

Preparation of Polypropylene Single-Polymer Composites by Injection Molding

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ABSTRACT: The hot compaction process is still the principle method to produce single-polymer composites (SPCs) at present, which limits the industrial mass production of SPCs. In this article, an approach of preparing SPCs by injection molding was demonstrated. Polypropylene (PP) SPCs were prepared at different temperature and pressure. The flexural properties of the PP SPCs were tested. The optimum processing parameters, injection pressure of 140 MPa and nozzle temperature of 220°C were obtained. Flexural strength of the PP SPCs reached up to 43.3 MPa, which is 54.2% higher than that of nonreinforced PP. Flexural strength can be promoted by raising the injection pressure and nozzle temperature. The morphology of the fibers in the PP SPCs was investigated by optical microscope and scanning electron microscopy techniques. The matrix melt was able to penetrate into fiber webs under appropriate temperature and pressure, good interfacial compatibility and bonding properties were obtained. As the injection and holding pressure increased, the penetration capacity could be improved and the arrangement of fibers tended to be closer. © 2013 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* 130: 2176–2183, 2013

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INTRODUCTION

Polymer composites are generally formed by embedding and orienting fibers (polymer, glass, carbon, etc.) in a thermoplastic matrix polymer, they are widely used in many applications in aerospace, automotive, electrical, microelectronics, infrastructure and construction, medical, and chemical industries.^{1,2} However, it is hard to fully recycle this kind of composites as a result of different compositions between fiber and matrix. In addition, polymer composites usually fail at the weak fiber/matrix interface, resulting from their chemical incompatibility.³ The development of single-polymer composites (SPCs) seems to be an alternative in this aspect. SPCs, made by integrating high-strength fibers into a matrix of the same polymer, do not contain mineral fillers and are characterized by improved adhesion quality between the matrix and the reinforcement. The future of SPCs looks promising because of continuing improvement in their preparation and properties, their market growth, and their recyclability.⁴

SPCs are first prepared and reported in 1975 by Capiati and Porter.¹ Since then, different thermoplastic polymers, including polyethylene (PE)^{5,6} polypropylene,^{7,8} polyethylene terephthalate,⁹ polyethylene naphthalate,¹⁰ polylactic acid,¹¹ polyamide,^{12–14} polymethylmethacrylate,¹⁵ and polytetrafluoroethylene,¹⁶ were

exploited to manufacture SPCs. However, in the case of true SPCs, the matrix and the fiber should originate from the same polymer, and hence should have same or similar melting temperatures. Therefore, it is difficult to combine the fiber with the matrix, without melting the oriented fiber and thus losing stiffness and strength developed in the process. The preparation methods for traditional composites are not suitable for SPCs. The difference between the melting temperatures determines the processing window. Researchers have developed different methods for preparing SPCs, including hot compaction of fibers or tapes,^{7–10,12,17} film stacking,^{18–21} combination of hot compaction and film stacking,²² co-extrusion.^{2,23–26} However, these extant methods to prepare SPCs are all limited in compaction process; the industrial mass production of SPCs has still been limited. The hot compaction technology has a few disadvantages such as long preparation cycle and products only with simple shapes. Therefore some new processes such as extrusion or injection molding need to be developed, which can achieve the goals to produce SPCs efficiently with large scales and complex geometry.

Injection molding is the most important industrialized manufacturing technique, which is widely used in the field of polymer composites preparation. Some advantages of injection

molding are high production rates, repeatable high tolerances, the ability to use a wide range of materials, low labor cost, minimal scrap losses, and little need to finish parts after molding. The fiber/resin mixture is fed into the hopper and transferred into the heated barrel, mixed, and forced into a mold cavity where it cools and hardens to the configuration of the mold cavity. However, the traditional injection molding process preparing polymer composites is not suitable for SPCs. As the fiber/resin mixture is from the same polymer, extensive fiber damage occurs when the fiber/resin mixture has been fed into the barrel because of the high barrel temperature, the intensive mixing with high-shear and passage through a narrow gate. Therefore, there have been no reports about preparing SPCs by injection molding till now.

Aiming at the limitation of existing production methods for SPCs, the feasibility of preparing SPCs by injection molding is investigated. PP SPCs are prepared. The flexural properties of PP SPCs are tested. The microstructure of the PP SPCs is analyzed by light microscopy using polished sections and by scanning electron microscopy (SEM) using cut surfaces. The influence of processing pressure and temperature is described.

