Jute Reinforced Polyolefines for Industrial Applications

Md. Siddiqur Rahman and Dr. Latifa Binte Lutfar International Jute Study Group (IJSG), Dhaka, Bangladesh

&

Mr. Martien Van den Oever Wageningen UR -AFSG, the Netherlands

<u>Abstract</u>

Since the 1980's natural fibre reinforced polymer composites have been extensively investigated. This has led to the use of various natural fibres like jute, kenaf, flax, sisal etc. in many polymer reinforced composites in a wide range of applications. However, significant use of natural fibres in polymer composites only started in the 1990's with wood polymer composite material.

It was apparent that there is a growing demand from the plastics compounding industry to find cheaper, lighter and eco-friendly alternatives to the use of glass fibre as reinforcing agents of plastics. Jute, having outstanding intrinsic mechanical properties, has the potential to compete with glass fibres as reinforcing agents in plastics. It has additional benefits of low cost, low density, renew-ability, recycle-ability and combustibility.

Jute reinforced polyolefines granules have been developed as an intermediate material for the production of various end-products to replace glass fibre reinforced Polypropylene (PP). Industrial trial application of the technology to produce jute based composites has been successfully conducted at industrial sites in Bangladesh and India. It has been observed that the new jute-based material has appropriate technical characteristics and economic potential for commercial exploitation in various end uses, particularly automotive parts, packaging and household articles.

The potential of jute based composites is based on its price/performance ratio, where the performance can reach that of glass fibre reinforced composites, but its price range is substantially lower. In addition, these composites can compete in a number of end uses with expensive engineering plastic products. In fact, by compounding plastic with jute the product price can substantially be lowered.

Key Words: Composite, Injection moulding, Granulation, Reinforcing fibre, Plastics, Biodegradable, Environment friendly.

1. Introduction

Jute is an annually grown natural fibre. It is extracted from the stems of plants belonging to the genus *Corchorus*, family Tiliaceae. About 40 species of *Corchorus* are known throughout the world but *C. capsularis* (White jute) and *C.olitorius* (Tossa jute) are the ones which are cultivated for their fibre (Kundu, 1956; Atkinson, 1965).

Jute and kenaf are versatile textile fibres. The fibres are biodegradable, environmentally, benign and renewable. Jute has wide range of usage. Besides being used as packaging materials worldwide it is now widely used as floor covering, home textiles, decorative fabrics, shopping bags, carry bags, handicrafts, cushion cover, curtains, blankets, nursery pots, insulation material, soil saver, etc. Also it is used in jute reinforced thermoset composites. It has the potential to be used in large scale as geotextiles in various applications like soil stabilisation, erosion control etc. It could be a good source of raw material for making pulp and paper. When used as a source of biomass fuel, jute and kenaf production helps to conserve tree cover and natural forests. Moreover, leaf and crop trash remains in the field to be recycled as organic materials, thereby reducing demand for supplementary chemical

fertilizers for subsequent crops. With the growing global awareness about a pollution free environment, jute is poised to be the fibre for the future for various end uses and applications.

2. Background

The traditional jute products' markets such as packaging materials for agricultural products (including sacks, bags, carpet backing cloth, packaging for fertilizers, cement and chemicals) are being eroded by synthetic substitutes.

To overcome the declining market of these conventional products of jute, new technologies have been evolved for bulk use of jute, as a raw material in the production of high value added and price competitive intermediaries or final products. A host of innovative new products generally termed as 'Diversified Jute Products' have been developed with high value-addition.

The process of diversification of the uses of jute has been the main thrust of global efforts and one alternative is the possible utilization of jute as a reinforcing agent in thermoplastics as the jute reinforced composites have better competitive advantages over glass fibre composites for various applications.

In fact, people have been using composite material from ancient times. The most primitive composite materials were straw and mud combined to form bricks for building construction. The advanced examples perform routinely on spacecraft in demanding environments. The visible applications pave our roadways in the form of both steel and aggregate reinforced asphalt concrete. Those composites closest to our personal hygiene form our shower stalls and bath tubs made of fiberglass. Solid surface, imitation granite and cultured marble sinks and counter tops are widely used to enhance our living experiences.

Since the 1980s, natural fibre reinforced polymer composites have been investigated extensively. This has led to the application of various natural fibres, like jute, flax, hemp, sisal, wood and likewise fibres in many polymer reinforced composites in a wide range of applications. Apart from wood panel and board products like MDF and Particle Board, significant use of natural fibres in polymer composites only started in the 1990s with wood polymer composite (WPC) material in the US and Japan, and recently also in Europe and China (**Table-1**). The main outlet for WPC material is decking, siding, fencing, profiles, etc. Key driver for the marketing of WPC materials in the US was lower lifetime costs due to expected maintenance benefits. The growth rate of WPC materials in the past years has been very high.

In European automotive industry, natural fibre mat reinforced thermoplastics (NMT), based on fibres other than from wood, have been applied since about 1995 (**Table-1**). One of the drivers for the developments in Europe was the sustainability aspect of natural fibres. For actual implementation, however, mainly lower cost appeared to be a driving force. Nearly 100% of the NMT materials are applied in the automotive sector.

Table-1: Production volumes of wood polymer composite (WPC) and other natural fibre reinforced thermoplastics materials worldwide (Carus, 2008a and 2008b).

