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Processing and Mechanical Properties of Long Bagasse Fiber Reinforced Composites with Recycled Polypropylene Matrix

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Mechanical properties of composites consisting of the recycled polypropylene matrix reinforced with up to 66% fiber cross-sectional area fraction of unidirectionally oriented long bagasse fibers have been evaluated. Processing parameters such as the molding temperature and forming pressure were varied with respect to the fiber area fraction with the aim of verifying the optimum manufacturing conditions. Improved mechanical properties were found to be prominent at 175°C and 10 MPa, above which the properties tend to deteriorate. Tensile strength tends to increase monotonically depending on the increase in cross-sectional fiber area fraction with the exception of pure bagasse fiber of which strength becomes rather lower. Factors responsible for such deviation were traced and discussed on the basis of the bagasse fiber characteristics. Microstructural analysis of the composites was conducted predominantly at the fiber/matrix interface using scanning electron microscopy (SEM) and the results were discussed in some detail.

Key words: Materials recycling, Composite material, Long natural bagasse fiber, Recycled polypropylene, Mechanical properties

1. INTRODUCTION

The recent environmental consciousness, legislative requirements and economic concerns are stimulating research for the re-utilization of agricultural wastes, particularly of the tropical non-woody fibrous wastes. A reasonable and integrated recycling system, which should optimize the utilization of these lignocellulosic wastes through converting them into the value-added new materials, has been developed in an earlier study [(1)].

Together with molasses, bagasse is a by-product of the sugar cane industry. About 50% of the total annual production of bagasse is usually burnt to generate steam in the sugar industries [(2)(3)]. Therefore, the surplus 50% should be fully utilized in manufacturing the value-added new materials such as composites.

The existing works on natural fiber – polymer matrix composites consist of mostly commercial/textile fibers such as flax, jute, sisal, etc. and thermosetting binders such as phenol formaldehyde and urea formaldehyde [(2)(4)(5)], whose emissions are subjected to very severe legislation. Hence, a shift of paradigm in developing the natural fiber-polymer matrix composites using the non-woody agricultural wastes that generates annually from agricultural practices in combination with commodity-based recycled polymer matrix is desperately required.

A recent study on the long maize fibers and virgin polypropylene (PP) composites without usage of any coupling agents proved a reasonable improvement in mechanical properties. The method of fiber extraction and sophisticated processing technique adopted has largely contributed in achieving the interesting results [(6)].

The aim of this work is therefore to both manufacture and evaluate the mechanical properties of the low cost and value-added new composite materials composed of the long bagasse fibers and recycled PP matrix without coupling agents. The microstructural aspect was especially investigated in some detail to determine the adhesion at the fiber/matrix interface of the composites.

2. EXPERIMENTAL

2.1 Characteristics of bagasse fiber

Bagasse, from the sugar cane stalks consists of the pith fraction derived from the thin-walled cells of the ground tissue dispersed throughout the interior of the stalk, the rind/sheath, which is a thick-walled, relatively long, fibrous fraction, and the epidermis which contains waxes and other materials, as shown by SEM in **Fig. 1.** Bagasse contains about 50% fibers, 30% pith or parenchyma cells, about 15% short, less resistant fiber bundles and vessels that are scattered throughout the interior pith portion, and about 5% epidermis [(7)].

The chemistry of bagasse reveals that it contains about 35% cellulose, 30% hemicellulose, and about 20% lignin. Bagasse also contains extractives, some residual sugars, pentosans, hexasans, and other reactive low molecular weight products [(8)]. **Table 1** [(7)] shows the chemical constituents of the whole bagasse together with the chemical analysis and reactivity.

2.2 Extraction and preparation of bagasse fiber

The bagasse was procured from the local subtropical island source in Kagoshima prefecture, Japan, during the spring 2001. The fibers were extracted and prepared according to the methodology denoted in the authors' previous work [(6)].

