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Processing and mechanical property evaluation of maize fiber reinforced green composites

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Abstract—Green composites composed of long maize fibers and poly ε-caprolactone (PCL) biodegradable polyester matrix were manufactured by the thermo-mechanical processing termed as 'Sequential Molding and Forming Process' that was developed previously by the authors' research group. A variety of processing parameters such as fiber area fraction, molding temperature and forming pressure were systematically controlled and their influence on the tensile properties was investigated. It was revealed that both tensile strength and elastic modulus of the composites increase steadily depending on the increase in fiber area fraction, suggesting a general conformity to the rule of mixtures (ROM), particularly up to 55% fiber area fraction. The improvement in tensile properties was found to be closely related to the good interfacial adhesion between the fiber and polymer matrix, and was observed to be more pronounced under the optimum processing condition of 130°C molding temperature and 10 MPa forming pressure. However, processing out of the optimum condition results in a deterioration in properties, mostly fiber and/or matrix degradation together with their interfacial defect as a consequence of the thermal or mechanical damages. On the basis of microstructural observation, the cause of strength degradation and its countermeasure to provide a feasible composite design are discussed in relation to the optimized process conditions.

Keywords: Materials recycling; biodegradable plastics; natural fibers; composite materials; mechanical properties.

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1. INTRODUCTION

Recently, green composites have emerged as attractive alternatives to conventional fiber reinforced composites (FRC), due to the rising awareness of environmental issues, global sustainability, industrial ecology, eco-efficiency, and depletion of petroleum resources coupled with stringent environmental regulations. Hence, a paradigm shift in favor of new materials and products that are compatible with the environment and independent of fossil fuels, especially for applications in composite structures, is being considered seriously. In this case, the use of epoxy, unsaturated polyester, or phenolics with reinforcing glass, carbon or aramid fibers for manufacturing composite structures are considered as critical [1]. Alternative materials from renewable sources that are deemed to offer significant contributions in support of the strongly desired global sustainability are regarded as possible replacements. For instance, the viability for the re-utilization of agro-based residues, particularly those derived from the non-woody fibrous crop wastes that are annually produced in abundance should be given high priority, because the majority of this material ends up being burned in the field or simply landfilled [2, 3]. However, researches that involve the re-utilization of crop wastes such as maize stalks, rice and wheat straws have been found to be grossly inadequate, and perhaps this is responsible for their grossly underdeveloped practical re-utilizations [2].

Except for the non-woody agro-based waste fibers, several publications on agro-based fiber–polymer composites could be found involving the extensive utilization of both thermoplastic and thermoset materials with mostly hemp, jute, flax, kenaf, sisal, bamboo, etc., that are referred to as hard or commercial fibers. These have an identified economic value and already established application areas, particularly in the automotive industry [4–7].

An attempt to strengthen the global sustainability scenario using a much more reasonable methodology that aims at maximizing the benefits obtainable from the agro-based wastes was developed by the authors. It suggests the utilization of their fibers in combination with other materials such as plastics to produce value-added composite materials [2]. The methodology was demonstrated in several papers by authors that feature maize and bagasse fibers in combination with both virgin and recycled polypropylene (PP) matrices [8–10]. The incorporation of such natural fibers in composites offers several advantages such as low density, good specific properties, non-abrasive nature, high level of filler loadings, availability, renewability and a safe working environment [3].

Since the non-renewable portion of the above composites precludes complete biodegradation, development of such completely biodegradable composites or green composites becomes quite indispensable. The aforementioned view has prompted many researches on the completely biodegradable composites with different biodegradable matrices and lignocellulosic fibers. Several examples of such green composites dealing with common biodegradable polymers such as poly(lactic acid) (PLA), poly(butylene succinate) (PBS), poly[(3-hydroxy-butyrate)-co-

(3-hydroxyvalarate)] (PHBV), etc. in combination with mainly commercial fibers such as jute, flax, etc., could be found in recent literature [11].

The main objective of this study, hence, is to establish the manufacturing methodology through looking for the optimum process condition on the basis of tensile property evaluation and failure analysis of the green composites comprising long maize fiber/PCL matrix without coupling agents for the fiber/matrix interface.

