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DIRECT LONG BIOFIBRE THERMOPLASTIC COMPOSITES FOR AUTOMOTIVE, AEROSPACE AND TRANSPORTATION INDUSTRIES

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ABSTRACT

Natural fibres such as flax, hemp, jute and wood are increasingly being used in various industries as reinforcing materials for composites to reduce weight, cost and environmental impact. These fibres can have the added benefit of producing equal or higher stiffness-to-weight ratios than glass fibres. However, processing natural fibres presents a number of challenges, some of which are common to other types of fibres, such as the ability to de-bundle, mix and uniformly distribute them throughout the entire volume of a composite part. One particular challenge for natural fibres is the processing temperature limitations determined by their propensity to thermally degrade after long exposure times. This paper deals with the challenges of using biofibres as reinforcing materials for thermoplastic resins. The research work involves the use of short flax fibres in a continuous compounding process and flax fibres in the form of rovings and slivers in a Direct-Long Fibre Thermoplastic (D-LFT) process. The materials were compounded and moulded to produce parts for characterization. Polypropylene (PP) was used as polymer matrix because of its proven performance in automotive applications. Flax fibres were chosen given their combination of good mechanical properties, availability and relative low cost compared to other bast fibres. Different formulations using heat stabilizers, antioxidants and coupling agents were implemented with the objectives of preventing material degradation and improving bonding between the fibres and the thermoplastic material. Formulations with PP and 20% wt. discontinuous fibres showed an increment of up to 30% in tensile strength and 50% in tensile modulus when compared with virgin PP. Experiments using commercial flax rovings and slivers (continuous fibres) in conjunction with glass fibres (i.e. hybridizing of fibres) on an industrial large scale D-LFT line showed the viability of the processing technique for the manufacturing of hybrid reinforced thermoplastic composite parts.

INTRODUCTION

The use of renewable sources for the production of composite materials has been the subject of intensive industrial and academic research in the last two decades. However, the first attempts to accomplish this can be traced back to 1941 when Henry Ford championed the idea of developing a car with body panels made with a natural fibre reinforced plastic. Records indicate that the body panels were made with soybean fibre in a phenolic resin with formaldehyde used in the impregnation[1]. The main motivation back in 1941 was the shortage of steel, but Ford was also driven by the prospects of combining agriculture and the automotive industry and the potential of composite materials to be safer than steel in crash scenarios. Unfortunately by the end of World War II the idea of a plastic car had fallen through the cracks due to efforts being directed towards war recovery plans[2]. There were many initiatives in the 1960s involving the development of natural fibre reinforced thermosets which have been thoroughly documented[3]. In the 1970s attempts were made to produce cellulose-filled thermoplastics with a number of serious challenges. It became evident that cellulosic fibres from wood and other natural sources did not disperse easily throughout the molten thermoplastic materials causing the end products to show lack of uniformity and in general poor properties when compared with other fibre reinforced plastics. It wasn't until 1988 when Roger Wittenberg perfected his production process, calling his material, a mix of shredded recycled plastic bags and sawdust, Rivenite. Wittenberg believed his product would perform well as a substitute for fire logs. However, the material did not burn easily, so other uses for the material needed to be considered. The acquisition of Rivenite by Mobil Chemical Co. in 1992 and subsequent developments led to the creation of Trex, the largest manufacturer of wood thermoplastic composites (also known as WPC) for decking products in the world. After many years of research and development the WPC decking industry has come to maturity with a technology based, for the most part, on the use of wood flour, with characteristic particle lengths of around 400 microns or smaller and aspect ratios no greater than 10. This choice of particle size and shape, the lessons learned about keeping wood flour moisture levels low, the use of coupling agents (mostly maleic anhydride grafted polyolefins), the use of proper amounts of lubricants and the widespread use of various twin screw extruder technologies for the processing of WPC materials have consolidated the technology for the decking market. After 20 years of development the industry has learned how to overcome the drawbacks of using natural fibres as reinforcement materials and has created solutions to the problems related with fibre dispersion, moisture absorption, incompatibility of natural fibres and thermoplastics, natural decay and others.

Despite the progress in WPC technology, the automotive industry requirement is pushing for even higher technological improvements. Automotive applications demand tighter tolerances, higher impact and flexural strengths and better surface quality than decking applications. While HDPE, PP and PVC have been the thermoplastics most commonly used for natural fibre composites (or WPCs), automotive applications have higher requirements of strength and high temperature resistance. PP is widely used in the automotive industry as a fundamental component for TPO materials used for fascias and other non structural parts as well as for structural components when combined with fibreglass reinforcements. PA6 and other engineering resins are used for applications under the hood where temperature requirements are higher.

In this study co-financed by Transport Canada and the National Research Council of Canada, data is presented from moulded composite samples using PP as the thermoplastic matrix and flax fibres in both continuous and discontinuous formats. Flax fibres have been chosen for this study because of their good strength-to-weight ratio, their availability and relative low cost compared to other bast fibres. The study involves varying processing conditions as well as matrix formulations in an attempt to quantify polymer/fibre interactions. Heat stabilizers, antioxidants and coupling agents were used to prevent matrix and fibre degradation and to increase compatibility between phases upon processing. A reactive additive (CaO) was used to absorb residual water in cellulosic fibre in order to improve the mechanical properties of the composite parts. Injection and compression moulded samples were produced and characterized by means of mechanical (tensile, flexion and impact), thermal (crystallization and degradation) and microstructural (distribution and wet-out) tests to determine the optimal formulations and processing conditions.

EXPERIMENTAL

Discontinuous biofibre/PP composites

The first step of the experimental work involved the use of a discontinuous version of flax fibres in a PP matrix. The flax fibres were pelletized with the purpose of allowing a consistent way of feeding the material into the extruder and ensure proper volumetric dispersion. This was done without a significant loss in fibre length. However, the pelletizing process helped splitting the technical fibres into elementary fibres. Flax fibres in pelletized form are easy to manipulate, do not need special auxiliary equipment for feeding into the extrusion compounding process, and provide important predictive information for the potential behaviour of long continuous fibres. The objective was to assess the different formulations by characterizing the material for:

- Mechanical properties (tensile and impact)
- Thermal properties: Differential Scanning Calorimetry (DSC) and Thermo-gravimetric Analysis (TGA)
- Morphological properties (SEM)

Materials

The polymer used was Ineos PP H38G-02, an injection moulding grade homopolymer PP with a melt flow index of 38 g/10 min. Flax pellets were produced by cutting the fibres to an approximate length of 6.5 mm and then pelletized using the following procedure: the cut fibres were uniformly wetted using a 1:1 water/flax weight ratio after which they were pressed through a die plate using two rotating roll mills. Figure 1 shows the physical aspect of flax fibres in their original state (as received) and after the pelletizing step. There was no thermal degradation or significant fibre shortening observed after the pelletizing step. The temperature during the process was kept constant at around 70°C. Two treatments were used for the fibres: unwashed and washed in 80°C water. The washing of fibres with water was performed to evaluate the impact of clean flax (without impurities such as wax or hemicellulose) on the final mechanical properties of the composite material. Following the pelletizing step pellets were dried at 80°C for 24 hours to minimize the moisture content. Two types of coupling agents were used for this study: acrylic based grafted PP and maleic anhydride grafted PP. The specific materials used were: PB1001, (acrylic-based grafted PP), PB3150, PB 3200 and E43 (MA-based grafted PP). 98% purity calcium oxide (CaO) from Laboratoire MAT Inc. was also used as a reactive filler, which has the role of absorbing moisture present in the fibres, neutralize the acidity in fibre impurities and minimize oxidation and degradation of fibres with the ultimate purpose of improving the mechanical properties of the composite (Ton-That et al.[4]). The complete composite formulations using PP and discontinuous fibre are shown in Table 1.

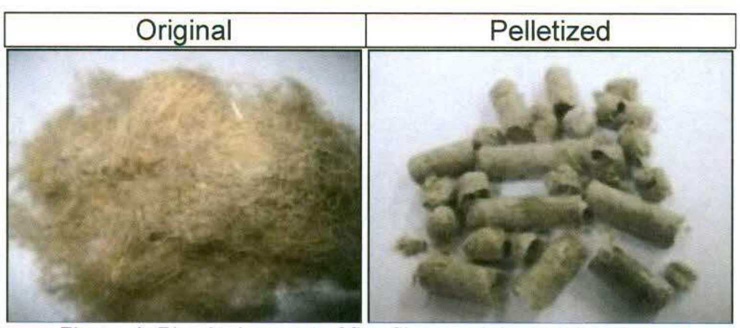


Figure 1: Physical aspect of flax fibres: original and pelletized

Extrusion setup for lab scale experiments

Mild extrusion conditions (low shear, low temperature) were used to ensure the lowest possible fibre and matrix damage in order to preserve the mechanical properties of the composites being produced. The screw configuration was specially conceived to achieve a good fibre wetting without compromising the fibre length and to avoid attrition* as much as possible. The extrusion temperature used was 180°C, which guarantees complete melting of the polymer matrix while preventing thermal degradation of the flax fibres. Compounding was carried out on a Buhler 20 mm clamshell co-rotating twin-screw extruder with an L/D ratio of 40. The flow rate was set at 2 kg/h. The screw configuration can be seen in Figure 2.

Table 1: Composite formulations using PP and discontinuous fibre

ID	PP	PB1001	PB3150	PB3200	E43	CaO (*)	Flax	Temp	Fibre treatment
		weight %						(°C)	
1	100.0							180	NA
2	90.0					10.0		180	NA
3	86.5	3.5				10.0		180	NA
4	86.5		3.5			10.0		180	NA
5	86.5			3.5		10.0		180	NA
6	86.5				3.5	10.0		180	NA
7	80.0						20.0	180	Unwashed
8	80.0						20.0	180	Washed
9	76.5	3.5					20.0	180	Washed
10	66.5	3.5				10.0	20.0	180	Washed
11	76.5		3.5				20.0	180	Washed
12	66.5		3.5			10.0	20.0	180	Washed
13	76.5			3.5			20.0	180	Washed
14	66.5			3.5		10.0	20.0	180	Washed
15	76.5				3.5		20.0	180	Washed
16	66.5				3.5	10.0	20.0	180	Washed

*These formulations are protected under US patent 7,041,716 B2 (2006) by T.Ton-That, F. Perrin-Sarrazin, J. Denault

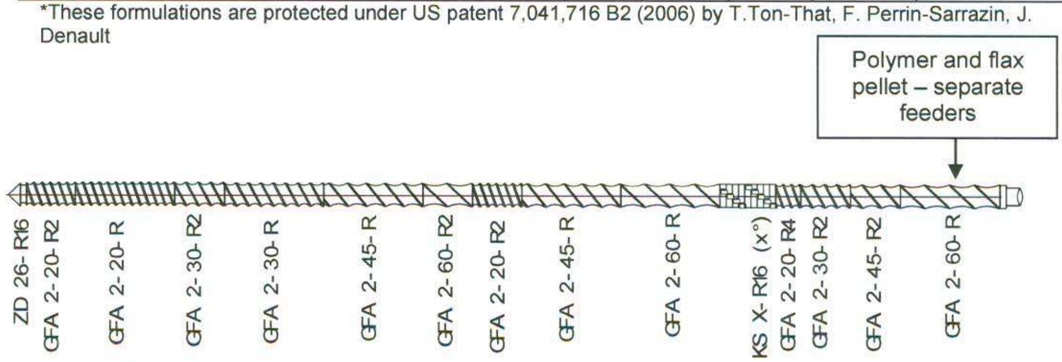


Figure 2: Screw configuration used for compounding on the 20 mm Buhler

* Reduction in size and integrity of the fibres caused by excessive shear.