2.3 Preparation of polypropylene for matrix

The recycled PP is of plain and commercial type with MFI of about 10g/10min, 0.9g/cm^3, which was supplied by a petrochemical wholesaler company. Thermal

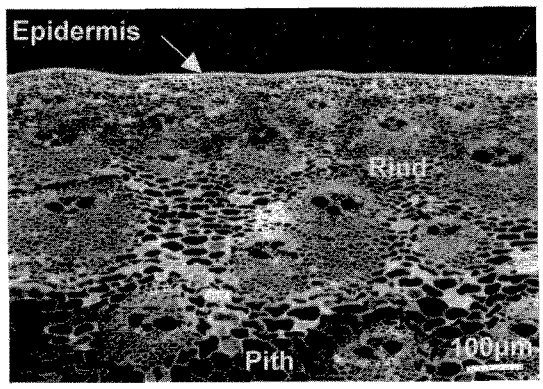

Fig. 1 Cross sectional microstructure of bagasse.

Table I Chemical analysis of bagasse and anatomic elements [7]

Constituents and reactivity	Whole bagasse	Depithed fiber	Pith only
Cellulose (%)	31.8	38.3	31.5
Pentosan (%)	27.7	30.7	33.0
Lignin (%)	21.3	21.1	20.0
Hot water solubility (%)	5.7	2.4	1.5
1% NaOH solubility (%)	33.9	28.8	30.8
Alcohol-Benzene solubility (%)	3.2	3.6	2.1
Ash (%)	5.4	2.0	3.3

investigation on the recycled PP and method of film preparation are described in the previous study [6].

2.4 Composite forming

The details of the composite manufacturing facility and the methodology of composites forming have been fully described in the previous study [6]. Composite specimens of ASTM Code D 638 (type V) geometry, with nominal gage length of 25mm, were manufactured with fiber loadings of 17, 31, 39 and 51 mass% corresponding to the fiber cross-sectional area fractions 22, 39, 50 and 66% respectively [6], as shown in **Fig. 2.** The thermo-mechanical processing parameters are listed in **Table II.**

2.5 Tensile testing

The mechanical behavior of the fiber, recycled PP and the composites were examined by the tensile testing. The bagasse fiber bundles specimens were prepared by carefully fixing their ends on stiff cardboard pieces with gage lengths of 50, 25 and 12.5mm apart. The actual outer and inner diameters of the fibers with central lumen were measured by SEM. For the outer diameter determination, an average of at least 5 readings were taken along the length of each fiber in order to cater for the diameter variation. Tensile testing of the bagasse 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 composites together with the recycled PP specimens with nominal gage length of 25mm were conducted for at least more than 2 specimens under the same condition at the crosshead speed of 0.3 mm/min using the same universal testing machine. Microstructural examination was conducted

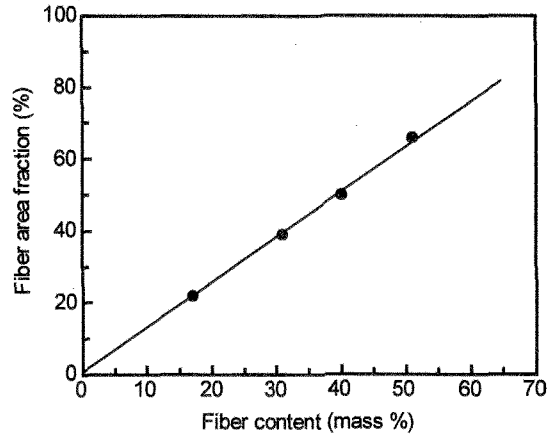

Fig. 2 Relationship between fiber content and fiber cross-sectional area fraction in the composites.

Table II Processing parameters for composite forming.

Molding temperature (°C)	Forming pressure (MPa)	Fiber area fraction (%)
160, 175, 190, 205	5 ~ 15	0 ~ 70

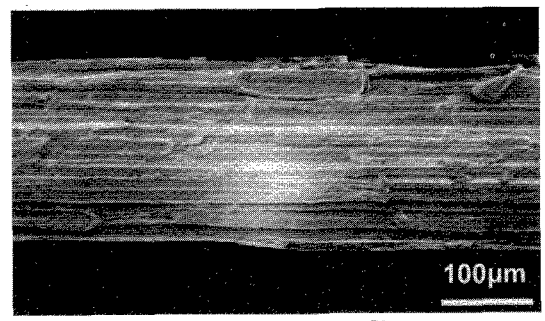

Fig. 3 Appearance of a bagasse fiber by SEM.

mainly at the fiber/matrix interface of the composites using SEM after gold sputting.