Grade	Area	Volume	Year	Change (year)
		(kton)		
WPC profiles	China	150	2007	+ 100% (2006)
WPC profiles	North America	600	2003	+ 100% (2000)
WPC profiles	Europe	120	2007	+ 20% (2006)
WPC profiles	Japan	50	2007	+ 25% (2006)
NMT for trim applications	Germany	30	2005	Stable
Natural fibre-polymer granules	EU	4	2006	Growing

Recently, industry started to pay attention to injection moulding grades of natural fibre-polymer composites [Carus, 2008a]. This includes both wood and other natural fibre based polymers. The advantage of granules for injection moulding applications is the large freedom of design and the low cost at high volumes of injection moulding processing. The volume of non-wood natural fibre reinforced polymer granules was estimated by Nova Institut in Germany to be 4 kton in Europe in 2006 [Carus, 2008a].

The amount of natural fibre based composites produced currently is still nearly a factor of 5 lower than the amount of glass fibre reinforced polymers, and far lower than the amounts of the pure bulk polymers produced (**Table-2**). The majority of the glass fibres used in composites in Europe finds application in SMC/BMC (25%), and lay-up and spray-up (31%). Other fields of application are RTM (10%), sheets (7%), pultrusion (4%) and pipes and tanks (12%). All of these technologies comprise thermosetting processing. With 8%, thermoplastic glass-PP is completing the 100%. The glass-PP finds mainly application in the automotive industry (98%). Growth rates of glass fibre reinforced plastics are small compared to natural fibre reinforced composites, in the US the market volume even reduced in 2007.

Grade	Area	Volume	Year	Change to previous	Reference
		(kton)		year (%)	
Glass-PP	Europe	100	2007	+10	Schemme, 2008
Glass-all	Europe	1,200	2007	+ 5.6	Witten, 2008
Glass-all	US	1,600	2007	- 10	Witten, 2008
PP	World	45,000	2007	Few %	Ebner, 2008, p.13
PVC	World	33,200	2006	Few %	Wunderlich, 2008, p.10

Table-2: Production volumes of glass-PP (GMT, D-LFT, LFG) and pure polymers worldwide

Nova Institut from Germany states that "the forecast for the use of natural fibre reinforced polymers shows a trend significantly going up" [Carus, 2008b].

It has been observed that there is a growing demand from the plastics compounding industry to find cheaper, lighter and eco-friendlier alternatives to the use of glass fibres as reinforcing agents of plastics. Jute, having outstanding intrinsic mechanical properties, has the potential to compete with glass fibres as reinforcing agents in plastics. Supplementary benefits include low cost, low density, renew-ability, recycle-ability and combustibility. In addition, they are less abrasive for the equipment during processing with thermoplastics and do not expose operators to potential safety or health problems.

In this backdrop the International Jute Study Group (IJSG) initiated and has successfully implemented a project entitled 'Jute Reinforced Polyolefines for Industrial Applications'. The main objective was to develop new jute-based materials having appropriate technical characteristics and economic potential for commercial exploitation in various end-uses.

The development of jute reinforced polypropylene (PP) granules as intermediate material for the production of various end-products to replace glass fibre (GF) reinforced PP was found suitable to be used to produce such products as crates and pallets, furniture, automotive parts and household goods.

3. Materials

Jute fibre was processed into a sliver, wound into a wheel and wrapped for shipment. Polypropylene (Stamylan P 17M10, homopolymer, $MFI_{2.16,230} = 10.5$) was obtained as granules from DSM (currently Sabic).

Polypropylene (D220.01, performance polymer, $MFI_{2.16,230} = 52$) was obtained as granules from Dow Chemical. Polypropylene (PHC27, block copolymer, $MFI_{2.16,230} = 14$) was obtained as granules from Sabic. Maleic anhydride grafted PP (Epolene G 3015) was obtained as granules from Eastman

Chemical Corp. A bio-stabilizing masterbatch Sanitized MB E 22-70 was provided by Sanitized AG (Burgdorf, Switzerland).

4. **Principle of composites**

There are two categories of constituent materials for a composite: matrix and reinforcement. At least one portion of each type is required. The matrix material surrounds and supports the reinforcement materials by maintaining their relative positions. The reinforcements impart their special mechanical and physical properties to enhance the matrix properties. A synergy produces material properties unavailable from the individual constituent materials, while the wide variety of matrix and strengthening materials allows the designer of the product or structure to choose an optimum combination. Engineered composite materials must be formed to shape. The matrix material can be introduced to the reinforcement before or after the reinforcement material is placed into the mold cavity or onto the mold surface. The matrix material experiences a melding event, after which the part shape is essentially set. Depending upon the nature of the matrix material, this melding event can occur in various ways such as chemical polymerization or solidification from the melted state. Most commercially produced composites use a polymer matrix material often called a resin solution. There are many different polymers available depending upon the starting raw ingredients. There are several broad categories, each with numerous variations. The most common are known as polyester, vinyl ester, epoxy, phenol, polyamide, polypropylene, and others. The reinforcement materials are often fibers but also commonly ground minerals.

A variety of molding methods can be used according to the end-item design requirements. The principal factors impacting the methodology are the nature of the chosen matrix and reinforcement materials. Another important factor is the gross quantity of material to be produced. Large quantities can be used to justify high capital expenditures for rapid and automated manufacturing technology. Small production quantities are accommodated with lower capital expenditures but higher labour and tooling costs at a correspondingly slower rate. Fibre reinforced composites can be divided into a number of groups: 1) Short fibre reinforced thermoset composites that are injection moulded or compression moulded. 2) Fibre mat reinforced thermoset composites which comprise the use of a fibre woven or non-woven. The techniques are called resin transfer moulding (RTM), vacuum infusion moulding (VA-RTM), hand lay-up, etc. 3) Fibre mat reinforced thermoplastic, usually PP. 4) Fibre reinforced thermoplastics for injection and compression moulding applications.

5. Experimental & Results

The Agrotechnology and Food Sciences Group at Wageningen UR (Wageningen UR - AFSG) of the Netherlands was the Project Executing Agency (PEA) of the project and developed an extrusion compounding technique to produce agro-fibre/thermoplastic compounds.