2. EXPERIMENTAL

2.1. Extraction and preparation of maize fiber

The maize stalks were acquired from a local source in Nagano prefecture, Japan, during the summer of 2005. A fiber extraction methodology that guaranteed minimized damage to the fibers with least chemical utilization, energy consumption and overall cost was adopted in order to optimize the fiber properties [4]. Furthermore, the chosen solvent of sodium hydroxide (NaOH) provides adequate surface treatment for the maize fiber [12].

The internodes of the maize stalk (hard outer layer) were separated from the pith, and heated for 90 min at 95°C under atmospheric pressure in 6 g/l of NaOH aqueous solution in a ceramic pot-like container with high liquor to material ratio (15–20:1). Finally, sheets of fibers were obtained based on the procedure described in the authors' previous study [8]. The fundamental physical and mechanical properties of the maize fibers could be summarized as follows: density of 1.5 g/cm^3, average diameter of 145 µm with a range of 60–220 µm, average tensile strength and elastic modulus of 141 MPa and 6.3 GPa, respectively [8].

2.2. Characterization and preparation of polymer matrix

The PCL, Celgreen ($PHBO_2$), manufactured by Daicel Co. Ltd., Japan [13], was adopted in this study. The principal physical and mechanical properties of the polymer matrix are summarized in Table 1. A table-type hot press was used to prepare the initial film with approximately 100 µm thickness from the pellets at 190°C molding temperature and 1 MPa forming pressure. Thermal investigations on the Celgreen was conducted by means of both differential scanning calorimetry (DSC) and thermogravimetry — differential thermal analyzer (TG-DTA), to determine the melting point and thermal decomposition characteristics, respectively. The melting and thermal degradation temperatures of the $PHBO_2$ grade of Celgreen was determined as 113°C and approximately 300°C, respectively, as shown together with other mechanical properties in Table 1.

2.3. Composite processing

A specially designed cylindrical heating unit type molding and forming facility was used in manufacturing the composite specimens with geometry in accordance with

Table 1.
Physical and mechanical properties of Celgreen [13]

Property	Standard (JIS)	Grade (P-HBO$_2$)
Density (g/cm^3)	–	1.21
MFR (190°C–2160 g) (g/10 min)	K7210	2.7
Melting point* (°C)	K7121	113.0
Tensile yield strength (MPa)	K7113	26.0
Tensile rupture strength (MPa)	K7113	36.0
Elongation at break (%)	K7113	360.0
Young's modulus (MPa)	K7113	265.0

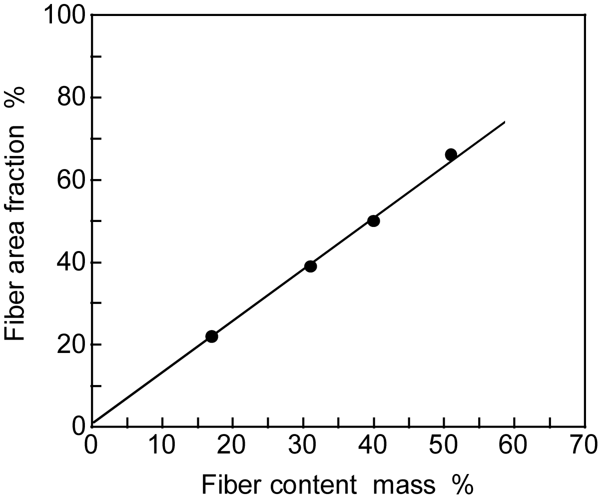


Figure 1. Relationship between fiber content and fiber cross-sectional area fraction in the composites.