The extruded composite pellets were dried at 80°C for 24 hours and injection-moulded on a BOY injection-moulding machine to simultaneously produce standard Type I dog-bone specimens (as specified on standard ASTM D638-91), and bars with a thickness of 3.1 mm for tensile and impact tests respectively (as specified on standard ASTM D259) .

Differential scanning calorimetry (DSC) was used to evaluate the thermal behaviour of the composites and the changes in the crystallization behaviour with the addition of 20%wt. flax, coupling agents and 10%wt. CaO. The analysis was applied using a Pyris 1 instrument calibrated with an indium standard. The samples were heated from 30 to 200°C at 20°C/min, kept at 200°C for 5 minutes to erase the thermal history, and cooled again down to 30°C at 20°C/min. The heat of fusion for PP (used for the calculation of Xc initial) is 207.1 J/g.

The thermal stability and decomposition temperature of the composites with selected formulations were carried out on a Mettler Toledo TGA/DTA 1 apparatus at a scan rate of 10°C/min from 30°C to 700°C under nitrogen.

Scanning electron microscopy (SEM) was carried out on polished composite surfaces coated with a gold/palladium alloy prior to the observation. JEOL JSM-6100 SEM at a voltage of 10 kV was used to analyze the dispersion of triticale fibres into the matrix using polished surfaces and the interface between flax and PP matrix using the fractured specimens that resulted from mechanical testing.

Continuous biofibre composites

Long fibre reinforcements produce a significant improvement in the strength of thermoplastic based composite parts (Bartus et al.)[5]. The main limitation for the production of long biofibre composites is the ability to feed them into the compounding system. Short fibres of about 3 mm or less can be pre-compounded and pelletized using batch or continuous systems which can be later feed into the hopper of a twin screw extruder. The fibres can also be pelletized using the method described earlier on this paper, but this is a labour intensive process with applicability at a laboratory scale. The goal is to devise a Direct Long Fibre Thermoplastic (D-LFT) process using continuous flax rovings. The flax rovings are characterized by their tex number, defined as the number of grams of material per kilometre of roving, and the tpi, defined as the number of twists per inch. The tex number is crucial in the determination of the percentages of fibre added to the composite material whereas the tpi is a parameter that affects the pulling strength, generally increasing as the number increases, but also affects the ability to de-bundle and disperse the fibres into the polymer melt. The selection of the right combination of tex and tpi numbers for the flax rovings has a significant impact on the effectiveness of the manufacturing technique. In order to attain the typical properties required for automotive components, natural fibres are used in conjunction with glass fibres. This has been referred to as hybridizing of natural and glass fibres by Vaidya[6].

Experiments were conducted at the Magna-NRC Composites Centre of Excellence (Concord, Ontario) using a PP/flax/fibreglass roving material system. The goals of the evaluation were to:

- Evaluate the processability of flax in a continuous form by means of assessing :
    - Feeding mechanisms
    - Distribution of fibres in the polymer matrix
    - Length of fibres (average size and size distribution) and their correlation with the final mechanical properties of the moulded part
    - Thermal degradation of flax fibres
- Evaluate the practical aspects of feeding and processing flax rovings in an industrial scale compounder. The main aspects are:
    - Roving spools arrangement
    - Initial feeding of the rovings into the twin screw section

- Verification of the accuracy in the estimation of flax weight percentage by post mortem analysis

Materials

The calculation of weight percentage poses a challenge for a quick estimation of the proportions of the materials being used since the equipment program performs an internal calculation of screw speed and pellet dosage based on three input parameters: weight percentage of fibre, fibre tex and total extruder throughput. Since two different fibres are being introduced simultaneously, the program cannot do the calculation automatically. To go around this limitation the feeding of 850 tex flax fibres was done in groups of three rovings (for a total tex of 2550) which approximately matches the fibreglass tex of 2400. This is a practical way to implement the weight percentages of both glass and flax fibres being fed into the extruder. The layout was three spools of flax surrounding every spool of glass. The procedure was done by tying three rovings of flax to one roving of glass being fed into the extruder. The glass roving pulled the flax rovings into the machine and once the flax rovings entered the screw section the glass roving was cut completing the transition.

The experiment started with twelve glass rovings being fed for 40%wt. glass content and then reduced to 30% wt. glass-10% wt. flax by replacing three rovings of glass with nine rovings of flax, then 20% wt. glass-20%wt. flax and so on until the percentages were 0% wt. glass and 40%wt. flax. Table 2 provides the materials and equipments used for processing and moulding the plaques. Table 3 shows the typical extruder temperature profile and Table 4 the experimental matrix for composites made with flax and glass rovings. Table 5 contains the experimental matrix for the flax sliver/glass roving trials. The cutting pattern for samples to be extracted from the plaque mould is provided in Figure 3. The plaques were labelled sequentially with the letter "T" followed by a number denoting the trial order. Ten plaques of each trial were tested for mechanical properties.

Table 2: Materials and equipment for experimental work

Extruder:	Coperion ZSK-70 (70 mm 35 L/D Co-rotatingTSE)
Compression press:	2500 Ton Dieffenbacher press
Compression mould	Load floor tool
PP resin	PP Ineos H35Z-00 MFI 35
Additive 1*:	ADD-VANCE 130
Additive 2*:	Priex 20097 (Maleic anhydride grafted PP)
Reinforcing Material:	Flax rovings, 850 tex/glass rovings (2,400 tex)
	Flax sliver, 10,000 tex, no twist/glass rovings (4,400 tex)

*Additives 1 and 2 are premixed and added at a 3% concentration

Table 3: Temperature zones ZSK-70 Coperion extruder

Temperature profile										Die	
Hopper	TC2	TC3	TC4	TC5	TC6(r)	TC7	TC8	TC9	TC10	TC11	TC12
18	220	230	240	240	240	225	225	230	230	235	235

Table 4: Experimental matrix for flax rovings (850 tex)

Trial	PP	MB-add	Flax	GF
T1	68.0%	3.0%	0%	30.0%
T2	68.0%	3.0%	7.5%	22.5%
T3	68.0%	3.0%	15.0%	15.0%
T4	68.0%	3.0%	22.5%	7.5%
T5	68.0%	3.0%	30.0%	0%

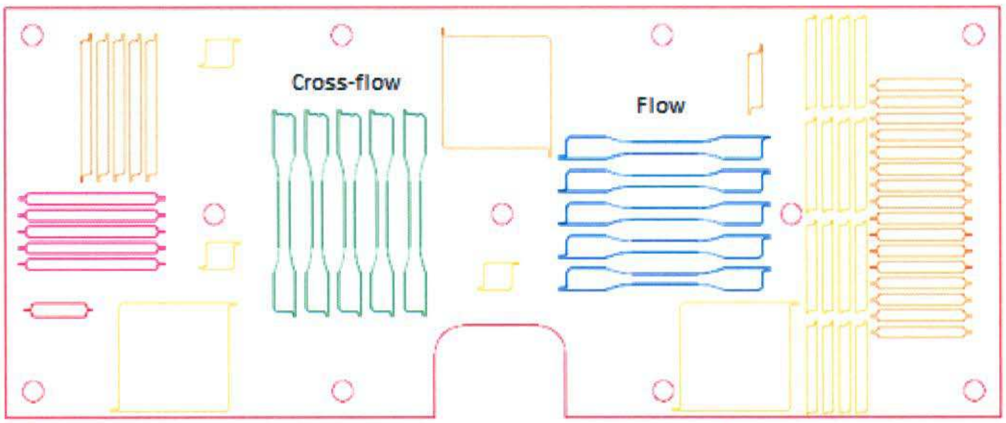


Figure 3: Compression moulding plaque and specimen cutting configuration. Green samples in the cross-flow direction; blue samples in the flow direction.

Table 5: Experimental matrix for flax slivers (10,000 tex)

Trial ID	PP	MB-add	Flax	GF
1	57.0%	3.0%	0%	40.0%
2	57.0%	3.0%	10.0%	30.0%
3	57.0%	3.0%	20.0%	20.0%
4	57.0%	3.0%	30.0%	10.0%
5	57.0%	3.0%	40.0%	0%

Extrusion setup for industrial scale experiments

For the industrial scale trials mild extrusion conditions (low shear, low temperature) were used to ensure the lowest possible fibre and matrix damage in order to preserve the integrity of the fibres. The screw configuration (Figure 4) was designed to achieve a good fibre wetting without compromising the fibre length and to avoid attrition as much as possible. The extrusion temperatures (Table 3) was set to higher levels than the pilot scale experiments as the fibre wet out is strongly affected by this and the compression moulding process requires the material log to be at a temperature that ensures proper flow and complete filling of the cavity. Industrial compounding experiments were carried out on a Coperion ZSK-70 co-rotating twin-screw extruder.

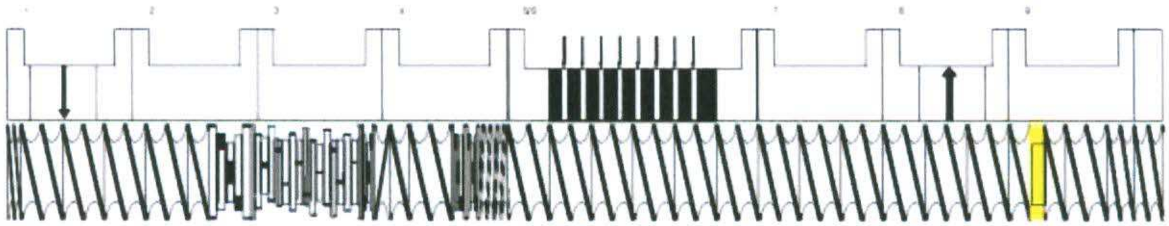


Figure 4: Screw configuration used for compounding on the 70 mm Coperion TSE.

