

Effect of Compatibilization on the Performance of Biodegradable Composites using Cotton Fiber Waste as Filler

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ABSTRACT: The usefulness of cotton waste as a source of reinforcing fibers for the preparation of cost-effective and biodegradable composites has been investigated. Biodegradable polyester (bionolle 3020) is melt-compounded together with cotton fibers. Maleic anhydride-grafted bionolle (bionolle-g-MA) is used as a compatibilizer. The grafting reaction is carried out in an intensive mixer in the presence of dicumyl peroxide as initiator. The effects of fiber and compatibilizer content as well as graft content are evaluated by mechanical property measurements and scanning electron

microscopy. The compatibilizer improved all mechanical properties significantly. Moreover, the water absorption and swelling of composites decreased, while the thermal stability increased slightly. Also, the biodegradation of the polyester bionolle 3020, as well as that of its composites with cotton fibers, were studied. © 2003 Wiley Periodicals, Inc. *J Appl Polym Sci* 88: 1825–1835, 2003

Key words: fibers; compatibilization; biodegradable; composites; polyesters

INTRODUCTION

The incorporation of natural fibers in thermoplastic polymers has recently attracted attention from scientists for two main reasons: fiber addition can lead to the reinforcement of the produced material and, as a substitute, fiber contributes to the solution of environmental problems caused by the disposal of large volumes of non-biodegradable materials.¹ Cellulose-based fibers are strong, light weight, low cost, abundant, renewable, nonabrasive, nonhazardous, and biodegradable.^{2–5} Their use in the production of composites has gained significant importance both in technical applications, such as the automotive industries,³ and in packaging industries for applications with minor strength requirements.⁶

The main disadvantage of natural-fiber-reinforced composites is the lack of good interfacial adhesion between fiber and matrix, which results in poor properties of the final material. Therefore, to develop such composites with good properties, it is necessary to decrease the hydrophilicity of the fibers by chemical modification or by the use of a compatibilizer. Chemical modification is usually obtained with reagents that contain functional groups that are capable of bonding to the hydroxyl groups of the fiber. Chemical treatments, such as dewaxing, acetylation, and chemical grafting, are used for modifying the surface prop-

erties of the fibers.^{7–11} Another effective way to improve the interface between fiber and matrix is the use of compatibilizers, which are usually graft copolymers of a polymeric matrix and an anhydride, such as maleic anhydride (MA). These reagents are compatible with the polymeric matrix and can react with the hydroxyl groups of the fiber to form covalent bonds. Both methods, modification of the fibers and use of compatibilizers, improve the stress transfer between the two components and lead to the improvement of mechanical and physical properties of the produced composites.

As already mentioned, the use of fully biodegradable polymers as a substitute for traditional nonbiodegradable synthetic polymers could contribute to the solution of the waste management problem. Biodegradable polymers constitute a loosely defined family of polymers that are designed to degrade through the action of living organisms.¹² Such biodegradable plastics that are commercially available include polycaprolactone (PCL), poly(lactic acid) (PLA), polyhydroxyalkanoates, poly(ethylene glycol) (PEG), and aliphatic polyesters, like poly(butylene succinate) and poly(butylene succinate-co-butylene adipate). The use of these materials is still restricted by their relatively high cost in comparison with commodity plastics, such as polyethylene and polypropylene. This situation makes necessary the use of low cost fillers as a way to reduce the cost of the end product. Many researchers have studied the incorporation of native and plasticized starch as filler in biodegradable polymers.^{13–16} Other than starch, natural fibers are also used as filler.

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TABLE I
Intrinsic Viscosity and Graft Content of Bionolle-g-MA Compatibilizers

Compatibilizer	Temperature (°C)	Reaction time (min)	MA (phr)	DCP (phr)	Graft content (wt%)	Intrinsic viscosity (dl/g)
C ₁	120	5	7	0.3	0.84	0.82
C ₂	140	5	7	0.7	1.72	0.68
C ₃	150	5	7	0.7	2.14	0.57

Although there are many studies concerning the use of fibers as filler for commodity plastics, their use with biodegradable matrices has not been extensively studied. Wollerdorfer et al. studied the influence of natural fibers on the mechanical properties of biodegradable polymers.¹⁷ They incorporated flax, ramie, jute, oil palm, and cellulose fibers in aliphatic polyesters, polysaccharides, and blends of corn starch with biodegradable polymers. Gatenholm et al. investigated the properties of composites of bacteria-produced polyesters reinforced with wood cellulose.¹⁸ Mohanty et al. studied the surface modification of jute and its influence on the performance of biodegradable jute/biopol⁶ and jute/polyester amide composites.¹² In addition, Nitz et al. tested the addition of wood flour and lignin in polycaprolactone and also the compatibilization of these composites with MA-grafted polycaprolactone (PCL-g-MA).¹⁹

The objective of this study is the production and characterization of cost-effective and totally biodegradable cotton fiber–aliphatic polyester (bionolle 3020) composites. The use of the copolymer bionolle-g-maleic anhydride as a compatibilizer and, in particular, the effect of its graft content and amount on the mechanical and physical properties of the composites are investigated.

EXPERIMENTAL

Materials

The commercial polyester bionolle® 3020 was kindly supplied by Showa Highpolymer Company, Ltd (Tokyo, Japan) [$d = 1.23 \text{ g/cm}^3$, $[\eta] = 0.96 \text{ dl/g}$]. Bionolle 3020 was used as the substrate of poly(butylene succinate-co-butylene adipate) (PBSA), which consists of succinic acid (S), adipic acid (A), and 1,4-butanediol (B), with an S/A/B composition ratio of 40:10:50.²⁰ Cotton cellulosic fibers, with fiber length of <1.8 cm, were a byproduct of a local spinning mill. Maleic anhydride (MA) (99%, Aldrich), dicumyl peroxide (DCP) (98%, Aldrich) and chloroform (Riedel-de-Haen) were used as received.

