Jute Fiber Reinforced Polypropylene Produced by Continuous Extrusion Compounding, Part 1: Processing and Ageing Properties

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ABSTRACT: This article addresses the processing and ageing properties of jute fiber reinforced polypropylene (PP) composites. The composite has been manufactured by a continuous extrusion process and results in free flowing composite granules, comprising up to 50 weight percent (wt %) jute fiber in PP. These granules have similar shape and diameter as commercially available PP granules. Rheological analysis shows that viscosity of the compounds follows the same shear rate dependency as PP and is on the same level as glass-PP compounds. The mechanical properties show very little variation and exhibit strength and stiffness values at the upper range of competing natural fiber reinforced compounds for injection molding. The mechanical performance reduces gradually upon prolonged thermal loading and immersion in water. The low water diffusion coefficient of the 50 wt % jute-PP composites indicates that the fibers are not forming a continuous network throughout the polymer. The jute fibers exhibit a stabilizing effect against ultra violet irradiation (UV) on PP polymer and, as a consequence, the mechanical properties of jute-PP composites hardly decrease during an accelerated UV ageing test. Bacteria, fungi, and garden mold grow easily on the compound material, but only have a limited effect on mechanical properties. The resistance to growth of bacteria on the materials surface can be increased using a biostabilizer. © 2008 Wiley Periodicals, Inc. J Appl Polym Sci 110: 1009–1018, 2008

Key words: jute fibers; polypropylene; extrusion compounding; processing; ageing

INTRODUCTION

During the last decade of the 20th century, agrofiber composites have gained much interest from both research and industrial communities. These research and development efforts have resulted in the industrial production of natural fiber mat reinforced thermoplastics (NMT). These allow the production of 2D applications like e.g., car door and seat panels. Recently, industry has been searching to implement agrofiber based compounds that are suitable for injection molding, which allow 3D applications.[1]

Most literature on nonwood agrofiber-based thermoplastic compounding is based on batch kneading processes.[2–13] Many publications based on nonwood agrofiber thermoplastic extrusion compounding do not specify the fiber feeding process, or apply a method that is not useful at an industrial processing scale.[5,6,14–23] Karnani et al.[24] use a gravimetric feeder for 1.58 mm chopped kenaf fiber without mentioning feeding problems. Huda et al.[25] apply a side feeder for feeding wood fiber with average size smaller than 850 μm into an extruder without discussion of fiber feeding problems. Bledzki and Faruk[19] state that for 4 mm wood fibers they apply "a high speed mixer due to its length." Gauthier et al.[26] state that a main problem of wood fiber feeding is "to maintain continuous and regular feeding of fibers, short fibers with a length ~ 400 μm present less difficulties."

Agrofibers have a finite length and poor free flowing properties. Therefore, feeding of natural fibers to an extruder is not trivial. Natural fibers, however, can be stapled like roofing tiles and carded into a continuous sliver, which usually is an intermediate product in a textile spinning process. Such a sliver can be fed continuously to an extruder with a sliver feeder designed and built at Wageningen UR (Fig. 1). The applied continuous extrusion process subsequently mixes the fibers intimately with the thermoplastic polymer and at the same time opens up the natural fiber bundles into strong elementary fibers (individual plant cells).[27–29] Free flowing compound granules are obtained by making use of an under-water pelletizer. This extrusion

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Figure 1 Photograph of the extrusion compounding set-up: polymer granules feeding (1), jute sliver feeding (2), actual compounding inside the barrel (3), de-gassing (4), under water pelletizing (5), granule collection (6).

compounding process is currently evaluated for industrial up-scaling.

One of the concerns, expressed by molders before performing injection molding trials with natural fiber polymer compounds, has been the degradation of natural fibers. Hot spots or emergency may cause fiber degradation and thus lead to blockage of their equipment. Also environmental influences such as moisture, fungi and microbes, and UV irradiation are expected to reduce strength properties of natural fiber composites over time. Previous studies confirm that water absorption negatively affects the mechanical performance of natural fiber composites.[12,13,30] The time it takes water to penetrate into the natural fiber composite however depends very much on fiber form. Long fiber mat composites[30] show far higher water diffusion rates than short fiber compounds.[12–14] The authors of the short fiber compounds conclude that water absorption in their composites follows Fickian diffusion. This conclusion however is not supported by the presented data.[12–14] Microbiological decay causes a huge drop in strength of a variety of natural fibers within a couple of weeks.[31] On the other hand, whereas natural fibers make the composites sensitive towards water and biological attack, the lignin in jute fibers may contribute to UV stability.[12,32]

This article addresses the processing and ageing characteristics of jute-PP composites manufactured with continuous extrusion compounding. The jute-PP compound has been developed specifically for injection molding applications. Since it is expected that composites with high fiber loading will exhibit the most pronounced degradation effects, both during processing as well as during the compounds lifetime, fiber content has been selected as 50 wt %. A 50 wt % natural fiber-PP composite still can be injection molded very well. Maleic anhydride grafted polypropylene (MAPP) was used as a compatiblizer. Composite morphology and rheology were evaluated. Further, the effects of (salt) water, UV, thermal degradation, microbial and fungal attack on both the dimensional stability and the mechanical performance of jute-PP compounds were studied. The effect of a biostabilizing agent on biodegradation was determined as well. Properties are compared with those of compound materials described in literature.