ASTM Code D 638 (Type V). Film stacking technique was used to produce the composites with fibers oriented unidirectionally. The process involves alternating the dried fiber sheets with Celgreen matrix film, which are then prepared and inserted into the die. Quantities (masses) of the dried fiber spread were measured to the composite loadings of 17, 30, 38 and 51 mass%, corresponding to the fiber cross-sectional area fractions at the gage lengths of 25, 39, 55 and 66% respectively, as shown in Fig. 1. Composites specimens were prepared on the basis of a technique termed 'Sequential Molding and Forming Process' that was newly developed by the authors and explained in detail in the previous study [8]. Pressure application is required at two different temperatures in this process, i.e. the pre-set molding temperature and the 'semi-melting state' temperature of the matrix material. First and foremost, the stacked matrix film–maize fiber alternate arrangements are inserted into the die prior to the heating process. Then, 1 MPa pre-pressure is applied after allowing the equalization and stabilization period of 5 min at the attainment of the pre-set molding temperature. The pre-pressure was determined experimentally and is considered to be sufficient to dislodge entrapped air and to facilitate the percolation of the matrix through the fibers while ensuring sufficient infiltration of polymer among the fibers and adequate contact at the fiber/matrix interface. The subsequent cooling process was begun after the application of the pre-pressure. The final and full forming pressure was applied continuously at the semi-melting stage of the polymer matrix, which lies within a temperature regime of *ca.* 100–90°C for the Celgreen matrix. The pressure is maintained until consolidation is attained *via* air-cooling at the rate of approximately 4°C/min. Using this technique, both the molding temperature and forming pressure were varied as relevant parameters for the composite manufacturing; 115, 130, 160 and 190°C, and 5 or 10 MPa, respectively.

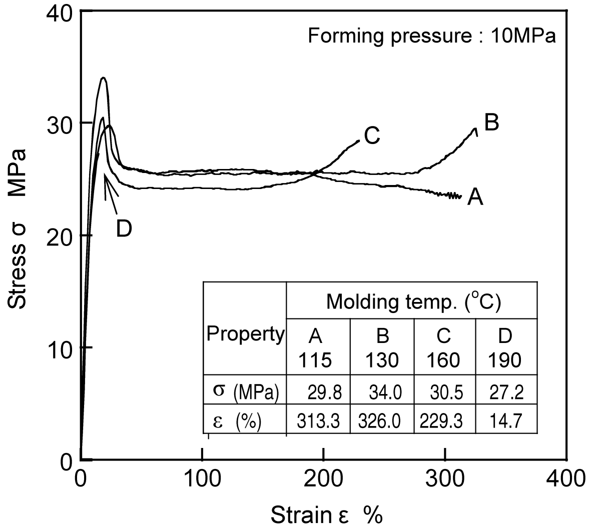
2.4. Tensile testing

A universal testing machine with a loading capacity of 10 kN was used in order to examine the mechanical behavior of the composites in the uniaxial direction together with the maize fiber and Celgreen matrix *via* tensile testing. In order to conduct tensile testing of the maize fiber, fibers with a gage length of 25 mm were securely glued between hard cardboard papers at both grip ends and allowed to dry perfectly before storing in desiccators. According to the preliminary testing, both single fibers and fiber bundles produced nearly the same results. Then, the testing was conducted for more than 15 specimens of the single maize fibers at a crosshead speed of 0.1 mm/min. On the other hand, 5 specimens with 25 mm gage length and 3.2 mm thickness were tested for the dumb-bell shaped composites and Celgreen matrix under the same condition except for 0.3 and 1 mm/min, respectively. Scanning electron microscopy (SEM) was conducted in order to reveal the microstructural characteristics of the fiber and composites failure. All the specimens were gold sputtered prior to the SEM examination in order to provide adequate electrical conductivity.

3. RESULTS AND DISCUSSION

3.1. Tensile properties of polymer matrix

Figure 2 shows stress–strain curves of the polymer specimen for the matrix formed under 10 MPa pressure and at molding temperatures 115, 130, 160 and 190°C. As seen in the figure, all specimens with the exception of D show greatly increased strains of more than 200% with extensive plastic deformation producing a yield strength of approximately 30–34 MPa, which is indicative that PCL matrix material behaves in a way that is consistent with other thermoplastic materials such as PP. However, D exhibited a fracture at low ductility with the lowest yield strength of about 27 MPa, which may be attributed at least partially to thermal degradation of the matrix.



Property	Molding temp. (°C)			
	A 115	B 130	C 160	D 190
σ (MPa)	29.8	34.0	30.5	27.2
ε (%)	313.3	326.0	229.3	14.7

Figure 2. Typical stress–strain curves of Celgreen matrices.