Two forms of flax were used for the experiments according to Table 2: 850 tex rovings and 10,000 slivers, as shown in Figure 5.

Figure 5: On the left two bundles of 10,000 flax sliver; on the right 850 tex flax roving spools.

RESULTS AND DISCUSSION

Discontinuous biofibre/PP composites

Differential scanning calorimetry results are shown in Table 6. These results indicate that during the first heat cycle, the melting temperature of PP formulations slightly decreased from 169.1°C to 167.4°C. The crystallinity of matrices varied from 38% to 44% with the addition of CaO, most likely due to its nucleating effect[†]. For the same reason, the crystallization temperature increased from 117°C up to around 125°C during the cooling cycle. A comparison of DSC results between pure PP and PP/20% unwashed flax and PP/20% washed flax without any other additives was performed. Results are presented on Table 7. On Tables 6 and 7, Tm refers to Melting temperature, ΔHm to Enthalpy of transition (melting), Xc to Degree of crystallinity after heat cycle, Tcc to Crystallization temperature (cooling), ΔHcc to Enthalpy of transition (crystallization during cooling), and Xcc to Degree of crystallinity (crystallization during cooling).
The composites processed using 20%wt. flax did not show a significant change in melting temperature, although there was a change of 2.4°C in the crystallization temperature for the washed flax sample. Crystallinity increased from 39.8% up to 46.1%. The increase in the degree

† Nucleating or nucleation is the process by which crystals are formed. Crystals form initially on minute traces of foreign substances that act as the nucleus.

of crystallinity and crystallization temperature during cooling are attributed to the nucleating effect of the flax fibres present in the compound. Almost no difference can be observed between the composites obtained with unwashed and washed flax. Even if the washing eliminates impurities from the fibre surface (as wax for example), this effect did not influence the composites thermal behaviour. The reason is that the hydrophilic cellulosic fibres are not compatible with the hydrophobic PP

Table 6: DSC results obtained for neat PP and PP formulations without flax fibres

	1st Heat			Cooling			2nd Heat		
	Tm	ΔHm	Xc	Tcc	ΔHcc	Xcc	Tm	ΔHm	Xc
	°C	J/g	%	°C	J/g	%	°C	J/g	%
1	169.1	82.4	39.8	116.7	96.6	46.7	164.7	90.5	43.7
2	168.1	79.7	42.8	119.8	90.9	48.8	164.4	89.7	48.1
3	168.7	82.5	44.3	122.1	90.9	48.8	164.4	89.8	48.2
4	167.4	75.8	40.7	124.4	91.1	48.9	164.4	90.2	48.4
5	168.1	80.0	42.9	125.1	90.0	48.3	164.8	93.1	50.0
6	167.4	76.9	41.3	121.4	89.0	47.8	163.4	89.6	48.1

Table 7: DSC results obtained for PP and PP formulations with flax fibres

	1st Heat			Cooling			2nd Heat		
	Tm	ΔHm	Xc	Tcc	ΔHcc	Xcc	Tm	ΔHm	Xc
	°C	J/g	%	°C	J/g	%	°C	J/g	%
1	169.1	82.4	39.8	116.7	96.6	46.7	164.7	90.5	43.7
7	168.1	72.1	43.5	117.1	74.6	45.0	168.8	71.2	43.0
8	166.1	76.3	46.1	119.1	77.6	46.9	164.1	75.9	45.8

When coupling agents were used to increase this adhesion between the flax fibres and polypropylene, (results not shown here), the obtained DSC results were very similar as those in Table 7 for the formulations 7 and 8. PP composites formulations proved that the crystallization behaviour is affected by the presence of fibres and of CaO. No important differences were observed between composites formulations processed with different coupling agents.

The thermal stability and decomposition temperature of the composites were analysed. All the composite formulations that did not include flax fibres showed similar performance with the onset of thermal degradation around 375°C (Figure 6). Regardless of the matrix formulation, the addition of 20%wt of flax resulted in a two step weight loss (Figures 7 and 8). The first step, which is very likely related to the degradation of the flax, started at around 250°C. It can be observed in Figure 7 that the degradation of composites obtained with unwashed flax took place more rapidly than that for the composites obtained with washed flax.

This first step in weight loss is attributed to the presence of impurities, waxes and small particles that degraded at a lower temperature than the rest of the materials in the composite. However, the start point of the degradation is about 70°C higher than the extrusion temperatures used for the experiments, so the presence of impurities was not expected to have a significant impact in the loss of properties due to degradation. For the compositions containing CaO and coupling agents, the rate of total weight loss was reduced. The presence of CaO causes a shift in the curve to the right as this inorganic compound does not degrade at the range of temperatures used for the TGA.

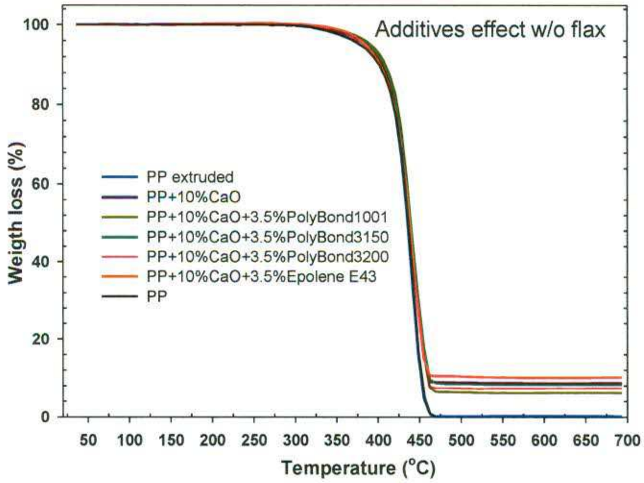


Figure 6: TGA Curves for neat PP and additives

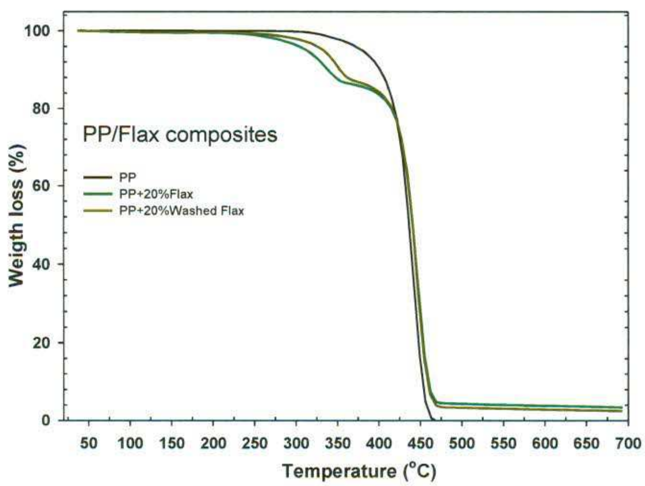


Figure 7: TGA curves for neat PP and discontinuous flax (unwashed and washed)

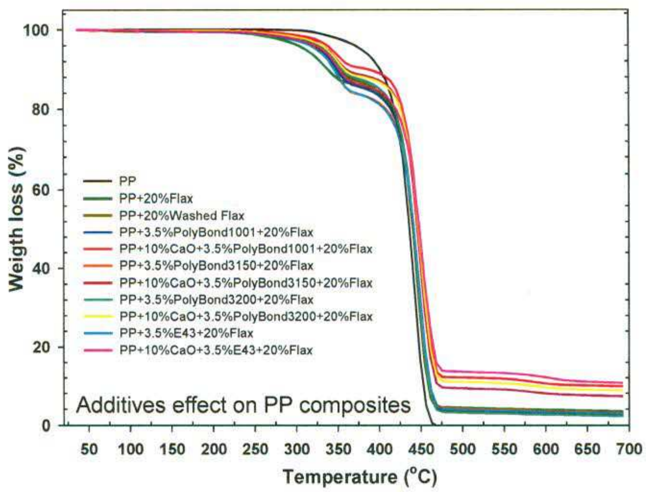


Figure 8: TGA curves of PP composites for different formulations

Figures 9 and 10 present the tensile strength and tensile modulus respectively of PP references (as received and extruded) and PP formulation (blue bars) in comparison with PP/flax composites (green bars). PP, extruded PP and PP formulations (coupling agents and CaO) all show a similar tensile strength of around 33 MPa. The addition of 20% washed (WF) and unwashed flax (UF) without the presence of coupling agents did not affect the value of the tensile strength. The lack of adhesion between the hydrophobic PP and hydrophilic flax fibres causes the composite material to fail at the interface between the two materials.

Coupling agents were added to enhance the adhesion between the natural fibres and PP and reduce the potential failure at the weak areas on the interfaces. The coupling agents that provided the highest increment in tensile strength were the maleic anhydride grafted PP grades (PB3150, PB3200 and E43). An increment of about 30% in comparison with the tensile strength of non-compatibilized composites was observed with these coupling agents. The incorporation of 10% of CaO caused an additional increase in tensile strength with PB3150 and E43. The increase is attributed to the reactive nature of CaO which could potentially improve the surface adhesion between the fibres and the PP. The tensile modulus almost doubled with the addition of 20%wt of flax fibres. However, it is interesting to note that the addition of different coupling agents had no apparent effect on the tensile modulus. As the tensile modulus is measured in the elastic region of the stress-strain curve, the increased rigidity imparted by the flax fibres is independent of the presence of coupling agents and the enhanced fibre-matrix adhesion. All the composites containing CaO showed higher modulus than their counterparts. This is attributed to the significantly higher stiffness of the CaO, compared with PP, which acts as a filler in the composite material.