This article only addresses 50 wt % jute-PP compounds. The mechanical and fatigue properties of jute-PP compounds with fiber content ranging from 30 to 50 wt % will be addressed in a subsequent paper, including routes to improve the impact strength.

EXPERIMENTAL

Materials

Jute fiber was based on 64% Bangla Tossa D and 36% Bangla Tossa E and was obtained from Bangladesh. Jute is the bast fiber from *Chorchorus spp*, which is a crop growing very well in e.g., Bangladesh and India. The jute was processed into a sliver, wound into a wheel (Fig. 1) and compressed and wrapped for shipment. The jute contained batching oil and had a moisture content of (8 $\pm$ 1)% during compounding. Polypropylene (Stamylan P 17M10, homopolymer, $MFI_{2.16,230} = 10.5$) was obtained from DSM (currently Sabic). Maleic anhydride grafted PP (Epolene G 3015) was obtained from Eastman Chemical. A biostabilizing masterbatch Sanitized MB E 22-70 was kindly provided by Sanitized AG (Switzerland).

Extrusion compounding

Compounds containing 50 wt % jute, 5 wt % MAPP, and 45 wt % PP were produced according to WO patent 9956936 using a Berstorff ZE 40- 38D twin screw extruder.[27] In the biostabilized compound, 3 wt % of PP has been replaced by biostabilizing masterbatch. The PP and MAPP granules and if applicable biostabilizing additive were premixed and fed upstream to the extruder by gravimetric feeding (Fig. 1). The continuous jute sliver was unwound and fed downstream to the extruder with an in-house built sliver feeder. A vacuum degassing unit was positioned between the compounding section and the die. Free flowing compound granules were obtained using a Gala under-water pelletizer. The total flow was 8 kg/h. By accident, due to very humid weather, also a batch of jute containing

(10 ± 1)% moisture was produced into jute/PP compounds.

The granules were dried overnight at 80°C with predried air and injection molded into flexural/impact test bars with dimensions 80 × 10 × 4 mm^3 using a Demag ERGOtech 25-80. Mold shrinkage was determined in three dimensions using a caliper. All composite specimens were conditioned for at least 7 days at 20°C and 50% relative humidity (RH) before further analysis.

Scanning electron microscopy

Scanning electron microscopy (SEM) was performed using a Jeol JSM-5600 LV scanning electron microscope. The samples analyzed included cryo-fractured surfaces of jute-PP granules and of injection molded test bars of both compositions. Prior to the analysis the fracture surfaces were covered with a 10 nm layer of platinum using an Oxford CT1500 sputter coater.

Capillary rheometry

The jute-PP granules were dried and rheological analysis was performed at 200 and 220°C on a Rosand Capillary Rheometer RH7-2 with two 12 mm barrels. The die was 16 mm in length and 1 mm in diameter. Prior to analysis, the material was heated in the barrel for 6 min while applying a pressure of 1 MPa at time $t = 0$ and $t = 3$ min. The analysis was repeated with a preheating time of 36 min to study the effect of thermal degradation on rheology. Pure PP is tested as a reference material.

Accelerated degradation and dimensional stability tests

Injection molded 50 wt % jute-PP test bars were irradiated according to ISO 4892-2 (2003) in a Heraeus Suntest CPS for 56 days at maximum power, being 765 W/m^2.

Injection molded test bars with and without biostabilizer were subjected to three biodegradation tests at Sanitized AG (Switzerland). Specimens were subjected to a bacterial resistance test for 24 h according to SN 195 920 using *Staphylococcus aureus* ATCC 6538 and evaluated for surface growth of bacteria. A separate set of specimens was subjected to a fungi resistance test at 28°C for 28 days according to ASTM G 21–96 and evaluated for surface growth of fungi. A third series of specimens was tested in garden mold at 29°C for 42 days according to EN ISO 846—Section D and evaluated for weight loss.

Thermal degradation was simulated by performing injection molding at 200°C with extended cycle time, the jute-PP compound thus being in the molten stage for a longer period. The cycle times were chosen as 5, 10, and 15 min.

Injection molded test bars were immersed in demi water and in an aqueous 3.5 wt % Sodium Chloride and a 3.5 wt % Red Sea salt solution at 40°C. The water absorption and thickness swelling of 10 specimens were monitored until a plateau level was reached. Injection molded pure PP was included as reference.

After all accelerated degradation tests, specimen warping was monitored, linear expansion was measured with a caliper and mechanical properties were determined.

Thermal expansion in length direction of molded specimens was determined at −20, 40, and 80°C, relative to the length at 20°C. Evaluation was performed in fivefold, using a caliper with accuracy 0.01 mm.

Mechanical performance tests

Before testing, specimens were conditioned at 20°C and 50% RH for 7 days. The specimens that were subjected to the 40°C water absorption test were cooled to 20°C in demi water for 24 h prior to testing.

The flexural properties were measured on a Zwick 1445 according to ISO 178. The support length was 64 mm and the crosshead speed was 2 mm/min for the modulus and 10 mm/min for the strength. The flexural strength and modulus were determined from five specimens per batch. The Charpy unnotched impact strength was determined using a Ceast pendulum impact tester according to ISO 179/1fU using an impact hammer of 4 J at a speed of 2.9 m/s. The Charpy impact strength was determined from eight specimens per batch.