5.1 Processing

The extrusion compounding process of jute-PP basically consisted of 3 steps: the feeding of jute fibre and polymer granules to a co-rotating twin screw extruder, actual compounding in the extruder and jute-PP granule formation (**Figure-1**). Compounds containing 30, 35, 40 and 50 wt. % jute in PP were produced using a Berstorff ZE 40-38D co-rotating twin screw extruder (**Figure-2**). The compounding consisted of homogeneously mixing the 2 components and at the same time refining the jute fibre to the level of its strong elementary plant cells. The jute should contain less than 8 wt. % of moisture while entering the extruder in order to enable stable extrusion processing.

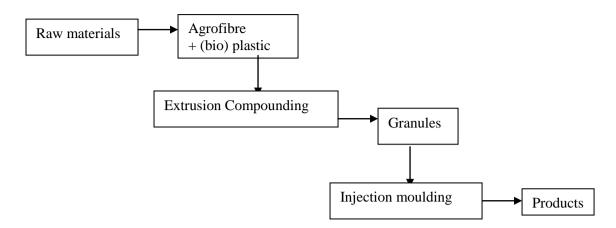


Figure-1: Schematic overview of extrusion compounding process, comprising feeding of fibre and polymer to the extruder, actual compounding and granule formation.

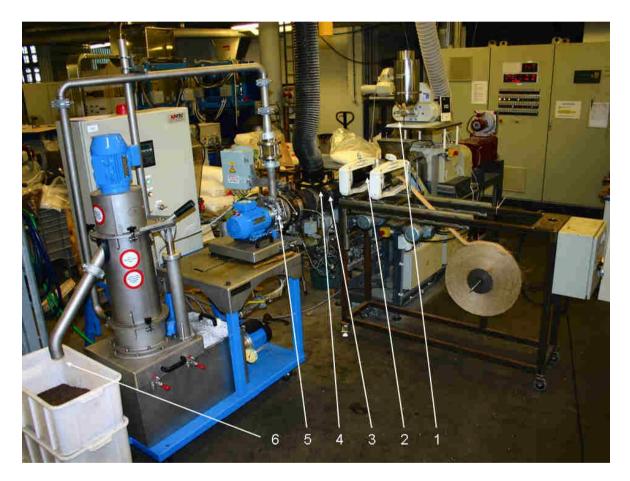


Figure-2: Experimental set-up for pilot scale extrusion: 1) polymer granule feeding; 2) jute sliver feeding; 3) compounding inside the extruder; 4) venting opening; 5 granule formation in underwater pelletizer; 6) granule collection.

To each 10 wt. % of jute fibre, 1 wt. % of MAPP was added. The PP and MAPP granules were premixed and fed upstream to the extruder by gravimetric feeding. The continuous jute sliver was unwound and fed downstream to the extruder with an in-house built sliver feeder (**Figure-3**). The sliver is an intermediate product of jute yarn production. A vacuum degassing unit was positioned between the compounding section and the die, to maximize moisture extraction. Free flowing jute-PP granules were obtained using a Gala LPU under-water pelletizer. The pelletizing device may be a different type than an underwater pelletizing system. The underwater pelletizing system has been successfully used in this project however, which means that free flowing granules have been obtained

that can be processed further in basically any injection moulding machine. These granules contain adhering and absorbed water and need further drying prior to packing/sealing into plastic bags.



Figure-3: Jute sliver feeder

The granules were dried overnight at 80°C and injection moulded into flexural/impact test bars with dimensions 80x10x4 mm³ using a Demag ERGO tech 25-80. Before further analysis, all composite specimens were conditioned for at least 1 week at 20°C and 50% relative humidity (RH).

Results

It was observed that feeding of the jute sliver is adequate when the sliver wheel can be unwound easily. Extrusion processing of 50 wt.% jute fibre in PP is stable with respect to the torque of the screws, the die pressure and the melt temperature when the moisture content in the jute fibre is below 8 wt.%. These conditions result in compact granules. When the moisture content of the jute is about 10 wt. %, the compound starts foaming and comes out of the venting opening in the extruder. Apparently, not all water can be released in the vacuum vent completely. 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. Under these conditions the torque of the screws and the die pressure is not constant and the obtained compound granules have many air inclusions. The trials with 30-40 wt. % jute fibre in PP exhibited fewer irregularities if moisture content in the jute was above 8%. The 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 moulding, however, the mechanical performance of the foamed granules is on the same level as the solid granules. The resulting granules have similar shape and diameter as commercially available PP granules. The injection moulding process exhibits no irregularities.

If the moisture content in the jute entering the extruder is low enough to allow it's evaporation in the venting opening, nice free flowing granules are obtained that are suitable for injection moulding in any conventional equipment. Excess moisture in the jute fibre results in poor processing and foamed granules, the mechanical performance after injection moulding is on the same level as for perfectly processed granules.

This process yields composite granules as intermediate products, which subsequently can be used in thermoforming processing techniques, such as injection moulding and compression moulding. The design of this process is such that during the continuous mixing the jute fibres are opened up to elementary fibres with a high aspect (length to diameter) ratio, which are homogeneously distributed in the polymeric melt. The addition of coupling agents has enabled the highly polar jute fibre surface

and the non-polar polymer to show good interaction. The process results in a compounded material with improved rigidity and strength.