Grafting procedure

The grafting reaction was carried out in a Haake–Buchler Rheomixer, model 600, with roller blades and

a mixing head with a volumetric capacity of 69 cm³. The three components (bionolle 3020, MA, and DCP) were mixed by hand before being fed into the mixer. The amount of the polymer was ~60 g. Mixing conditions are presented in Table I. Melt temperature and torque were recorded during the mixing period.

Determination of graft content

The grafted polymer was refluxed in chloroform for 4 h, and the hot solution was filtered into cold methanol. The precipitated polymer was washed with methanol to remove any unreacted reagents, and then dried in a vacuum oven at 75°C for 24 h. The weight percentage of grafting reaction was determined by titration of the acid groups derived from anhydride functional groups.²¹

Preparation of composites

The fibers and the polymer were mixed in a Haake–Buchler Rheomixer. Prior to mixing, fibers were dried in a vacuum oven at 75°C for 24 h to avoid the formation of porous products. Blending was performed at 160°C for 10 min with a rotor speed of 60 rpm.

Viscosity measurements

The intrinsic viscosity of 0.5% w/v solutions of grafted polymers was measured at 30°C in a constant-temperature bath with a Ubbelohde viscometer.

Fourier transform infrared measurements

Fourier transform infrared (FTIR) spectra were acquired with a Perkin-Elmer Spectrum GX FTIR spectrometer. For each spectrum, 64 consecutive scans with 4 cm⁻¹ resolution were co-added.

Thermogravimetric analysis measurements

Thermogravimetric analysis (TGA) measurements were performed with a Shimadzu TGA-50 thermogravimetric analyzer. Each sample was heated at a rate of 10°C/min to 500°C.

Water absorption and thickness swelling measurements

Water absorption was determined according to the ASTM D570 method. The measurements were performed by soaking the samples in distilled water. At regular time intervals, each sample was removed from the water tank, dried by wiping with blotting paper, and subsequently weighed to determine water uptake. The samples were placed back in water after each measurement. The water absorption was calculated as the weight difference and is reported as percent increase of the initial weight.

For thickness swelling measurements, the samples were immersed in distilled water for 10 days and, after drying the surface, the thickness was measured again. The thickness swelling was calculated according to the following formula:

$$\text{Thickness swelling} = \frac{T_{\text{wet}} - T_{\text{dry}}}{T_{\text{dry}}} \times 100$$

where T_{dry} is the thickness (mm) of the specimen before immersion and T_{wet} is the thickness (mm) of the specimen after immersion.

Mechanical properties

Tensile strength, Young's modulus, and elongation at break were measured on a Zwick mechanical tester, model 1445, according to the ASTM D638 method. The samples were prepared in a hydraulic press at 160°C. The crosshead speed was 5 mm/min. Six measurements were conducted for each sample, and the results were averaged to obtain a mean value.

Izod impact measurements were performed on a Tinius Olsen instrument according to ASTM D256. Eight measurements were conducted.

Prior to mechanical measurements, the samples were conditioned at $50 \pm 5\%$ relative humidity for 48 h at ambient temperature in a closed chamber containing a saturated H_2SO_4 solution in distilled water (ASTM E104).

Biodegradation

The biodegradation of the composites took place during soil burial for 3 months. To study the effect of the thickness on biodegradation, two samples with different thicknesses were prepared. Specifically, specimens with 1 and 3 mm thicknesses (2.0×2.0 cm, width \times length) were prepared by hot pressing at 160°C and 200 bar. After preparation, the specimens were dried at 75°C for 24 h to remove any traces of moisture, weighed, and placed in a well for soil burial.

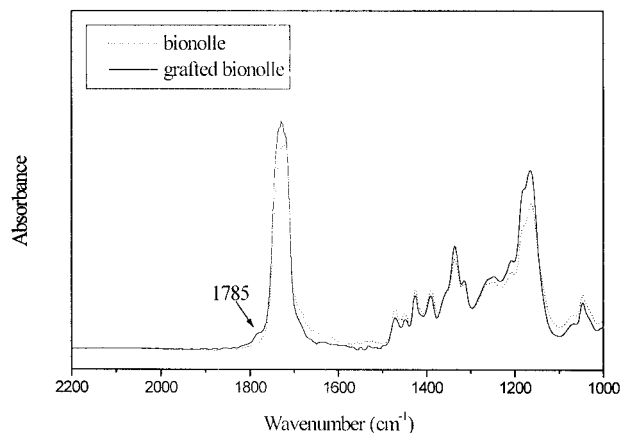


Figure 1 FTIR spectra of unmodified and grafted bionolle after extraction.

Scanning electron microscopy (SEM)

The impact specimens were fractured, and the exposed surfaces were observed with a scanning electron microscope (JEOL, model JSM-840A). Prior to the analysis, the samples were coated with graphite to avoid charging under the electron beam.

RESULTS AND DISCUSSION

Synthesis and characterization of compatibilizers

The melt grafting of bionolle with MA at different graft contents was performed in a Haake-Buchler Rheomixer. Three compatibilizers with different graft content were prepared to find the optimum one and the appropriate amount of this compatibilizer in the composites. The mixing conditions, the initial concentrations of DCP and MA, as well the graft content and the intrinsic viscosity of the produced compatibilizers are listed in Table I.

The graft content of bionolle-g-MA depends on many factors, such as monomer and initiator concentration, temperature, rotor speed, and residence time.^{21, 22} From the data in Table I it is clear that both temperature and initiator concentration are important factors in the grafting reaction. As the temperature and the initiator concentration were increased, a noticeable increase in the graft content was achieved. Moreover, this increase was accompanied by a reduction in intrinsic viscosity of the grafted product, due to a chain scission/degradation process.

