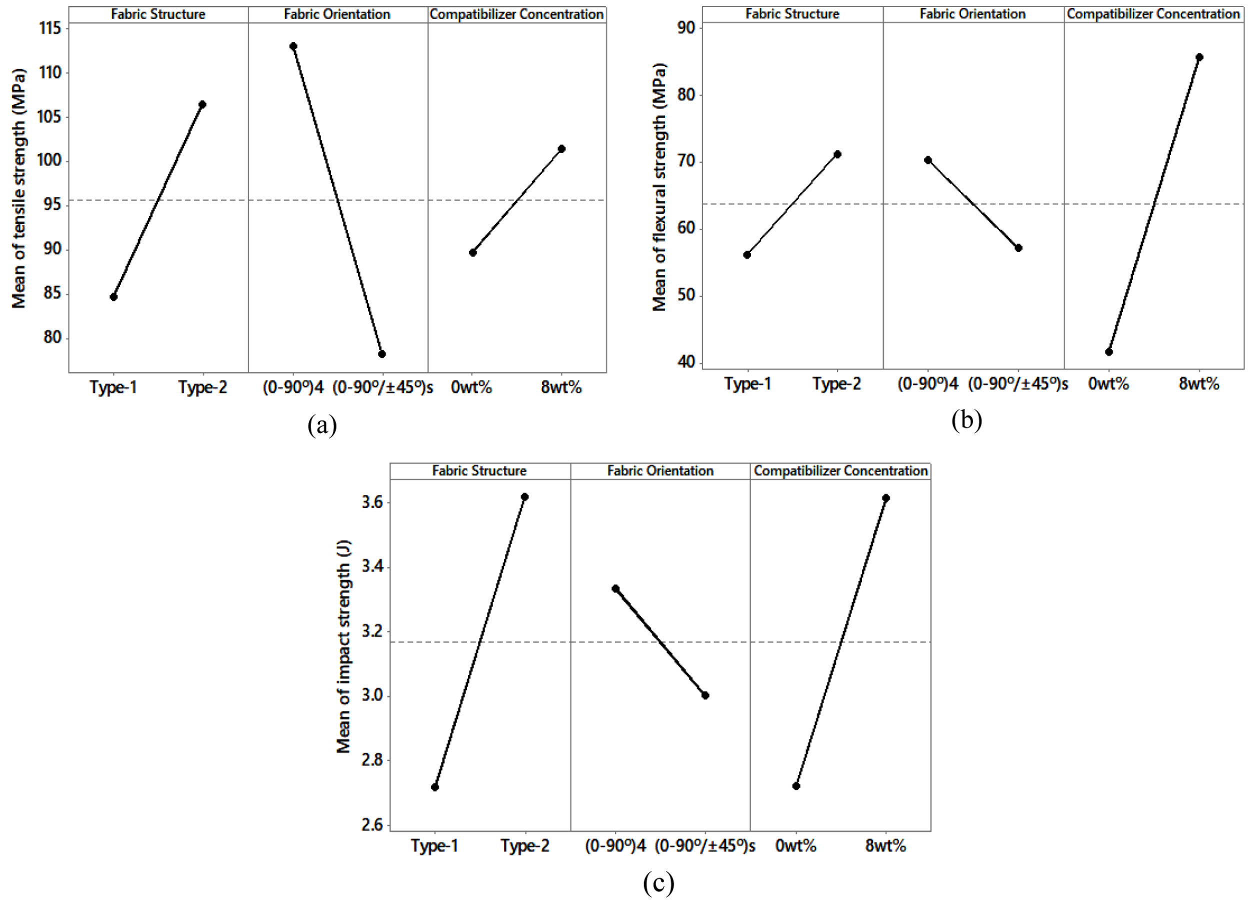


Figure 2. Main effects plots for (a) tensile strength; (b) flexural strength; (c) impact strength.