3. RESULTS

3.1 Tensile properties of bagasse fiber and recycled polypropylene

Like other natural fibers, the surface of the bagasse fiber is characterized by a general uneven morphology as shown in **Fig. 3.** The diameters of the fibers, when considered as perfectly spherical (solid diameter) range from approximately 125 to 235μm providing difference in the tensile strength from 80 and 186 MPa with an average value of 136 MPa, as depicted later.

The relationship between the outer and inner hole diameters determined from the cross-sectional SEM measurements conducted on more than 30 bagasse fibers is summarized in **Fig. 4.** This suggests a linear relationship between the both diameters even though with relatively large scatter band. The inner diameter was found to be small and less dependent on the outer diameter as compared with the case of maize fiber [6].

The typical stress-strain curves of bagasse fiber together with the recycled PP are shown in **Fig. 5.** The recycled PP is characterized by low strength with very high ductility. Contrarily, the stress-strain curve of the fiber is characterized by an initial linearly increasing region followed by an extended deformation. The overall

behavior of the bagasse fibers otherwise manifested their quasi-elastic nature that can be explained as follows. Increase in the applied stress brings about straining of the hitherto intact fiber bundle. As the stress gradually increases, at first, the weak primary cell wall of the weaker fibers tends to collapse prematurely and decohesion of cells occurs owing to the decohesion of the cellulosic and non-cellulosic molecules chiefly through weak links and imperfections. The applied stress also causes the uncoiling and extension of the crystalline fibrils in the secondary walls of cells.

The tensile strength, as also noted in Fig.5, did not show any remarkable increase even if the diameter is corrected to encompass the hollow central lumen based on the linear equation of the trend line, shown as curve b

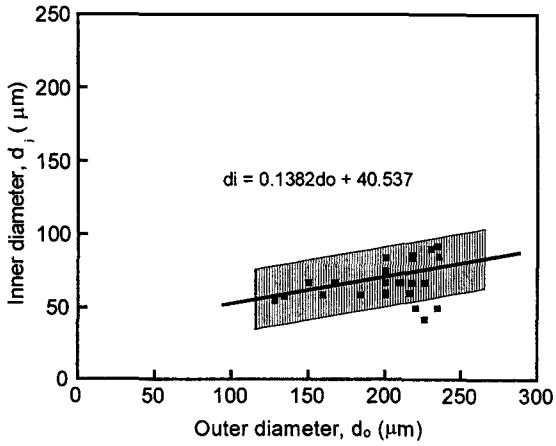

Fig. 4 Relationship between outer and inner diameters of bagasse fibers.

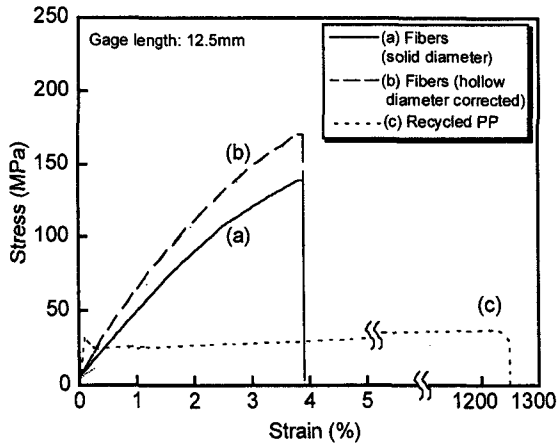

Fig. 5 Typical stress-strain curves of bagasse fiber and recycled polypropylene matrix.

Table III Mechanical properties of maize and other natural fibers.

Material/ Property	Tensile Strength (MPa)	Elastic Modulus (GPa)	Elongation at Break (%)	Ref.
Bagasse	169 **	5.5 **	3.8	Present work*
Maize	256	7.5	2.5	6
Jute	318	27.0	2.4	9
Wheat Straw	43	9.3	-	10

* Average value of 6 specimens
** Values for the diameter corrected by Eq. in Fig. 4

in Fig. 5. This suggests that only a little improvement in the strength should be expected if the hollow diameter is taken into account. The mechanical properties of the bagasse fiber are summarized in **Table III**, in comparison with other plant fibers [6][9][10].

3.2 Tensile properties of composites

Figure 6 shows the typical stress – strain curves of the composites. As can be seen, the composites yield after the initial short range of linear regime, and the molecules slip past one another resulting in a higher strain rate as indicated by the gradual curvatures. Such a behavior is more pronounced in C and D with higher fiber fraction, possibly due to the increased fiber-to-fiber interaction, stretching of bent and misaligned fibers. Increase in the tensile strength of the composites with higher fiber content seems to be associated with the increased elongation at break except for that with the lowest fiber fraction, since curves of B and C tends to trace up to half way the curve D. This suggests that an advantage of increasing the fiber fraction in composite is directly related to the improved rupture ductility.

