Effect of Needle Density on the Mechanical Properties of Fiber-Reinforced Polypropylene Composites

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ABSTRACT: The partial impregnation textile preform consisting of chopped-strand, long glass fiber and nonwoven polypropylene (PP) has been prepared by needle-punching to improve fiber–matrix distribution before processing. These unconsolidated textile preforms were then preheated and hot-pressed for consolidation and formation. A multichannel recorder was used to determine the completion of impregnation on multilayer glass fiber-reinforced PP, which could significantly reduce the required consolidation time. The effect of needle density on their impregnation has studied by scanning electron microscopy and optical microscopy, along with mechanical analysis. The increasing needle density up to 400 st/cm^2 has increased the flexural modulus, but the impact strength decreased. The optimal needle density contained proper flexural and impact properties is 50–100 st/cm^2, consistent with the observations from scanning electron microscopy and optical microscopy. A similar phenomenon is also observed by using nonwoven maleic-anhydride-modified polypropylene (mPP) instead of unmodified PP. However, the effect of needle punching on flexural and impact properties is not significant in mPP, which is probably due to better adhesion between glass fiber and mPP matrix. © 1999 John Wiley & Sons, Inc. J Appl Polym Sci 73: 2169–2176, 1999

Key words: thermoplastic composite; polypropylene; needle punching; flexural property; impact property

INTRODUCTION

Thermoplastic composites have received increasing interest due to their great manufacturing flexibilities, high strength, and stiffness. They also offer distinct advantages over the conventional thermosetting composites, for example, short molding cycle processability, excellent damage tolerance and impact resistance, unlimited shelf life, and good thermal stability.[1–6] The manufacturing costs can also be reduced due to high-speed production because they do not need to be cured during consolidation into composites, whereas thermosetting composites require extra time for chemical reactions to occur.[7] In addition, the cost of waste is reduced because they can be reused, reformed, or recycled.

Despite the above advantages, thermoplastics at their processing temperature have viscosity 500–5000 Pa s compared to thermosets that possess values less than 100 Pa s. The high viscosity along with poor fiber wetting or impregnation imposed problems in the manufacturing process of thermoplastic composites. During the recent decade, substantial efforts have been made to overcome the difficulties in impregnation with thermoplastic resin. Typically, some innovative technologies have been developed to construct thermoplastic matrix composite from intermedi-

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ate forms representing some kind of a partial impregnation, such as film stacking, commingled or intermingled fibers, and powder-impregnated fiber bundles.[8–12] The latter two intermediate material forms have brought fibers and matrix together in that the reinforcing fibers and the matrix resin may already have a good distribution before processing. Based on these new material forms, several new techniques have been attempted for manufacturing thermoplastic composites.[13–16] Due to those developments of good impregnation and interfacial characteristics, thermoplastic composites offer a combination of high toughness and good environmental resistance.[17]

The manufacturing process for thermoplastic composites may be divided into different categories, according to the state of preimpregnation and preconsolidation of the preforms. Among the manufacturing techniques currently used, the following three main categories may be distinguished by different processing steps: using preimpregnated tape and tow, preconsolidated sheet, and postshaping impregnation.[18] The main step in the processing cycle for preimpregnated tape or tow can be divided into three stages: (1) heating and melting, (2) consolidation and (3) cooling solidification. On the other hand, the main step in the processing cycle for preconsolidated sheets is (1) preheating and melting, (2) transfer to mold, (3) forming and consolidation and (4) removal from mold. Compared to the shaping process for preimpregnated tape and tow, the main difference in the shaping process by using preconsolidated sheets is that the time-consuming consolidation step may be reduced or eliminated. The preconsolidated sheet can be preheated in an external oven before being transferred to the mold. By this sequence of operations, the cycle time can be considerably reduced, as the time-consuming thermal cycling of the mold is eliminated.

