

Processing and Mechanical Properties of Long Maize Fiber Reinforced Polypropylene Composites

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Maize fibers have been incorporated in a polypropylene matrix to form the uniaxially reinforced composites containing up to 55% fiber area fraction. Variations of the processing parameters such as molding temperature and forming pressure were conducted in order to determine the optimum manufacturing conditions. It was found that an improvement in the mechanical properties is pronounced in the molding at 175°C and forming under 10 MPa, above which the properties tend to either level off or at the best show only slight improvement. The tensile strength is increased monotonically depending on the increase in fiber area fraction, especially at 39 and 55%, suggesting in general to obey the rule of mixture (ROM). The properties were found to be generally decreased at lower molding temperature, forming pressure and fiber area fraction. Examination of the microstructural morphologies by scanning electron microscopy of the composites in particular at the fiber/matrix interface suggests that with appropriate pressure, the fibers are possible to contact intimately with the polymer matrix, provided for the molding temperature above 175°C.

Key words: Materials recycling, Long natural maize fiber, Thermoplastic polymer, Composite material, Mechanical properties

1. INTRODUCTION

Recently, the re-utilization of agricultural waste, especially of the tropical non-woody fibrous waste, has emerged as an important research area judging from both environmental protection and materials recycling viewpoints⁽¹⁾⁽²⁾. The rationale of re-utilizing such wastes was highlighted in previous authors' work⁽¹⁾, which also proposed a reasonable and integrated recycling system that should convert the wastes into the value-added new materials. The use of these renewable resources is intended to offer significant opportunities for the improved materials with enhanced support for the global sustainability.

One of the promising alternative methods for achieving this is by combining the fibers of the lignocellulosic wastes with materials of different characteristics such as plastics, to come up with a composite material having the improved properties. In this case, the fibers offer a number of advantages as reinforcing fillers in the plastic composites, such as high specific strength and/or stiffness, low weight, low cost, and lower abrasion to the processing equipment⁽³⁾. Such synergistic combination of natural fibers, wood, thermoplastics and their composites, synthetic fibers, can meet the demanding requirements for construction and transportation industries, namely for motor vehicle components⁽⁴⁾.

It was observed that while wood continues to dominate as the major fiber raw material especially in pulp and paper industries, agro-based fiber usage will expand well into the 21st century. However, cereal straws, sugarcane bagasse, and annual fiber crops will be the feedstocks that will account for much of the future

expansion⁽⁵⁾. Some reports indicate that, of these fibers, jute, ramie, flax and sisal (hard fibers) are the most commonly used fibers for polymer composite. On the other hand, natural fibers in the form of wood flour are often used for the preparation of natural fiber composites than the so-called hard fibers⁽²⁾.

The major drawbacks in the application of natural fibers in composites are however in the high moisture absorption and poor wettability with polymer matrix due to the hydrophilic nature of the fiber leading to insufficient interfacial adhesion with the hydrophobic polymer matrix. Therefore, most of the research articles on natural fiber/polymer matrix composites are related to the improvement of mechanical properties by enhancing the interfacial adhesion using the processing aids and coupling agents with additional reaction and costs⁽⁶⁾⁻⁽¹⁰⁾. To the best of our knowledge, however, there is only a few report about the usage of maize fibers as reinforcing materials for thermoplastic composites, especially with polypropylene (PP)⁽¹¹⁾.

This study presents about both the development of a suitable processing technique and the mechanical property evaluation of the low cost and value-added new composite materials composed of the long maize fibers and PP without coupling agents. In particular, the microstructural aspect namely for the adhesion at the fiber/matrix interface of the composites was investigated in some detail.

2. EXPERIMENTAL

2.1 Characteristics of maize fiber

Corn stalks, like many agricultural fiber sources, consist of a pithy core (parenchyma) with an outer layer

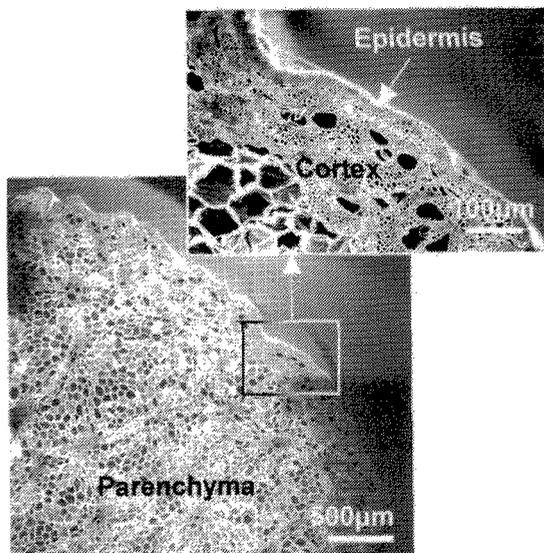


Fig. 1 Cross sectional microstructure of maize stalk.

of long fibers or cortex, as shown in Fig. 1. The total maize stem contains about 50% fibers, 50% parenchyma cells and vessels, and only about 1% epidermis cells⁽¹¹⁾.