The influence of the fiber area fraction on the tensile strengths of composites processed under 175°C – 10 MPa are shown in **Fig. 7**, together with pure recycled PP and bagasse fiber that are not subjected to thermo-mechanical treatment. The tensile strength increases

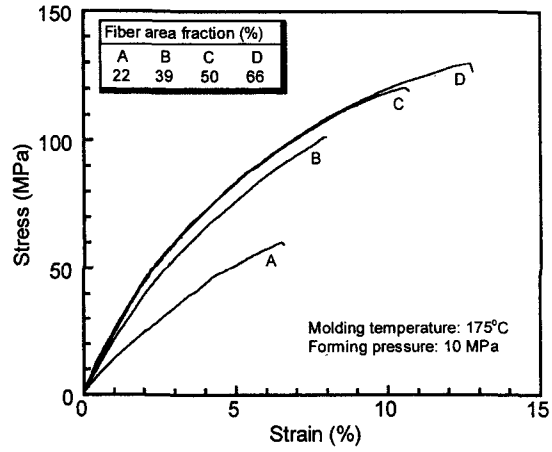

Fig. 6 Nominal stress-strain curves of composites.

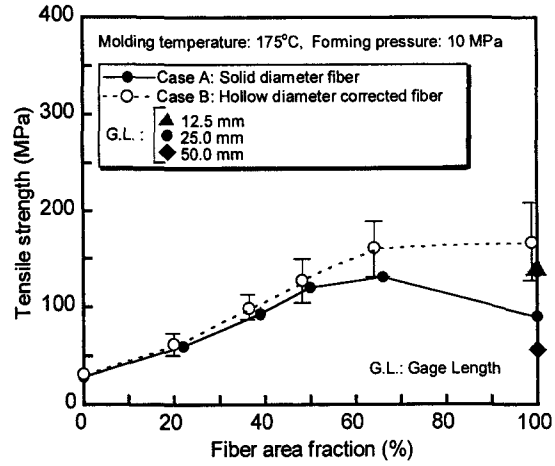

Fig. 7 Dependence of bagasse fiber strength on length.

monotonically with the increase in fiber area fraction up to 50% of fiber fraction. However, further increase in the fiber fraction was found to result in the strength leveling off or rather degradation, which gives rise to the much lowered strength of the pure fiber. Pure fiber tends to exhibit a relatively higher strength with small scatter band when the shorter gage length of such as 12.5 mm is adopted. As noted in the figure, the strength of the bagasse fiber should depend strongly on the length rather than the hollow diameter correction. The reason can be explained as follows; the numerous micro-fibers having a mean length of about 2 mm and nodes dividing the fibers into individual cells are interconnected by lignin to form a long strand of fibers. The regions of interconnectivity and flaws or defects that are intrinsically present in the fiber are generally weak, thus the longer the gage length, the more the number of such imperfections which translates to higher probability of failure. This phenomenon is observed when the gage length is increased from 12.5 to 25 and 50 mm. The hollow diameter correction (Case B in Fig. 7) produced no significant improvement in the tensile strength of the fiber by virtue of the small central lumen, with a data scatter that depends strongly on the fiber area fraction in the composites. The inherently non-uniform fiber surface topography provides disparity in fiber diameters, which increases with increase in fiber area fraction.

Figure 8 shows the overall effect of fiber area fraction on the tensile properties of composites. Hereafter, nominal stress derived from the solid fiber diameter will be denoted, since the strength increment by considering the central lumen of fiber is less pronounced. In the case of same forming pressure of 10 MPa, both tensile strength and elastic modulus increase almost steadily with increasing fiber area fraction up to 50%. However, apart from the molding temperature of 175°C, which showed a slight improvement beyond the 50% fiber area fraction, the tensile strength leveled off despite the increase in fiber area fractions to 66% for the other molding temperatures. This indicates that the molding temperature 175°C probably provides sufficient heating for the polymer to flow and penetrate among the fibers without inducing thermal degradation for them. On the other hand, no elastic modulus was influenced by the variation in the molding temperature, especially for composites containing 50 and 66% fiber area fractions. When the same molding temperature of 175°C is considered, a similar trend as in the forming pressure was noted. Except for the 10 MPa forming pressure that produces an improvement in the tensile strength beyond 50% fiber area fraction, both tensile strength and elastic modulus increase steadily with increase in the fiber area fraction up to 50% for all forming pressures. The tensile strength either levels off or shows only an insignificant improvement beyond 50% fiber area fraction for composites formed under 5 and 15 MPa, suggesting inadequacy of compaction and fiber damage, respectively.