In the present article, we have prepared a new material preform of fiber-reinforced thermoplastic polypropylene (PP) by the combination of the film stacking and intermingled fibers methods using needle punching to improve fiber–matrix distribution. This material is classified as preimpregnated tow. A thermoforming process is used during the preheating step to reduce the manufacturing cycle time. A multichannel temperature and pressure recorder is used to determine the completion of consolidation for multilayer PP composites. The effect of needle density on the impregnation has studied by scanning electron microscopy (SEM) and optical microscopy, along with flexural and impact property analysis. Moreover, the effect of maleic-anhydride-modified polypropylene (mPP) instead of PP on the impregnation is also discussed.

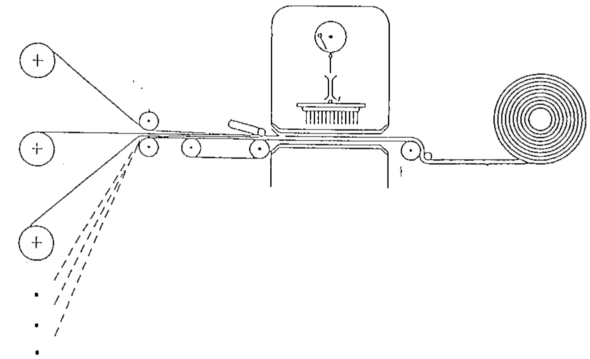

Figure 1 Schematic of the manufacturing of the nonconsolidated, needle-punched, composite preform.

EXPERIMENTAL

Materials

Polypropylene (PP) and glass fiber (GF) were supplied by Taiwan Polypropylene Comp. Ltd. and Intex Ind. Ltd.. Maleic-anhydride-modified polypropylene (mPP) pellets were supplied by Nytex Composites Comp. Ltd. and drawn into fiber in our laboratory. The densities of PP, mPP, and GF were 0.90, 0.90, and 2.56 g/cm^3, respectively. The materials examined in this study were prepared from preforms consisting of stacked nonwoven PP layers and chopped strand random glass fiber individually, held together by needle punching. Detail processing is shown in Figure 1. In order to study the effect of needle density on the impregnation of fiber-reinforced PP composites, several needle densities in the range of 50–500 st/cm^2 are applied in this study. For comparison, PP–GF sandwich structure without needle punching is also prepared. Multilayer PP–GF with dimension 20 by 20 cm could be preheated in an external oven for melting and consolidation. The materials were then transferred to a lower temperature mold for forming under different pressure, temperature, and closing time.

The weight fraction of fibers was determined by conducting burn-off tests in a high-temperature furnace heated to 450°C for 4 h. The specimens were cut and weighted before and after the 4-h cycle.

Mechanical Analysis

Tensile and flexural tests were performed on an Instron test machine 4505 model according to the ASTM D-3039 and D-790 standard. Flexural strength and modulus were both determined using 0.25-in. diameter support and loading pins. Center span deflections for flexural tests were measured directly using an LVDT deflectometer. The drop-weight impact properties of these composites were assessed using a Instrumented Impact Tester. All measurements were made at room temperature.

Thermal Analysis

The melting temperature (T_m) and crystallization temperature (T_c) were determined using a Perkin–Elmer DSC7 with a heating rate of 20°C/min to 200°C and held for 3 min. The samples were then cooled to 40°C at different cooling rates in the range of 5–50°C/min.

Scanning Electron Microscopy

Specimens of fiber-reinforced PP with different needle densities after Izod impact tests were mounted on A1 stubs with double-sided tape. The specimens were coated with approximate 5 nm of gold in order to minimize charging. SEM was performed on either JSM-6400 instrument operating at 10 kV in the secondary electron imaging mode.

Optical Microscopy

Specimens of fiber-reinforced PP with different needle densities after Izod impact tests were mounted between two glasses. Optical microscopy was performed on Wild M32 type-S instrument in reflection mode.