Table 8. Analysis of Variance for Tensile Strength

Process parameter	Degrees of freedom	Sum of squares	Mean sum of squares	F-ratio	P-value	Contribution (%)
Fabric structure	1	946.1	946.13	16.00	0.016	24.42
Fabric orientation	1	2415.1	2415.12	40.85	0.003	62.34
Compatibilizer concentration	1	276.1	276.13	4.67	0.097	7.14
Error	4	236.5	59.13			6.10
Total	7	3873.9				100.00

Table 9. Analysis of Variance for Flexural Strength

Process parameter	Degrees of freedom	Sum of squares	Mean sum of squares	F-ratio	P-value	Contribution (%)
Fabric structure	1	450.0	450.0	1.36	0.308	7.53
Fabric orientation	1	338.0	338.0	1.02	0.369	5.65
Compatibilizer concentration	1	3872.0	3872.0	11.74	0.027	64.74
Error	4	1319.5	329.9			22.08
Total	7	5979.5				100.00

Table 10. Analysis of Variance for Impact Strength

Process parameter	Degrees of freedom	Sum of squares	Mean sum of squares	F-ratio	P-value	Contribution (%)
Fabric structure	1	1.6290	1.6290	623.55	0.000	47.17
Fabric orientation	1	0.2211	0.2211	84.64	0.001	6.40
Compatibilizer concentration	1	1.5931	1.5931	609.80	0.000	46.13
Error	4	0.0105	0.0026			0.30
Total	7	3.4537				100.00

(46.13%) and fabric orientation (6.40%).

Effects of Process Parameters on Tensile Strength. Details of the tensile strength and modulus of GFRTP composites are presented in Figure 3, which reveals both the tensile values as higher for GFRTP-5 and GFRTP-6 composites (i.e.) Type-2 glass fabric with $[0\text{-}90]_4$ orientation composites. Compared to Type-1 glass fabric composites, the interfacial bonding between the constituent materials in Type-2 glass fabric composites was very strong due to the presence of a gap in the fabric. The flow of molten PP along with MAPP, through the glass fabric gaps contributed to a substantial improvement to the effectiveness of *in-situ* impregnation, which in turn leads to the enhancement in the tensile properties of Type-2 glass fabric reinforced composites.

A significant reduction was seen in the load carrying capacity of the composite, when the glass fabrics were placed at an angular orientation of ±45° with respect to the direction of the weft. The composite laminates with 8 wt% MAPP concentration exhibited a higher tensile strength value than the composite laminates without MAPP concentration. This enhanced tensile property could be attributed to the improvement in the interfacial adhesion between the silane treated glass fabric and the polypropylene due to compatibilizer interactions.

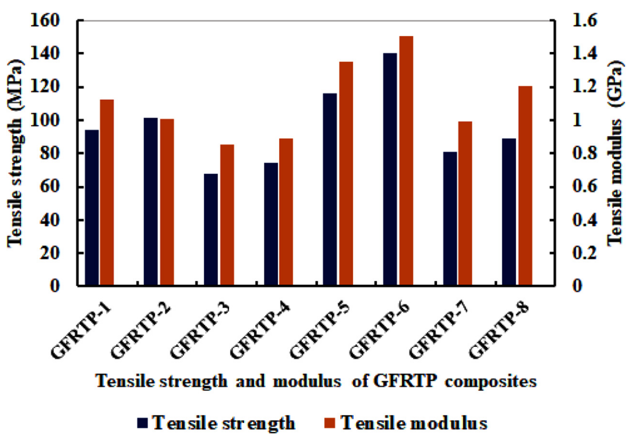


Figure 3. Tensile strength and modulus of GFRTP composites.

There was an increase in the tensile strength of GFRTP composites from 89 to 140 MPa, when the fabric orientation changed from $[0\text{-}90/\pm45°]_s$ to $[0\text{-}90]_4$ at an optimum fabric structure (Type-2) and optimum compatibilizer concentration (8 wt%) levels. This change in fabric orientation resulted in a 57.3% improvement in the tensile strength of GFRTP composites. Likewise, changes in fabric structure and compatibilizer concentration at an optimum parameter levels, triggered an improvement in the tensile strength of composites by 37.3% and 20.7%, respectively. Compared to the other two process parameters, the change in fabric orientation largely influenced the tensile performance of the composites. The conclusion was that the enhancement in tensile properties was achieved by orienting the woven fabrics along the weft direction of the composites.

Figure 3 helps understanding the variations from 68 to 140 MPa in the tensile strength of GFRTP composites and the difference in modulus from 0.85 to 1.50 GPa. The tensile strength and tensile modulus of GFRTP-6 composite were increased by 106% and 76.5% respectively, compared to GFRTP-3 composite which gave lower tensile values. The results show the highly significant effect of the process parameters on the tensile properties of the composites.