EXPERIMENTAL

Materials

PP (model number: K8303) granules with a density of 0.9 g/cm^3 were provided by Beijing Yanshan branch of China Petroleum & Chemical Corporation. Its melt flow rate is 1.0–3.0 g/10 min. The PP fiber cloth was supplied by Innegrity LLC (Simpsonville, SC). The PP cloth was weaved in a plain structure and each yarn is consisted of 225 continuous filaments with a diameter of $48 \mu\text{m}$. The density of the weft and warp is 4.3 and 6 threads/cm, respectively. Thickness of the PP cloth is 1 mm.

Differential Scanning Calorimetry

A differential scanning calorimeter (DSC-60, Shimadzu) was applied to study the melting and crystallizing process of PP granules and PP fabrics. They were heated from 40°C to 200°C at a rate of $10^\circ\text{C}/\text{min}$ and held for 10 min at 200°C in order to erase thermal and mechanical history, and then cooled to 40°C at the cooling rate of $10^\circ\text{C}/\text{min}$. The flow rate of Nitrogen here was 60 mL/min.

Sample Preparation

The samples of the PP SPCs (No. 1-1 to 1-5) are molded into rectangle shape with a length of 63.5 mm, a width of 12.7 mm and a thickness of 6.35 mm. Figure 1 shows the schematic illustration of the samples. The fiber volume fraction is about 16% for the samples (No. 1-1 to 1-5).

A reciprocating screw injection machine manufactured by GSK CNC was used to prepare PP SPCs samples. The setting temperature from the feed-inlet to the barrel frontier was 50°C , 195°C , 220°C , respectively. The temperature of the barrel frontier was changed with different values of 210 – 235°C in different injection molding cycles. The nozzle temperature was also changed from 195°C to 230°C . PP granules were added to the plastication system where they were melted under the effects of heat and shear force provided by the screw and barrel, at the same

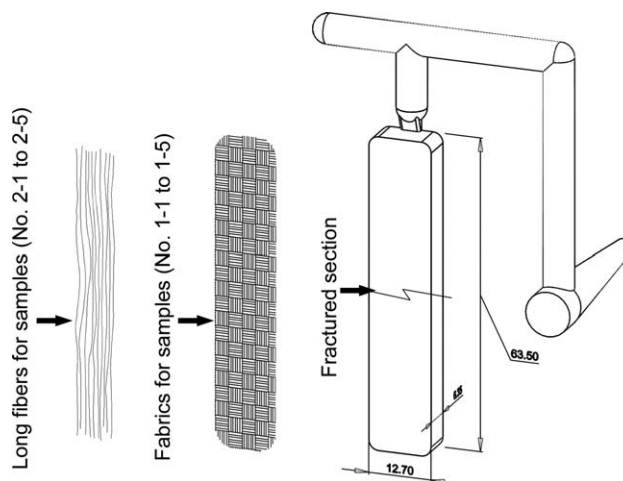


Figure 1. Schematic illustration of the samples.

time the melt was conveyed forward. The PP fiber fabric was preplaced into the inner surface of the mold cavity following by closing the mold. Then the PP melt was injected into the cavity within 1 s and under the injection pressure of 100–200 MPa following by packing for 10 s under the holding pressure of 85–180 MPa. The back pressure here was 2 MPa and the mold was at room temperature. The material in the cavity was cooled and solidified for 10 s. Finally the products of SPCs could be removed by opening the mold. The key processing parameters of five SPCs samples are listed in Table I. Nonreinforced PP samples were also produced under each group of process conditions for comparison.

In addition, several long fibers instead of fiber fabric were preplaced into the inner surface of the mold cavity then PP melt was injected into the cavity to prepare samples (No. 2-1 to 2-5). These samples were used in SEM to investigate the influence of the processing pressure and temperature on the morphology of fibers in the PP matrix. The processing parameters of five samples with several long fibers are listed in Table II.

Flexural Test

The flexural test was carried out on a universal testing machine (XWW-20Kn) made by Beijing Jinshengxin Testing Machine. In the process of bending test, the span length was 40 mm and the loading velocity was 50 mm/min. Although the samples were

Table I. Processing Parameters of Five PP SPCs Samples (Nos. 1-1 to 1-5)

No.	Injection pressure (MPa)	Holding pressure (MPa)	Nozzle temperature ($^\circ\text{C}$)	Barrel frontier temperature ($^\circ\text{C}$)
1-1	100	85	195	210
1-2	120	100	195	210
1-3	120	100	210	220
1-4	140	120	220	225
1-5	200	180	230	235

Table II. Processing Parameters of Five Samples with Several Long Fibers (Nos. 2-1 to 2-5)

No.	Injection pressure (MPa)	Holding pressure (MPa)	Nozzle temperature (°C)	Barrel frontier temperature (°C)
2-1	150	130	210	220
2-2	150	130	220	225
2-3	150	130	230	235
2-4	170	150	210	220
2-5	190	170	210	220

not standard transects for flexural test, the comparable flexural properties of the materials (PP SPCs and nonreinforced PP) could be analyzed.