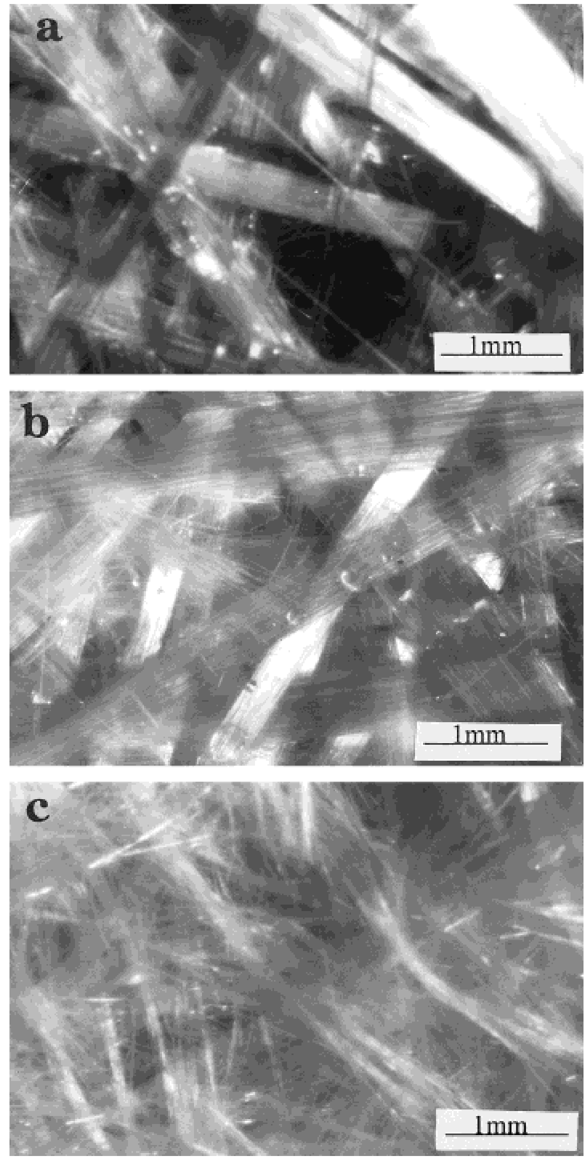

Figure 2 Photographs of the surface of PP–GF composites with different needle densities after burn-off tests. The needle densities are (a) 0, (b) 100, and (c) 400 st/cm^2, respectively.

RESULTS AND DISCUSSION

Figure 2 shows the photographs of optical microscopy for random PP–GF composites with different needle densities after burn-off tests. The needle densities of fabrication condition were in the range of 0–500 st/cm^2. Using needle-punching to mix PP and GF together can improve PP–GF distribution. The effect of needle density on the impregnation is dependent on PP–GF distribution during fabrication of PP–GF nonconsolidated composite preform. As seen in this figure, the bundles of glass fiber are tightened together [Fig. 2(a)] and gradually become loose with increasing needle densities [Fig. 2(b) (c)]. This observation implies that the higher the needle density, the better the PP–GF distribution. Therefore, using needle punching to improve PP–GF distribution can be enhanced the wetting or impregnation between the fiber–matrix during the manufacturing process. However, the higher needle density may break down the glass fiber and then reduce the aspect ratio between the PP and GF. Fiber break-

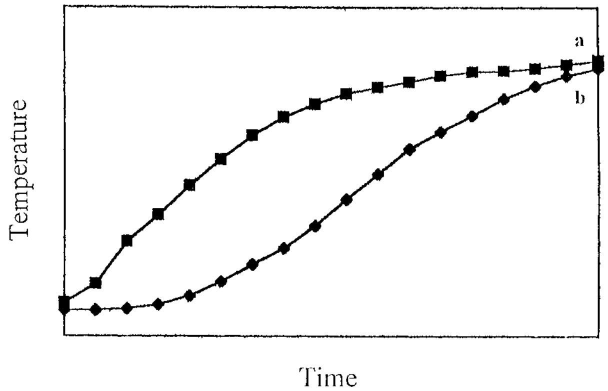

Figure 3 Time–temperature distribution (a) on the surface and (b) in the center for multilayer PP–GF during the preheating processing.