Effects of Process Parameters on Flexural Strength. The flexural properties of GFRTP composites reported in Table 4 lead to the understanding that the flexural strength increased from 37 to 124 MPa, when the compatibilizer concentration varied from 0 to 8 wt% at an optimum fabric structure (Type-2) and fabric orientation ($[0\text{-}90]_4$) levels. This change in compatibilizer concentration resulted in a 235.14% improvement in the flexural strength of the composite. This is due to the reaction of MAPP with amine group of silane treated GF and the development of a strong covalent bonding, which would provide a better stress transfer between the constituent materials through the interface. Hence, the presence of

MAPP compatibilizer had clearly a positive effect on both flexural load and flexural strength of GFRTP composites.

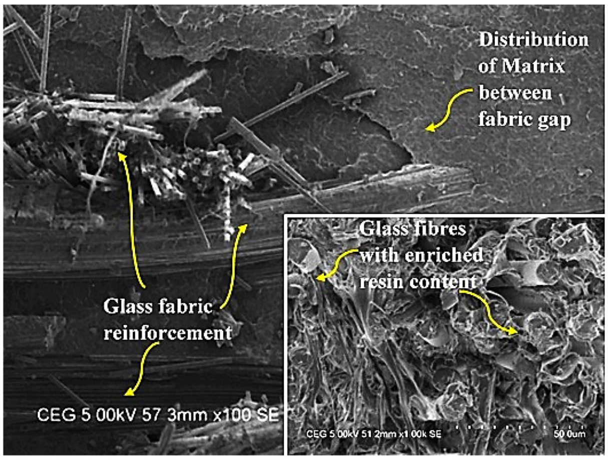
As with changes in compatibilizer concentration, changes in fabric orientation and fabric structure at an optimum process parameter levels improved the flexural strength of composites by 67.6% and 61%, respectively. The change in compatibilizer concentration has a great influence on the flexural performance of the composites than the other two parameters. The effective *in-situ* impregnation between the constituent materials has made a significant contribution to the huge improvement in the flexural performance of GFRTP composites. As with tensile strength, the flexural strength was maximum at Type-2 glass fabric structure, $[0\text{-}90]_4$ fabric orientation and 8 wt% compatibilizer process parameter level.

Effects of Process Parameters on Impact Strength. The impact strength indicates the energy absorbed by the specimen at the time of fracture. It is also an indication of the crack initiation and propagation in the specimen. It depends on the matrix properties and the adhesion between reinforcement and matrix. The energy absorbed for the plastic deformation leads to an increase in the impact strength of the composites. The details of the impact strength of GFRTP composites presented in Table 4 show the variation in strength from 2.12 to 4.23 J and reaching the maximum value at the optimum parameter values. The change in fabric structure from Type-1 to Type-2, at an optimum fabric orientation ($[0\text{-}90]_4$) and optimum compatibilizer concentration (8 wt%) levels, resulted in the increase in impact strength from 3.37 to 4.23 J. This change in fabric structure helped an improvement of 25.5% in the impact strength of the composite. Similarly, a change in compatibilizer concentration and fabric orientation at an optimum parameter levels, improved the impact strength of GFRTP composites by 25.5% and 9.9%, respectively. The results indicated above demonstrate the great influence of the fabric structure and compatibilizer concentration on the impact performance of the composites. This finding confirms the ability of MAPP compatibilizer in improving the interfacial adhesion between GF and PP, which could be attributed to the enhancement in the impact strength of composites.

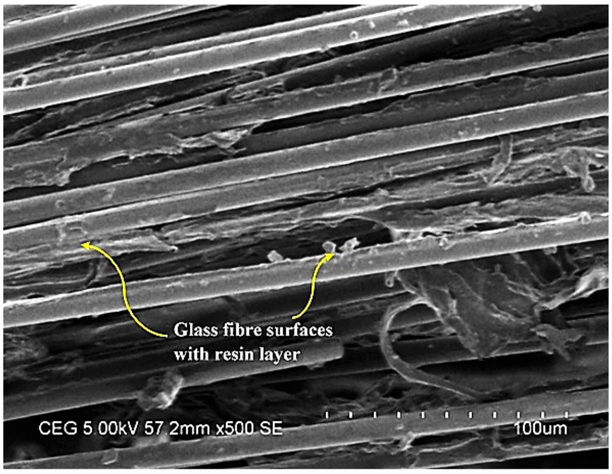
Table 11 presents a comparison of the mechanical properties of the various fibre reinforced PP composites with the findings of this study GFRTP-6 composite. The presence of higher mechanical properties in the GFRTP-6 composite than in the composites of the other types was clearly seen. Hence, the developed GFRTP-6 composite could well be suited for the structural and automotive applications.