Except for the specimen at 190°C, all the specimens elongate by more than 200%, especially for 130°C which extends beyond 300%, suggesting an important property of matrix materials that aids uniform load transfer function in the composites.

3.2. Tensile properties of composites

Figure 3 shows the typical stress–strain behavior as a function of fiber area fraction for the composite specimens manufactured at the presumed most optimum and attractive processing condition; 130°C molding temperature and 10 MPa forming pressure. Although the highest strength was obtained in the composite containing the highest fiber area fraction of 66%, its rupture ductility lagged behind the composite containing 55% fiber area fraction. This behavior may be caused by mechanical damage inflicted on the fiber on account of high forming pressure at high fiber loading in the composite.

The mechanical properties of the composites together with pure Celgreen matrix and maize fiber are summarized in Table 2. The increased fracture strain of the composites, which is far beyond the fiber fracture strain, is not very clear to the authors at the moment. However, possible reasons may involve the consequence of the methodology used for strain determination that was based on the crosshead displacement. Furthermore, substantial strain was absorbed at the initial stage of the tensile loading of composites, in which stage the softer matrix concerns dominate the strain increment, whereas fibers experience alignment and straightening without contributing to the load bearing until sufficient strain is attained. The unified and simultaneous stress bearing ability of both the fiber and matrix in the composite specimens should have appeared after the apparent strain, which occurred at the initial stage of the tensile loading. Consequently, the net strain experienced in

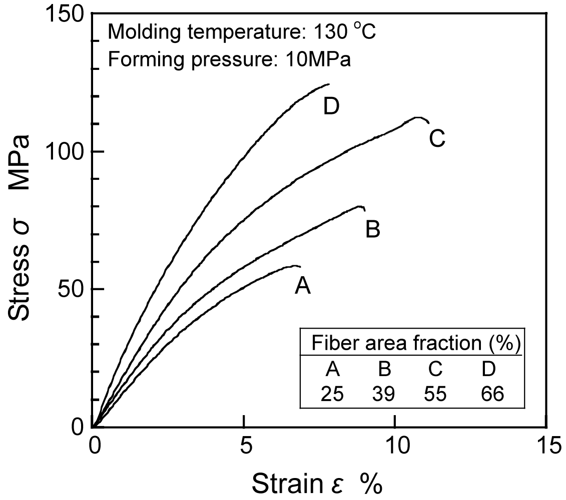


Figure 3. Nominal stress–strain curves of composites.

Table 2.
Some mechanical properties of composites with different fiber area fractions (molding temp. 130°C and forming pressure 10 MPa)

Fiber area fraction (%)	Tensile strength[**] (MPa)	Elongation[**] (%)
0 (Pure PCL)	26.0	422.0
25	68.1	8.6
39	110.0	12.9
55	114.5	13.0
66	121.5	8.4
100 (Fiber only)[*]	121.0[*]	3.3[*]

[*] Av. value of 17 specimens.
[**] Av. value of 5 specimens.

the composites may not be as high as the indicated values. A typical example of the influence of the fiber area fraction on the tensile strength of the composites that were processed with molding temperature 115°C or 130°C and forming pressure 5 MPa is shown in Fig. 4. Maize fiber exhibited the highest strength but with a large scatter band, which is acceptable in the realm of natural resources. It is also indicative from the figure that all the composite specimens showed gradual improvement in both stress and strain as the fiber area fraction increases in accordance with the rule of mixtures (ROM), especially up to 55% fiber area fraction. The slight decline in rupture ductility observed at 66% fiber area fraction may be associated with poor fiber wettability by the minority polymer matrix at the maximum fiber loading, hence probably resulting in poor load transfer function. It is worth noting that even though both fiber and composites were tested with the same gage length, the strength level attained at a higher fiber content of more than 55% nearly equals the