The grafted polymers were totally soluble in chloroform, indicating that there was no crosslinking reaction occurring during the grafting process. Crosslinking reactions are possible when high amounts of initiator are used.²¹ The absorption at 1785 cm^{-1} in the FTIR spectrum (Fig. 1) of extracted grafted polymers, which is characteristic for succinic anhy-

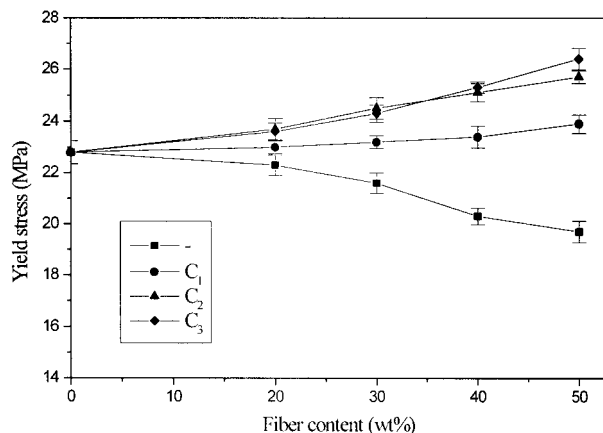


Figure 2 Effect of graft content on yield stress of bionolle-cotton composites.

drude groups, confirms the performance of the grafting process.

Mechanical properties

Effect of cotton content on mechanical properties

As seen in Figures 2–4, the incorporation of cotton fibers into the polymer matrix resulted in a significant change in all mechanical properties. In particular, the addition of cotton fibers led to a decrease of yield stress, an increase of Young's modulus, and a significant decrease of elongation at break and impact strength.

The decrease of yield stress with increasing fiber content (Fig. 2) is not surprising because other studies have also indicated that the incorporation of filler into a thermoplastic may not necessarily increase the strength of a composite.^{23, 24} This result is probably due to the poor adhesion between the two phases (i.e., fiber and matrix).²⁵ Moreover, fiber incorporation was associated with a significant increase of Young's mod-

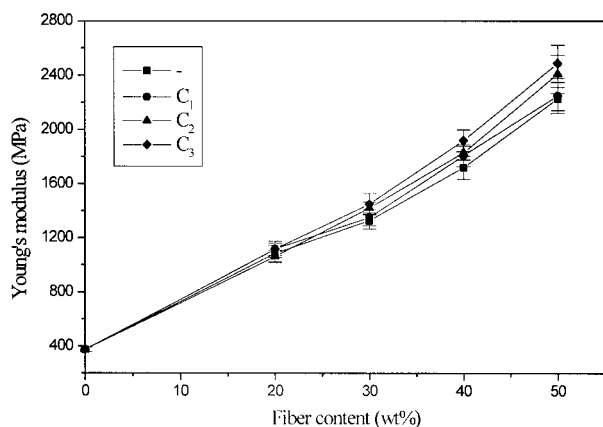


Figure 3 Effect of graft content on Young's modulus of bionolle-cotton composites.

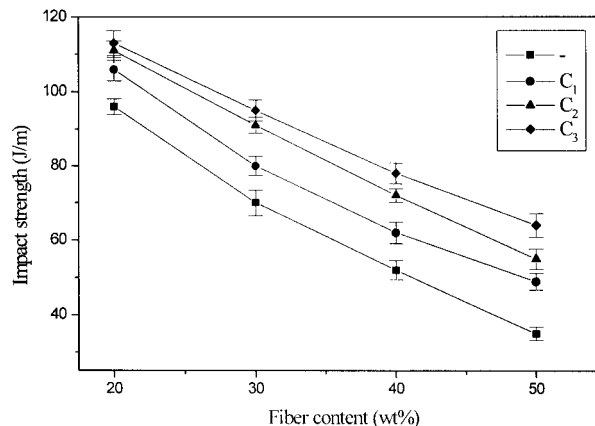


Figure 4 Effect of graft content on impact strength of bionolle-cotton composites.

ulus (Fig. 3) and material embrittlement, as suggested by the low impact strength values of the composites (Fig. 4). The Young's moduli of bionolle and cotton are 375 MPa and 5.5–12.6 GPa,¹¹ respectively. The significantly high Young's modulus value of cotton fiber explains the gradual increase in stiffness of the produced composites as the fiber content increases. The reduction of matrix amount as fiber loading is increased contributes to the decrease of impact strength because the matrix is primarily responsible for the absorption of the impact energy.

The incorporation of fibers into the polymeric matrix drastically reduced the elongation at break of the composites. In particular, the elongation of bionolle is > 350%, whereas the incorporation of even a low amount of cotton fiber resulted in materials with elongation values of <5%. This observation is rather common for almost all composites in which elongation decreases monotonically with addition of more fibers to the polymer.²³ Additionally, the significant reduction in mechanical properties (yield stress and impact strength) at high fiber content might be due to the presence of many fiber ends in the composites, which could cause crack initiation and, hence, potential material failure.¹²

The production of composites with <20 wt % cotton content was accompanied by the appearance of increased fiber agglomeration into bundles, and the consistent measurement of the properties was difficult.

Effect of compatibilizer graft content on mechanical properties

The most important factor for obtaining good fiber reinforcement in a composite is the strength of adhesion between polymeric matrix and fiber.¹² Because of the difference in the polarity of hydrophilic cotton fibers and hydrophobic polymer matrices, a weak interfacial bonding between the two components is ob-

tained. Therefore, to develop composites with improved mechanical properties, the use of a compatibilizer is necessary. To achieve optimum results, the effect of graft content as well as the effect of compatibilizer content on the properties of the material have to be studied.