Although the maximum nominal throughput of the pelletizer is 180 kg/h, at 15-20 kg/h throughput, a kind of pulsed flow occasionally causes 'freezing' of the molten compound at the die, thus blocking the flow and eventually requiring a fresh start-up of the process. It is expected that a modified fibre feeding system and composite granulation method will increase the throughput to a level of over 100 kg/h, which is supposed to make the process economically feasible. In this project focus was on injection moulding applications because of far higher relevance for the plastics processing industry in India and Bangladesh. The resulting granules are suitable for use in any conventional injection moulding equipment, as long as processors take notice of a few aspects. Key requirement is that the jute-PP granules are dried prior to injection moulding to achieve proper moulding and to avoid degradation of the jute during processing. Furthermore, processing should be performed at lowest temperature and shear forces possible and during shortest melting time possible, actual parameter limits to be determined for each specific product design. The granules are in principle also suitable for compression moulding.

5.2 Morphology: Morphology addresses the structure of a material.

Scanning electron microscopy (SEM) was performed on cryo-fractured surfaces of an injection moulded test bar based on 50% jute-PP using a Jeol JSM-5600 LV scanning electron microscope. Prior to the analysis the fracture surfaces were covered with a 10 nm layer of platinum using an Oxford CT1500 sputter coater.

Results

The injection moulded jute-PP composite contains mainly elementary fibres and bundles of a few elementary fibres that are still glued together (**Figure-4**). The fibre pull out lengths in general are shorter than the fibre diameter and fibres are covered with matrix material, which suggests that the fibre-matrix adhesion is good.

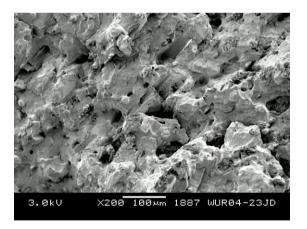


Figure-4: SEM picture of a cryo-fractured injection moulded test bar containing 50% Jute in PP.

5.3 Rheology: addresses the flow of a (reinforced) plastic

Granules based on 50% jute-PP were dried and rheological analysis was performed at 200°C 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 minutes while applying a pressure of 1 MPa at time t=0 and t=3 minutes. The analysis was repeated with a pre-heating time of 36 minutes in order to study the effect of thermal degradation on rheology. Pure PP was tested as a reference material.

Results

The Bagley-correction for pressure drops at the entrance and exit of the die has been determined for natural fibre extrusion compounds during earlier projects. 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 $200 - 220^{\circ}$ C. The slope of the viscosity curves was not affected. Therefore, the rheology data in this study have not been Bagley-corrected.

Increase of the temperature from 200°C to 220°C resulted in a reduction of the viscosity. Also an extended residence time of 30 minutes at these high temperatures resulted in a lower viscosity, although the drop in viscosity is very minimal at 200°C. 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. The addition of the jute fibres 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 (Nanguneri et al³.

A flow additive used in natural fibre reinforced profile extrusion (Licomont ET 141 of Clariant GmbH) has been evaluated for its effect on rheology. However, whereas flow improved to some extent, the flexural and impact strength of the composites dropped dramatically, from 85 to 55 MPa and from 13 to 6 kJ/m^2 respectively.

Partial replacement of the PP by a PP grade with lower viscosity (higher Melt Flow Index, MFI) have been analysed as well. Replacement of 33% of the standard PP with a high MFI grade PP resulted in a slightly better flow (10% lower injection pressure) and a better performance, from 85 to 89 MPa flexural strength and from 13 to 15.4 kJ/m² Charpy impact strength. Using just a PP grade with a slightly higher MFI, 14 vs. 10.5, also resulted in a 10% lower injection pressure (better flow), but with less good strength performance, going down from 85 to 68 MPa.

Conclusions

The addition of the jute fibres 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 composites. An increase of the processing temperature, for instance as a result of hot spots, causes a reduction of the viscosity. Also an extended residence time at typical processing temperatures results in a lower viscosity, thus indicating that no blocking of equipment due to natural fibre degradation has to be expected during injection moulding. The flow of jute-PP composites may be improved by partial replacement of the PP by a PP grade with a very low viscosity.

5.4 Flexural properties

The flexural properties of the specimens conditioned at 20° C and 50% RH for 1 week 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 for 5 specimens.

Results

Figure-5 shows the effect of jute content on the mechanical properties of the extrusion compounds. The flexural modulus is proportional to the fibre fraction. The flexural strength seems to level off above 40% fibre, whereas the strain goes down above 40% fibre content. The mechanical properties of un-degraded jute-PP compounds show very little variation (standard deviation error bars have been included in the graph). The mechanical performance of pure PP is 1.3 GPa flexural stiffness, 43 MPa flexural strength and 6.9 % strain at maximum stress.

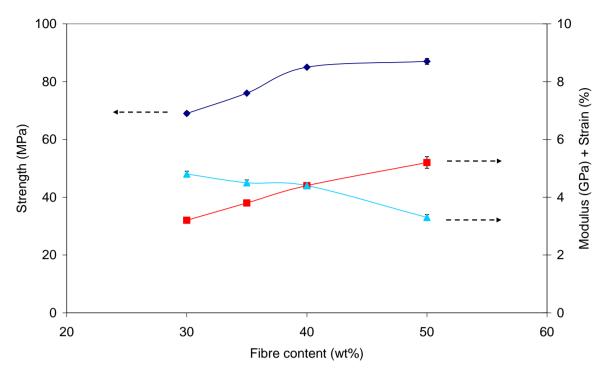


Figure-5: Mechanical properties of jute/PP extrusion compounds versus fibre wt. fraction: flexural strength (\blacklozenge), flexural modulus (\blacksquare) & strain at fracture (\blacktriangle).

Conclusions

Incorporation of 50% jute in PP results in a 4 fold increase of flexural modulus and a 2 fold increase in flexural strength. The strain at fracture decreases upon increasing jute fibre loading.