The basic constituents of maize are cellulose (mostly present in the fiber), hemicellulose and lignin which binds the fibers together. Cellulose and hemicellulose are present in the form of holocellulose amounting to about 40% of the total constituents. Table I summarizes the chemical characteristics of maize⁽¹²⁾.

2.2 Extraction and preparation of maize fiber

The maize stalks were obtained during the summer 2000 from local source in Nagano prefecture, Japan. The pulping method adopted was to ensure the minimized damage to the fibers at the reduced chemical, energy and cost. In addition to these benefits, the chosen solvent of sodium hydroxide (NaOH) solution should be considered to provide an adequate surface treatment/modification for the maize fiber⁽¹³⁾.

The maize stalk (hard outer layer) were separated from the pith, and heated for 1.5 hours at 95°C under atmospheric pressure in the 6g/l of NaOH aqueous solution in a ceramic pot-like container with high liquor to material ratio (15 ~ 20: 1). The hitherto loose and softened stalks are then hand-scrubbed carefully, discarding the pith portions to expose and separate only the fibers. The fibers were again washed thoroughly in water, hand pressed into thin unidirectional sheets of

flattened fiber bundles depicting prepreg and then air-dried sufficiently to store in a dessicator.

2.3 Preparation of polypropylene for matrix

The virgin PP used is J-2021 GR (MFI 22g/10min, 0.9g/cm³, manufactured by Idemitsu Petrochemical Co. Ltd, Japan). In addition, the commercially available recycled PP also was adopted for a quality check.

Approximately 100µm thick films were prepared from both virgin and recycled PP pellets using a table type hot press. Thermal investigation on the PP was performed by means of the differential thermogravimetric analyzer equipped with the controlling and programming unit, which can operate in air atmosphere. The softening, peak melting and complete melting temperatures were determined as 134, 154 and 166°C respectively for the virgin PP, while the values were 141, 155 and 170°C for the recycled one.

2.4 Composite manufacturing facility

The facility used is a specially designed hot press. It is composed of the 350°C variable temperature-controlled cylindrical heating chamber with about 100mm diameter, and the variable 15 MPa hydraulically operated press which has upper and lower plungers capable of moving up and down in and out of the heating chamber, as schematically shown in Fig. 2. The specifications and the criteria for the design were adopted after extensive literature review of the existing molding methods and facilities⁽¹⁴⁾⁽¹⁵⁾. For instance, the facility is able to combine the multiple functions including fiber drying, matrix melting, pressure application and the subsequent ejection of the die forming the composite specimen.

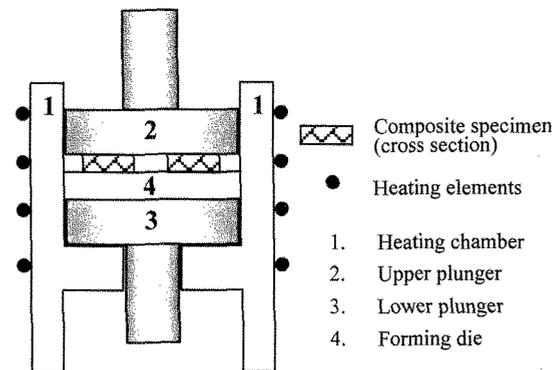


Fig. 2 Schematic drawing of the manufacturing facility.

Table I Chemical analysis of maize and anatomic elements⁽¹²⁾

Constituents	Whole stalks	Stalks without leaves and pith	Nodes only	Leaves only	Pith only
Cellulose (%)	45.5	47.7	41.4	44.2	47.6
Pentosan (%)	27.1	26.2	29.0	28.3	26.0
Pentosan in the cellulose (%)	11.0	12.6	13.1	9.4	13.7
Lignin (%)	16.7	16.4	19.8	14.6	13.3
Hot water solubility (%)	14.9	12.7	14.9	10.6	18.8
1% NaOH solubility (%)	47.6	40.2	-	-	-
Alcohol-Benzene solubility (%)	7.0	6.6	6.3	2.8	10.9
Ash (%)	6.9	6.0	6.2	10.3	5.8
SiO ₂ in ash (%)	63.2	39.9	29.4	72.0	24.1

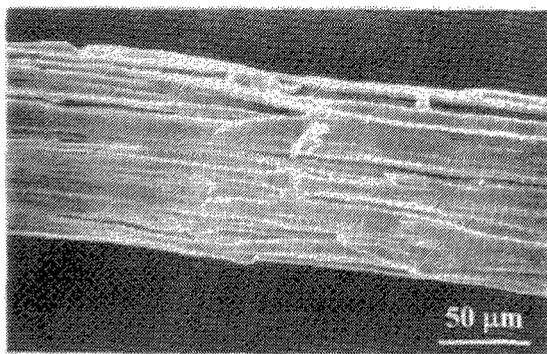


Fig. 4 Appearance of a maize fiber by SEM.