Three compatibilizers with different graft content were prepared (Table I). The compatibilizers are products of the melt grafting reaction of bionolle with MA, as previously mentioned. The graft contents of the three compatibilizers, C_1 , C_2 , and C_3 , were 0.84, 1.72, and 2.14 wt %, respectively. The amount of the compatibilizer was kept constant at 10 phr, based on the fiber. The results in Figures 2–4 show that all properties were improved with the use of compatibilizer. These agents can modify the interface by interacting with both the fiber and matrix, thus forming a link between the two phases of the composite.

The excellent performance of bionolle-g-MA as a compatibilizer in cotton–bionolle composites could be attributed to the following two factors: (1) the ability of the MA to react with the hydroxyls of the cotton fibers, and (2) the great compatibility of the grafted copolymer bionolle chains with the main bionolle phase.

As expected, an improvement in the tensile strength was observed as the graft content was increased. This improvement is attributed to an increment of the functional MA groups on the compatibilizer, leading to the formation of more ester groups with hydroxyl groups at fiber surface, and thus to improved adhesion between the two phases. As seen in Figure 2, there was a significant improvement even with the use of low graft content compatibilizer (C_1), whereas the best results were observed with the higher graft content compatibilizers C_2 and C_3 . The yield stress of the composites with 50 wt % fiber content increased to values of 23.9 and 26.4 MPa with the addition of C_1 and C_3 compatibilizers, respectively, compared with the value of 19.7 MPa for the noncompatibilized composite.

Furthermore, the addition of the compatibilizers led only to a slight improvement in Young's modulus (Fig. 3). Generally, the improved adhesion between matrix and fiber does not affect the Young's modulus to a great extent.²⁴ However, a remarkable increase in impact strength values was noticed (Fig. 4). Interfacial adhesion, in addition to increasing the strength of the composite (because stress transfer from matrix to fiber becomes more effective), is associated with energy absorbing mechanisms. Impact strength is a measure of the energy needed for failure, and the results generally demonstrate that more energy is needed to break composites with high graft content compatibilizer. Finally, the elongation at break was not affected significantly by the addition of the compatibilizer.

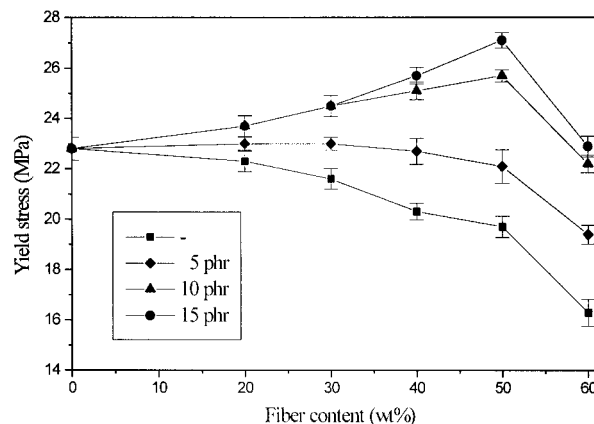


Figure 5 Effect of the amount of compatibilizer on yield stress of bionolle–cotton composites.

The results in Figures 2–4 also show that the use of the high-graft-content compatibilizer, C_3 , did not lead to significantly improved properties compared with those obtained with the compatibilizer C_2 . So, further studies were carried out using compatibilizer C_2 .

Effect of compatibilizer content on the mechanical properties

Three ratios (5, 10, and 15 phr based on fiber) of compatibilizer C_2 were added to the composites. From the results, it is apparent that the composite strength increased with increasing compatibilizer content. As seen in Figure 5, the increase of compatibilizer content led to a prominent improvement of yield stress, which was more remarkable at higher fiber content. Specifically, for 15 phr compatibilizer, the yield stresses for 30 and 50 wt % fiber content composites increased to 24.5 and 27.1 MPa, respectively, compared with the values of 21.6 and 19.7 MPa, respectively, for the noncompatibilized composites. This increase is probably because the addition of sufficient compatibilizer leads to improved interfacial adhesion and the fiber reinforcement effect.

The results in Figure 5 show that for a cotton content of 50 wt %, the yield stress was maximal. At higher fiber content, the property decreased, possibly because of incomplete fiber wetting, void generation, and increased fiber–fiber contact.²⁶

Furthermore, a slight improvement in Young's modulus (Fig. 6) was noticed as the compatibilizer content increased.

The results in Figure 7 show that the composite impact strength increased with increasing compatibilizer content. The impact strength for 50 wt % cotton content and 15 phr compatibilizer increased to 62 J/m compared with 35 J/m for the noncompatibilized composite. The effect of compatibilizer becomes more pronounced with increased fiber content, as already suggested by Felix.²⁴ The improvements for 30 and 50

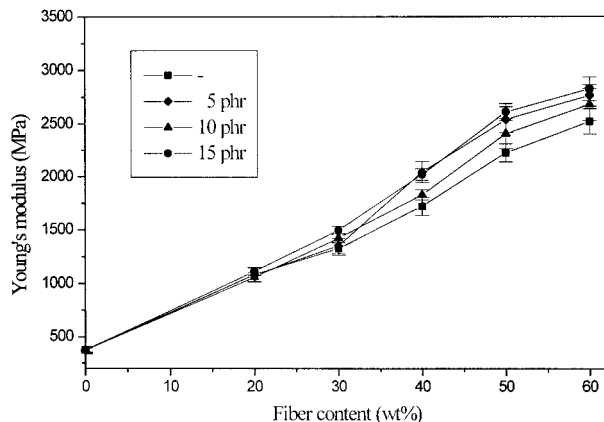


Figure 6 Effect of the amount of compatibilizer on Young's modulus of bionolle-cotton composites.

wt % cotton content composites were 31% (from 70 to 92 J/m) and 77% (from 35 to 62 J/m), respectively, with the incorporation of 15 phr compatibilizer. This result confirms the previous statement.