Metallographic Observation

The microstructure of PP SPCs samples was observed by a metallomicroscope (Zeiss Axio Observer). The samples were solidated by denture powder and denture water. Then they were preliminary grinded using a series of sand papers, with a granularity of 600, 800, 1000, 1200, and 1500, respectively. After that the samples were polished on a polishing machine with grinding paste whose granularity was 1.5 and 0.5 aiming to eliminate the nicks on the samples. Finally the samples could be observed under the metallomicroscope.

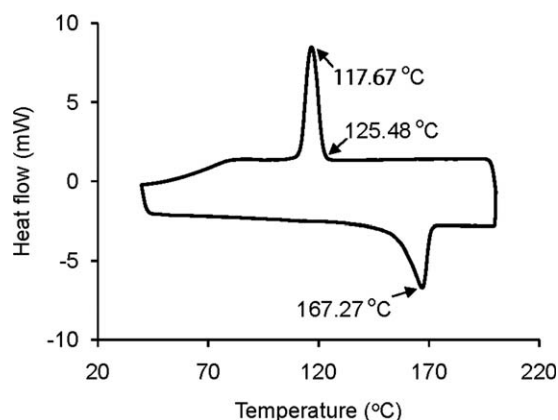
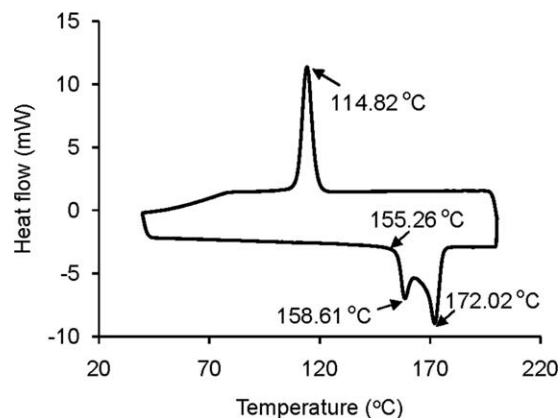
Scanning Electron Microscopy

The PP SPCs samples with several long fibers (No. 2-1 to 2-5) after bending test were quenched breaking in liquid Nitrogen. The fractured section in the middle part of the sample was gold coated and its morphology was observed using a SEM Model no. JSM-7401F scanning microscope with an accelerating voltage of 10 kV.

RESULTS AND DISCUSSION

Thermal Analysis

The thermal study of PP granules and PP fibers was carried out by differential scanning calorimetry (DSC). Figure 2 shows the

**Figure 2.** DSC thermogram of PP granules determined at heating and cooling rates of 10°C/min.**Figure 3.** DSC thermogram of PP fibers determined at heating and cooling rates of 10°C/min.

DSC thermogram of PP granules. It exhibits that the melting point of PP matrix is 167.27°C. The PP matrix will not begin to crystallize until it is cooled to 125.48°C. So it has a large supercooling degree⁵ of more than 43°C (126 to 167°C). Preparation of PP SPCs within this temperature window can maintain not only the fluidity of the matrix but also the morphology and strength of the fibers. Figure 3 gives the DSC thermogram of PP fibers. The fibers begin to melt at 155.26°C and there are two melting peaks (158.61°C and 172.02°C) in the heating curve. Schwenker et al.² reported that the stretched PP fiber has two melting peaks at the temperature of 158°C and 173°C. They considered that the disorientation of crystallization lead to the first peak and the melting of the crystallization contributed to the second peak at 173°C. Therefore the melting temperature of it is 173°C. It is known that the second melting peak at 172.02°C is the real fusion point of PP fibers.