The degraded side of the UV irradiated specimens was positioned to the nose side for flexural testing and to the blow side for impact testing.

RESULTS

Processing

Feeding of the jute sliver is adequate when the sliver wheel can be unwound easily. This is the case for the outer part of the wheel. The fibers of subsequent layers in the inner part of the wheel are entangled to such large extent that the sliver disrupts regularly during mechanical unwinding. The entanglements are a result of compressing during wrapping for shipment. The material produced within 5 min of disruption has been excluded from further processing and analysis. The rotating screws in the extruder swallow the amount of sliver that is offered by the feeder. Extrusion processing of 50 wt % jute fiber in

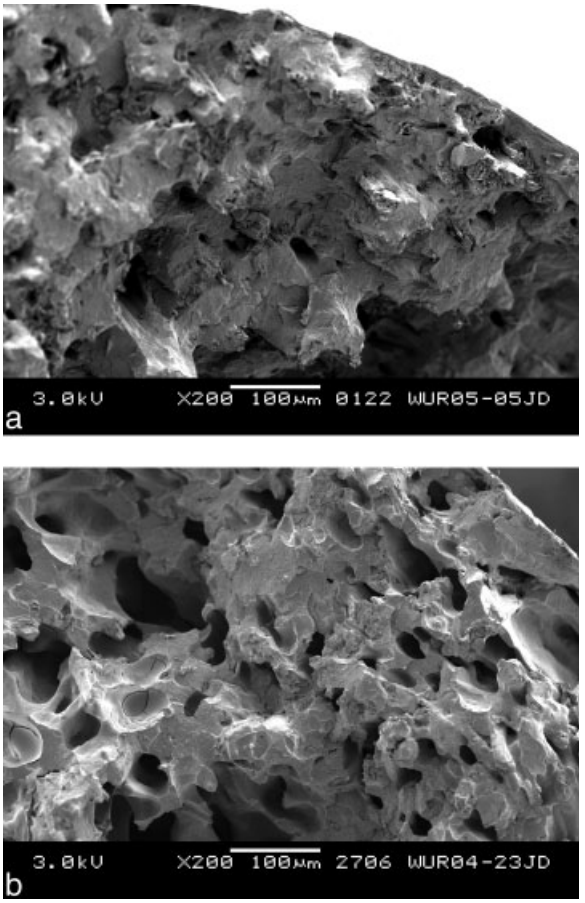

Figure 2 SEM micrographs of fracture surfaces of 50 wt % jute-PP extrusion compound granules from a stable (A) and unstable (B) process.

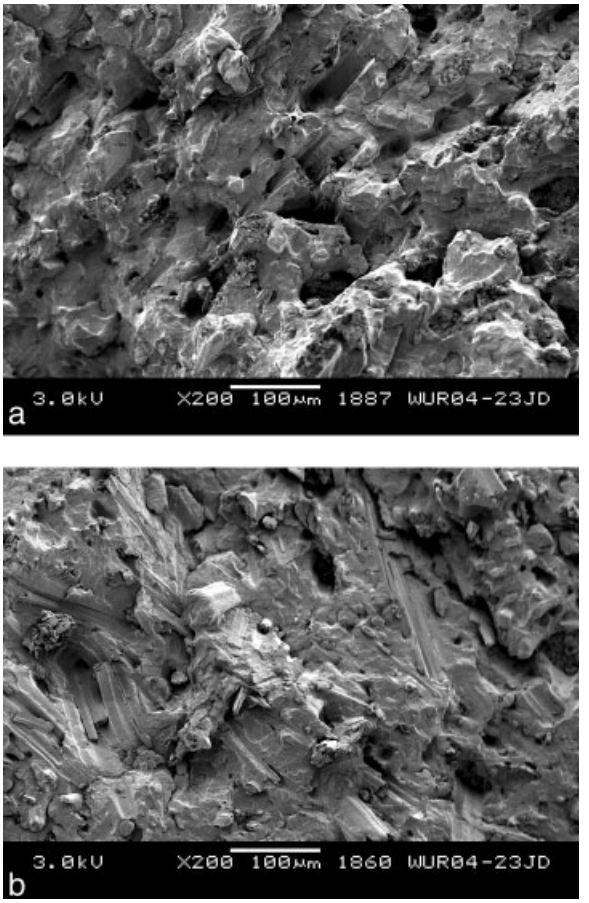

Figure 3 SEM micrograph of fracture surface of injection molded 50 wt % jute-PP compound, without (A) and with (B) biostabilizer.

PP is stable with respect to the torque of the screws, the die pressure and the melt temperature during a period of at least 30 min when the moisture content in the jute fiber is below 8 wt %. These conditions result in compact granules [Fig. 2(A)]. When the moisture content of the jute is $\sim$ 10 wt %, the compound starts foaming and comes out of the venting opening in the extruder. Under these conditions the torque of the screws and the die pressure is not constant and the obtained compound granules have many air inclusions [Fig. 2(B)].

The resulting granules have similar shape and diameter as commercially available PP granules. The injection molding process exhibits no irregularities. Also at cycle times of 15 min at 200°C the molding process is stable and the molded samples easily release from the mold.