Interfacial properties

The state of the fiber-matrix interface was investigated by SEM. The SEM micrographs reveal that there was a remarkable difference in the fiber-matrix interaction between the compatibilized and noncompatibilized composites. Composites containing compatibilizer showed better fiber dispersion, a more effective fiber wetting by the matrix, and an improved adhesion between the two phases.

It is clear from the results in Figure 8(a) that in noncompatibilized composites, fibers aggregated into bundles, resulting in an uneven distribution. It can also be seen that there was no wetting of fiber surfaces by bionolle, probably because the surface energy of fibers and polymeric matrix are significantly different (i.e., the cotton surface is hydrophilic, whereas the polyester surface is hydrophobic²⁷). On the contrary, according to the results in Figure 8(b), the addition of compatibilizer led to wetting of the fibers by bionolle-bionolle-g-MA. In this case, the anhydride groups of the compatibilizer reacted with the hydroxylic groups of the fiber, and the fiber surface energy was lowered to a level much closer to that of the matrix.

The fiber surfaces of untreated and treated fibers are shown in Figures 8(c) and 8(d), respectively. There is no matrix adhesion on the surface of the untreated fiber. In contrast, the treated fiber is covered by layers of matrix material being pulled out together with the fiber. In general, it is known that weak interfacial interactions result in poor composite mechanical properties and vice versa. Therefore, the SEM studies support the previous discussion.

Water absorption

Effect of cotton content on water absorption and thickness swelling

Cellulose-based fibers absorb water, causing reversible and irreversible swelling. This swelling can result in undesirable dimensional changes of the final products.²⁸ The sorption of water by hydrophobic polymer matrices containing filler depends mostly on the nature of the filler. For hydrophilic cellulose-based fillers, such as cotton fibers, an increase in water sorption is expected.

The uptake of water by noncompatibilized composites as a function of time for various fiber contents is shown in Figure 9. It is evident that the weight increase of the noncompatibilized composites depends on exposure time and cotton content. The water uptake increased with fiber loading because of the increased cellulose content. The water absorption increased linearly at first and then gradually, and finally reached a plateau. It is apparent that the initial rate of water absorption and equilibrium uptake of water increased with increasing fiber content. Exactly the same trend for thickness swelling is observed in Figure 10. As the fiber content increases, there is a noticeable increase in thickness swelling.

Water absorption obviously affects the mechanical properties of the composites.²⁹ The adhesion between matrix and fiber becomes weak when the composite is wet. The fiber-matrix interface moisture may reduce the strength of adhesion by breaking the bonds. Specifically, the absorption of water may cause rapid debonding, delamination, and loss of structural integrity.³⁰ Moreover, intermolecular adhesion between cellulose and water molecules leads to deterioration in the material properties. Thus, as the immersion time increases, the composite tensile strength decreases. Mechanical damage caused by moisture-induced swelling may also lead to property changes.³⁰

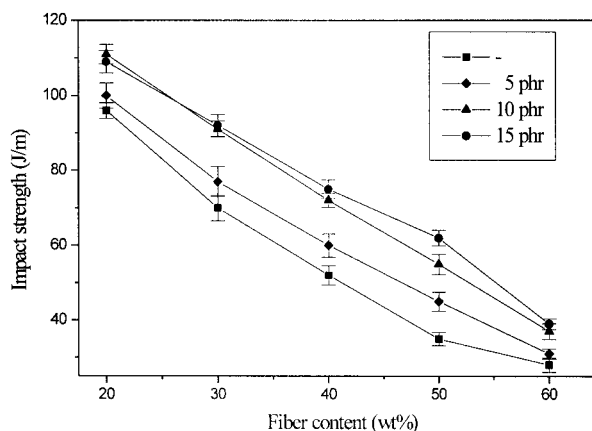


Figure 7 Effect of the amount of compatibilizer on impact strength of bionolle-cotton composites.

