Influence of Natural Fiber Type in Eco-Composites

Miriam García, Izaskun Garmendia, Javier García

CIDEMCO Technological Research Centre, Pol. Ind. Lasao, Area Anardi 5, 20730 Azpeitia (Guipúzcoa), Spain

Received 23 March 2007; accepted 5 September 2007
DOI 10.1002/app.27519

ABSTRACT: This study concerns the preparation of eco-composites based on natural fibers coming from wood and subproducts (rice husks) and products (kenaf) of annually grown plants. The matrices used were of two types: a biopolymer (PLA) and a petroleum-derived polymer (HDPE). Results showed that natural fibers markedly increase the tensile and flexural properties of both polymers by extending the field of application of these materials with less use of nonrenewable resources. The properties obtained are comparable to commercially available fiber-filled composites. Moreover, processing can easily be carried out in one step below a critical fiber volume. Fire and durability performance of the composites can be also improved by adding typical fire retardants and pigments. © 2007 Wiley Periodicals, Inc. J Appl Polym Sci 107: 2994–3004, 2008

Key words: composites; biofibres; biomaterials; reactive processing; mechanical properties

INTRODUCTION

Natural fiber composites are environmental-friendly materials, since reinforcement comes from renewable resources such as wood or annually grown plants.[1] This contributes to the easier degradation of the composites and, thus, to sustainability. Moreover, these fibers involve less environmental impact on their production[2] compared with both glass fibers and polymers, because they require lower energy (9.55 MJ/kg for flax fiber mat vs. 54.7 MJ/kg for glass fiber mat) and produce fewer emissions. Natural fiber composites can evidence performances as good as glass fiber composites[2] but with lower specific weight, machine wear and price, at the expense of lower impact strength. Wood fiber composites compete in properties with talc and mica composites, but have better environmental performance.

The global market of natural fiber composites was 771 million kg in 2002, with a value of 645 million euros.[3] The natural fibers come from the stalks, the leaves, and the seeds. Thirty million tons of natural fibers are produced annually, of which 20 million are cotton, and between 2 and 3 million tons are wood and jute. One of the most important fields of application is the car industry. In the year 2000,[4] this sector consumed 23,000 tons of natural fiber and it is estimated that its consumption will rise to 100,000 tons by 2010. The most used fibers in this sector include jute, kenaf, hemp, sisal, and flax. Besides the car industry, it is foreseen that these fibers will become widely used in packaging, consumer items and the construction industry.

Eighty five percentage of the market corresponds to wood composites (WPC).[3] Construction products (decking, cladding, window, and door profiles) cover 80% of the market share in North America and an annual increase in consumption by 14% is foreseen. Nowadays, 8% of the decking is made of this material, which is worth 390 million euros. Infrastructures occupy second position and transport the third.

The market of natural fiber composites is much smaller in Europe, with a market share of 105 million euros in 2002. However, increasing expectations about these materials are very positive.[5] The UK-based Hackwell Group[6,7] suggests an annual production and consumption growth rate of 10% per year for WPC, rising from 99,288 tonnes in 2005 to nearly 145,000 tonnes by 2009, which is worth about 290 million euros. Moreover, faster growth is likely to occur if larger companies with more resources enter the market.

The car industry is the biggest user of WPC in Europe at the moment, with well over half the total consumption. However, this amount corresponds to 7% of WPC consumption worldwide. In future, the Hackwell Group[6,7] expects a few European car suppliers to be diverted to other natural fibers. On the other hand, there is a great scope for growth in WPC sales in construction and furniture.[8] Construction is already the second largest sector after the car industry. Little furniture made of WPC is still available in the market, but more and more companies such as Ikea or IPT are developing furniture compo-

---

Correspondence to: M. García (miriam.garcia@cidemco.es).
Contract grant sponsor: EU.

Journal of Applied Polymer Science, Vol. 107, 2994–3004 (2008)

nents made of this material. The infrastructure industry is also a good potential consumer in applications such as highway sound barriers and waterfront fixtures.

The advantages of natural fiber composites are their good dimensional stability and durability against fungi and insects compared with wood.[9–12] Both aspects make these materials can have a working life of even 25–30 years with very low maintenance requirements. Moreover, even if they can content high amounts of fiber, they can be processed like plastics. The most popular resin[7] for natural fiber composites in Europe is virgin polypropylene (PP), whereas the global preference is for polyethylene, often recycled. PVC is also used by some companies.

The addition of natural fibers to polymers markedly increases the stiffness of the resulting material and also the strength, specially in terms of low fiber content.[13–15] However, due to the usual lack of proper fiber/matrix compatibility, the interfacial adhesion is not good enough, leading to a reduction in the mechanical properties. Thus, a compatibilizing agent is required.[13,15,16] Despite the fact that several compatibilizing methods exist, one of the most used is the addition of maleic anhydride grafted polymer for reactive blending with the polymer and the fiber during transformation.[13,16–18] The improvement of the interfacial adhesion markedly increases the mechanical performance of the composites because debonding is hindered.[13,15,19]

There exist various studies related to injection molding of WPCs.[18,20–24] Some of them analyze the effect of a compatibilizing agent[18,21] on the mechanical properties while others compare the mechanical performance of the composites after different processing techniques.[23,24] However, little is said about the effect of the structure of the wood on the WPCs. This has been taken into consideration in this work, together with the effect of injection molding parameters on the mechanical performance of the WPCs.