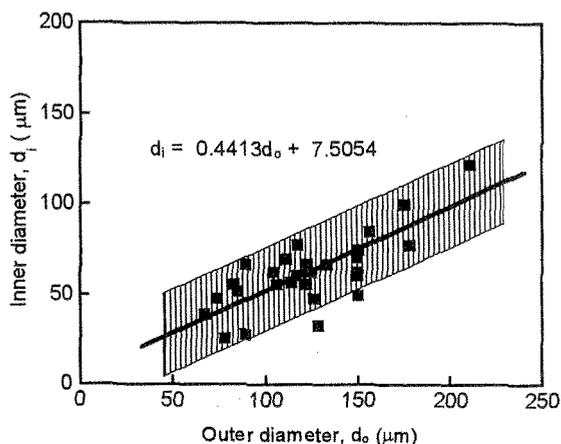


Fig. 5 Relationship between outer and inner diameters of maize fibers.

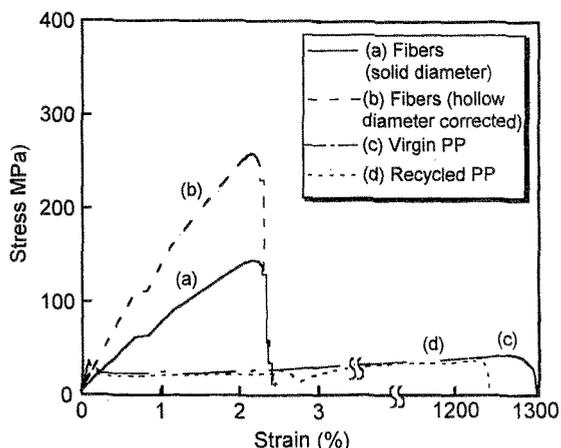


Fig. 6 Typical stress - strain curves of maize fiber and polypropylene matrices.

diameters in spite of the relatively large scatter band. The outer diameter was found to be approximately twice the inner diameter as noted by the trend linear equation in the figure.

Figure 6 shows typical stress-strain curves of a maize fiber together with the pure and recycled PP. Both the virgin and recycled PP exhibit almost similar tensile behavior with essentially equivalent tensile properties. On the other hand, the stress-strain curve for the fiber is characterized by an initial linearly increasing region followed by a short plateau region and then again

Table III Mechanical properties of maize and other natural fibers

Material/Property	Tensile Strength (MPa)	Elastic Modulus (GPa)	Elongation at Break (%)	Reference
Maize	256**	7.5**	2.5	Present work*
Jute	318	27.0	2.4	15
Wheat Straw	43	9.3	-	17

* Average value of 15 specimens

** Values for the diameter corrected by Eq. in Fig. 5

increasing stress - strain region. An occurrence of the unusual short plateau region in all the fibers tested might have been caused by slipping of the cardboard papers between the grips (chucks) of the tensile testing machine prior to an extended deformation. The overall behavior of the fibers is otherwise in accordance with their quasi-elastic nature, which can be explained as follows. As the applied stress increases, the weak primary cell wall collapses and decohesion of cells occurs because of the decohesion of cellulosic and non-cellulosic molecules mainly through weak links and imperfections. The applied stress also causes the uncoiling and extensions of the crystalline fibrils in the secondary walls of cells.

As noted in the preceding figure, the tensile strength of the fiber also approximately doubles when the diameter is corrected to encompass the hollow central lumen based on the linear equation of the trend line, as shown as curve b in Fig. 6. This suggests that higher strength should be expected if the inner hollow diameter will be taken into account. The mechanical properties of the maize fiber can be summarized in Table III in comparison with the other plant fibers⁽¹⁵⁾⁽¹⁷⁾.

3.2 Tensile properties of composites

The typical stress - strain behavior of the composites is shown in Fig. 7. It is observed that after the initial linear region, the composites yield and the molecules slip past one another resulting in a higher strain rate as indicated by the curvatures of the curves, which are more pronounced in B and C, probably due to the increased stress bearing capacity with higher fiber contents. These experimental values are however the nominal case. Thus, the true strength of the composites should be higher than the indicated values if the hollow diameter correction will be taken into account.

Figure 8 shows the effect of fiber area fraction on the tensile strengths of composites formed under 175°C - 10 MPa as well as pure PP and maize fiber subjected to no thermo-mechanical treatment, along with the data corrected by the hollow diameter consideration. Apparently, the tensile strength of composites tends to increase almost linearly with increase in the fiber area fraction, in accordance essentially with the rule of mixture (ROM). Pure fiber exhibits the highest strength but with large scatter band. The data scatter associated with the hollow diameter correction (Case B) depends strongly on the fiber area fraction. The higher the fiber area fraction, the more the disparity in fiber diameters due to the inherently non-uniform topography of the fiber surface. For evaluation of the true or net strength of the fibers, actual outer and inner diameters have been determined on the basis of the measurements conducted on the 30 maize fibers. Consequently, if the variation of