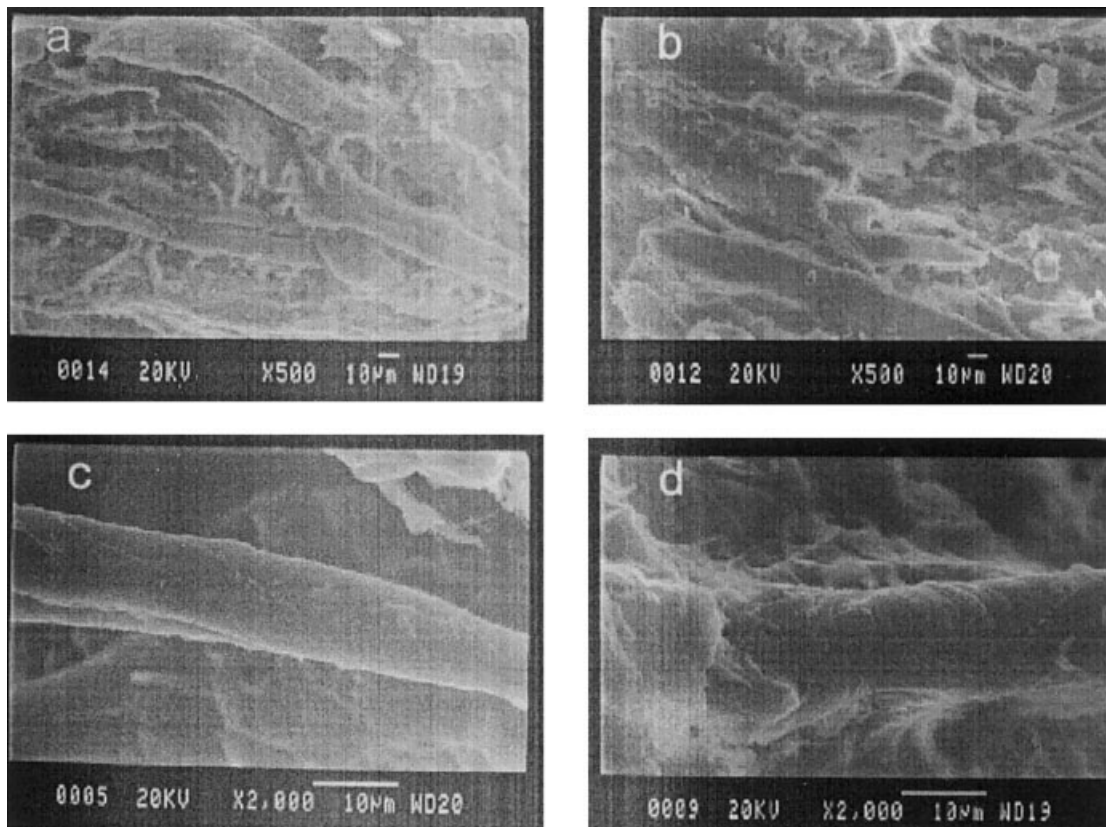


Figure 8 SEM micrographs of fracture surfaces of bionolle-cotton composites (30 wt % fiber content): (a) and (c) without compatibilizer; and (b) and (d) with 10 phr compatibilizer C₂.

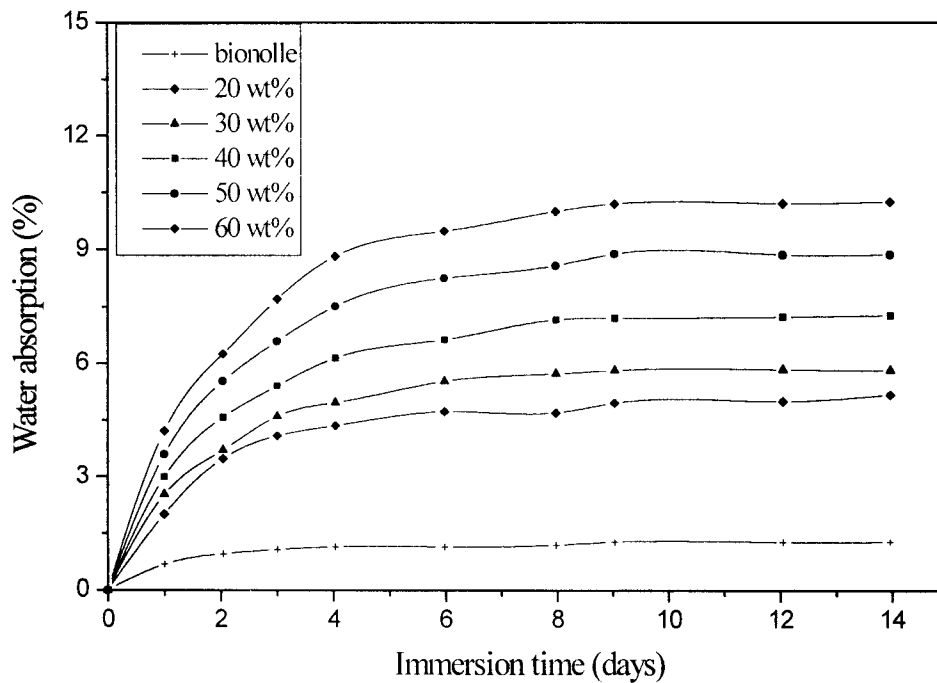


Figure 9 Water absorption curves showing the uptake of bionolle-cotton noncompatibilized composites with time for different fiber contents.

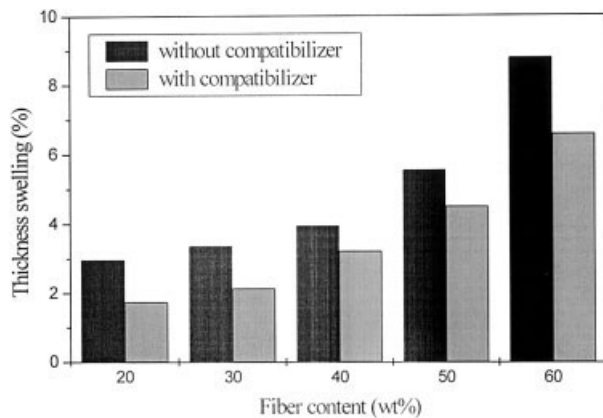


Figure 10 Thickness swelling of composites with (C_2 , 15 phr based on fiber) and without compatibilizer.

Effect of compatibilizer on water absorption and thickness swelling

The addition of compatibilizer reduced the water uptake of composites (Fig. 11). The reduction in moisture uptake has been attributed to the formation of covalent bonds between the functional groups of MA and the hydroxyl groups at the surfaces of cotton fibers.³¹ In addition, as the amount of the compatibilizer increases, less water is absorbed. Undoubtedly, this result occurs because there are more functional MA groups as compatibilizer content increases, so more bonds can be formed between matrix and fibers. The same conclusion is drawn from the use of different grafting content compatibilizers. Thickness swelling

of cellulosic materials occurs when the cell wall is bulked by water. The results in Figure 10 clearly show that composites with compatibilizer showed lower thickness swelling compared with noncompatibilized composites, and the explanation for this result is similar to that discussed in the water absorption results.