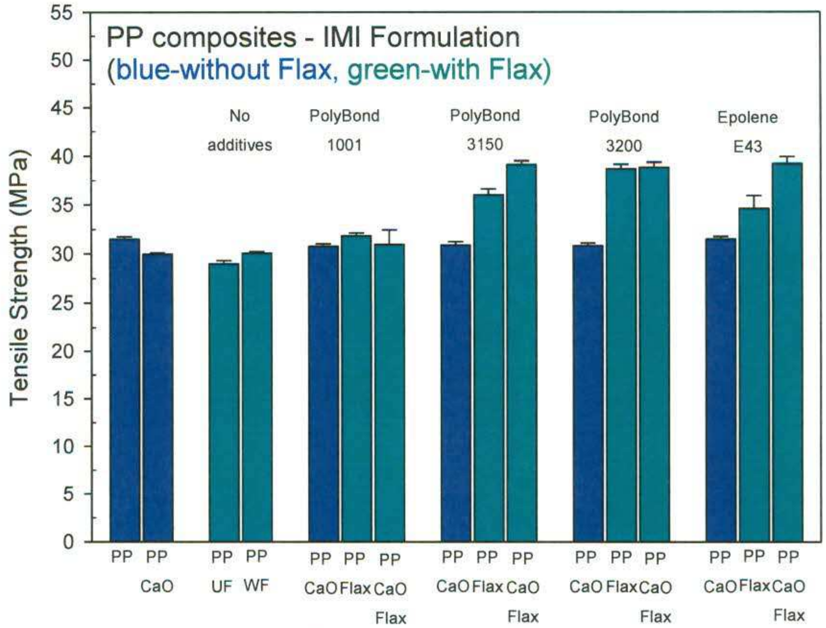


Figure 9: Tensile Strength of PP composites

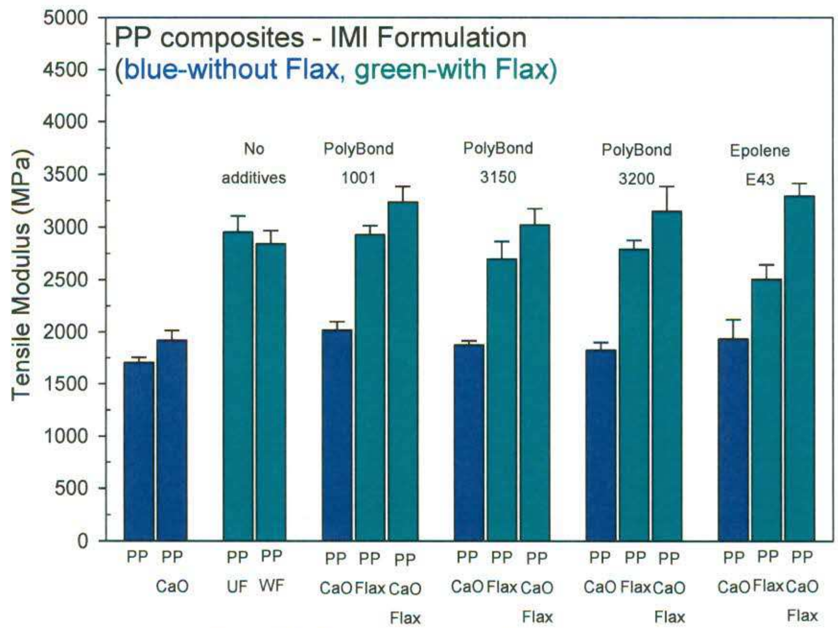


Figure 10: Tensile modulus of PP composites.

Figure 11 presents the impact strength of PP and PP formulations without fibres (in blue) and PP/flax fibre composites formulations (in green). It can be observed that the impact strength of flax fibre reinforced PP is about 75% higher than the neat PP samples, with slight variations depending on the coupling agents used.

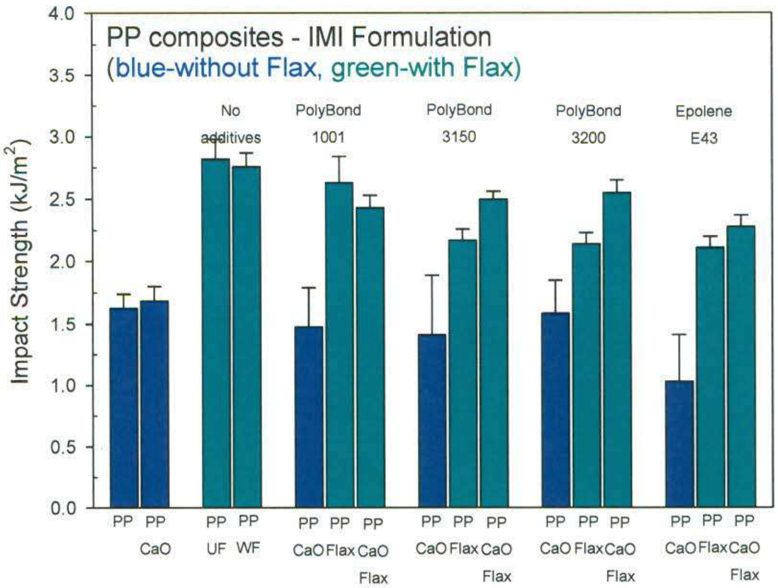


Figure 11: Impact strength of PP composites.

Table 8 presents Scanning Electron Microscopy (SEM) images of the fracture surface morphologies for PP/flax short fibres composites. The fracture surfaces of composites with unwashed and washed flax fibres without coupling agent show a significant number of fibres pulling out of the polymer matrix, which corroborates the inherent lack of adhesion between hydrophilic flax fibres and hydrophobic PP. The composites containing PB1001 (acrylic based coupling agent) also showed this lack of adhesion at the fibre interface. In the case of PB3150, 3200 and Epolene E43 composites, the fibre pull-out was noticeably reduced. These images show a qualitative explanation of the mechanism behind the gain in tensile strength observed for the samples with maleic anhydride grafted PP coupling agents. The two SEM in Table 9 are higher magnification micrographs of PP/flax fibres with and without coupling agents and CaO confirming the improvement on fibre-matrix adhesion in the presence of coupling agents and CaO. The enhanced compatibility reflects on a better bonding and ultimately on unequivocal improvement in tensile strength.

The best formulations for PP/discontinuous fibre composites in terms of mechanical properties were those containing 20%wt. flax, coupling agents (PB 3150, 3200 and E43) and CaO as reactive additive, with up to 30% increment in tensile strength and 50% in tensile modulus when compared with virgin PP.

Table 8: SEM of fracture surfaces of PP composites

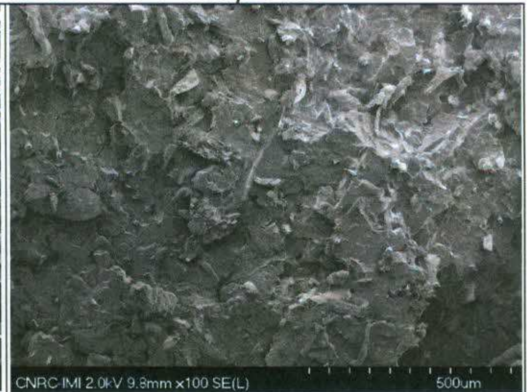
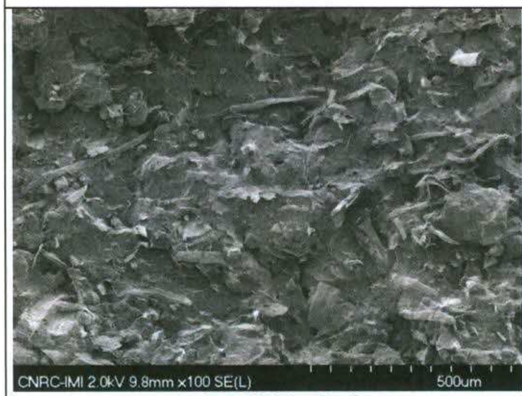
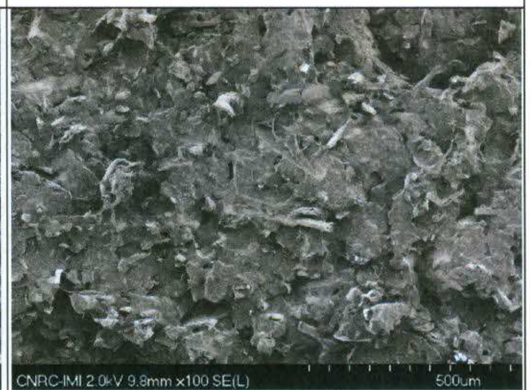
 7-unwashed flax	 13-PB3200
 14-PB3200+CaO	 16-E43+CaO

Table 9: Magnification of PP/flax composites with and without coupling agents and CaO

 PP/flax fibres without coupling agents or CaO. Note the gap between the fibre and the polymer matrix.	 PP/flax fibres with coupling agents and CaO. Fibres are embedded and tightly bonded to the polymer matrix

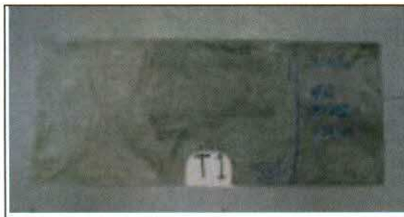
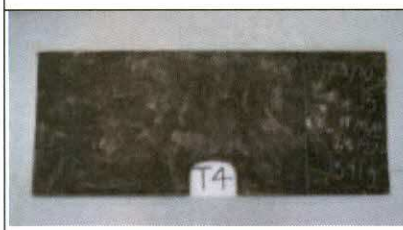
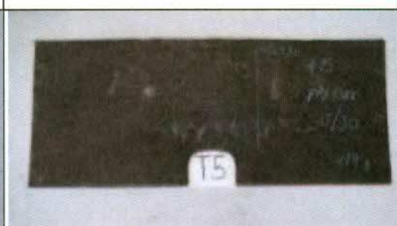
Continuous biofibre composites

Composites with 850 tex flax rovings

Table 10 presents the images of moulded parts using flax rovings. For all formulations, natural PP was used to allow for a visual examination of the fibres present in the moulded parts. Natural

PP, although not completely transparent, is inherently translucent. The experiments showed that 850 tex flax rovings with low twist can be successfully fed without problems into the fibre feeding section of an intermeshing co-rotating twin screw compounder in similar fashion as for the feeding of fibreglass rovings. Visual examination allows the qualitative evaluation of fibre distribution, which is an indication of the compounding equipment's ability to separate the fibre rovings and distribute the fibres homogeneously throughout the entire moulded part. The colour of the samples got darker as the amount of flax fibres increased. The fibre rovings distributed homogeneously throughout the entire part. Within each plaque, fibre-rich and fibre-poor regions could be observed. In all cases, individual rovings could be identified, indicating that fibre bundles were not opened as a result of the roving twist. The visual examination also allowed identifying dry rovings reaching the surface of the plaques, indicating the lack of fibre wet-out within unopened fibre bundles.

Table 10: Images of moulded parts using flax rovings.