Morphology

The injection molded jute-PP compound contains mainly elementary fibers and bundles of a few elementary fibers that are still glued together [Fig. 3(A)]. The fiber pull out lengths in general are shorter than the fiber diameter and fibers are covered with matrix material. The addition of the biostabilizer to the compound results in similar fiber

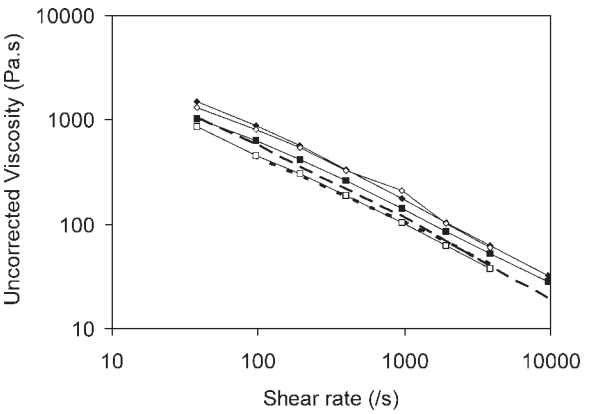

Figure 4 Uncorrected viscosity versus shear rate of 50 wt % jute-PP compound at 200 (◆,◇) and 220°C (■,□), measured without delay (filled symbols) and after 30 min heating at given temperatures (open symbols). The dashed lines represent the data for pure PP (short dash) and for 30% glass fiber/PP from Nanguneri et al.[34] (long dash), both at 200°C.

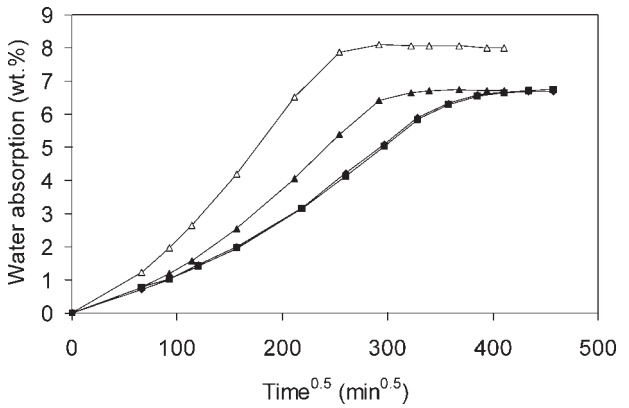

Figure 5 Water absorption curves for extrusion compounded 50 wt % jute-PP in water at 40°C: Demi water after predrying at 105°C (△), Demi water (▲), 3.5 wt % Red Sea salt water solution (◆), 3.5 wt % NaCl water solution (■).

diameter distribution and fiber pull out lengths [Fig. 3(B)].

Rheology

The uncorrected viscosity of 50 wt % jute-PP compounds versus shear rate is given in Figure 4. Increase of the temperature from 200 to 220°C results in a reduction of the viscosity. Also an extended residence time of 30 min at these high temperatures results in a lower viscosity, although the drop in viscosity is very minimal at 200°C. The viscosity of the jute-PP compound after 30 min at 220°C is similar to the viscosity of the pure PP at 200°C at a standard residence time.

Accelerated degradation

The (salt) water absorption of injection molded 50 wt % jute-PP test bars at 40°C as a function of time is presented in Figure 5. The water absorption levels off (M_∞) after ~ 60 days to a value of 8.1 (±0.06) wt % for dried composites in demi water. The composite specimens that were not dried prior to water absorption reach maximum values of 6.7 (±0.03) wt % after ~ 90 days in demi water and of 6.7 (±0.04) wt % after ~ 120 days in both the aqueous 3.5 wt % Red Sea salt and 3.5 wt % NaCl solutions. The variation in data was so small for all four absorption tests that incorporation of the standard deviation in Figure 5 would interfere with the dots.

Pure PP did not show water absorption in 40°C water during the period investigated.

The observations of the biodegradation tests are summarized in Table I. After 1 day of bacterial incubation, the standard 50 wt % jute-PP compound was fully covered with bacteria. Addition of 3 wt % of the biostabilizer Sanitized MB E 22-70 restricted bacterial growth to 5% of the specimen surface. The biostabilizer hardly contributes to the resistance to fungi and garden mold.

The mechanical properties of the undegraded jute-PP specimens are summarized in Table II. The mechanical properties as a function of water absorption, UV irradiation, and thermal degradation time are presented in Figures 6–8. The mechanical properties of undegraded specimens have been normalized to 100%. The flexural strength and stiffness and Charpy impact strength of the jute-PP compounds gradually decrease during (salt) water absorption and thermal degradation, while the properties of PP are hardly affected. Absorption of demi water and salt water have a similar effect on the mechanical properties of the jute-PP compounds. On the other hand, UV irradiation has virtually no effect on the flexural properties of the jute-PP compound and limited effect on its impact properties, whereas the flexural and impact properties of pure PP reduce dramatically after 56 days of UV irradiation. UV irradiation causes bleaching of the very top layer of the compound from dark brown to light brown.

The biodegradation tests virtually cause no reduction in flexural strength and stiffness. The Charpy impact strength reduces with 8%, independent of the use of biostabilizer.