A significant improvement in the elongation at break was observed for 50% fiber area fraction for all the forming pressures, especially for 10 MPa. The improvement must have resulted partly from the improved interfacial adhesion between the fibers and the recycled PP matrix, and partly from the stretch of the entangled fibers embedded in the matrix. However, the

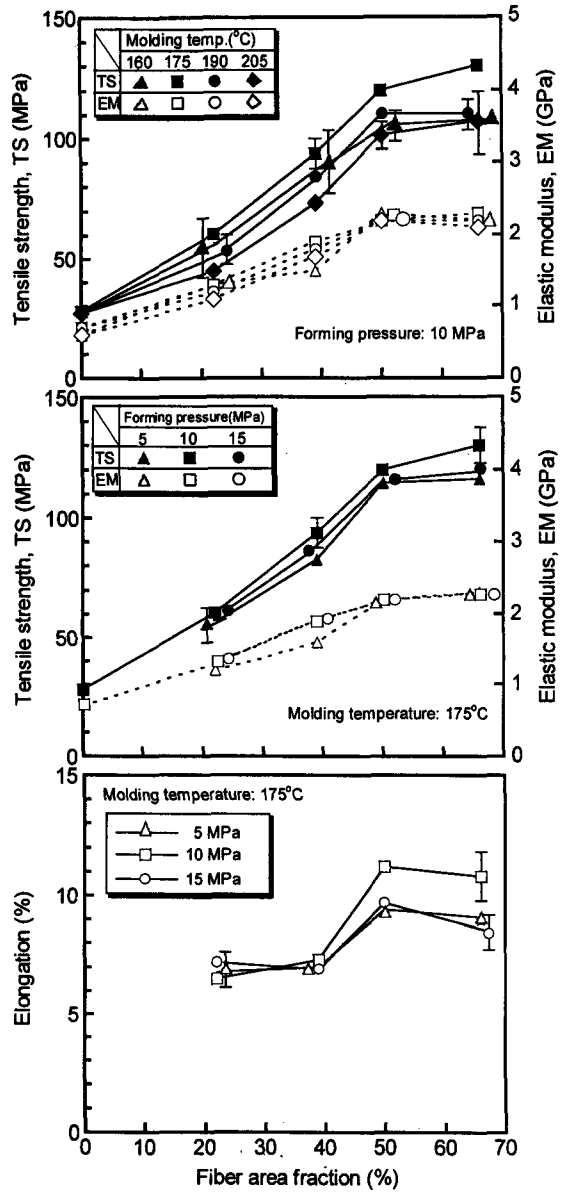

Fig. 8 Effect of fiber area fraction on the tensile properties of composites.

elongation showed rather decreasing tendency beyond the 50% fiber area fraction. Such a reduction is probably caused by insufficient fiber impregnation by the recycled PP matrix, resulting in the inadequate interfacial adhesion with less load transfer.

Figure 9 represents the effect of the processing conditions on the mechanical properties of the strongest composites containing up to 66% fiber area fraction. The most improved tensile strength was revealed to be obtainable for such a dense fiber composite under the processing condition of 175°C molding temperature and 10 MPa forming pressure. Contrarily, the elastic modulus shows almost equivalent value irrespective of the molding temperature.