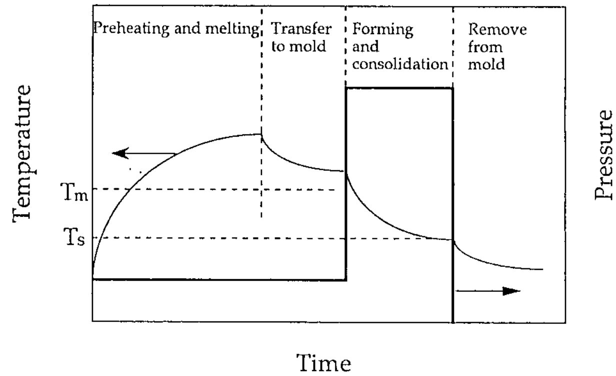

Figure 5 Principal stages of pressure and temperature in the center in the processing of multilayer PP–GF.

age and fiber pullout are two major mechanisms to affect the flexural and impact properties. In this case, it is very important to find an optimal needle density containing relatively higher flexural and impact properties in the fabrication of PP–GF composites.

In order to measure the mechanical properties for PP–GF composites, the unconsolidated textile preform needs to be processed. Differential scanning calorimetry (DSC) thermal analysis can provide the information about preheating and molding temperatures for processing fiber-reinforced PP. The melting temperature for PP–GF is about 160°C, and the crystallization temperatures for PP–GF are in the range of 100–120°C, depending on their cooling rates. The preheating temperature is generally around 20–40°C higher than the melting temperature, and the molding temperature is around 20–40°C lower than the softening temperature. Therefore, the preheating temperature and mold temperature for the processing of PP composites are in the range of 180–200°C and 60–100°C, respectively.

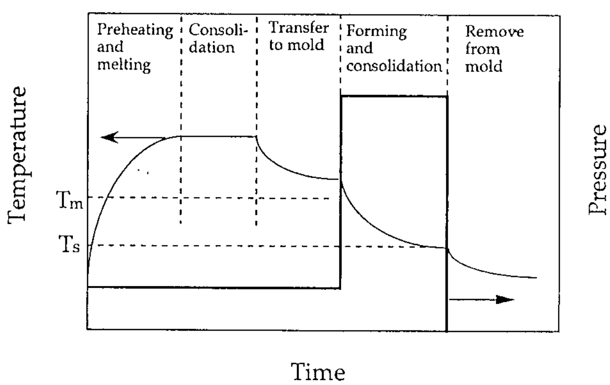

Figure 4 Principal stages of pressure and temperature on the surface in the processing of multilayer PP–GF.

As mentioned earlier, the main step of the processing cycle for preimpregnated tow, the textile preform of studied material, requires time-consuming consolidation, which is not desirable for commercial manufacturing. Therefore, a high-temperature thermoforming processing technique along with low-temperature compression molding is used on this preimpregnated tow. These processing steps are similar to those for the preconsolidated sheet. In this process, multilayer PP–GF sheets are preheated in an external oven to its softening temperature, while it is held horizontally. Thermoforming is quite different from most plastic processing methods in that the material is not converted to a liquid, but is only converted to a pliable, semirigid form. The pretreated sheet is then transferred to another lower temperature mold for forming by pressure. Figure 3 shows the trends of temperature–time distribution measured from first–second and second–third layers of five-layer PP–GF composite during preheating. The shortest preheating time for five-layer PP–GF composites can be easily determined by multichannel temperature and pressure recorder. The main step in the processing cycle for thermoplastic composites on the surface can be divided into the following five stages, as Figure 4 indicates:

1. Preheating and melting;
2. consolidation;
3. transfer to mold;
4. forming and consolidation; and
5. removal from mold.

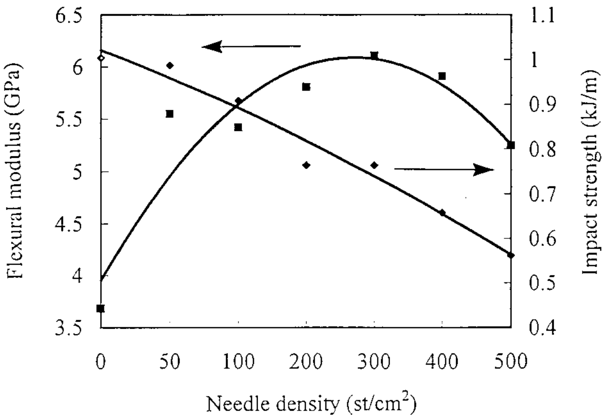

Figure 6 Flexural and impact properties versus needle densities of PP–GF composites.