Dimensional stability

None of the test bars showed warping after the different accelerated tests, viz. (salt) water absorption, UV degradation, and bacterial and fungi attack. Also after the thermal elongation test no bending has been observed. The thickness of the compounds has

TABLE I
Effect of Biostabilizer Sanitized MB E 22-70 on Biodegradation of 50 wt % Jute-PP Compounds

Test method	Incubation time (days)	Jute-PP compound	Jute-PP comp. + 3% Biostab	Cotton control
Bacterial resistance test (% of surface covered)	1	100	<5	
Fungi resistance test	7	2	2	1
Grading:	14	3	2	3
0 = no growth	21	4	4	4
4 = ≥60% surface growth	28	4	4	4
Garden mould resistance test (% weight loss)	42	0.45	0.36	

TABLE II
Flexural and Charpy Impact Properties of Undegraded 50 wt % Jute-PP Compound and Pure PP, Standard Deviation is Indicated Between Brackets

	Strength (MPa)	Stiffness (GPa)	Strain (%)	Impact (kJ/m^2)
50% jute/PP	86 (1)	5.1 (0.1)	3.2 (0.1)	15 (1)
PP	43.3 (0.4)	1.31 (0.02)	6.9 (0.1)	Not broken

not been affected by the UV irradiation and biodegradation tests.

The swelling of conditioned 50 wt % jute-PP compounds in 40°C (salt) water levels off after circa 80 days to values around 3% for thickness and width swelling and to 0.3–0.4% swelling in length direction (Table III). The values are averages of 10 specimens. Pure PP shows no swelling in 40°C water during the period investigated.

The thermal elongation in length direction of the test bars shows a linear trend in the temperature range −20 to +80°C and is 2.8 × 10^{-5} mm mm^{-1} °C^{-1} for 50 wt % jute-PP compounds and 6.8 × 10^{-5} mm mm^{-1} °C^{-1} for pure PP (Fig. 9).

Mold shrinkage of the 50 wt % jute-PP test bars is 0.003, 0.005, and 0.02 in longitudinal, width, and thickness direction, respectively.

DISCUSSION

Processing

When the jute fibers enter the extruder with a moisture content above 8 wt %, this water can not completely be released in the vacuum vent. The remaining trapped water causes foaming of the compound at the exit of the die, where the polymer melt has a temperature of around 210°C. The resulting foamed granules easily take up water during underwater pelletizing and water contents in the granules of up to 30 wt % have been determined. Such granules will require more energy to dry. Furthermore, transportation of foamed granules is a disadvantage. After injection molding, however, the mechanical performance of the foamed granules is on the same level as the solid granules. To obtain the compact granules, jute fibers have to be dried to at least below 8 wt % moisture content before compounding, and the extrusion process requires at least one degassing zone. Trials with 30–40 wt % jute fiber in PP showed that the extrusion compounding process runs more smoothly with reduced fiber content. A following paper, Part 2, will address jute-PP compound properties as function of fiber content.

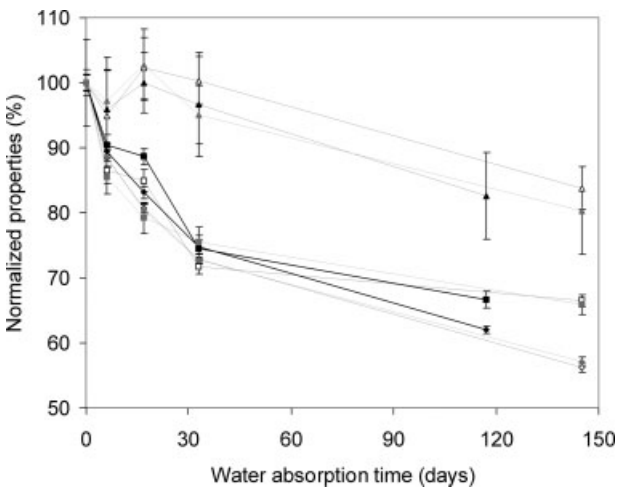

Figure 6 Mechanical properties of 50 wt % jute-PP compound versus water absorption time: flexural strength (◆), flexural modulus (■), Charpy impact strength (▲); demi water (solid symbols), 3.5 wt % Red Sea salt solution (open symbols), 3.5 wt % NaCl solution (gray symbols). The mechanical properties of undegraded specimens have been normalized to 100%.

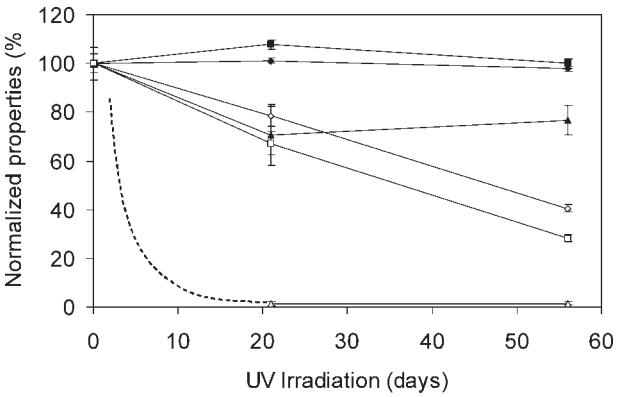

Figure 7 Mechanical properties of pure PP and 50 wt % jute-PP compound versus UV irradiation time: flexural strength (◆), flexural modulus (■), Charpy impact strength (▲); PP (open symbols), 50 wt % jute-PP (solid symbols). The mechanical properties of undegraded specimens have been normalized to 100%.