According to the thermal results of PP granules and PP fibers, the barrel temperature and the nozzle temperature can be determined as mentioned in the sample preparation. The processing temperature is usually set up to 20°C more than the melting point of the polymer. Temperature history in an injection molded part is a cooling process with time. When the PP melt get into the mold cavity, it will be cold abruptly. The melt that contacts with the inner surface of the mold cavity is cooled first. In the injection molding process of SPCs, the preplaced fiber fabrics are set on the inner surface of the mold cavity. There is heat conduction between the fibers and the matrix when the melt from the nozzle is injected into. So the surface fibers are melted partially or fully because of high temperature of the matrix more than 172°C (melting point of the fiber), at the same time the matrix is cooled fast at a lower temperature. Then supercooling²⁷ of the matrix occurs during the wetting phase, it protects melting of the inner fibers. We can only control the melt temperature inside the mold cavity by nozzle temperature indirectly. Therefore, different nozzle temperature in the range from 195°C to 230°C (see Table I) was used to determine temperature variation of the materials in the mold cavity. The influence of nozzle temperature could be analyzed through morphological properties of the final SPCs samples.

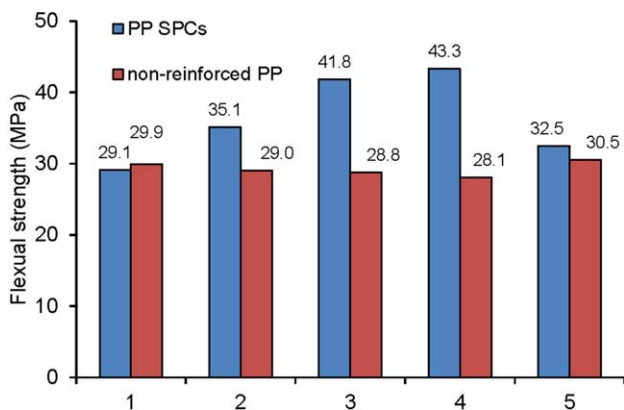


Figure 4. Flexural strength of PP SPCs and nonreinforced PP. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Flexural Properties

Figures 4 and 5 show the flexural properties of PP SPCs and nonreinforced PP under different process conditions. It is found that the flexural strength and modulus of the PP SPCs were obviously improved than that of nonreinforced PP. It demonstrates that the flexural strength and modulus of PP SPCs increased first and then decreased as the injection pressure and nozzle temperature increased. But the strength and modulus of the nonreinforced PP were almost not changed with different processing conditions. The flexural strength reached a maximum of 43.3 MPa, which is 54.2% higher than that of nonreinforced PP prepared under the same conditions when the injection pressure and nozzle temperature were 140 MPa and 220°C, respectively.

Both temperature and pressure play an important role to help the penetration capacity. In experiment No. 1-1 and 1-2 (see Table I), nozzle temperature of 195°C and barrel frontier temperature of 210°C were kept constant, but injection pressure and holding pressure were different. Comparing the flexural strengths of sample No. 1 and 2, it is found that increasing injection pressure can improve the flexural strength of PP SPCs.

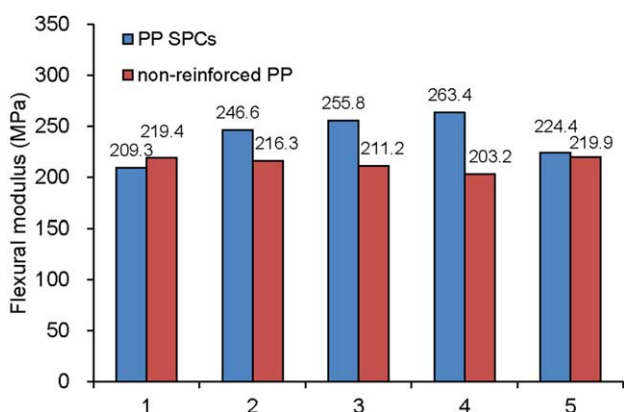


Figure 5. Flexural modulus of PP SPCs and nonreinforced PP. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

The flexural strength was improved by approximately 20% within 20 MPa change in injection pressure. As higher injection pressure is able to facilitate the flow into minute spaces among fibers and improve the wetting property of PP matrix. In addition, the holding pressure increases with the increasing injection pressure, it leads to a good adherent strength between fibers and the matrix.

In experiment No. 1-2 and 1-3, nozzle temperature and barrel frontier temperature were different, but injection pressure of 120 MPa and holding pressure of 100 MPa were kept the same values. It is demonstrated by comparing sample No. 1-2 and 1-3 that the flexural strength can be promoted by raising the nozzle temperature when keeping the injection pressure constant. The flexural strength was improved by approximately 19% within 15°C change in nozzle temperature. The reason is that the nozzle temperature determines the temperature of molten matrix injected into the cavity. As the temperature of nozzle increases, the viscosity of PP melt decreases and the mobility increases, which can improve the wetting and bonding capacity between the fiber and the matrix.