		
T1: 68% PP, 30% fibreglass, 2% additive masterbatch	*T2: 68% PP, 22.5% fibreglass, 7.5% flax, 2% additive masterbatch*	*T3: 68% PP, 15% fibreglass, 15% flax, 2% additive masterbatch*
		
T4: 68% PP, 7.5% fibreglass, 22.5% flax, 2% additive masterbatch	*T5: 68% PP, 30% flax, 2% additive masterbatch*	

Figures 12 to 17 show the results for the evaluation of tensile and flexural and impact properties of samples extracted from the plaque as illustrated on Figure 3 and produced according to Table 4. Simultaneous feeding of flax and fibreglass rovings (hybridizing) was achieved using standard equipment for fibreglass rovings. Parts moulded with flax and glass fibres showed a homogenous distribution of the flax fibre rovings throughout the parts. This demonstrates the ability of the twin screw compounder to distribute the fibre bundles homogenously. However, visual examination of the samples and the implementation of a dissolution technique (i.e. PP dissolution in xylene at 160°C) showed that the individual fibres in the rovings are not being separated and distributed within the polymer melt by the twin screw elements of the compounder. The result is a lack of proper distribution of the individual technical fibres in the final part. However, roving de-bundling can be improved by optimizing the screw design in the twin screw compounding system. Optimization of screw configuration will be the subject of future research.

Figure 12 and 13 present tensile strength (TS) and modulus (TM) of plaques obtained using flax rovings and the samples are in the same order as in the Table 4. The blue bars correspond at the samples cut in the flow direction and the green bars in the cross-flow direction. The tensile properties of the reference virgin PP are TS = 30 MPa and TM = 1450 MPa as it can be seen in the Figure 9 and 10. Results for tensile strength indicate that the introduction of flax fibres as a substitute for glass fibres for composite materials produced a significant reduction in strength and a moderate reduction in modulus. The reduction in strength was expected since flax fibres have a lower specific strength than glass fibres. However, the moderate reduction in modulus might be associated to the poor dispersion of the flax fibres rather than the intrinsic modulus of elasticity of the flax. The high standard deviation observed is attributed to the inadequate distribution of the technical fibres throughout the moulded parts.

The tensile strength in the flow direction is higher for the higher concentrations of glass whereas the trend is reversed as the flax content increases up the maximum value on trial 5 for which the formulation has no glass content. The higher values of tensile strength in the flow direction are associated to the preferred orientation of the glass fibres in the predominant flow direction. For the compression moulding technique, the predominant direction of flow is determined by the size, shape and initial orientation of the material log and by the mould geometry. However, this orientation is not as strong as for the injection moulding process where the material is forced at very high pressure through narrow paths and the orientation is much more pronounced. Visual observation of the flax fibres indicate that they are uniformly distributed throughout the entire volume, even though the fibre rovings were not de-bundled to separate the technical fibres. In general, the substitution of fibreglass by an equivalent proportion of flax fibres produced a reduction in tensile strength. The complete substitution of fibreglass by flax produced a reduction of 52% in the average tensile strength for the direction of flow and a reduction of 29% for the cross flow direction. The standard deviation did not show a significant differences between samples with fibreglass, hybrid and flax reinforced samples. This indicates that the magnitude of the variations is associated with the process rather than the reinforcing material.
The tensile modulus did not show a decrease for the cross-flow samples (Figure 13, green bars). It has been reported in literature[7] that flax fibres have comparable specific stiffness to that of glass fibres. As the glass fibres were substituted for an equivalent amount in weight of flax fibres for the experiments run during this investigation, this result validates the assertion of specific stiffness equivalence for the two fibres. However, the flow direction results did show a decrease in the value of modulus from 4.6 to 2.9 GPa. This difference is attributable to differences in fibre orientation as the amount of glass was substituted by flax fibres. The standard deviation observed for the tensile modulus remained consistent for all the conditions.

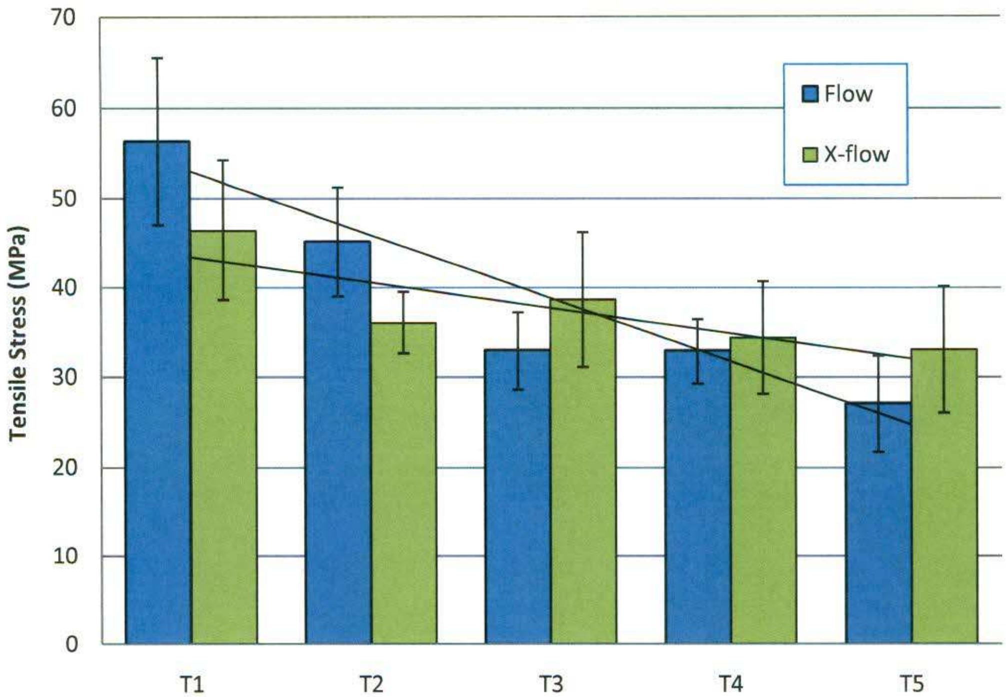


Figure 12: Comparison tensile strength for compression moulded flax roving composite specimens in the flow and cross-flow directions

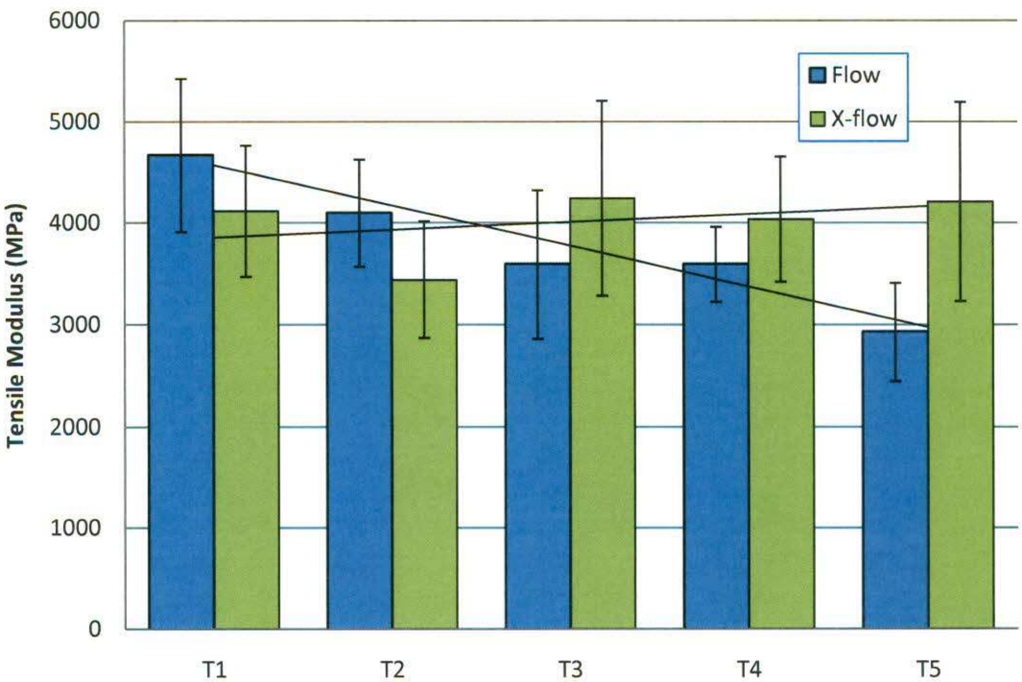


Figure 13: Comparison tensile modulus for compression moulded flax roving composite specimens in the flow and cross-flow directions

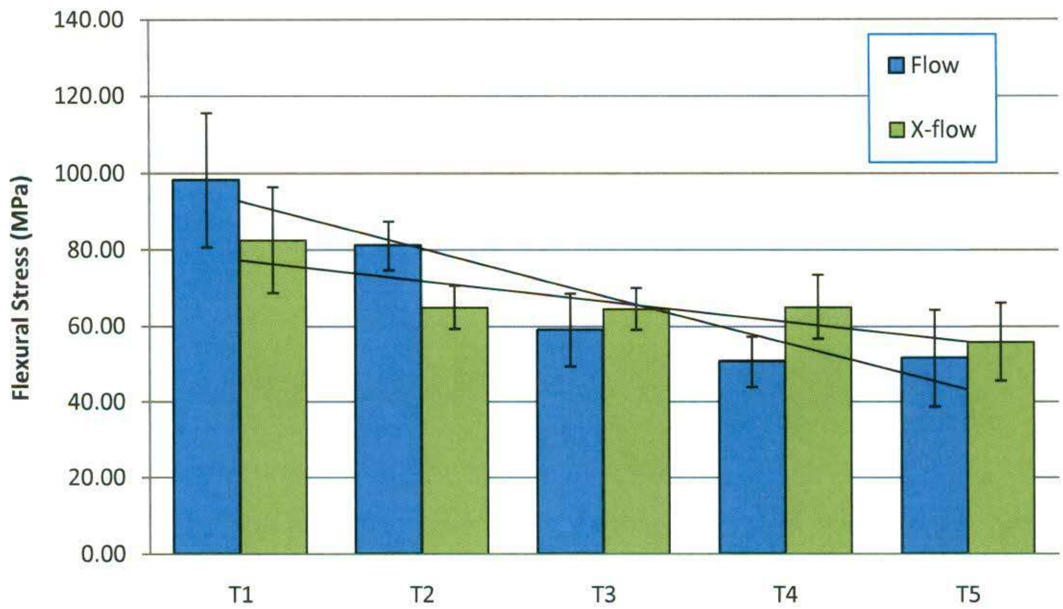


Figure 14: Comparison flexural strength for compression moulded flax roving composite specimens in the flow and cross-flow directions