Table 11. Comparison of Various Fibre Reinforced PP Composites with the Composite in the Present Study

S. No.	Fiber/Matrix form	Fibre loading (wt%)	Tensile strength (MPa)	Flexural strength (MPa)	Remarks
1	Woven GF fabric/PP sheet	40	140	124	Mechanical properties varied based on the process parameters [Findings of present study].
2	Short corn fiber/PP pellets	0-25	26.35	49.82	Addition of MAPP moderately increases the properties which decreases after 5wt% fibre loading [4].
3	Short CF (10 mm)/PP pellets	0-30	58	120	MAPP and modified fibre had a good synergistic effect in strengthening the interfacial adhesion [5].
4	Woven GF fabric/PP film (hybrid)	45 vol%	--	179	Different stacking configurations improved the flexural strength and modulus [6].
5	Short GF (4-8 mm)/PP pellets	70	11.2	58.85	Increase in fibre length increases the mechanical properties [9].
6	Horn fibre powder/PP pellets	5-20	32.48	37.14	Fibre loading decreases the tensile strength while slightly increases the flexural strength [15].
7	Long fiber (3-12 mm)/PP pellets	5-15	43	63	Increase in fibre length and fibre content considerably increases the both tensile and flexural properties [17].
8	Glass wool/PP pellets	30	37	-	APTE silane coupling agent along with epoxy film former improved the tensile strength of composite [21].
9	Continuous GF/PP film	75	101	-	GF surface modification is a suitable method for mechanical properties improvement in GF/PP composites [22].
10	Bacterial cellulose/PP	3	38.21	-	The addition of MAPP (up to 7wt%) increased the mechanical properties of the composites [27].



(a)



(b)

Figure 4. SEM micrographs of GFRTP-6 composite: (a) before tensile test; (b) after tensile test.

Morphological Analysis. The GFRTP-6 composite with higher mechanical properties was considered for the SEM analysis. The SEM morphology of GFRTP-6 composite before and after the tensile testing is presented in Figure 4(a-b), which reveals the nature of the adhesion between the constituent materials.

Figure 4(a) leads to the observation of the uniform distribution of the PP matrix over the GF fabric gaps. The SEM micrograph in Figure 4(a) subset, reveals the presence of a strong interfacial bonding between the hydrophilic glass fibre surfaces and the hydrophobic resin content. In order to demonstrate the effect of MAPP on fibre/matrix interface,[27] the SEM morphology of fractured tensile specimen has been analyzed. The SEM micrograph of fractured surface of GFRTP-6 composite as shown in Figure 4(b), reveals the presence of a resin layer throughout the glass fibre surface. This confirms the existence of a strong interfacial adhesion between them, even after the failure. Such an improved adhesion leads to a higher mechanical performance of the composite, which is in good agreement with the above mechanical properties.

Conclusions

Glass fabric reinforced polypropylene composites were fabricated by hot compression molding. The effects of the process parameters on the mechanical properties of GFRTP composites were investigated. The following conclusions are derived from this study:

· The optimum process parameter levels such as fabric structure [Type-2], fabric orientation [$(0\text{-}90)_4$], and compatibilizer concentration [8 wt%] help achievement of improved mechanical performance characteristics of GFRTP composite.

· The changes in process parameters have improved the tensile, flexural and impact strength by 106%, 235% and 100% respectively.

· The investigation showed the fabric orientation (62.34%) as a prominent factor in achieving the highest tensile strength (140 MPa) of GFRTP-6 composite, followed by fabric structure (24.42%) and compatibilizer concentration (7.14%).

· The compatibilizer concentration (64.74%) acted as the most significant factor for flexural strength (124 MPa), followed by fabric structure (7.53%) and fabric orientation (5.65%).

· The fabric structure (47.17%) is a dominant factor for impact strength (4.23 J), followed by compatibilizer concentration (46.13%) and fabric orientation (6.40%).

· The SEM micrograph of fracture surface has revealed the presence of a resin layer over the glass fibre surfaces, confirming the enhanced interfacial adhesion between GF and PP in GFRTP-6 composite.

· The test results show the ideal suitability of the developed GFRTP-6 composite for automotive applications such as the door module carrier, dashboard carrier, and structural carriers.

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