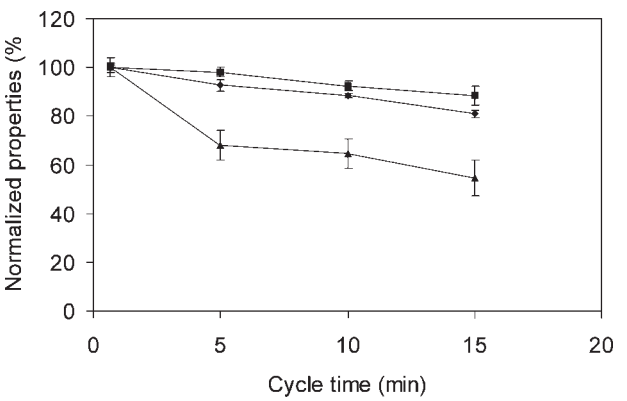

Figure 8 Mechanical properties of 50 wt % jute-PP compound versus cycle time in the injection molding equipment at 200°C: flexural strength (◆), flexural modulus (■), Charpy impact strength (▲).The mechanical properties of undegraded specimens have been normalized to 100%.

TABLE III
Swelling of Injection Molded 50 wt % Jute-PP Specimens after Saturation in 40°C (salt) Water, Standard Deviation is Indicated Between Brackets

	Demi water (%)	3.5 wt % Red Sea salt solution (%)	3.5 wt % NaCl solution (%)
Thickness	2.7 (0.2)	3.0 (0.1)	3.0 (0.1)
Width	2.7 (0.1)	2.8 (0.1)	2.8 (0.2)
Length	0.31 (0.02)	0.38 (0.04)	0.42 (0.03)

The use of 5 wt % MAPP in the 50 wt % jute-PP compound is based on previous research.[18]

Morphology

The fiber-matrix adhesion is good, which can be concluded from the short pull out lengths and the fibers being covered with matrix material (Fig. 3). Most of the jute fibers have been separated into the strong elementary fibers, although less fibers seem to have been refined to elementary fibers than in flax/PP compounds obtained during earlier extrusion trials.[22,29]

Rheology

The Bagley-correction for pressure drops at the entrance and exit of the die has been determined for natural fiber extrusion compounds during earlier experiments.[33] The correction resulted in a small shift of the viscosity to lower values in the shear rate range 20–5000 s^{-1} and in the temperature range 200–220°C. The slope of the viscosity curves was not affected. Therefore, the rheology data in this study have not been Bagley-corrected.

The reduction of viscosity at increasing processing temperature and at extended heating times is probably mainly due to the lower melt viscosity of the PP polymer. This indicates that processing becomes easier in case *hot spots* arise or in case processing equipment is not able to keep to short cycle times. A suggested blocking effect from natural fiber degradation has not been observed.

The addition of the jute fibers increases the viscosity of the PP but does not change the basic plastic character of its viscosity-shear rate dependency, as has also been observed for glass-PP by Nanguneri et al.[34] The viscosity of 30 wt % glass fiber reinforced PP compound as presented by Nanguneri has been incorporated in Figure 4 and shows the same slope as the 50 wt % jute-PP compounds from this study, although at somewhat lower viscosity values.

Two meter of compound has been pressed through the die and the pressure has risen to 2.7 MPa during the extended (36 min) preheating for the rheology tests at 220°C. This pressure may be caused by water or other components being formed during the preheating. The presence of water would result in a vapor pressure of 2.3 MPa at 220°C and 2.7 MPa at 227°C. The compound has been dried prior to rheological analysis and no pressure build up has been observed during the first 15 min of the preheating period. The presence of water can therefore only be explained as a result of degradation of components in the jute fiber. Alternatively, the pressure of 2.7 MPa and the extruded 2 m of compound presumably would be caused by formation of other degradation products from the jute fibers at 220°C or by water formed from condensation reactions of these degradation products.

Dimensional stability and accelerated degradation tests

Thermal expansion of the jute-PP compound in longitudinal direction of the test bars is 2.4 times lower than of the pure PP over the temperature range evaluated. If thermal expansion is linear in the temperature range 20–200°C, mold shrinkage values of 0.005 and 0.012 for the jute-PP and the PP respectively, are expected. Experimentally determined longitudinal mold shrinkage for the jute-PP and the pure PP was 0.003 and 0.016, respectively, indicating a less than linear expansion for the jute-PP compound above 80°C and a more than linear expansion for the pure PP above 80°C. The experimental value for jute-PP is comparable to the value of 0.003 reported for 50 wt % kenaf-PP by Caulfield et al.[4]

The length swelling of the injection molded jute-PP test bars in 40°C water is a factor of 8–9 lower than the thickness and width swelling (Table III). Mold shrinkage was seven times larger in thickness direction than in length direction. These observations suggest that the fibers have a preferred orientation perpendicular to the thickness direction of the

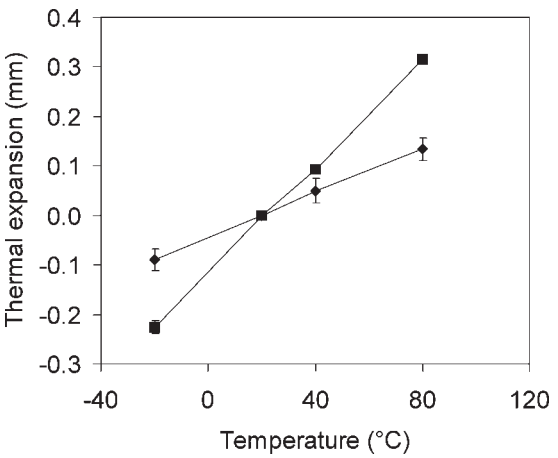

Figure 9 Thermal expansion of 50 wt % jute-PP compound (◆) and pure PP (■) relative to specimen length at 20°C.