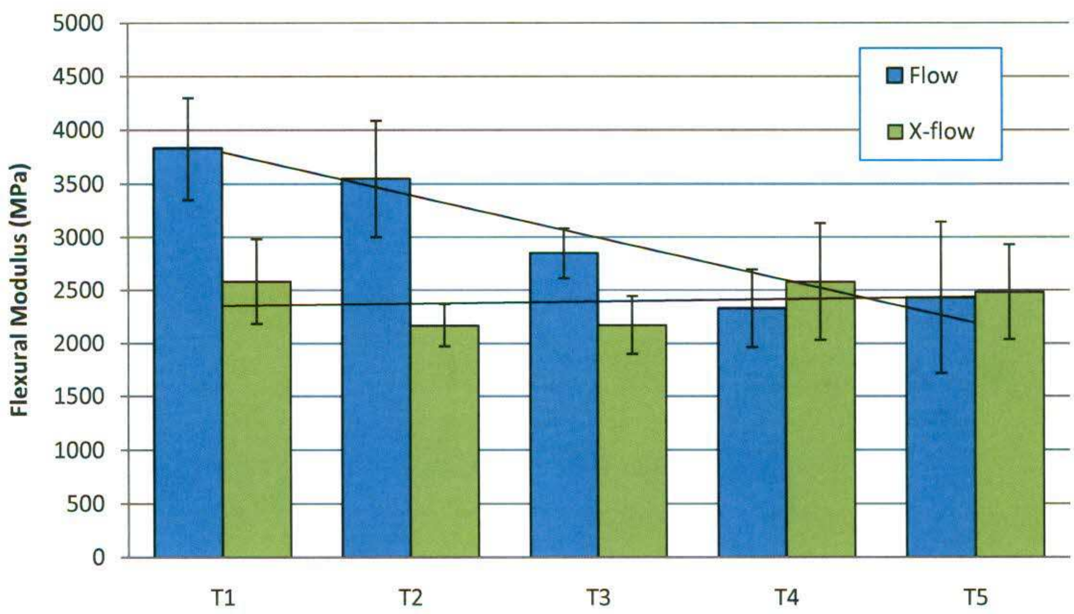


Figure 15: Comparison flexural modulus for compression moulded flax roving composite specimens in the flow and cross-flow directions

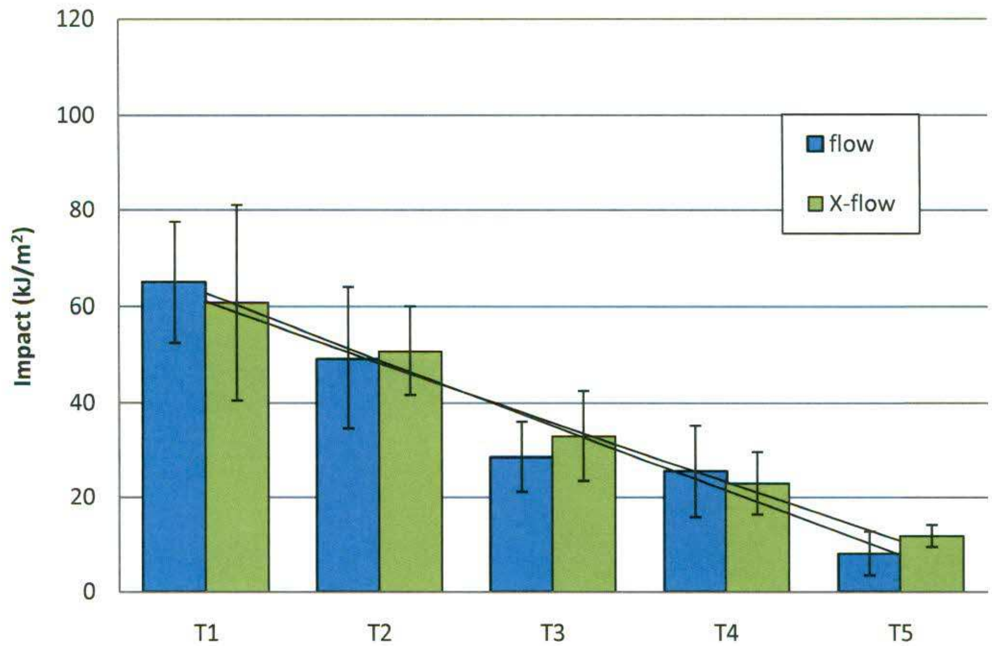


Figure 16: Comparison of notched impact strength for compression moulded flax roving composite specimens in the flow and cross-flow directions

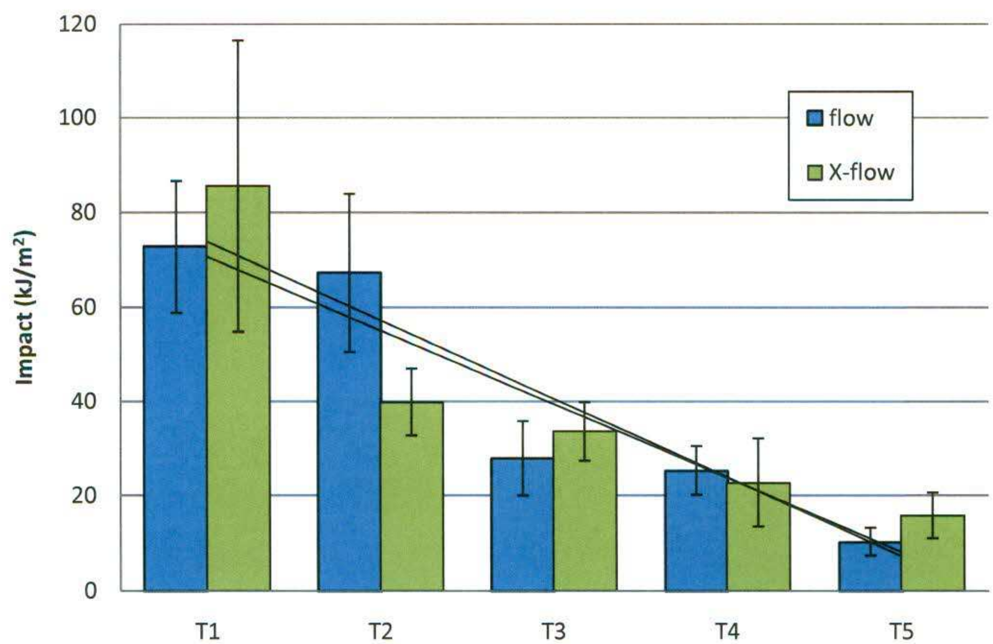


Figure 17: Comparison of un-notched impact strength for compression moulded flax roving composite specimens in the flow and cross-flow directions

Figures 14 and 15 present flexural strength (FS) and modulus (FM) of plaques obtained using flax rovings and the samples are in the same order as in the Table 4. The blue bars correspond at the samples cut in the flow direction and the green bars in the cross direction. The flexural properties of the reference virgin PP are FS = 45 MPa and FM = 1150 MPa. The results for flexural strength show a similar pattern as those for tensile strength with a linear reduction as the glass content is replaced by flax. The complete substitution of fibreglass by flax produced a reduction of 47% in the average flexural strength for the direction of flow and a reduction of 35% for the cross flow direction. It is interesting that the maximum stress for trials 1 and 2 is higher for the flow direction than for the cross direction specimens, but for trials 3, 4 and 5 the results are reversed and the cross flow values are higher. In general the cross flow flexural stress values remained almost constant for all trials except for trial 1 where the stress value was 81 MPa, the highest of all the trials.

The cross flow flexural modulus values show once again a tendency to remain constant at different combination of glass and flax reinforcements, as expected for materials with equivalent specific stiffness. The values of flow direction flexural modules show a noticeable decrease as the amount of flax is increased and replacing glass. This is consistent with the results obtained for tensile strength. It appears that at high fibreglass concentrations the orientation of the fibres plays an important role in the strength of the parts, whereas as the flax content is increased the orientation is not as critical. The following explanation is suggested: as the individual glass fibres are better dispersed than the flax fibres, these former tend to follow the flow of the viscous molten polymer more closely making them to align with the flow streamlines. This would produce a more noticeable dependency of the mechanical properties on fibre orientation and hence of the orientation of the specimens extracted from the moulded parts.

Results for notched and un-notched izod impact tests are presented in Figures 16 and 17. The notched impact values showed to be independent from the orientation of the specimens extracted from the moulded plaques. The un-notched impact values, as opposed to the notched sample results, appear to be affected by the orientation of the specimens. The impact values decreased as the percentage of flax increased in a linear fashion. The un-notched izod values for T1 (40% weight fibreglass) are very high (more than six times the values for neat PP) which initially suggests the presence of long fibres. A post-mortem evaluation of fibre lengths confirmed the presence of numerous fibres around 50 mm in length. The notched impact strength for T2 (22.5% fibreglass, 7.5% flax) was 49 kJ/m^2. This performance confirms the viability of using hybrid flax/glass reinforcements for composite parts. The impact strength for all hybrid reinforced samples (T2, T3 and T4) showed all values over 20kJ/m^2. The largest standard deviation occurred for the plaques containing only glass as reinforcement.

Composites with flax sliver

Figures 18 to 21 correspond to the experimental results for tensile and flexural properties from samples produced with flax slivers (see left side picture on Figure 5) according to Table 5. The values of tensile strength shows a clear decreasing trend in both the flow and cross flow directions as the percentage of glass decreased and the amount of flax increased. The main difference between the results of experiments using flax rovings and slivers is the clear difference in the flow and cross flow values. The cross flow values are consistently lower than the flow direction values. A visual examination of the plaques shows a better distribution of flax fibres as the slivers have no twist and the fibres are more readily separated and dispersed by the screw elements within the twin screw extruder. The linear trend for both flow and cross flow samples show parallel lines suggesting that glass and flax fibres follow similar flow patterns, as opposed to the flax rovings that appear to have a different pattern than the glass fibres due to the inability of the twin screw to de-bundle the twisted fibres. The flow direction values of maximum tensile stress varied from 77 MPa for the 40%wt. fibreglass sample to 33 MPa for the 40%wt. flax sample for a reduction of 57%. This reduction is comparable with the 52% measured for the flax roving plaques. The tensile modulus, on the other hand, showed as expected a much smaller percentage reduction of 29%. Both results in the flow and cross-flow directions for the tensile modulus show a rather irregular behaviour, even though the overall trend points to a decrease in value as the glass is replaced by flax. However, these results show once again that the equivalent specific stiffness reflects on relatively small changes in the tensile modulus as the glass is replaced by flax fibres.