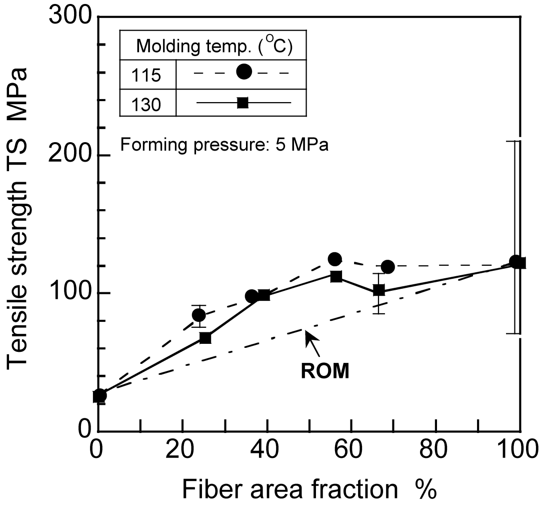


Figure 4. Summary of the tensile strength of composites together with maize fiber.

average strength level of the fiber. The exact reason why such higher fiber content composites have tensile strength that is comparable to that of the fiber is under investigation. However, there is a possibility that pure fiber rupture prematurely, especially at the weakest points that are usually found around the nodes made up of cork and silica cells. In composites, however, such nodes are well accommodated and bound by the matrix material in the composites thereby allowing fracture only at higher strain.

Figure 5 presents the summary of the overall effect of fiber area fraction on the tensile properties of composites. It is noteworthy from Fig. 5(a) that the composite manufactured at 130°C showed steady improvement in the rupture strength up to the maximum fiber area fraction of 66%. With such an exception of the composite manufactured at the 130°C molding temperature, almost all composites manufactured at the same forming pressure of 10 MPa showed increase in both tensile strength and elastic modulus with increasing fiber area fraction up to 55%, beyond which the properties either diminish or show minimal improvement. In general, the strength improvement was noted between 39 and 55% fiber area fractions for all composites; the improvement is most pronounced in composites manufactured at 130°C and to a lesser extent at 115°C, which also competed closely but the improvement significantly declined at 66% fiber area fraction. On the other hand, composites manufactured at 160°C and 190°C proved to be rather less competitive as compared to the previous ones. The elastic moduli of the composites displayed almost the same behavior as the tensile strengths, where increase in fiber area fraction is associated with increase in the elastic modulus up to 55% fiber area fraction. However, non-uniformity of the results was exhibited at the maximum fiber area fraction of 66%, which is inevitable from lowering of the ductility. Based on these results, in spite of the large data scatter exhibited by the composites