2.5 Composite forming

Composites were formed in relation with the investigated physical properties such as melting points of the polymer matrices, and the high temperature sensitivity of maize fibers. The die capable of producing 2 specimens at a time was designed to fit the specimen geometry of the ASTM Code D 638 (type V). Film stacking technique was used to produce the composites. In this process, the dried fiber sheets, which composed of the spread maize fibers prepared in section 2.2, were cut to the dimension of the die. Quantities (mass) of the dried fiber spread were measured to the composite loadings of 17, 30 and 38 mass %, corresponding to the fiber cross-sectional area fractions 25, 39 and 55% respectively, as shown in Fig. 3. The cut fibers were then arranged alternately with PP films in the specimen grooves contained in the forming die as shown in Fig. 2. The forming die was then inserted into the heating chamber on top of the lower plunger. The PP pellets were generously and evenly spread on top of the forming die to cover the alternately stacked fiber/PP films. The lower plunger was caused to gradually descend to the bottom of the heating chamber, followed by the upper plunger, which sandwich the forming die and cover the whole set up.

The heating process begins while the temperature of the fiber/PP films was monitored via thermocouples slotted onto the lower portion of the upper plunger (upper metal die). Stabilization period of 5 minutes is allowed when the maximum molding temperature was attained. Afterthen the pre-pressure of 1 MPa, which was determined experimentally to be sufficient, is applied to enable the entrapped air to escape and also to facilitate the impregnation or permeation of PP matrix among the fibers. This molding process is followed by air-cooling process at the rate approximately 4°C/min.

Full pressure is applied and held consistently when the upper die temperature lowers to 130°C, which seems to be so-called "semi-melting state" of PP matrix. This

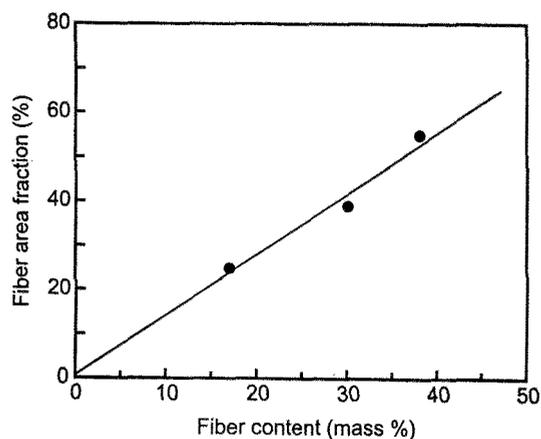


Fig. 3 Relationship between fiber content and fiber area fraction in the composites.

Table II Processing parameters for composite forming

Molding temperature (°C)	Forming pressure (MPa)	Fiber area fraction (%)
160, 175, 190	5 ~ 15	0 ~ 55

pressurizing temperature was determined as an appropriate temperature for the composite forming through a series of trial experiments, and probably lies in the real temperature range between 160 and 140°C for PP matrix⁽¹⁶⁾. It was revealed that full pressuring at temperatures higher than this brings about a leakage of the PP matrix, while the contrary results in the voids and uneven surface of the composite specimen due to the premature solidification of the PP matrix. The fitting tolerance selected and the extent of pressure application can minimize an excessive damage to the fibers. The pressure can be transmitted to the fibers indirectly via the softened PP. A small amount of the excess PP that comes out through the gap between the upper die and the heating chamber solidifies as it comes in contact with the cooling air to prevent further over-flow, because of the pressure exerted on the composites. The specimens were removed by splitting the forming die when the temperature falls to ambient. The thermo-mechanical processing parameters adopted in this study are summarized in Table II.

2.6 Tensile testing

The mechanical behavior of the fiber, PP and the composites were examined by the tensile testing. For the maize fibers, specimens were prepared by carefully fixing the ends of single fibers on stiff cardboard pieces 50mm apart. SEM photographs of the outer and inner diameters were taken in order to measure the actual diameters of the fibers. Since the fiber diameter varies along the length, an average of more than about 5 readings were taken for the diameter determination. Tensile testing of the maize fibers was carried out at 0.1 mm/min crosshead speed by using the universal testing machine with a loading capacity 10kN, which is connected to a computer that reads the output for subsequent data treatment.

Tensile tests for the PP and composites were conducted of at least more than 2 specimens under the same condition except for the crosshead speed of 1mm/min and 0.3 mm/min, respectively, using the same universal testing machine. Scanning electron microscopy (SEM) was conducted for examining the microstructural characteristics of the fiber and composites. All specimens were gold sputted prior to the SEM examination for giving the sufficient electrical conductivity.

3. RESULTS

3.1 Tensile properties of maize fiber and polypropylene

Figure 4 shows the surface appearance of the maize fiber indicating both non-uniformity of the cross-section and variation of diameter along the axis, which suggests the generally uneven surface topography characterized by the natural fiber. Diameter of fibers, when considered as perfectly spherical (solid diameter), was found to vary approximately from 65 to 210µm, which provides wide differences in the tensile strength between 81 and 211 MPa, in average 141 MPa, as denoted later.