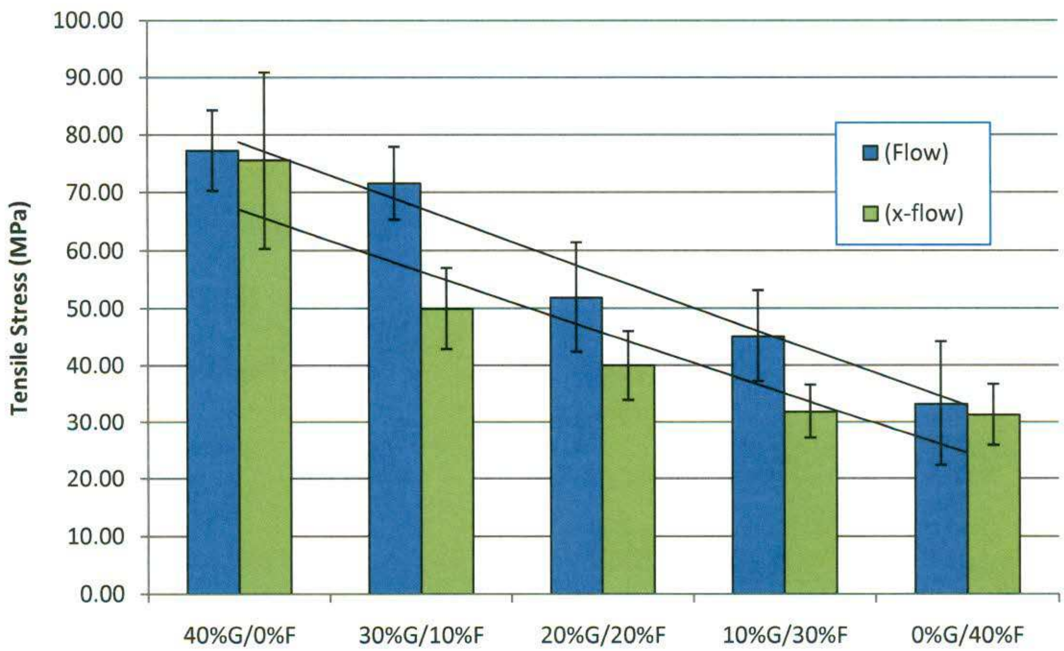


Figure 18: Tensile strength for compression moulded flax sliver composite specimens in the flow and cross-flow directions

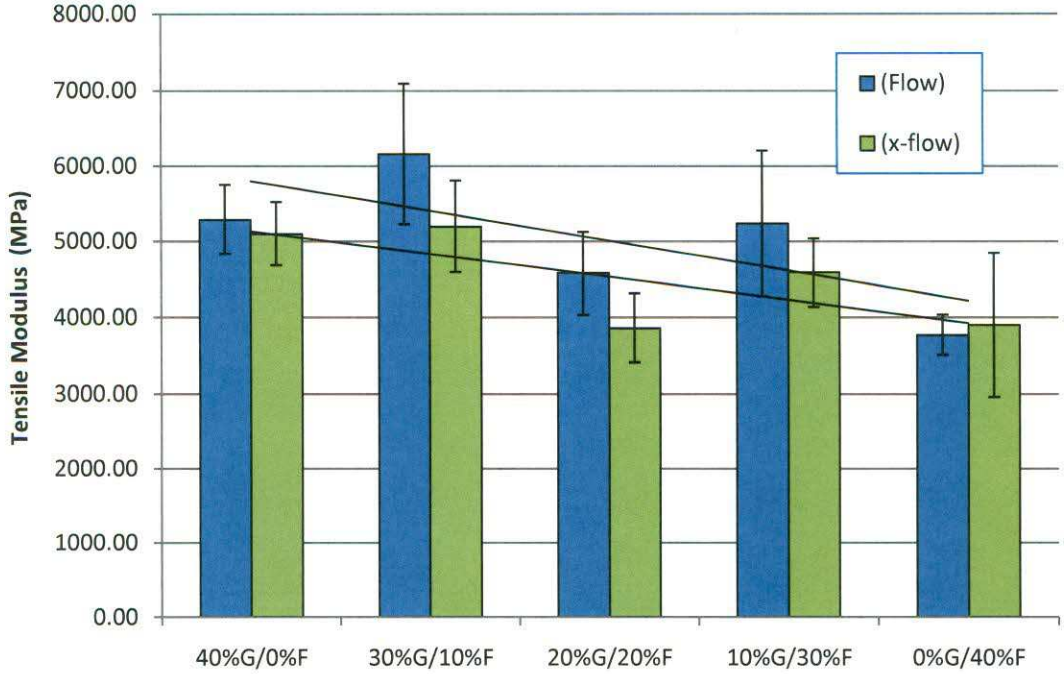


Figure 19: Tensile modulus for flax sliver compression moulded composite specimens in the flow and cross-flow directions

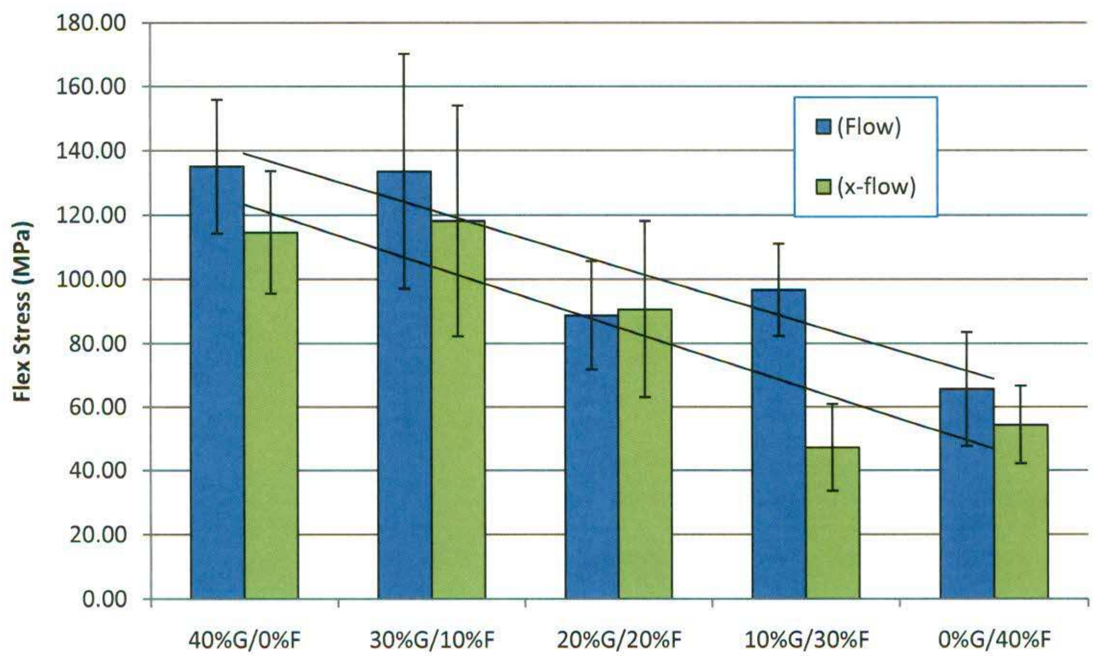


Figure 20: Flexural strength for using flax sliver compression moulded composite specimens in the flow and cross-flow directions

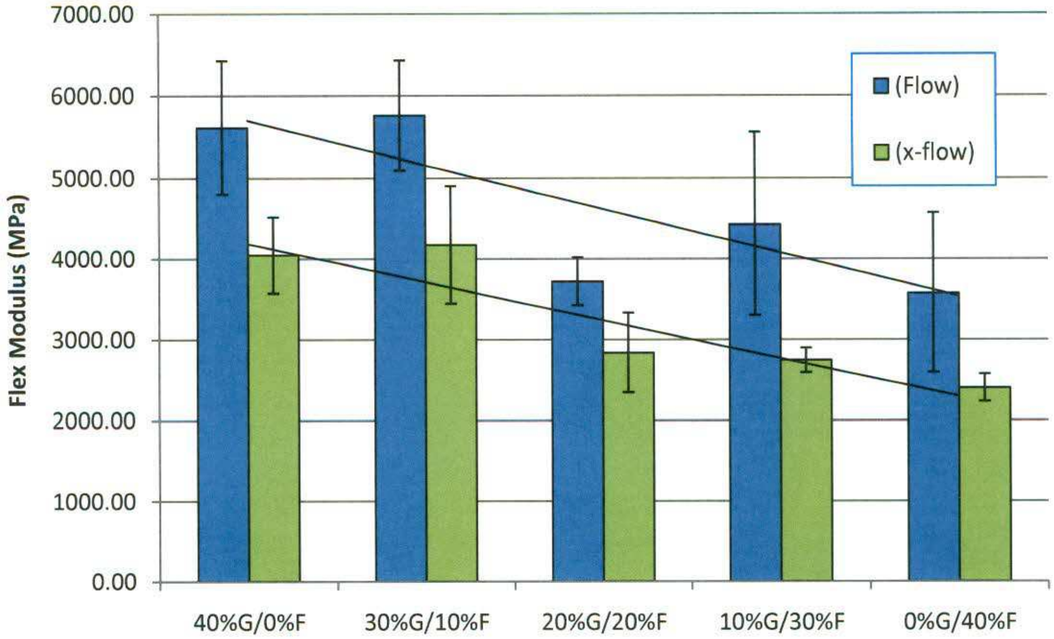


Figure 21: Flexural modulus for using flax sliver compression moulded composite specimens in the flow and cross-flow directions

The flexural strength results show a similar trend than the tensile strength result, with regression lines showing a clearly linear behaviour and a reduction of both stress and modulus of elasticity values as the glass fibres are replaced by flax fibres. The parallel trend lines suggest that the orientation plays an important role in the mechanical properties. The flow direction values of maximum stress and modulus of elasticity are consistently higher than the cross-flow values. The good dispersion of flax fibres with the use of flax slivers produced a much more consistent relation between the flow direction and the cross flow samples. While the flexural strength in the flow direction dropped 52% from the 40% glass content plaques to the 40% flax plaques, the flexural modulus decreased by 37%.