TABLE IV
Diffusion Kinetic Parameters *D*, *n*, and *k* for 50 wt % Jute-PP Compounds

	D (10^{13} m^2/s)	n	k (g/g $days^n$)	k (g/g h^n)	k (g/g min^n)
Demi water, predried at 105°C	11.2	0.714	0.069	0.0037	0.00038
Demi water	8.72	0.719	0.051	0.0027	0.00027
3.5 wt % Red Sea salt solution	6.07	0.657	0.049	0.0033	0.00041
3.5 wt % NaCl solution	5.91	0.637	0.051	0.0038	0.00050

test bar, since for natural fibers it is known that the anisotropy in longitudinal direction is larger than in transverse direction.[35]

Jute fiber polymer composites absorb water due to the hydrophilic nature of the fibers. Flexural test bars have been used for evaluation of water absorption, to enable subsequent mechanical performance evaluation as well. An absorption value of 8.1 wt % for a 50 wt % jute-PP compound indicates that the jute fibers absorb $\sim$ 16 wt % water. This low value is probably due to tight encapsulation of the jute by the polymer matrix, which on its turn is a result of the use of MAPP compatibilizer. Karmaker et al.[36] show water absorption values of 20 wt % for 40 wt % jute fabric-PP composites.

The curvatures in the initial stages of the sorption curves in Figure 5 suggest that water diffusion in the jute-PP compounds is dependent on concentration.[37] In fact, water sorption rate increases with proceeding sorption time, which corresponds to sigmoid sorption as defined by Crank.[37] Sigmoid sorption refers to special cases in which diffusion flow and changes in surface concentration occur at comparable rates.[37] The change in effective water concentration at the specimen surface may depend on water repellent substances being present on the specimen surface and being washed away progressively upon sorption time. These water repellent substances may be processing aids present in the PP, which usually concentrate at the specimen surface during injection molding. Although water diffusion in the jute-PP composites depends on water concentration, still an average diffusion coefficient, D (m^2/s), can be defined for the more or less linear part of the curve, which relates to the maximum water absorption rate for the composites studied[37]:

$$D = \pi^*(h\theta/4M_\infty)^2 \quad (1)$$

where h is the initial sample thickness in meters, θ is the maximum slope of the absorption curves versus the square root of time in seconds, and M_∞ is the moisture content at equilibrium conditions.

The diffusion coefficient of water in 50 wt % jute-PP compounds is a factor of 1.5–2 larger for demi water than for salt water (Table IV). The salt appears to hinder water diffusion. As expected it has no effect on the equilibrium moisture content in the composite. The D-values are in the same order of magnitude as the values found for 30 wt % sisal-PP kneading compound by Espert et al.[13] and for 30 wt % flax pulp-PP kneading compound by Retegi et al.[14] and a factor of 20 larger than presented for 30 wt % sisal-PP extrusion compound by Joseph et al.[12] The water absorption rates in the jute-PP compounds are six orders of magnitude lower than determined for flax mat reinforced PP by Stamboulis et al.[30] This large difference in diffusion coefficient suggests that up to 50 wt % jute fiber content in PP, fibers are mainly isolated and individually surrounded by PP and are not forming a network of fibers, as is present in fiber mat reinforced thermoplastics. Water diffusion is likely to be affected by occasional defects in the composite materials, since the data of Espert et al.[13] and Retegi et al.[14] do not show an unambiguous relation between the diffusion parameter D and the fiber content.

From Figure 5 it can be concluded that water sorption in the jute-PP composites is not following Fickian diffusion.[37] Several authors on water diffusion in natural fiber-PP composites, however, do conclude that Fickian diffusion occurs.[12–14] To evaluate the water diffusion mechanism in natural fiber reinforced polymer composites, the following equation has been used[12–14]:

$$\log(M_t/M_\infty) = \log k + n \log t \quad (2)$$

where M_t is the moisture content at time t, M_∞ is the moisture content at equilibrium conditions, and k and n are constants. For Fickian diffusion, the value for n is equal to 0.5. Figure 10 shows the diffusion plots and Table IV summarizes the values for n and k. Fitting of the initial slope results in R^2 of over 0.99 for all four absorption cases, which is a good fit. Values for n are significantly larger than 0.5, indicating that water diffusion is not purely Fickian.[37] Values for n presented in literature are in the range 0.235–0.724 for PP compounds comprising 10–30 wt % of different (lingo)cellulosic fiber grades.[12–14] The authors' conclusion that natural fiber reinforced PP compounds show a tendency to approach Fickian diffusion appears to be uncorrect.