Figure 5 shows the resulting relationship between the outer and inner hole diameters determined through the cross-sectional measurements for more than 30 maize fibers, suggesting the linear relation between the both

the diameters of the fibers is taken into account, the true or net strength of the maize fiber should fall within the scatter band indicated in the figure.

In general, an average error of the data resulted from the tensile testing of the composites stood at 5.3% when more than 2 specimens were used under the same testing condition. While for only a few condition yielded a relatively large data scatter beyond 10%, then additional specimens were adopted for testing under the same condition; these data are then represented with scatter band in the figures. Apparent elastic modulus, which can be obtained from the gradient of an initial linear stage of the stress-strain curve, may be used where necessary.

The overall effect of the fiber area fraction on the tensile properties of composites is summarized in Fig. 9. Afterthere, the nominal stress derived from the solid fiber diameter will be denoted. In the case of same forming pressure of 10 MPa, an increase in the fiber area fraction results in the increase of both tensile strength and elastic modulus, especially at 175 and 190°C molding temperatures for composites containing 39 and 55% fiber area fractions. When the same molding temperature of 175°C is considered, both the tensile strength and elastic modulus increase with increase in the fiber area fraction, particularly under the forming

pressure of 10 and 15 MPa. A remarkable increase in the elongation at break is noted for 55% fiber area fraction in the case of 10 and 15 MPa. This should be attributed to the improved interfacial adhesion between the fibers and PP matrix.

Figure 10 illustrates the effect of molding temperature and forming pressure on the mechanical properties of composites. Under the same forming pressure of 10 MPa, an increase in the molding temperature is accompanied by an increase in the tensile strength, especially at 175°C, but it tends to level off in spite of further increased molding temperature. This signifies that 175°C is sufficient for the composite manufacturing. Preliminary experiments also revealed the thermal degradation of fiber at temperatures above 190°C, which is mostly followed by a reduction in the tensile strength, as also reported by the others⁽¹⁵⁾⁽¹⁸⁾.

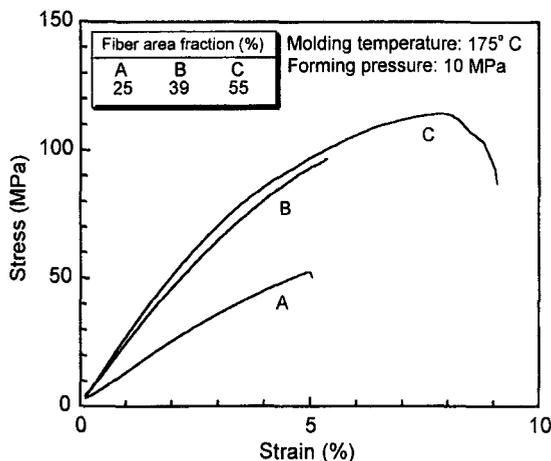


Fig. 7 Nominal stress - strain curves of composites.

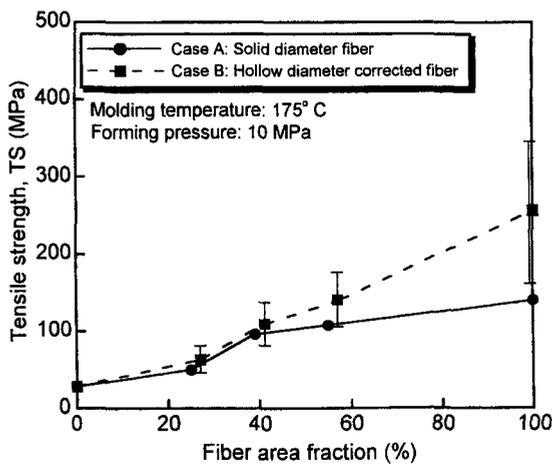


Fig. 8 Effective strength of maize fiber.