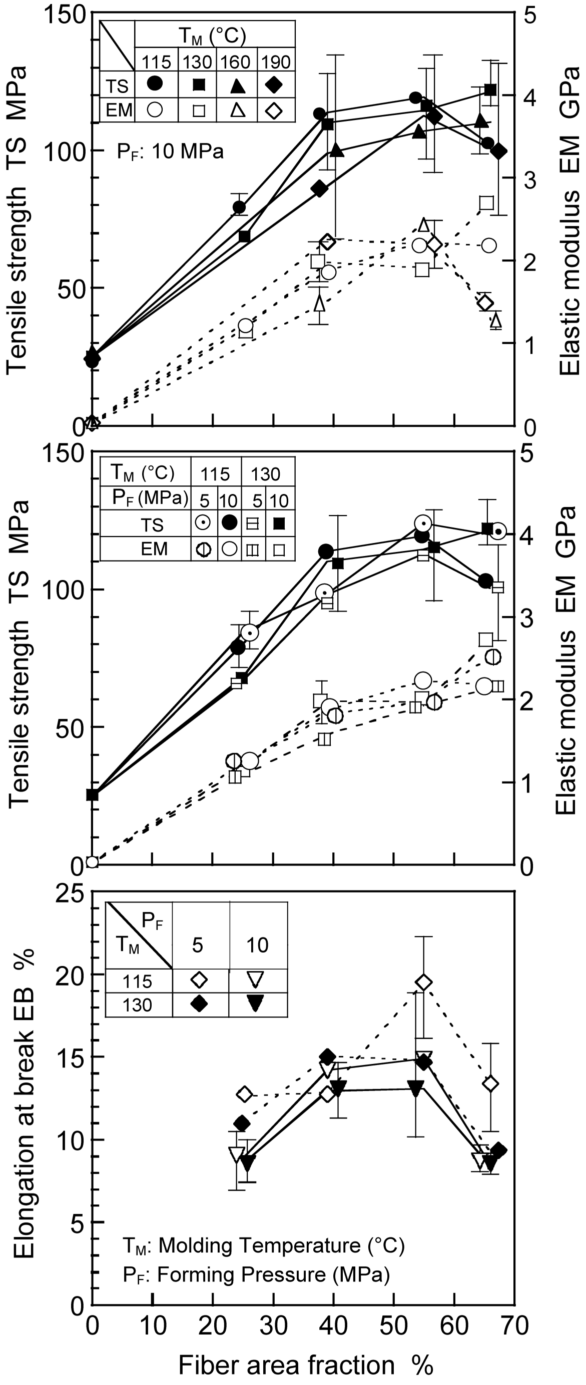


Figure 5. Effect of fiber area fraction on the tensile strength of composites.

manufactured at the maximum fiber area fraction, reasonable mechanical properties could be obtainable with the 66% fiber area fraction provided for the improved manufacturing methodology.

In the case of the competitive molding temperatures of 115 and 130°C, both tensile strength and elastic modulus increase monotonically with increasing fiber area fractions up to 55%, especially for composites manufactured at 115°C molding temperature and 5 and 10 MPa forming pressure, as shown in Fig. 5(b). However, only the composite manufactured at 130°C and 10 MPa showed continued property improvement beyond 55% fiber area fraction, even though tensile strength drastically falls in the remaining composites. The elastic moduli of all composites, however, showed steady increase with increase in fiber area fractions except for the composites molded at higher temperature more than 160°C, as shown in Fig. 5(a).

On the other hand, all composites exhibited higher elongation at break, especially at 39 and 55% fiber area fractions before slumping at the maximum fiber area fraction of 66%, as shown in Fig. 5(c). The trend is not unexpected since the highly loaded composites displayed relatively low load transfer function, thereby permitting fibers to slip off during plastic deformation. The composite manufactured under 5 MPa forming pressure and 115°C molding temperature showed rather complex and abnormal behavior, producing the highest elongation at 55% fiber area fraction. This behavior may be related to fiber orientation in the composite that requires longer displacement to straighten or align the bend fibers and/or the continued load transfer function that simultaneously allow stronger fibers to withstand load prior to final failure. It is obvious that manufacturing composites at higher temperatures such as 190°C will promote thermal degradation of not only the Celgreen matrix material as noted in Fig. 2, but also for the maize fibers, which often results in the decline of tensile strength, as previously reported by other researchers [1]. In contrast, applying forming pressure above 10 MPa will inflict mechanical damage on the fibers in the form of distortion or collapse of the lumen, as noted in the previous authors' work [8] and other studies [14].

3.3. Optimization of the processing condition of green composites based on failure analysis

The strengthening methodology for the composites of higher fiber contents is considered in order to optimize the processing condition by unveiling the degradation factors and suggesting possible counter-measures. Figure 6 shows the scenario illustrated on the basis of the microstructural examination and failure analysis conducted primarily at the fiber/matrix interfacial region by means of optical and scanning microscopes.

Ideally, fibers should be well embedded in the matrix with no gaps existing and with proper uniform distribution as noted in Fig. 6(a), where the fibers maintain their fibrous identity. Under current processing condition, fiber distribution is fairly heterogeneous so as to prevent the uniform load transfer function by matrix, even if the composite is prepared at the optimum process conditions of 130°C and