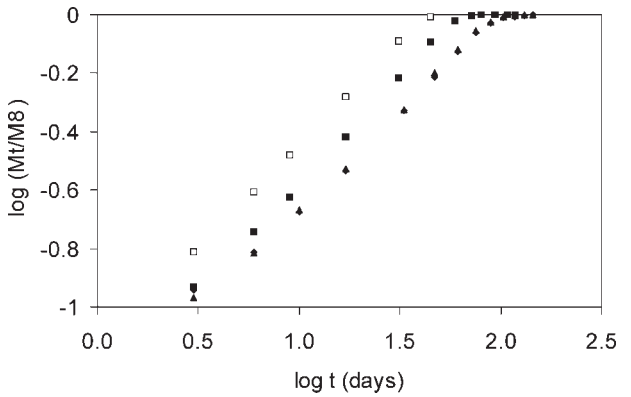

Figure 10 Diffusion case fitting plot for 50 wt % jute-PP compound in (salt) water at 40°C: demi water (■), demi water after drying of compound (□), 3.5 wt % Red Sea salt solution (▲), 3.5 wt % NaCl solution (◆).

Values for k in the present study are fitted, using the n values as derived above, and they are some factors lower than those found in literature. Retegi et al.[14] do not indicate the dimension of k, although this has a large effect on the value as indicated in Table IV. Joseph et al.[12] and Espert et al.[13] show k values calculated from eq. (2) and assuming a value for n of 0.5 while the related n values deviate as much as a factor of 1.66 from 0.5.

Water gradually enters the fibers throughout the whole material, and the relatively short lignocellulosic fibers become plasticized and their contribution to composite strength and stiffness will reduce.[4] Furthermore, the water causes the cellulosic fibers to swell and it is suggested that the swollen fibers develop shear stresses at the fiber-matrix interface, thus favoring fiber-matrix debonding, which in turn causes a reduction in composite strength.[12] The limited reduction in impact strength after water absorption may be caused by plasticization of the natural fibers, which are usually brittle after melt compounding in thermoplastics. Plasticization will reduce the crack initiating properties of the otherwise brittle fiber. Furthermore, plasticized fibers will have higher failure strain, thus also allowing the polymer matrix to absorb more energy until the higher failure strain. Strength and stiffness reduction of the 50 wt % jute-PP compounds at equilibrium moisture uptake is circa 40%, which is similar to the circa 20–40% reduction in stiffness and strength observed for 30 wt % sisal-PP compounds after saturation with water as presented by Joseph et al.[12] and Espert et al.[13]

It must be noted that PP strength improves by 22% after the water absorption test, probably as a result of crystallization of the polymer. Stiffness, strain, and impact of the PP remain unaffected after the water absorption test.

The UV irradiated pure PP specimens are very brittle and weak. Obviously, the PP itself has not been stabilized against UV well. On the other hand, UV irradiation hardly affects the mechanical properties of the jute-PP compounds. This is attributed to the jute fibers containing circa 12% of lignin, which is known for its UV stabilizing effect.[32,38] The jute fibers at the specimen surface absorb the UV irradiation—visualized by the fading top layer of the compounds—and thus protect the rest of the material against UV. Joseph et al. present similar data for UV irradiated sisal-PP compounds,[12] pure PP looses 95% of its strength after 12 weeks of UV radiation, whereas the strength of 30% sisal-PP composites decreases less than 25%.

The effect of thermal loading during injection molding was evaluated as well. Within the range of frequently used injection molding cycle times, i.e., within 5 min, reduction of flexural strength and stiffness is small. Impact strength, however, starts to reduce already at cycle times shorter than 5 min. The x-axis of Figure 8 indicates cycle time. However, we know that the residence time of the compound in the heated zone was circa three shots. The values on the x-axis in Figure 8 can thus be multiplied by circa 3 to obtain the actual residence time at 200°C.

The Sanitized biostabilizer inhibits bacterial growth to a major extent, whereas fungal growth was hardly inhibited. The high level of fungal growth is not in accordance with data by Richter for 70 wt % wood-PP composites with similar amounts of the same biostabilizer.[39] The reason for this is not yet elucidated and application of the 50 wt % jute-PP compounds in fungi sensitive conditions requires further investigation.

The mechanical properties of undegraded jute-PP compounds show very little variation (Table II) and exhibit strength and stiffness values, normalized for density differences, at the upper range of competing natural fiber reinforced compounds for injection molding.[1]

CONCLUSIONS

As far as the authors are aware, this is the first time that jute has been compounded into PP by a continuous extrusion process. The designed sliver feeder works well to get the jute sliver into the extruder. The obtained free flowing granules with up to 50 wt % jute in PP can be injection molded very well using conventional equipment. The viscosity of the compound goes down with increasing processing temperature and extended residence time at high temperature. Therefore the important conclusion can be made that no accelerated blocking effects inside processing equipment are to be expected.

The mechanical properties show very low variation and exhibit strength and stiffness values at the upper range of competing natural fiber reinforced

compounds for injection molding. The low water diffusion rate suggests that the fibers in the 50 wt % jute-PP composite are individually surrounded by PP to a large extent, and do not form a continuous fiber network. The water absorption by the compounds is not following Fickian diffusion. The jute fibers exhibit a UV stabilizing effect on PP. The mechanical performance gradually declines upon prolonged thermal loading and immersion in water. Bacteria, fungi, and garden mold hardly have a negative effect on mechanical properties.

This article has addressed processing and ageing properties of continuously produced jute-PP compounds. The actual use of jute-PP compounds in various applications will depend on the long term mechanical performance of the material and further up-scaling of the process. These topics are being worked on.

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