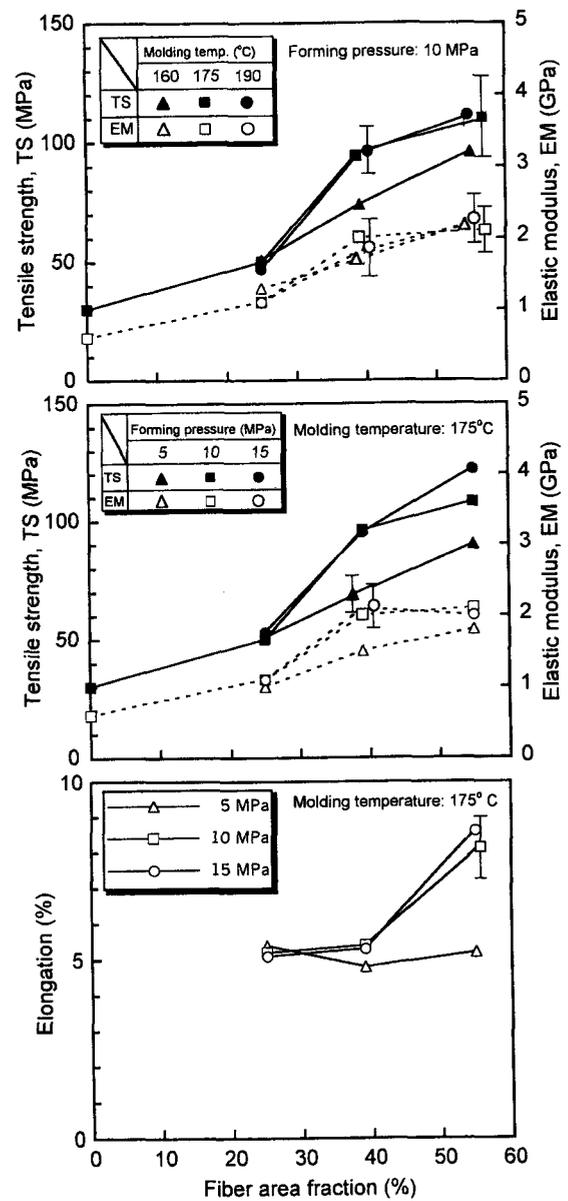


Fig. 9 Effect of fiber area fraction on the tensile properties of composites.

In the case of same molding temperature of 175°C, similarly, both the tensile strength and elastic modulus tend to increase with an increase in the forming pressure except for the composite with minimum fiber area fraction of 25%. The results suggest that a beneficial effect of the molding temperature and forming pressure are pronounced only for the composites with dense fiber content of more than 39% area fraction.

As regards the effect of PP matrix origin, there is no

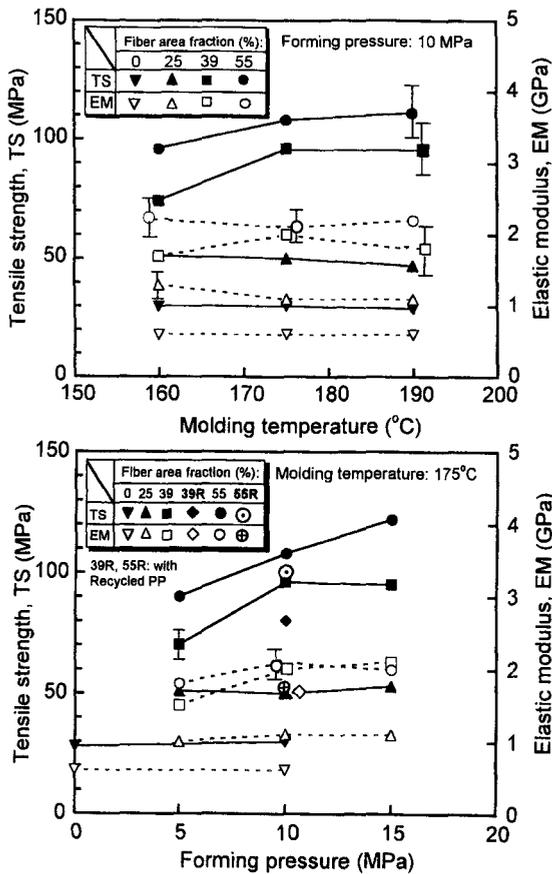


Fig. 10 Effect of molding temperature and forming pressure on strength and modulus.

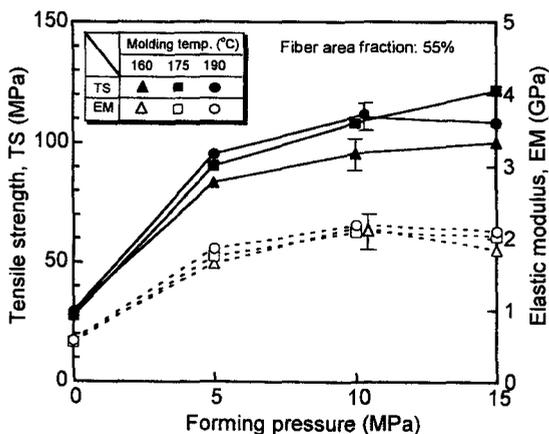


Fig. 11 Effect of forming pressure and molding temperature on strength and modulus of the composites with 55% fiber area fraction.

significant difference in the both tensile strength and elastic modulus between the composites with virgin PP and recycled one which has been denoted with "R" at the corresponding fiber area fraction in Fig. 10. Therefore, it should be noted for the recycled PP also to be available competitively as the matrix material for the high performance composite.

The effect of the processing conditions on the mechanical properties of the strongest composites with 55% fiber area fraction is summarized in Fig. 11. Tensile strength increases effectively with increase in the forming pressure, particularly for the composites molded at 175 and 190°C, suggesting the possible sufficient temperature regime for manufacturing the property-enhanced composites. However, an appreciable fall in the tensile strength is noted for the composite prepared under the 190°C molding temperature and 15 MPa forming pressure, which is presumably due to an excessive fiber damage. On the other hand, an elastic modulus shows almost equivalent value regardless of the molding temperature.