Figures 22 and 23 show the results for notched and un-notched Izod impact tests respectively. The impact strength improves considerably from the values for neat PP with the addition of long glass fibres and the levels are sustained for the 30% glass/10% flax formulation. Impact strength starts to reduce for the 20% glass/20% flax formulation, but it stays at around 20 kJ/m^{2,} which is very good for automotive applications. However, there is a very large variability observed for this test results. The variability is associated to the dispersion of fibres in the moulded part. Figure 24 depicts a typical situation for specimens extracted from different areas of the compression moulded part. The specimen on the top had very low flax and glass fibre content and it failed at a very low impact energy number. The specimen at the bottom extracted from a different area of the plaque had a very high concentration of fibres and the impact strength was reported to be more than 10 times the value of the sample on the top of the picture. Both the notched and un-notched impact values, as for the experiments with flax rovings, showed to be independent from the orientation of the specimens extracted from the moulded plaques. The impact values decreased as the percentage of flax increased in a linear fashion. The high variability is associated to screw configuration and rpm. The process needs to be optimized to achieve maximum homogeneity of fibre dispersion to guarantee uniform properties and high levels of impact strength.

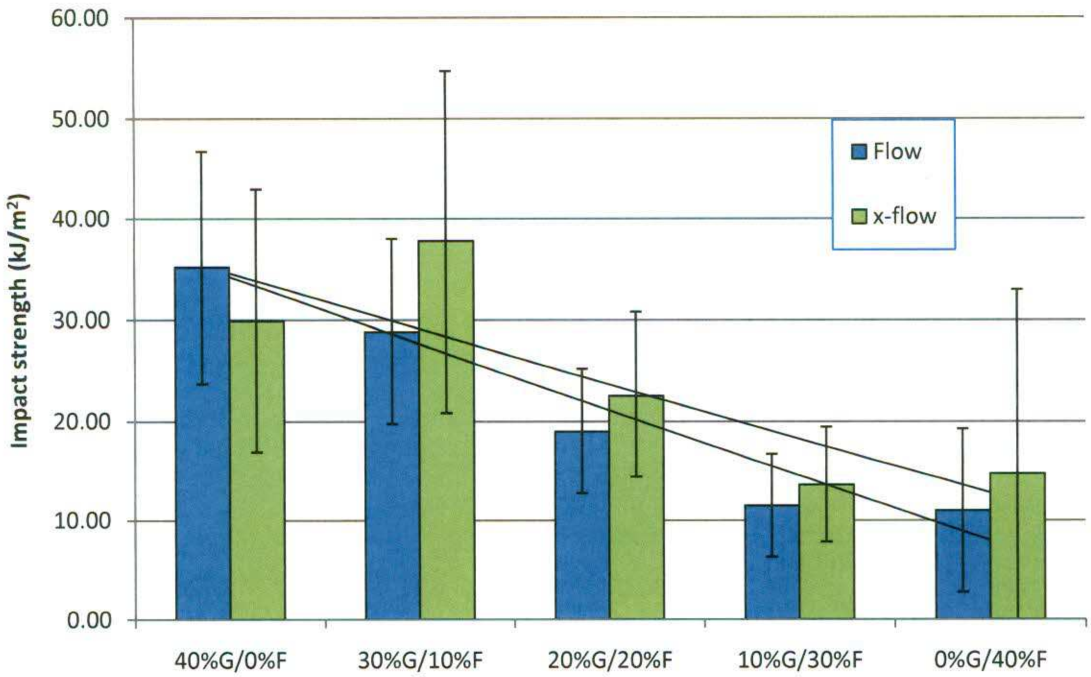


Figure 22: Notched impact strength for flax sliver compression moulded composite specimens in the flow and cross-flow directions

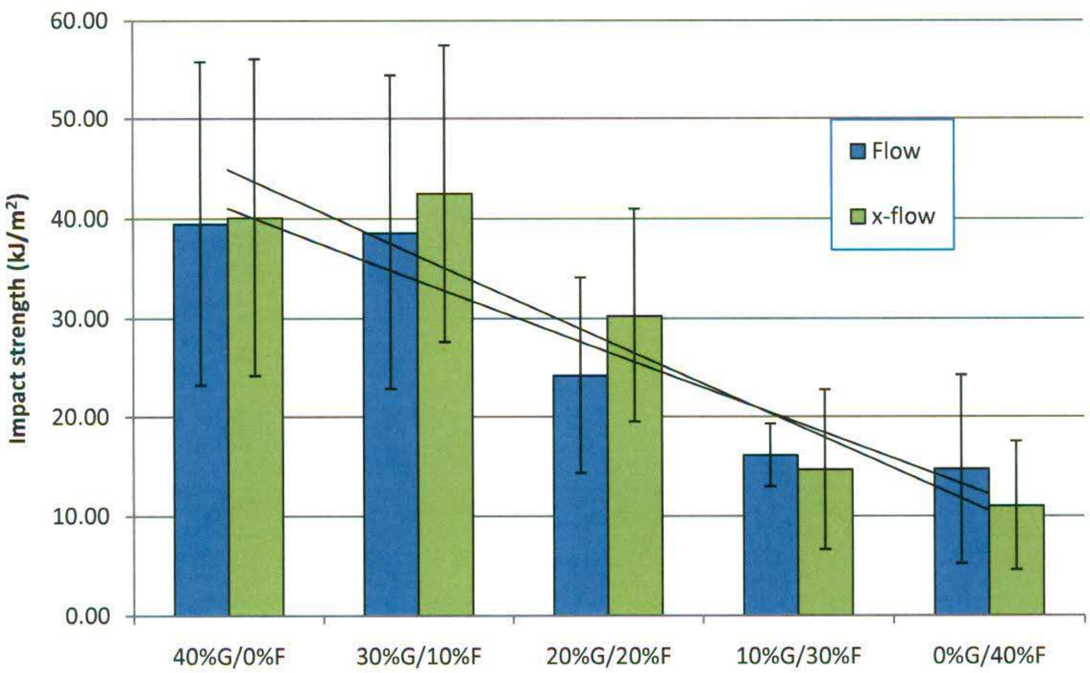


Figure 23: Un-notched impact strength for flax sliver compression moulded composite specimens in the flow and cross-flow directions.

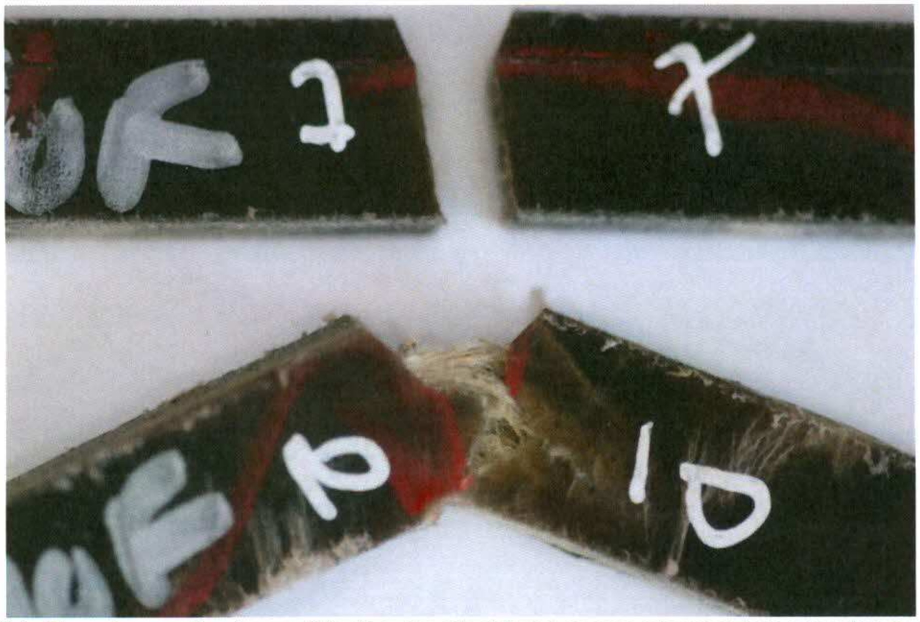


Figure 24: Specimens for impact test extracted from same plaque; top specimen is fibre-depleted, bottom specimen has large fibre loading.

Conclusions

Composite parts using PP as a thermoplastic polymer matrix and both discontinuous and continuous versions of flax fibres were produced using a twin-screw extruder at laboratory scale and a Coperion ZSK-70 respectively.

Composites with PP and 20% weight short flax fibres were obtained using flax in pelletized form. The best formulations for PP/20% discontinuous fibre composites in terms of mechanical properties were those containing MA grafted PP coupling agents and CaO as reactive additive, with up to 30% increment in tensile strength and 50% in tensile modulus when compared with neat PP. SEM micrographs showed the positive effect of MA grafted PP on the compatibilization of the polar (hydrophilic) flax fibres with the non-polar (hydrophobic) PP matrix. The enhanced compatibility reflects on a better bonding and ultimately on significant improvements in both tensile strength and modulus. TGA analysis confirmed the processability of flax fibres at the conventional processing temperatures used for PP homopolymers.

850 tex flax rovings with low twist was successfully fed into the fibre feeding section of an intermeshing co-rotating twin screw compounder in similar fashion as for the feeding of fibreglass rovings. Simultaneous feeding of flax and fibreglass rovings was achieved using standard equipment for fibreglass rovings. Parts moulded with flax and glass fibres showed a homogenous macroscopic distribution of the fibres throughout the parts. This demonstrated the ability of the twin screw compounder to distribute the fibre rovings homogenously.

Visual examination of the samples shows that the individual fibres in the rovings were not thoroughly separated and distributed within the polymer melt at a microscopic scale by the twin screw elements. The result is a lack of uniform distribution of the individual technical fibres in the final part. However, the obtained results suggest that roving de-bundling could be obtained by an optimization of the screw element configuration in the twin screw compounding system.

The feasibility of feeding and compounding flax fibre slivers in a conventional industrial D-LFT system has been demonstrated. Experiments using the 10,000 tex no-twist flax slivers produced a significantly improvement in the distribution of individual fibres when compared to the flax roving moulded samples. A visual examination of the plaques showed a more homogeneous distribution of flax fibres as the slivers have no twist and the fibres are more readily separated and dispersed by the screw elements within the twin screw extruder. Furthermore, the consistent relation between mechanical flow and cross-flow properties suggests the de-bundled fibres followed the flow trajectories in a similar fashion as the glass fibres.

The use of hybrid reinforcement fibres using the D-LFT technology with glass and flax fibre rovings and flax slivers presents itself as an interesting approach to produce parts that meet the performance requirements with a potential reduction in weight and possible improvements in sound and vibration damping.

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