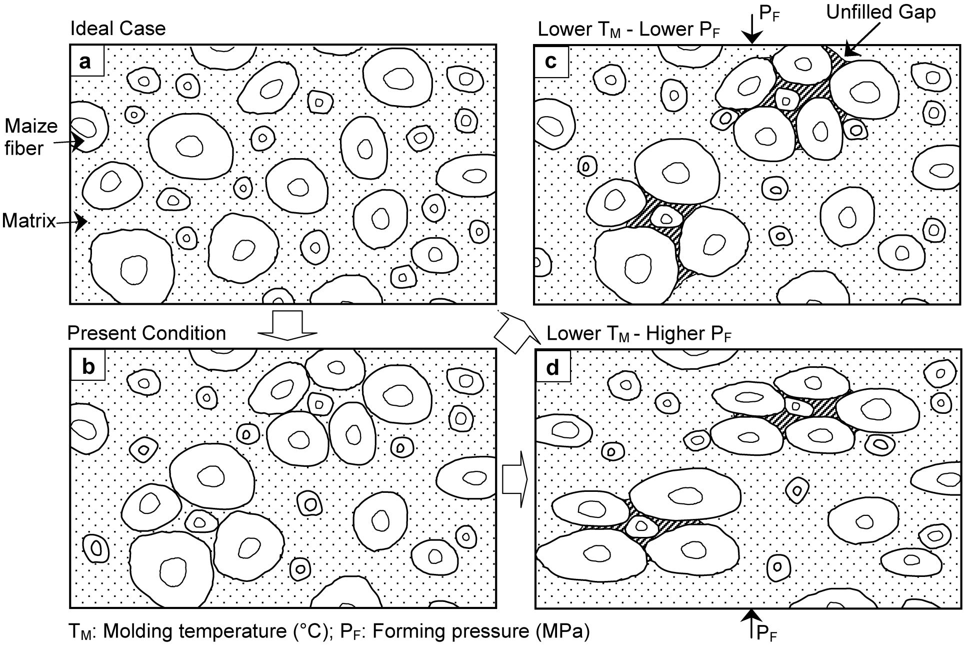


Figure 6. Schematic illustration depicting the influence of processing condition on the microstructures and mechanical properties in the case of higher fiber contents green composites.

10 MPa, as shown in Fig. 6(b). It has been confirmed that the matrix has sufficiently melted and infiltrated through the fibers, adequately wetting the fiber surfaces, which signify the existence of good interfacial adhesion with no gap existing at the fiber/matrix interface, provided for the sufficient molding temperature and forming pressure.

On the other hand, the difficulty in attaining the ideal condition is further manifested in composites prepared at lower molding temperature and forming pressure. In this case, even though the fibers appear to be unaffected, several gaps exist at the fiber/matrix interface, possibly due to the inadequate melting of the polymer matrix that prevents full infiltration of the polymer matrix at the lower molding temperature and insufficient compaction of fibers at the lower forming pressure, as shown in Fig. 6(c). This negates the suitability for application of the composites manufactured at 115°C and 5 MPa despite the fairly good rupture strength obtained. Similarly, processing composites at lower molding temperature and higher forming pressure results in poor matrix permeation through the fibers coupled with high compaction, which proved to be detrimental to the fibers, as shown in Fig. 6(d). The fibers appeared to have been crushed as a result of the direct action of the high forming pressure without fiber wettability, especially at the central portion, hence leaving some gaps at the interfacial regions, as observed in composites manufactured at 115°C and 10 MPa. This suggests that the fiber/matrix interface show poor adhesion that may prohibit its reliability for application.

The aforementioned problems associated with the uniform distribution of fibers, maximizing fiber contents and adequacy of load transfer function for rupture strength improvement in the composites must be tackled in order to design composites with reasonable properties. For instance, a new methodology for composite processing that will achieve efficient fiber distribution in the composite as against the hand lay-up technique should be considered. Furthermore, to solve the problem involving insufficient matrix permeation at lower molding temperature, the equalization and stabilization period should be prolonged prior to the pre-pressure application.

4. CONCLUSIONS

The study successfully dealt with the processing and mechanical properties evaluation of green composites incorporating long maize fiber and Celgreen biodegradable plastic using a thermo-mechanical technique termed ‘Sequential Molding and Forming Process’ that was developed by the authors. The conclusions drawn are summarized as follows:

(1) In general, both tensile strength and elastic modulus of composites tend to increase with increase in the fiber area fraction, especially up to 55% fiber area fraction, in accordance with the rule of mixtures (ROM).

(2) Optimization of composites with improved properties that will permit high fiber loading, especially up to 66% fiber area fraction, could be achieved after modifying the processing methodology, especially to achieve fiber distribution, etc.

(3) The overall results revealed the optimum conditions for the manufacture of the green composites to be around 130°C molding temperature and 10 MPa forming pressure.

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