Such an explicit difference in the behavior between the tensile strength and elastic modulus with respect to the processing conditions should be explained by considering the character of both properties that the tensile strength is structure-sensitive, while the elastic modulus is rather structure-insensitive and depends dominantly on the content of the maize fiber.

3.3 Microstructural examination of composites

Microstructural examination using SEM technique was conducted mainly at the interfaces of composites having 55% fiber area fraction, formed under 10 MPa at different molding temperatures, in order to investigate the actual interaction between the maize fibers and PP matrix. This result is shown in Fig. 12.

The high value of the tensile strength for composite specimen molded at 175°C, as shown in the previous figures, may be resulted from a combination of the improved fiber wettability by the polymer matrix and appropriate forming pressure, as seen in Fig. 12 (b). Careful observation of the fiber/matrix interface revealed an intimate contact between them. Conversely, insufficient melting of the polymer matrix by molding at 160°C resulted in a poor interfacial adhesion between the fiber and matrix with a number of gap, as shown with arrow in Fig. 12 (a). On the contrary, fiber damage resulting from thermal degradation is noticeable for the composite by 190°C molding, as shown in Fig. 12 (c), in where large gap exists probably due to a thermal shrinkage on solidification.

Furthermore, an effect of the forming pressure variation on the integrity of composite also was examined for those containing 55% fiber area fraction, molded at the same 175°C molding temperature. The result is indicative that the composites formed under 5 MPa contains gaps at the fiber/matrix interface due to the inadequate compaction, while fiber damage resulting from an excessive pressure application was observed in the case of composites formed under 15 MPa.

Figure 13 shows a SEM photomicrograph of the typical fracture surface of the composite specimen prepared under the optimum conditions. The rough fiber surface depicts for the fiber to have been pulled out less

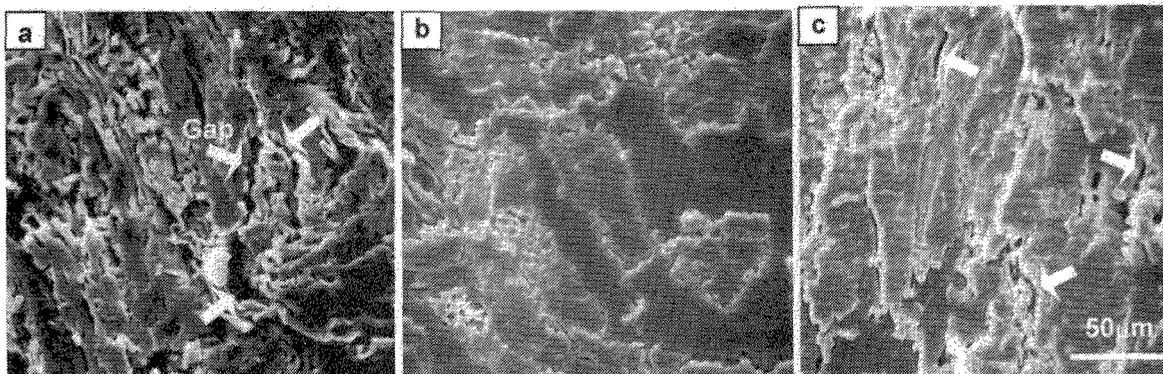


Fig. 12 SEM photomicrographs of the cross section of composite specimens with 55% fiber area fraction, molded at (a) 160°C, (b) 175°C and (c) 190°C (forming pressure: 10 MPa). Arrow in the photograph indicates a presence of gap.

easily from the PP matrix, providing a clear evidence of enough load transfer between the fiber and PP matrix. Similarly, careful examination at the fracture surface of specimen revealed no gap at the base of protruding fibers, which also provides an evidence of good adhesion. These facts support that the maize fibers are possible to contact intimately with PP matrix, provided for an appropriate thermo-mechanical processing.

In summary, the microstructural examination also signifies that an optimum condition for manufacturing the maize fiber/PP composites lies around the molding temperature 175°C and the forming pressure 10 MPa.

4. DISCUSSION

There may be two factors principally responsible for the improved strength properties of the maize fiber/PP matrix composites. Firstly, an improved strength of the maize fiber is closely related to the fiber extraction methodology adopted. The NaOH utilization imparts a surface modification (activation) of the fibers; it leads to the formation of a rough surface that can improve the mechanical interlocking between the fiber and PP matrix. There is an evidence that NaOH treatment removes the massive lignin and hemicellulose that affect the tensile characteristics of the fibers⁽¹⁹⁾. Fibrils rearrange themselves along the direction of the tensile deformation when the hemicellulose is removed. While a removal of lignin makes the middle lamella that joins the ultimate cell wall more plastic and homogeneous. Hence, the maize fibers can improve effectively the stiffness and strength of pure PP matrices. The fibers are main load bearing components of the composites, and the fiber strength dominates normally the strength of whole composite. The PP matrix is responsible for holding the maize fibers in place and, more importantly, transferring evenly the load from fiber to fiber through the interface⁽²⁰⁾.