Such an explicit behavioral differences between the tensile strength and elastic modulus with regard to the processing condition should be explained by considering the character of both properties that the former is structure-sensitive, while the latter is rather structure-

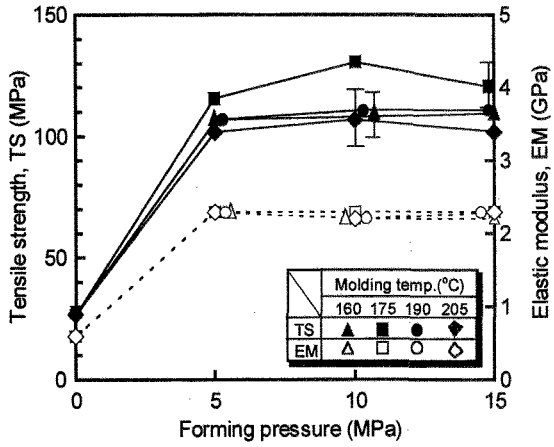

Fig. 9 Effect of forming pressure and molding temperature on strength and modulus of the composites with 66% fiber area fraction.

insensitive hence depends dominatingly on the bagasse fiber content.

3.3 Microstructural examination of composites

The microstructural examination conducted mainly at the fiber/matrix interface using SEM is shown in **Fig. 10.**

As noted in the previous figures, the composite specimen molded at 175°C produced the highest tensile strength. Such an improvement may be attributed to the sufficient wetting of the fiber by the recycled PP matrix in combination with an appropriate forming pressure, as shown in Fig. 10 (b). On the contrary, insufficient melting of the polymer matrix at 160°C imparts only poor adhesion between the fiber and the matrix, hence producing a number of gaps, as indicated with arrows in Fig. 10 (a). Also, fiber damage resulting from the thermal degradation must have been responsible for the drop in the tensile strength for composites molded at 190 and 205°C, where large gaps exists presumably due to the thermal shrinkage on solidification as shown in Fig. 10 (c, d). It is noteworthy that the fibers in Fig. 10 (d) have almost lost their fibrous structure due to thermal destruction; burning, encountered during processing.

Further investigations with respect to the forming pressure variation was also conducted on the composite with 66% fiber area fraction, molded at the same 175°C molding temperature. It was apparent that composites formed under 5 MPa hardly attained an adequate compaction thereby revealing numerous gaps at the fiber/matrix interface. Applying much higher forming pressure such as of 15 MPa, on the contrary, resulted in the mechanical destruction of fiber.

A SEM photograph of the typical fracture surface for the composite specimen prepared under the optimum conditions is shown in **Fig. 11.** The rough fracture surface of fibers indicates that a substantial work had been done as the fibers have been pulled out during the fracture process. This signifies that there was sufficient load transfer between the fiber and recycled PP matrix. However, meticulous examination at the fracture surface of the specimen revealed some gaps at the base of the protruding fibers, which suggests that despite the evidence of good load transfer, the adhesion between the fiber and the matrix is still inadequate.

The above microstructural examination agrees with the earlier tensile testing results that validate the molding temperature 175°C and forming pressure 10 MPa as the optimum condition for manufacturing the bagasse fiber/recycled PP composites.

4. DISCUSSION

Bagasse fiber/recycled PP composites produced the apparently improved tensile properties presumably by means of the fiber extraction and composite manufacturing methods adopted, which impart less damage to the fibers, as also observed previously with