The processing steps for PP–GF composites between layer and layer are as follows:

1. Preheating and melting;
2. transfer to mold;
3. forming and consolidation; and
4. removal from mold (Fig. 5).

Figure 6 shows the results of flexural and impact properties versus needle density. These results are also summarized in Table I. The flexural modulus is relatively low for PP–GF composite without needle punching and sharply increases using needle density of 50 st/cm^2. In the range of 100–400 st/cm^2 needle density, the flexural strength gradually levels off and then decreases at 500 st/cm^2. On the other hand, the izod impact strength goes the opposite way. They gradually decrease with increasing needle density. The optimal needle density to control the proper flexural and impact properties of composite is in the range of 50–100 st/cm^2. Under these conditions, the flexural modulus increases 50% compared to those without needle punching, but impact strength only decreases 24%.

Table I Effect of Needle Density on the Physical Properties of PP–GF Composite

Needle Density (st/cm^2)	Fiber Content (%)	Impact Strength (kJ/m)	Flexural Strength (MPa)	Flexural Modulus (GPa)
0	39	1.004	82.4	3.68
50	43	0.988	122.6	5.55
100	40	0.908	113.8	5.29
200	37	0.764	120.6	5.37
300	39	0.764	125.5	6.11
400	39	0.657	124.5	5.91
500	40	0.561	104.0	5.25

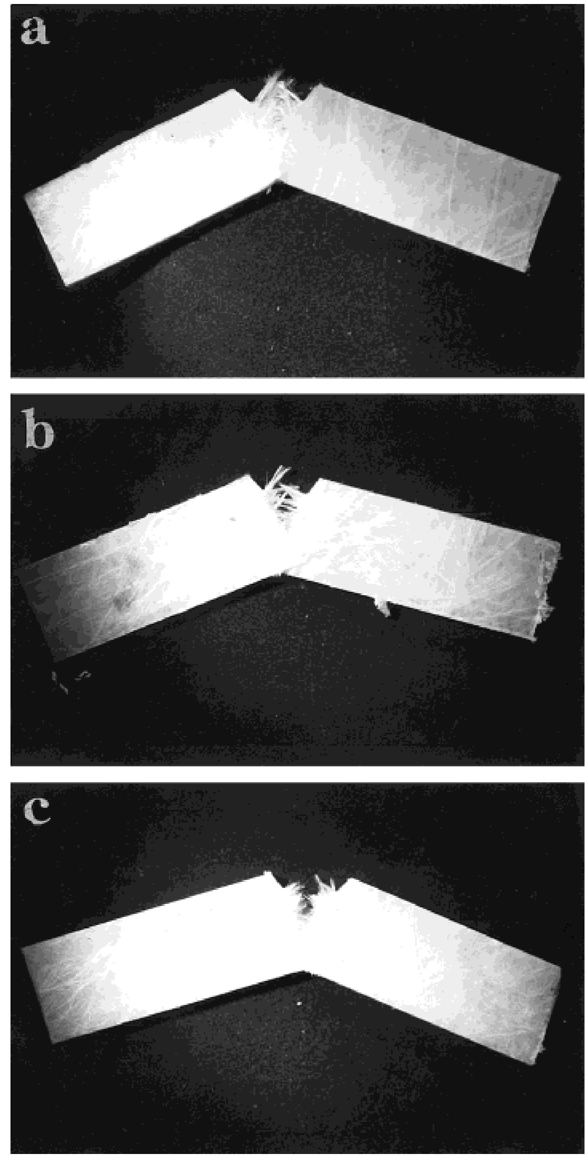

Figure 7 Optical microscopy of the interface of PP–GF composites with different needle densities after Izod impact tests. The needle densities are (a) 0, (b) 100, and (c) 400 st/cm^2, respectively.