Secondly, the processing methodology used for manufacturing the composites must have immensely contributed in acquiring such good composite properties. For example, the application of 1 MPa pressure, pre-pressuring, when the preset molding temperature is reached enables the permeation of the PP matrix through the fibers. Afterthen, full pressurization when the molding temperature lowers to about 160~140°C exerts the pressure with inducing little damage to the fibers.

Generally, the mechanical properties of the maize fiber/PP composites increase with increasing the fiber

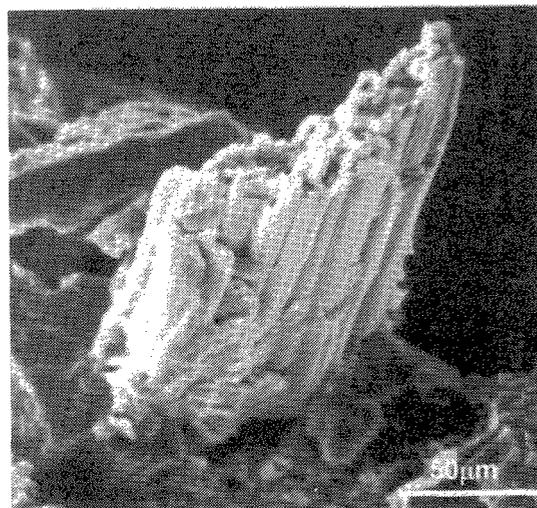


Fig. 13 Typical SEM photomicrograph of the fracture surface for composite specimen with 55% fiber area fraction, molded at 175°C and formed under 10 MPa.

contents, and were found to be invariably independent of temperature and pressure increase at low fiber contents. The properties were also found to be generally low at the lower temperatures and pressures; likewise, they also tend to drop at the higher temperatures and pressures. Such a strength reductions may be related to the fiber deformation due to the thermo-mechanical influences. A gap between the maize fiber and PP matrix also should cause the strength reduction by virtue of insufficient load transfer.

On a general note, an optimum condition for manufacturing the high performance composites as well as the associated degradation factors are summarized in Fig. 14. As experimentally determined from the foregone tensile tests results and microstructural examination, the composites with favorable properties should be manufactured at 175°C molding temperature under 10 MPa forming pressure, which specifies the optimum conditions. Preparing composites outside the optimum conditions may result in any of the following scenarios. At lower molding temperatures, insufficient melting of the PP matrix results in the inadequate permeability of PP among the fibers to be poor formability of composites. On the other hand, the

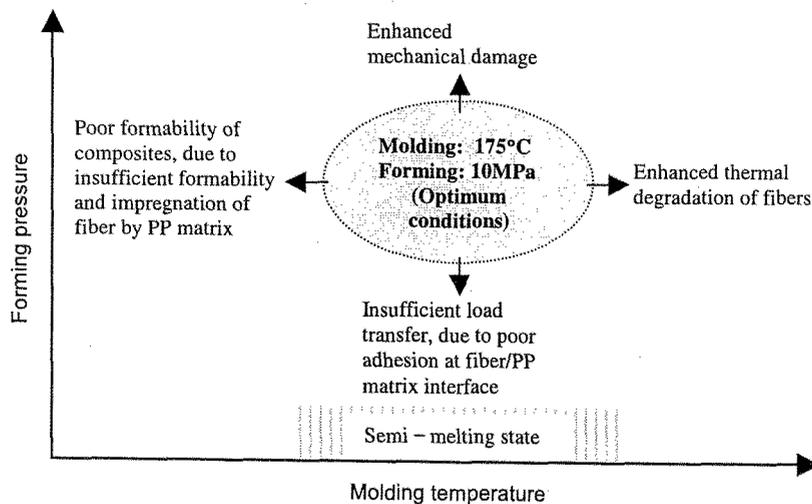


Fig. 14 Optimum condition and degradation factors for composite manufacturing.

molding at higher temperatures should bring about an enhanced thermal degradation of fibers, such as burning and excess shrinkage on solidification, as well as partly of the remaining lignin. Similarly, forming under low molding pressures results in the insufficient load transfer due to poor adhesion at the fiber/PP matrix interface. However, an enhanced mechanical damage such as crushing of fibers may also occur when composites will be molded under higher pressures.

5. CONCLUSIONS

- (1) Long maize fiber/PP composites with reasonable mechanical properties have been successfully manufactured using specially designed hot press machinery.
- (2) Two factors were traced to be responsible mainly for the favorable mechanical properties of the composites. Thus, the use of NaOH for fiber extraction was found possible to improve the fiber wettability with PP matrix, and the method of composite processing that incur little or no damage to the fibers was established.
- (3) The optimum conditions for the high performance composite manufacturing have been specified to be around 175°C and 10 MPa.
- (4) Only little difference in the tensile properties was observed for the composites with recycled PP and virgin PP, suggesting a possibility of manufacturing the composites with equivalent mechanical performance even by using the recycled PP instead of virgin one.

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