Thermal analysis

As seen in Figure 12, fiber incorporation in the polymeric matrix incurred the reduction of the thermal stability of the produced material. The thermal stability of the composites decreased because the degradation temperature of cotton fibers starts at $\sim 245^\circ\text{C}$, whereas the degradation temperature of bionolle 3020 is $\sim 300^\circ\text{C}$ (Fig 12). The results in Figure 13 show that the addition of compatibilizer led to a slight increase of composite thermal stability, especially at high temperatures. It is probable that the improved adhesion between fiber and matrix restricted the access of the oxidizing gas to their interface and thus slowed down the oxidation of the material. Other studies have also shown that the thermal stability of the produced material increases with fiber modification and improved adhesion.³²

Biodegradation

The photographs of the remaining samples of bionolle-cotton fiber composites after different degradation periods and for different cotton content are presented

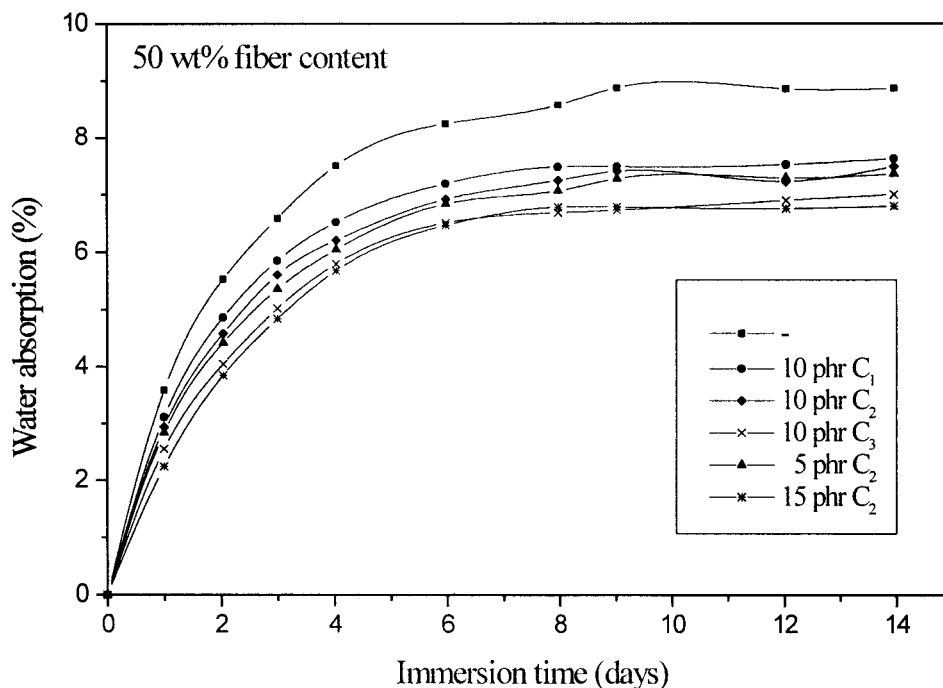


Figure 11 Effect of graft content and the amount of compatibilizer on water absorption (50 wt % fiber content).

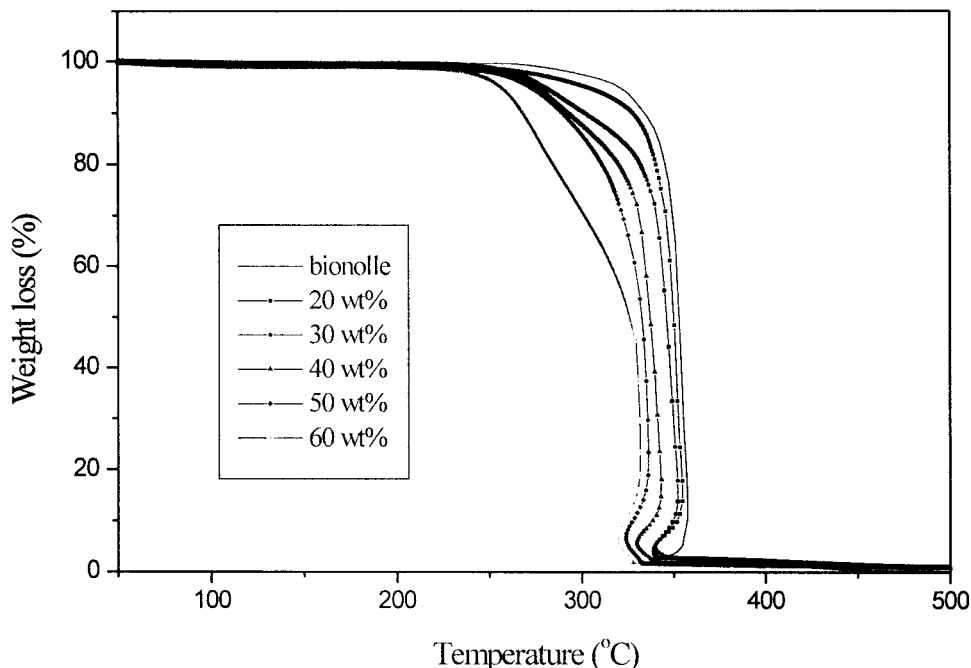


Figure 12 Effect of fiber content on thermal stability of the composites.

in Figure 14. The surface of the composites was discolored from brown to white after 2 months of biodegradation and damaged by microorganisms thereafter. It is clear from Figure 14 that the composites degraded faster than the pure matrix. This difference is because cotton fibers are highly hydrophilic and they can transport water in the composite by the capillary effect.³³ The humid environment promotes the growth of the microorganisms and, consequently, the hydrolysis of the ester groups of the polyester increases. These conditions also explain the increase of biodegradation rate with increasing fiber content.