Figure 7 shows the photographs of the interface of PP–GF composites with different needle densities after izod impact tests. Bundles of glass fibers pull out of the interface of PP–GF composite without needle punching. Increasing needle densities can separate the bundle of glass fiber, and, then, single fiber pulling out of the matrix is observed. Fiber dispersion can improve flexural

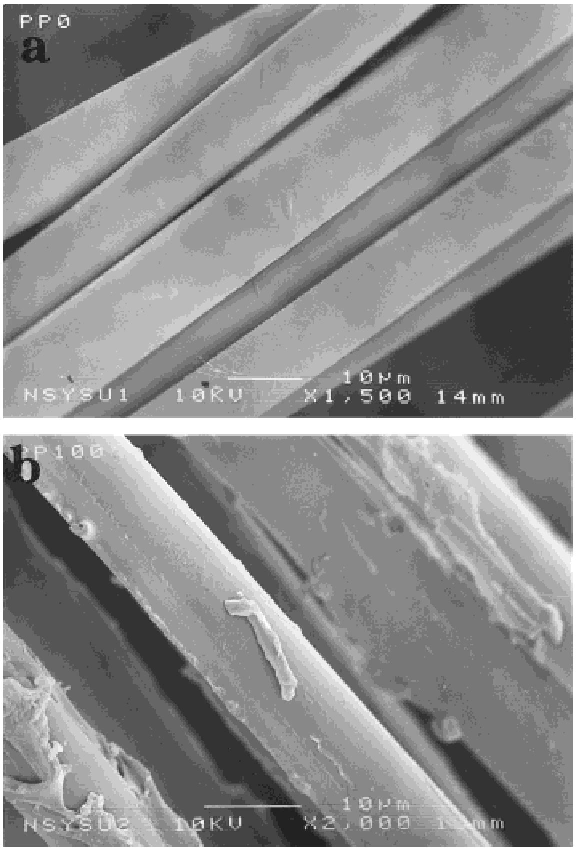

Figure 8 SEM micrographs of the cross section of PP–GF composites with needle densities of (a) 0 and (b) 100 st/cm^2.

property, but fiber pullout or debonding can cause poor impact property. These observations are generally supported with the results of increasing flexural properties and decreasing impact proper-

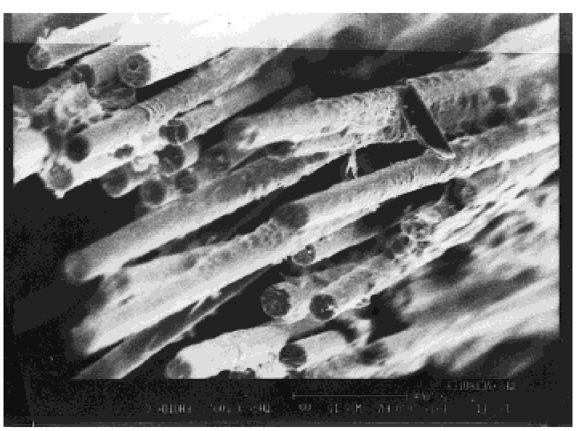

Figure 9 SEM micrographs of the cross section of mPP–GF composites with needle densities of 100 st/cm^2.

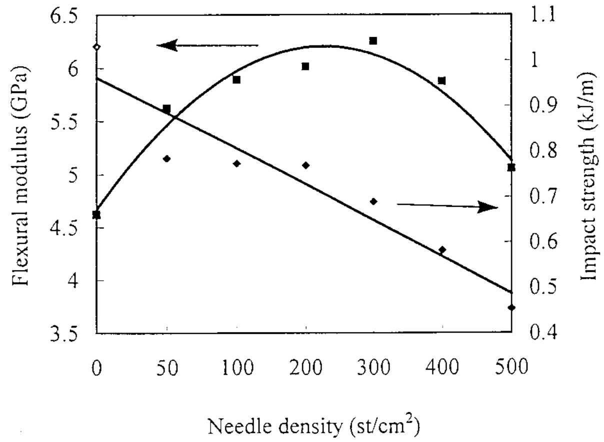

Figure 10 Flexural and impact properties versus needle densities of mPP–GF composites.

ties by using needle punching. The cross section of above specimens is further identified by SEM shown in Figure 8. The micrograph of PP–GF composite without needle punching shows pure glass fiber surface without any trace of PP matrix on the surface. No adhesion of PP on GF surface is observed. Poor adhesion between PP and GF is mainly due to nonreactive chemical structure of PP. Increasing needle density to 100 st/cm^2, the SEM micrograph shows some PP matrix is adhered to the GF surface, pointing to the better adhesion for PP–GF composite after needle punching fabrication. This is probably due to the surface roughness or heterogeneous nucleation caused by the glass fiber breakage using needle-punching fabrication, therefore, enhancing the PP–GF interfacial bonding.