5.5 Impact strength

The Charpy unnotched impact strength of the specimens conditioned at 20°C and 50% RH for 1 week 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 for 8 specimens.

Results

Once the PP contains jute fibres, the Charpy impact strength seems not to be much affected by fibre content, reaching values between 15 for 50% jute and 18.4 kJ/m² for 40% jute, whereas pure PP does not break in the Charpy test. The scatter in data is quite small. The impact strength may be improved by addition of impact modifiers. **Figure-6** presents the effect of 15wt. % of different impact modifiers on Charpy impact strength. For compounds including cotton, rayon and PAN fibres, the jute fibre content was reduced from 50% to 35%. The SBS impact modifiers Kraton and Tuftec H yield the best improvement in Impact. The strength and stiffness, however, reduce significantly. Rayon fibre, and to a less extent cotton fibre, increased both impact and flexural strength significantly, leaving the stiffness at nearly the same level. PAN fibre resulted in a reduction of all 3 mechanical properties.

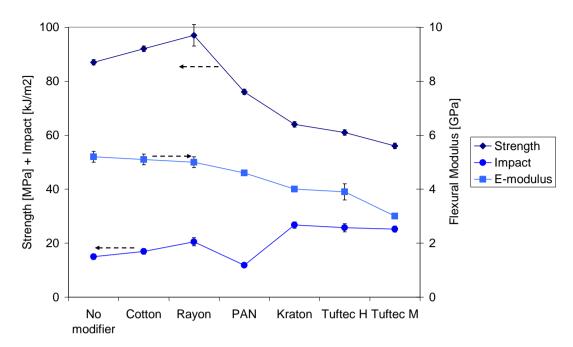


Figure-6: Effect of impact modifiers (15wt. %) on Charpy impact strength, flexural strength and modulus.

Conclusions

The Charpy impact strength of PP decreases upon incorporation of jute fibres. The impact can be improved by using impact modifiers. The strength and stiffness however as a result may decrease. Rayon fibre appears to be the best option found thus far.

5.6 Heat deflection temperature (HDT)

HDT is the temperature at which a material shows a specific deflection under a specific load. It was observed that Heat deflection temperature increases with increase in the jute fibre loading.

5.7 Accelerated degradation

5.7.1 UV irradiation

Injection moulded 50 wt. % jute-PP test bars were irradiated according to ISO 4892-2 (2003) in a Heraeus Suntest CPS for 8 weeks at maximum power, being 765 W/m^2 . 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

The mechanical properties of the un-degraded jute-PP specimens are summarized in Table 3.

Table-3: Flexural and Charpy impact properties of un-degraded 50% jute-PP composite and pure PP.

	Stiffness [GPa]	Strength [MPa]	Strain [%]	Impact [kJ/m ²]
PP	1.3	43	6.9	Not broken
50% Jute/PP	5.1	86	3.2	15

The mechanical properties of 50% jute-PP and pure PP versus UV irradiation time have been presented in **Figure-7**. The mechanical properties of un-degraded specimens have been normalized to 100%. 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 fibres containing circa 12% of lignin, which is known for its UV stabilizing effect. The jute fibres 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. [2002] present similar data for UV irradiated sisal-PP compounds, 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%.

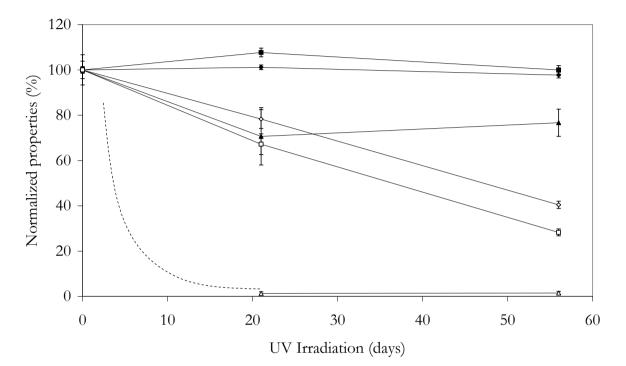


Figure-7: Mechanical properties of pure PP and 50 wt.% jute-PP compound versus UV irradiation time: flexural strength (◆), flexural modulus (■), Charpy impact strength (▲); PP (open symbols), 50 wt.% jute-PP (solid symbols). The mechanical properties of un-degraded specimens have been normalized to 100%.

5.7.2 Biodegradation

Injection moulded test bars with and without bio-stabilizer were subjected to 3 biodegradation tests at Sanitized AG (Switzerland). The content of bio-stabilizer was 3% of the total composition, added as replacement for PP. Specimens were subjected to a bacterial resistance test for 24 hours 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 4 weeks according to ASTM G 21-96 and evaluated for surface growth of fungi. A third series of specimens was tested in garden mould at 29°C for 6 weeks according to EN ISO 846 – Section D and evaluated for weight loss.

Results

The observations of the biodegradation tests are summarized in **Table-4**. After 1 day of bacterial incubation, the standard 50 wt. % jute-PP compound was fully covered with bacteria. Addition of 3 wt.% of the bio-stabilizer Sanitized MB E 22-70 restricted bacterial growth to 5% of the 3 wt.% specimen surface. The bio-stabilizer hardly contributes to the resistance to fungi and garden mould. The high level of fungal growth is not in accordance with data by Richter [2004] for 70 wt.% wood-PP composites with similar amounts of the same bio-stabilizer. 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 biodegradation tests virtually cause no reduction in flexural strength and stiffness. The Charpy impact strength reduces with 8%, independent of the use of bio-stabilizer.

Results

Table-4: Effect of bio-stabilizer Sanitized MB E 22-70 on biodegradation of 50% jute-PP composites.

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

5.7.3 Thermal degradation

Thermal degradation was simulated by performing injection moulding 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 minutes.