The flexural strengths of sample No. 1-4 proved the benefits of higher pressure and temperature further. However, it will create unfavorable impression on the flexural strength of PP SPCs when the injection pressure and nozzle temperature are too high just like sample No. 1-5. The reasons are as follows. In the experiment No. 1-5, the molten matrix was injected from one side of the cavity, so there was a displacement of PP fabric on the impact of matrix melt with pressure. As a result, the matrix cannot infiltrate and bond with the fibers in the drape that would turn into a weak point and finally influence the strength of PP SPCs. In addition, if the holding pressure is too high, it will diminish the spaces among fibers and increase the flow resistance of matrix melt, it will also exert an adverse effect on wetting. Furthermore, if the nozzle temperature is too high, it will bring excessive heat into the cavity leading to the melting possibility of fibers and decreasing of fiber strength.

Morphological Properties

Besides temperature referred in the thermal analysis, pressure and pressure history are key factors in the process, as pressure can control the shrinkage of the fibers and promote densification. The pressure history is complicated, and the pressure distribution inside the mold cavity changes with distance from the inlet gate. It is difficult to see the temperature and pressure history in the mold cavity, but we can deduce the influence of the temperature and pressure by microstructures of the final parts.

Optical micrographs of polished transversal cross-sections of the PP SPCs samples (No. 1-4 and 1-5) are reported in Figures 6 and 7 respectively. There are two types of PP fibers. The ones with circular shape are the fibers perpendicular to the section, and the others with a shape of long strip are the fibers parallel to the section. It shows that the matrix melt was able to penetrate into fiber webs by injection molding, and the SPCs samples had good interfacial compatibility and bonding properties. As shown in Figures 6 and 7, the fibers on the surface layer of the fabrics were melted easily because they first contacted the melt with high temperature, but the most fibers can still

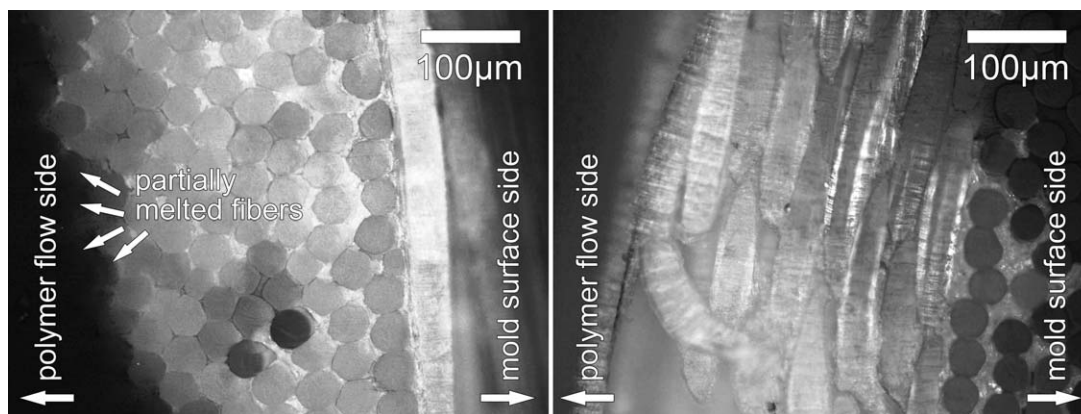


Figure 6. Optical micrographs (20 times magnification) of PP SPCs sample (No. 1-4) processed at injection pressure of 140 MPa and nozzle temperature of 220°C.

maintain its original morphology. It confirms the prediction in the paragraph of thermal analysis. There are more partial melted fibers in No. 1-5 than in No. 1-4. In Figure 7, more fibers were almost molten into matrix because of higher temperature. In addition, the arrangement of some fibers tended to be closer, and some fibers had been flattened into an oblong or hexagon shape under higher injection and holding pressure.

There are fibers with different size and geometry in different areas. Figure 8(a–c) shows the microstructures in the similar area of the sample No. 1-1, 1-4, and 1-5, respectively. Most fibers in No. 1-1 are bigger than those of No. 1-4 and 1-5. This is because that the sample No. 1-1 was molded with lower temperature and pressure thus the fibers of No. 1-1 scarcely melted. The diameters of fibers in Figure 8(b,c) are all less than the original value (48 μm) because of the partial melting of the fibers. It can be apparently seen that the fiber diameter became smaller as the nozzle temperature increased. However, comparing sample No. 1-4 and 1-5 in Figures 6 and 7, there are still many fibers that have similar diameter even the nozzle temperature is the highest in No. 1-5. This may be because of the larger injection and holding pressure that can provide fibers with a kind of

physical suppression action to constrain the relaxation of molecular chain and to achieve a state of overheating⁷ of fibers.