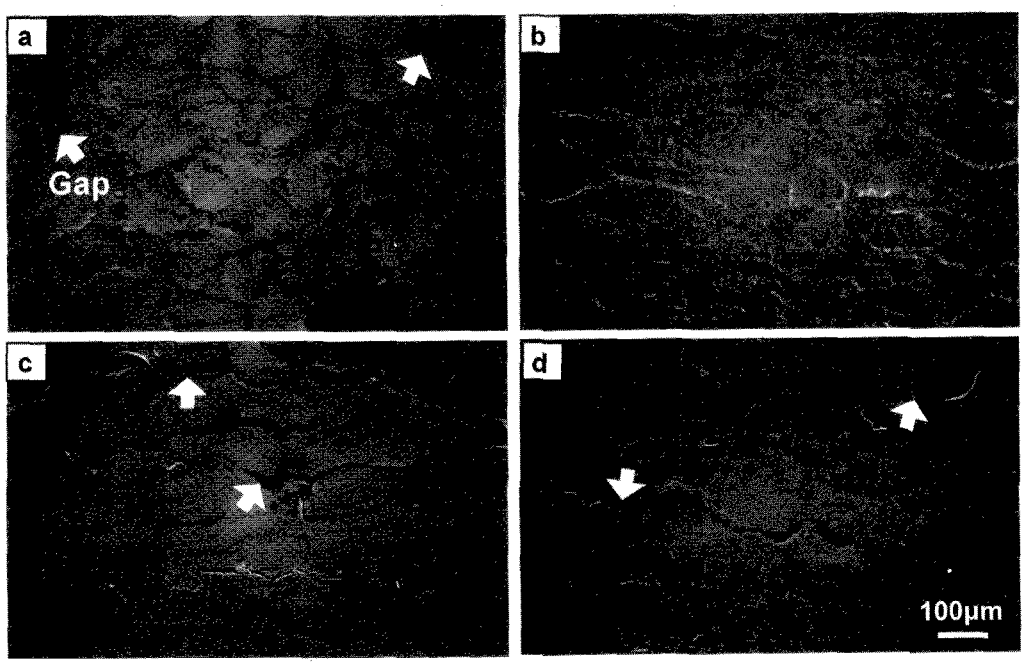

Fig. 10 SEM photographs of the cross section of composite specimens with 66% fiber area fraction, molded at (a) 160°C, (b) 175°C, (c) 190°C and (d) 205°C (forming pressure: 10 MPa). Arrow in the photograph indicates a presence of gap between the fiber/matrix interface.

Fig. 11 Typical SEM photograph of the fracture surface for composite the specimen with 66% fiber area fraction, molded at 175°C and formed under 10 MPa. Arrow indicates gap.

damage to the fibers, as also observed previously with maize fiber/virgin PP composites [6]. It was noted that the mechanical properties of lignocellulosic fibers could be increased by about 40% by reducing the damage resulting from fiber processing or extraction [11].

The strength of bagasse fiber was found to depend strongly on its length rather than the hollow diameter correction. The reason will be explained as follows. Bagasse fiber appears to have larger diameter and relatively shorter fiber cells [7] with numerous lignin-rich interconnecting nodes and flaws. Therefore, the increased strength resulting from the shorter gage length may be linked with the fewer number of the intrinsically present defects in the fiber and reduced number of such weak nodes, which possibly reduces the probability of failure.

The properties of the recycled PP matrix were immensely improved by incorporating the bagasse fibers. The combine effect of the load bearing capacity of the bagasse fibers held in place by the recycled PP matrix produced successfully even transfer of the load from fiber to fiber through the interface.

The tensile strength of the composites indicates the most improved strength at the fiber area fraction of about 50 and 60%, above which deterioration of strength was observed. Therefore, 50% fiber area fraction should be the optimum fiber loading for the composite system. Inadequate impregnation of the recycled PP matrix among the fibers due to the incorporation of high fiber area fraction in the composite reduces the load transfer ability of the composite system, which consequently affects the strength.

The mechanical properties of the bagasse fiber/recycled PP composites were generally found to increase with increase in fiber area fraction. The improvement at low temperature and pressure may be accredited to the exclusive nature of the matrix material used. The recycled PP itself contains some impurities, and the addition of the fibers must have contributed in altering its flow and thermal characteristics.

The insufficient interfacial adhesion between the fiber and the matrix as noted in Fig. 11 may be caused by the low melt flow index of the recycled PP. This problem could be solved by either increasing the stabilization period from 5 to 10 or more minutes for molding time during composite manufacturing or by using some sort of coupling agent.

The foregone tensile test and microstructural analysis results suggest that the composites with enhanced properties should be manufactured at the optimum conditions of 175°C molding temperature under 10 MPa forming pressure. Composites formed outside these conditions will experience any of the following setbacks, as also noted in the case of maize fiber/virgin PP composites [6]. Then, molding at lower temperature is accompanied by insufficient melting of the recycled PP matrix thereby affecting the impregnation of the matrix among the fibers, which consequently results in poor formability of composites. Conversely, an enhanced fiber damage through the thermal degradation such as burning of fibers and partly of the residual lignin, excess shrinkage on solidification will characterize composites molded at higher temperatures. Similarly, the negative influence of forming at lower pressure will result in insufficient load transfer due to poor compaction. On the other hand, molding at higher pressures will boost mechanical damage such as crushing of the fibers.

5. CONCLUSIONS

(1) Value-added composites consisting of long bagasse fiber and recycled PP have been successfully manufactured at the optimum conditions of molding temperature 175°C and 10 MPa forming pressure

(2) The method of fiber extraction and sophisticated composite processing methodology adopted in this study confirmed their suitability and adaptability for manufacturing of the value-added composites.

(3) Composites of competitive mechanical properties could be manufactured using recycled PP instead of the virgin one.

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