Results

The mechanical properties of un-degraded specimens have been normalized to 100%. The flexural strength and stiffness and Charpy impact strength of the jute-PP compounds gradually decrease during thermal degradation, while the properties of PP are hardly affected. Within the range of frequently used injection moulding cycle times, i.e. within 5 minutes, reduction of flexural strength and stiffness is small. Impact strength, however, starts to reduce already at cycle times shorter than 5 minutes. The x-axis of **Figure-8** indicates cycle time. The injection moulding process, even after cycle times of 15 minutes at 200°C, exhibited no irregularities and moulded samples could be released easily from the mould.

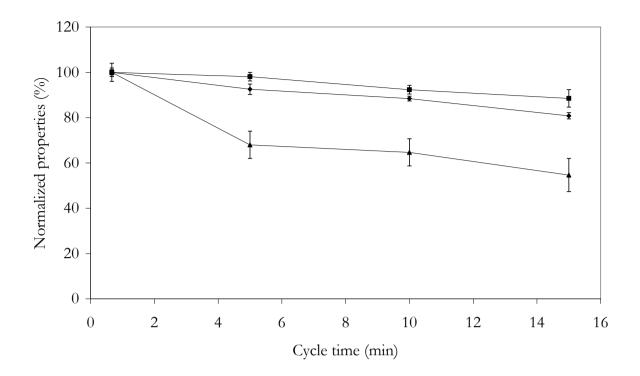


Figure-8: Mechanical properties of 50 wt.% jute-PP compound versus cycle time in the injection moulding equipment at 200°C: flexural strength (◆), flexural modulus (■), Charpy impact strength (▲). The mechanical properties of un-degraded specimens have been normalized to 100%.

5.7.4 (Salt) Water absorption

Injection moulded 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 moulded pure PP was included as reference. The specimens were cooled to 20°C in demi water for 24 hours prior to flexural and Charpy impact testing.

Result

Jute fibre polymer composites absorb water due to the hydrophilic nature of the fibres. Flexural test bars have been used for evaluation of water absorption, in order to enable subsequent mechanical performance evaluation as well. The (salt) water absorption of injection moulded 50 wt.% jute-PP test bars at 40°C as a function of time is presented in **Figure-9**. The water absorption levels off after ca. 60 days to a value of 8.1 ± 0.06) wt.% for dried composites in demi water. An absorption value of 8.1 ± 0.06 wt.% jute-PP compound indicates that the jute fibres absorb ca. 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. [1994] show water absorption values of 20 wt.% for 40 wt.% jute fabric-PP composites. The composite specimens that were not dried prior to water absorption reach maximum values of 6.7 ± 0.03) wt.% after ca. 90 days in demi water and of 6.7 ± 0.04) wt.% after ca. 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 9 would interfere with the dots.

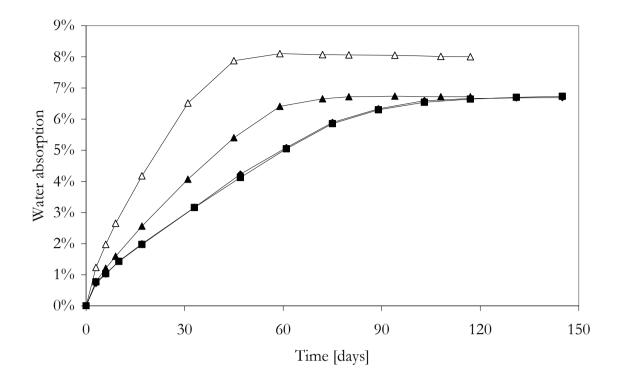


Figure-9: Water absorption curves for extrusion compounded 50 wt. % jute-PP in water at 40°C: Demi water after pre-drying at 105°C (△), Demi water (▲), 3.5 wt. % Red Sea salt water solution (♠), 3.5 wt. % NaCl water solution (■).

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

Water sorption rate increases with proceeding of sorption time. 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 moulding.

The length swelling of the injection moulded jute-PP test bars in 40°C water is a factor of 8–9 lower than the thickness and width swelling (**Table-5**). Mould shrinkage was 7 times larger in thickness direction than in length direction. These observations suggest that the fibres have a preferred orientation perpendicular to the thickness direction of the test bar, since for natural fibres it is known that the anisotropy in longitudinal direction is larger than in transverse direction.

Table-5:	Swelling of injection moulded 50% Jute-PP composite specimens after saturation in
	40°C (salt) water. Standard deviation is indicated between brackets:

	Demi water	3.5 wt. % Red Sea	3.5 wt. % NaCl
	[%]	salt solution [%]	solution [%]
Thickness	2.7 (0.2)	3.0 (01)	3.0 (0.1)
Width	2.7 (0.1)	2.8 (0.2)	2.8 (0.2)
Length	0.31 (0.02)	0.38 (0.04)	0.42 (0.03)

The flexural strength and stiffness and Charpy impact strength of the jute-PP compounds gradually decrease during (salt) water absorption (**Figure-10**). Absorption of demi water and salt water have a similar effect on the mechanical properties of the jute-PP compounds. Water gradually enters the fibres throughout the whole material, and the relatively short lignocellulosic fibres become plasticized and their contribution to composite strength and stiffness will reduce. The limited reduction in impact

strength after water absorption may be caused by plasticization of the natural fibres, which are usually brittle after melt compounding in thermoplastics. Plasticization will reduce the crack initiating properties of the otherwise brittle fibre. Furthermore, plasticized fibres 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 Espert et al. [2004] and Joseph et al. [2002].