The samples with several long fibers (No. 2-1 to 2-5) were produced. Although there was no improvement in flexural strength because of few fibers, it is useful to investigate the morphology of fibers in the PP matrix. SEM was used to monitor the fracture surface of the composites after quenching the samples in liquid nitrogen. Figures 9–12 are the micrographs of the samples produced with different injection pressure and nozzle temperature. Same injection pressure but different nozzle temperature was used to produce samples No. 2-1, 2-2, and 2-3 as shown in Figures 9, 10, and 11, respectively. Parts of matrix can be clearly seen on the surfaces of some fibers. As the nozzle temperature increased, the parts of matrix on the surfaces of fibers became larger. In Figure 9, some fibers were pulled out from the matrix because of poor adhesion between the fiber and the matrix. At higher nozzle temperature of 220°C, Figure 10 shows that the fibers were broken and the fiber and the matrix were closely associated, but fibers were still pulled out. In the case of Figure 11, fibers were broken and not pulled out when the nozzle temperature increased at 230°C. This is an

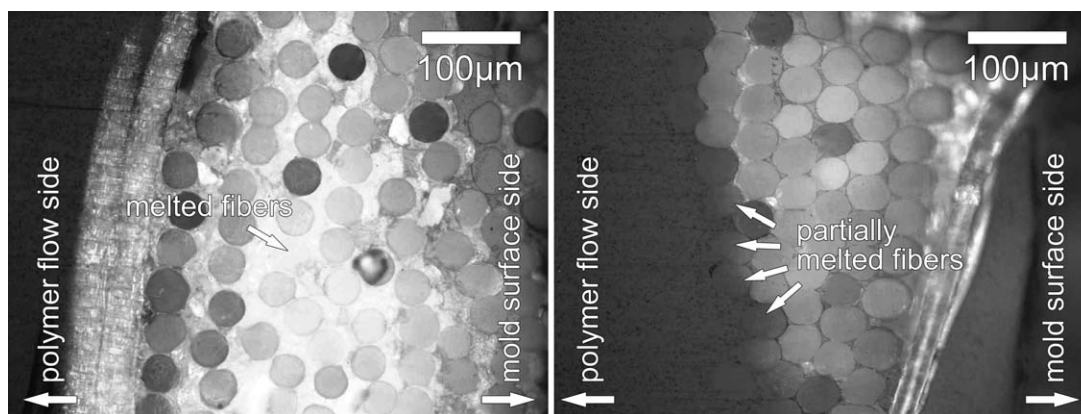


Figure 7. Optical micrographs (20 times magnification) of PP SPCs sample (No. 1-5) processed at injection pressure of 200 MPa and nozzle temperature of 230°C.

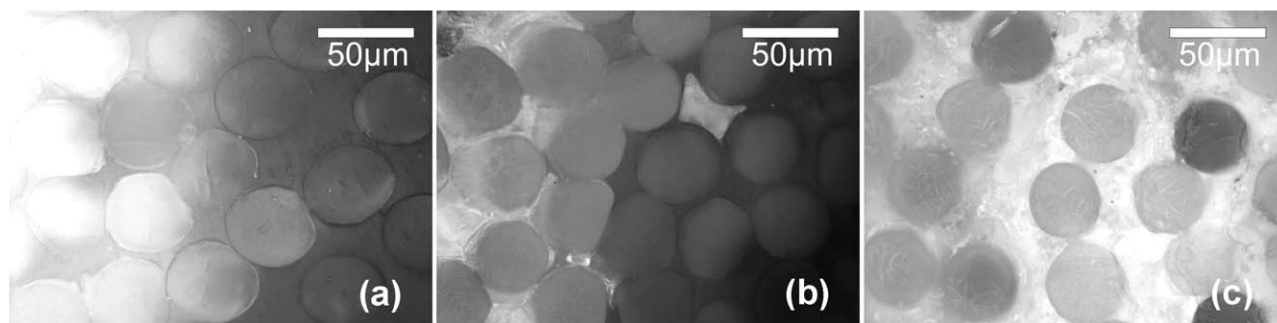


Figure 8. Optical micrographs (50 times magnification) of sample No. 1-1 (a), No. 1-4 (b), and No. 1-5 (c).