The weight loss of polyester bionolle 3020 and its composites with 40 wt % fiber content is shown in

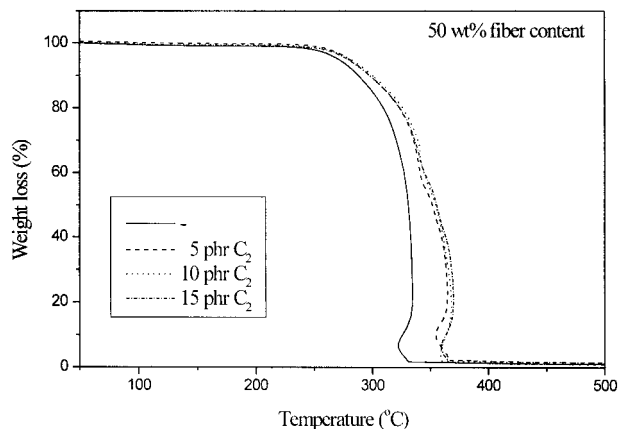


Figure 13 Thermal analysis of compatibilized composites.

Figure 15 as a function of sample thickness. The biodegradation rate dropped as the sample thickness increased. This result could be anticipated because the rate of water absorption of bionolle–cotton composites decreases with decreasing sample thickness. The rate of water absorption is higher for thinner samples because the diffusion of water in the composite begins from the surface and afterwards proceeds to the internal and the core of the material.

Moreover, it is important to mention that the addition of compatibilizer in composites led to a slight

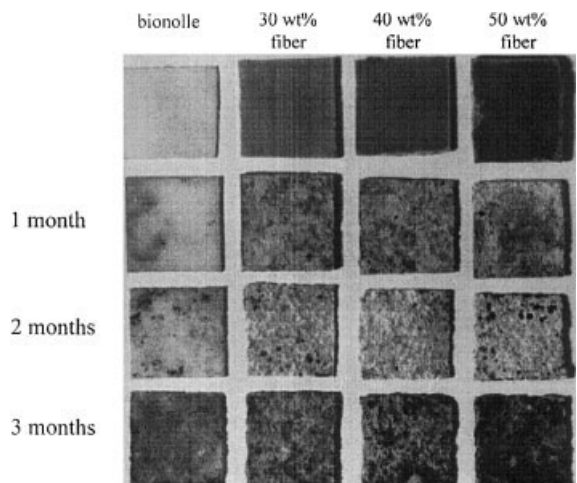


Figure 14 Photographs of bionolle and bionolle–cotton composites showing dependence on degradation time.

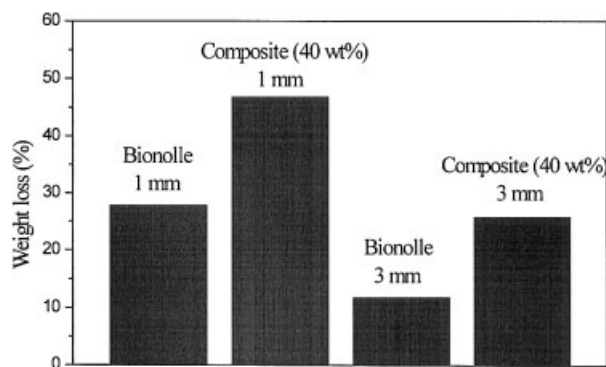


Figure 15 Weight loss values of bionolle and bionolle-cotton composites as a function of sample thickness.

decrease of the biodegradation rate as a result of the hydrophilicity reduction due to the modification of the fibers.

CONCLUSIONS

Biodegradable plastics have been introduced with the aim of fulfilling new ecological requirements regarding effective waste management. However, widespread application of these polymers is restricted by their high cost. So, an attempt was made to reduce the final cost of the material by incorporating a low cost filler such as cotton fiber waste.

The incorporation of cotton fibers in the polymeric matrix, without the use of a compatibilizer, resulted in a reduction of the yield stress (reflecting the poor adhesion between the two components), a drastic reduction of impact strength, and a significant increase of Young's modulus. The addition of even a low amount of bionolle-g-MA compatibilizer improved all mechanical properties of the produced composites. An increase in either graft or compatibilizer content led to a significant improvement of the mechanical properties. Apparently, the presence of an increased number of anhydride groups in the material led to improved interfacial adhesion and, thus, improved mechanical properties. The effect of compatibilizer became more pronounced with increased fiber content. SEM micrographs proved that there is a significant difference in the fiber-matrix interaction between compatibilized and noncompatibilized composites. Composites containing compatibilizer showed better fiber dispersion, a more effective fiber wetting by the matrix, and a better adhesion between the two components.

The water absorption of the composites increased with fiber loading because of increased cellulose content. Compatibilizer addition reduced the water uptake as a result of covalent bond formation between the functional groups of compatibilizer and the hydroxyl groups of cellulose. The formation of these

bonds eliminates the chances of cellulosic hydroxyl groups to come in contact with water molecules. Thickness swelling of the composites also increased with fiber content, whereas the addition of compatibilizer resulted in its reduction.

The thermal stability of the composites seems to be improved with the addition of the compatibilizer, probably because of improved adhesion between cotton and polymer, which restricts the access of the oxidizing gas to their interface.

The incorporation of cotton fibers in the polymeric matrix significantly increased the biodegradation rate. The thickness of the sample was a determinant parameter of biodegradation rate; that is, as the thickness of the sample increased, the biodegradation rate decreased. On the other hand the effect of the compatibilizer on biodegradation was negligible.

The cost of the resulting materials is very low compared with the expensive matrix. Moreover, the material is highly biodegradable and, in the presence of a compatibilizer, it exhibits good mechanical properties.

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