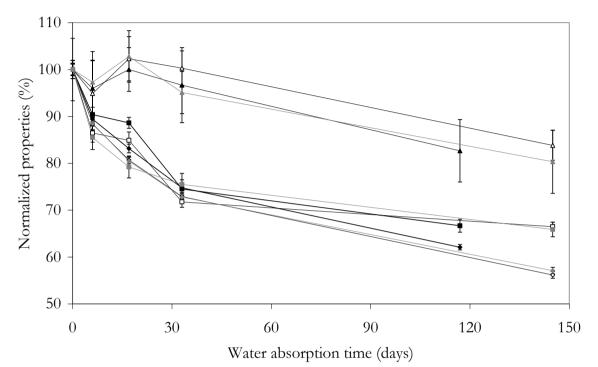


Figure-10: Mechanical properties of pure PP and 50 wt.% jute-PP compound versus water absorption time: flexural strength (◆), flexural modulus (■), Charpy impact strength (▲); demi water (solid symbols), 3.5 wt.% Red Sea solution (open symbols), 3.5wt.% NaCl solution (grey symbols). The mechanical properties of un-degraded specimens have been normalized to 100%.

Conclusions

The low water diffusion rate suggests that the fibres in the 50 wt.% jute-PP composite are individually surrounded by PP to a large extent, and do not form a continuous fibre network. The jute fibres exhibit a UV stabilizing effect on PP. The mechanical performance gradually declines upon prolonged thermal loading and immersion in water. Bacteria, fungi and garden mould hardly have a negative effect on mechanical properties.

5.8 Dimensional stability

Thermal expansion in length direction of moulded specimens was determined at -20, 40 and 80 °C, relative to the length at 20°C. Evaluation was performed in 5-fold, using a caliper with accuracy 0.01 mm. Mould shrinkage was determined in 3 dimensions by using a caliper.

Results

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 (**Figure-11**). If thermal expansion is linear in the temperature range 20–200°C, mould shrinkage values of 0.005 and 0.012 for the jute-PP and the PP respectively are expected. Experimentally determined longitudinal mould shrinkage for the

jute-PP and the pure PP was 0.003 and 0.016, respectively. The experimental value for jute- PP is comparable to the value of 0.003 reported for 50 wt.% kenaf-PP by Caulfield et al. [1999].

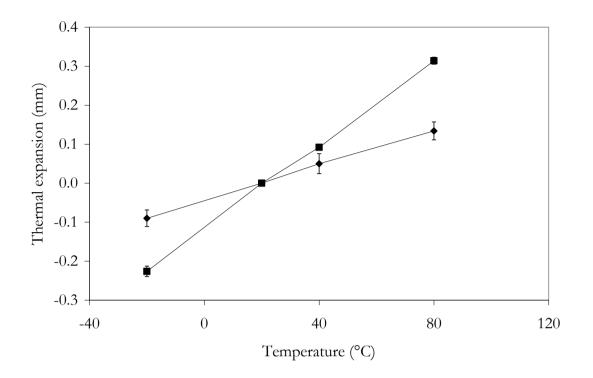


Figure-11: Thermal expansion of 50 wt.% jute-PP composite (♦) and pure PP (■) relative to specimen length at 20°C.

Conclusions

Thermal elongation of jute-PP composites is lower than for pure PP, which makes it more dimensionally stable, but which also should be taken into account when designing moulds for injection moulding.

5.9 Fogging and odour

These aspects are of particular importance for application in automotive industry.

Odour tests were performed according to VDA 270 C3 of the German 'Verband der Automobilindustrie'. Injection moulded specimens were heated in a glass beaker at 80°C for 2 hours and subjectively evaluated by 3 selected persons. Evaluation was done on a 6-point scale (1 is low odour level, 6 is very high odour level) and values were averaged.

Fogging tests were performed according to DIN 75201 B. Three injection moulded specimens were put in a glass beaker that was immersed in an oil bath heated at 100°C. The top cover of the beaker was a pre weighed aluminium sheet which was cooled at 21°C. After 16 hours of heating, the aluminium foil is removed, dried over a desiccant for 4 hours and weighed again.

Results

The results of the fogging trials and odour evaluations are presented in **Table-6**. A 50% jute-PP composite, comprising jute with batching oil, exceeds the fogging limit set by the European automotive industry, whereas the odour keeps just within limits. Jute without batching oils performs far better in the fogging trial and meets the limit. No odour evaluation on a specimen containing jute without batching oil was performed.

Table-6:Fogging and odour characteristics of extrusion compounded and injection moulded Jute-
PP; between brackets the standard deviation is presented.

	50% jute, with batching oil	5	Limit [*]
		batching oil	
Fogging [mg]	4.5 (0.24)	1.2 (0.01)	≤2
Odour [-]	3	-	≤3

* Limit as set by European automotive industry

Conclusions

The absence of batching oil has a positive effect on the fogging characteristics of injection moulded specimens. Specimens containing jute without batching oil meet the requirements of the European automotive industry.

5.10 Colour

Jute-PP granules containing 50 wt.% of jute fibre were dried overnight at 80°C with pre-dried air and mixed 50:50 with pure PP and additional 2 wt.% of a range of pigments. These compositions were injection moulded into flexural/impact test bars with dimensions 80x10x4 mm³ using a Demag ERGOtech 25-80.



Figure-12: Flexural test bars with 25% of jute fibre and household items with 40% of jute fibre, all containing 2% of pigment.

Results

Whereas jute-PP has basically a brown colour, by addition of pigments a whole range of colours can be obtained. The samples from (**Figure-12**) contain 2% of pigment each. For more bright colours, larger amounts of pigment need to be used, thus adding to the cost.

Conclusions

The brown basic colour of jute-PP composite material can be coloured to any dull colour.