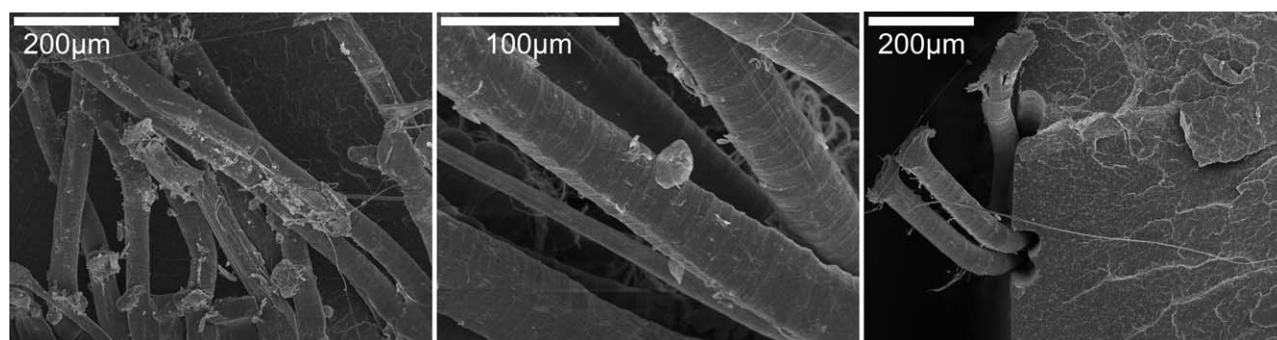


Figure 9. SEM micrographs of sample No. 2-1 processed at injection pressure of 150 MPa and nozzle temperature of 210°C.

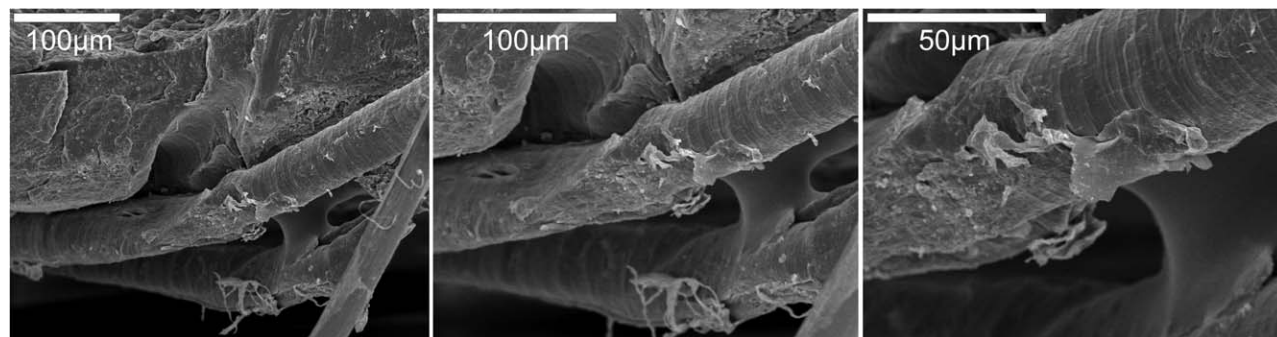


Figure 10. SEM micrographs of sample No. 2-2 processed at injection pressure of 150 MPa and nozzle temperature of 220°C.

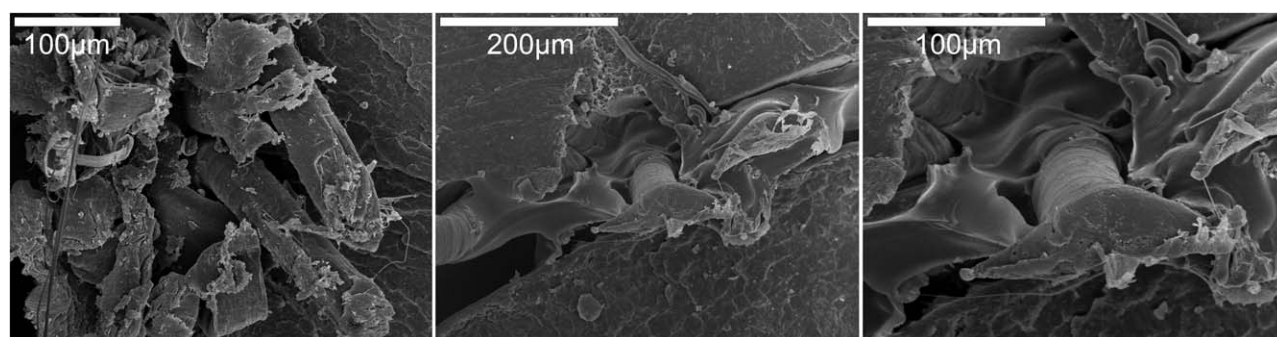


Figure 11. SEM micrographs of sample No. 2-3 processed at injection pressure of 150 MPa and nozzle temperature of 230°C.