Other composites are prepared by using maleic-anhydride-modified polypropylene (mPP) instead of PP matrix to enhance the fiber–matrix

Table II Effect of Needle Density on the Physical Properties of mPP–GF Composite

Needle Density (st/cm^2)	Fiber Content (%)	Impact Strength (kJ/m)	Flexural Strength (MPa)	Flexural Modulus (GPa)
0	39	1.031	102.0	4.62
50	37	0.785	126.5	5.62
100	35	0.774	143.2	5.89
200	36	0.769	130.4	5.58
300	36	0.689	142.0	6.25
400	38	0.582	128.5	5.87
500	38	0.454	105.9	5.05

Table III Physical Properties of PP–GF and mPP–GF Composites

Properties	Average Properties of Composite					
	PP–GF			mPP–GF		
Thickness (mm)	3.3	3.3	3.3	3.3	3.3	3.3
Fiber content (%)	20	30	40	20	30	40
Density (g/cm^3)	1.03	1.10	1.16	1.04	1.10	1.16
Tensile strength (MPa)	46.1	83.5	86.7	61.6	94.7	97.8
Tensile modulus (GPa)	2.74	5.29	5.68	3.59	5.94	6.16
Tensile elongation (%)	2.9	2.6	2.6	3.1	2.8	2.8
Flexural strength (MPa)	86.4	114	122	98.3	143	149
Flexural modulus (GPa)	2.69	4.84	5.23	3.27	5.89	6.12
Izod impact strength (kJ/m)	0.4	0.91	1.01	0.37	0.77	0.89
Dynatup impact energy (J)	11.1	18.0	23.3	14.3	21.7	25.8
HDT (264 psi, °C)	114	145	151	124	149	155

HDT: heat distortion temperature.

adhesion. Figure 9 shows the SEM micrograph of cross section of mPP–GF composite without needle punching after impact test. There are a lot of mPP adhered to the GF surface, indicating better adhesion between mPP and GF. This is probably due to the presence of functional groups in mPP induced to chemical bonding on the surface of fiber–matrix. Figure 10 shows the flexural and impact properties of mPP–GF versus needle density. These results are listed in Table II. The flexural properties sharply increase with increasing needle density and then level off. On the other hand, the impact properties gradually decrease with increasing needle density. The optimal needle density of mPP–GF is 50–100 st/cm^2, in which the flexural strength increases 40% compared to those without needle punching, but the impact strength decreases 33%. However, this effect of needle punching on flexural and impact properties is not significant in mPP–GF composites. This is probably due to better mPP–GF adhesion, which induced better flexural and impact properties without any mixing mPP and GF together by needle punching.

Table III shows the tensile, flexural, impact, and thermal properties of glass fiber-reinforced PP composites with glass fiber content in the range of 20–40%. It can be seen clearly that these mechanical and thermal properties sharply increase from 20 to 30% glass fiber content and gradually levels off at glass fiber content at 40%.

CONCLUSIONS

A new material type combined the film stacking and intermingled fiber using needle punching to improve the fiber–matrix distribution before processing has been produced in our laboratory. The increasing needle density up to 400 st/cm^2 has increased the flexural modulus, but the impact strength decreased. The optimal needle density contained proper flexural and impact properties is 50–100 st/cm^2, in which the flexural strength increases 50% more than those without needle punching, but impact strength decreases 24%. This is probably the effect of needle punching caused by mixing fiber–matrix together or some breakage of glass fiber during fabrication. The similar result is also observed for nonwoven mPP, in which the flexural strength increases 40% more than those without needle punching but impact strength decreases 33%. The effect of needle punching on flexural and impact properties is not significant in mPP, which is probably due to better adhesion between glass fiber and mPP matrix.

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