5.11 Comparison to competing materials

Jute-PP composite granules exhibit a mechanical properties profile that is close to that of glass fibre-PP composites (**Table-7**). Jute-PP granules outperform jute non-woven based PP sheets on flexural strength and stiffness and come close to its HDT performance. Apart from impact strength, it outperforms PP and talcum filled PP.

Type of Product	Modulus (GPa)	Strength (MPa)	Charpy (kJ/m ²)	HDT-A (°C)
PP	1.3	43	Not Broken	55-60
50% Jute-PP (this	4-5.4	65-90	18-27	107-115
project)				
Jute-PP Sheet, non-	3.9-4.2	48-50	>8.5	110-135
woven based**				
Woodstock sheet ^{**}	2.5-3.9	30-48	>4	
30% Glass fibre-PP,	5.3	113	31	134
from India ^{**}				
40% Talcum-PP	2.3		Not broken	75
(Rutsch 2008)				

 Table-7:
 Composite performance of jute-PP granules and competing materials

** Experimentally determined in this project

Conclusions

Jute-PP composite granules can compete very well with pure PP, talcum filled PP and glass fibre reinforced PP in specific applications, in particular if stiffness and strength is required. Whereas in this project only 2 jute grades were evaluated, from trials with flax fibres in previous projects it has become clear that fibre quality has minimal effect on the composites performance.

6. **Products**

Free flowing jute-PP granules were produced at Wageningen UR - AFSG in pilot scale quantity and used for moulding trials at several industrial moulders in Dhaka and Kolkata. Manufactured products include different automotive door panel items; step of a lorry; cone for yarn spinning; electric extension cable cover; motorcycle handle. Examples of moulded products obtained after: poor drying & insufficient drying are presented in **Figure-13**, products after adequate drying are presented in **Figure-14**.





Poor drying

Insufficient drying

Figure-13: Poor drying results in a 'burnt like product' (left) and insufficient drying results in fair looking product (right).



Figure-14: Products made from 40 wt.% Jute-PP after adequate drying: Back plate of magazine cover (left), Electric cable cover (middle) & Bobbin (right).

7. Potential

Currently, India and Bangladesh produce ca 1,600 and 1,000 kton of jute fibre per annum (kton/a), respectively. This is a huge amount compared to the estimated jute consumption of one typical commercial production line of ca. 0.2-0.75 kton/a. As a consequence, during the start up of commercialization of jute-PP granules, no effect on the jute fibre market is expected. At the same time it may be mentioned that during the past many years, jute in Bangladesh and India is cheaper than natural fibres with similar performance in e.g. Europe. On average, jute prices/kg are half that of flax, hemp and sisal, making jute very competitive in the global natural fibre reinforced plastics arena. In principle, target products could be in the following sectors: automotive; (electronic) consumer items; construction and housing; storage and handling items; diverse. The market for potential replacement by jute-PP is large compared to the output of a foreseen commercial production unit, being 0.36-1.44 kton/a, although jute-PP will only meet the requirements of a fraction of the glass fibre composite and pure PP markets. The annual world consumption of pure PP being 45,000 kton/a and the consumption of glass fibre reinforced composites, both thermoplastic and thermo-set, being estimated to be 2,800 kton/a in Europe and US only of which ca 100 kton/a of glass fibre reinforced plastics are being used by European automotive industry.

8. Conclusion

Jute reinforced materials could be initially introduced in the market segments like automotive industry (door panels, dashboard, instrument panels), Packaging: both for inland transport and export of agricultural products (crates, inlays for crates, pallets, boxes, and cases) and Consumer products (housing for computer screens, refrigerators, etc).

The potential of jute based materials is based on its price/performance ratio, where the performance can reach that of glass fibre reinforced composites, but its price range is substantially lower. In addition, these composites can compete in a number of end uses with expensive engineering plastic products. In fact, by compounding plastic with jute the product price can substantially be lowered.

The fibre quality is not expected to have an effect on composite performance, which means that cheap low quality jute can be used for making jute-PP granules. A market study was conducted which shows that jute based composites have market potential as they are cost competitive. It is, thus, expected that Jute Reinforced Polyolefines (JRP) would find industrial applications worldwide.

References

- Atkinson, R.R, Jute-fibre to yarn, B.I. Publications, Bombay India, 1965
- Carus M., Reinforced Plastics, 52 (4), 2008, p.18-25.
- Carus M., Proceedings of 6th N-FibreBase Conference, presentation P3, 22-23 September 2008b, Essen (Germany).
- Caulfield et al., Angewandte Makromolekulare Chemie, 272, 1999, p.57.
- Ebner M., KunstStoff Trends, 8 (4), 2008, p.12-13.
- Espert et al., Composites Part A, 35, 2004, p.1267.
- Joseph et al., Composites Science and Technology, 62, 2002, p.1357.
- Karmaker et al., Journal of Applied Polymer Science, 54, 1994, p. 1803.
- Kundu, B.C., Jute-World's foremost bast fibre, 1. Botany, agronomy, pests and diseases, Econ. Botany, **10**, 103 133 (1956)
- Nanguneri et al., Rheol Acta, Vol. 26, 1987, p. 301
- Richter, Additives for wood plastic composites, 5th Global Wood and Natural Fibre Symposium, Kassel, Germany, 27-28 April 2004
- Rutsch, H. W. Proceedings of 6th N-Fibre Base conference, Presentation C4, 22-23 September 2008, Essen (Germany).
- Schemme M., Reinforced Plastics, 52 (1), 2008, p.32-39.
- Witten E., Proceedings of 6th N-FibreBase Conference, Presentation P2, 22-23 September 2008, Essen (Germany).
- Wunderlich W., KunstStoff Trends, 8 (4), 2008, p.10-11.