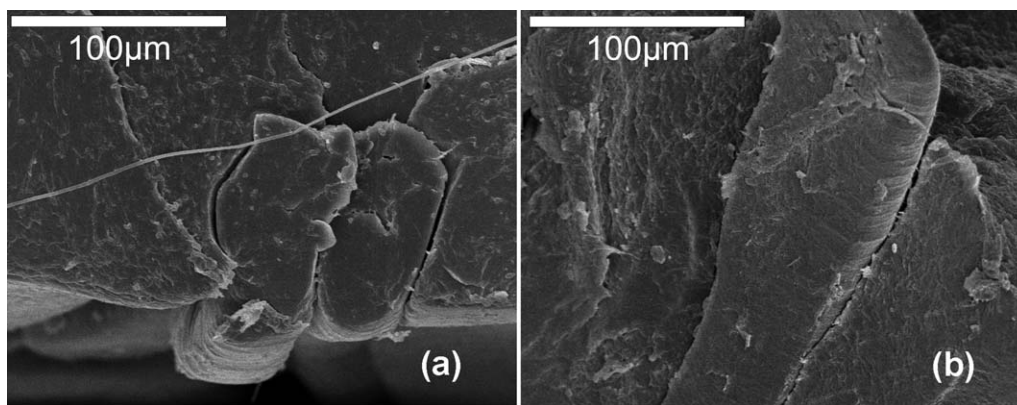


Figure 12. SEM micrographs of (a) sample No. 2-4 and (b) No. 2-5 processed at injection pressure of 170 and 190 MPa, respectively, and same nozzle temperature of 210°C.

indication of better adhesion between fiber and matrix. From these results, it can be concluded that higher temperature has improved interfacial adhesion properties.

Samples No. 2-1, 2-4, and 2-5 were produced with same nozzle temperature but different injection pressure. The SEM images are shown in Figure 12. Because lower nozzle temperature was used, it is also possible to observe distinct gaps between the fibers and the matrix indicating poor adhesion. The gaps were smaller at higher pressure, indicating that higher pressure could improve fibers densification then higher strength could be obtained. At higher pressure, fibers were pressed into ellipse geometry. It demonstrates that too high pressure would diminish the gaps between the fibers that increases flow resistance of matrix melt.

These above structures mainly depend on the processing conditions. High pressure and temperature benefits the penetration of matrix into the fibers. But much higher temperature will melt the fibers then destroy their strength function in the composites; much higher pressure will change the arrangement and geometry of fibers exerting an adverse effect on wetting.

CONCLUSIONS

The feasibility of preparing SPCs by injection molding was investigated. PP SPCs were successfully prepared by presetting the fabrics like an insert in the mold cavity. PP SPCs sample molded at an injection pressure of 140 MPa and a nozzle temperature of 220°C can reach a maximum flexural strength of 43.3 MPa, which was improved by 54.2% than that of nonreinforced PP. Both pressure and temperature are the key parameters. The results of flexural test showed that flexural strength can be promoted by raising the injection pressure and nozzle temperature. But they cannot be set too high so as to avoid fiber melting as well as the change of fiber arrangement and geometry. The microstructure of PP SPCs observed by metallomicroscope showed that the matrix melt was able to penetrate into fiber webs by injection molding under appropriate temperature and pressure, and the SPCs samples had good interfacial compatibility and bonding properties. The fiber will partially melt if the temperature of molten matrix is too high but this

phenomenon might be avoided by increasing the injection and holding pressure owing to a physical constraint action. SEM analysis confirmed the benefits of higher temperature and pressure in the injection molding of SPCs.

Injection molding is probably the most complicated method for processing thermoplastics, especially for the melt filling and holding phase in the mold cavity. From the appearance of the samples in this article, the side with fiber fabrics was difficult to be penetrated totally by the matrix in sample No. 1-1 and 1-2. Therefore, in a future study, it is better to set the fabrics in the center of the mold cavity like an insert part, then the melt could be injected and penetrate the fabrics from both sides of the fabrics. In addition, an experimental mold with pressure and temperature sensors should be designed to know the real temperature and pressure in the mold cavity.

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