Recently, attention is being focused on biodegradable polymers and composites.[19,25–29] The blending of biodegradable polymers and natural fibers offer the possibility of obtaining a new type of eco-friendly and fully degradable composites. Poly(lactic acid) (PLA) is one of the most studied biodegradable polymer. There exist several papers about composites of PLA with natural fibers such as flax,[30,31] jute,[32] and cellulose.[33] On the other hand, kenaf fiber has been blended with different polymers.[34,35] However, very little has been published about PLA/Kenaf eco-composites.[28,29,36] The studies developed are related to crystallization processes,[36] properties which are specific for electronic applications[29] and mechanical properties of extruded films.[28] As regards PLA/rice composites, Yew et al.[37] have studied some physical properties (water absorption and enzymatic degradation) of these composites. To understand better the performance of such composites, additivated PLA/Kenaf composites were analyzed in the present work.

Thus, the aim of this work was to analyze the effect of different types of fiber and polymers in the performance of eco-composites. Two types of composites were prepared: those based on polyethylene and wood fibers and those based on biopolymer and annually grown plant fibers. Polyethylene/wood fiber composites were prepared by direct injection molding and the effect of different fiber types and contents on the mechanical properties was analyzed. The strategy behind this approach was to obtain high quality WPCs in order to replace polyolefins in several applications with easy-to-obtain economical eco-composites with better performance. PLA was blended with kenaf and rice husks to prepare the composites. They were firstly compounded and then injected and finally their properties were measured. The effect of a pigment and a fire retardant on PLA/natural fiber composites was also studied. This approach was used in order to obtain completely biodegradable high performance materials which may be used in applications such as construction or the car industry.

EXPERIMENTAL

The work was divided into two sets. In the first one biodegradable poly(lactic acid) (PLA) and annually grown plant fibers (kenaf and rice husks) were blended and the effect of the nature of the fibers on various properties was measured. In the second set, high density polyethylene (HDPE) was blended with wood fibers of different length and the properties of the composites were compared.

PLA with rice husks and kenaf

The PLA used was Biomer L-9000. The maleinizated poly(lactic acid) (MA-PLA) used as compatibilizer was prepared and kindly supplied by the Institute of Chemistry and Technology of Polymers from the Italian National Research Council (Italy).

Kenaf short fibers were kindly supplied by KEFI S.p.A. (Italy) and rice husks (RS) were obtained from Macedonia as an agricultural subproduct. The kenaf had a density of 57 g/L and rice husks 104 g/L measured at 23°C and 50% relative humidity. Moisture content of the fibers was not determined. Fibers were not chemically treated prior to processing.

The fire retardant used was aluminium hydroxide and the pigment used was iron oxide.

Firstly, and after drying the PLA, kenaf and rice husks at 80°C for 12 h, the components were blended

TABLE I
Notched and Unnotched Charpy Impact Strength of the Composites

Fiber content (%)	Unnotched impact strength (J/m)		Notched impact strength (J/m)	
	Kenaf	Rice husks	Kenaf	Rice husks
0	76 ± 6 (17)	76 ± 6 (17)	7.4 ± 0.3 (1.7)	7.4 ± 0.3 (1.7)
20	47 ± 4 (11)	33 ± 7 (8)	11.5 ± 0.9 (2.6)	9.0 ± 1.6 (2.1)
30	52 ± 3 (12)	36 ± 4 (8)	12.2 ± 0.8 (0.3)	6.8 ± 1.0 (1.6)

Figures in brackets indicate incertitude of the measurements.

in a three blade high intensity mixer for 10 min in order to obtain homogeneous physical blends. Then, the blend was compounded by extrusion in order to obtain a good enough compatibility level between the phases, together with a homogeneous fiber distribution. The extrusion process was carried out in a corrotating double screw extruder at a screw speed of 250 rpm and temperature profile of 170–175°C. The injection process was performed in a Battenfeld PLUS 30/75 injection machine. ISO 527-2 ''Plastics-Determination of tensile properties-Part 2: Test conditions for molding and extrusion plastics'' Type 5 dog bone-shaped tensile specimens and 80 × 10 × 4 mm^3 flexural and impact specimens were obtained. The injection processing parameters were the following:

- Temperature profile: 175–185°C
- Injection speed: 24.5 cm^3/s
- Injection pressure: 2000 bar

PLA/MA-PLA/Kenaf and PLA/MA-PLA/RS composites with 5% MA-PLA and 20 or 30% of fibers were obtained.

Tests were conducted according to the standards proposed by CEN/TC 249/WG 13 ''Plastics-Wood-plastic composites (WPC)-Part 1: Test methods''. At least 5 specimens were tested in tensile, flexural, fire and durability tests; 8 specimens in impact tests and 3 specimens in Vicat temperature measurements. The data given is the arithmetic mean of the test values together with the typical standard deviation. Typical incertitude of the measurements is given in tables where mechanical properties are reported (Tables I and III). The typical incertitude is the incertitude of the measurement expressed as typical deviation. Type A evaluation method was used for the calculation of the typical incertitude. This method evaluates the incertitude of the measurement by means of statistical analysis of a number of observations.

Tensile and flexural tests were conducted in an Instron machine, model 5569 (Bucks, UK). Impact tests were carried out in an ATS15 Charpy impactometer. Tests were carried out at 23°C and 50% relative humidity. Tensile tests were performed according to EN ISO 527 ''Plastics Determination of tensile properties'' (at 5.0 mm/min with type 5 specimens), flexural tests according to EN ISO 178 ''Plastics-Determination of flexural properties'' (at 2.0 mm/min with 80 mm × 10 mm × 4 mm specimens) and notched and unnotched Charpy impact tests according to EN ISO 179-1 ''Plastics-Determination of Charpy impact strength-Part 1: Noninstrumented impact test'' (80 mm × 10 mm × 4 mm specimens). Test conditions were selected from the values recommended in ISO standards. Notches were performed in an ATS Charpy-Izod notcher, model 1013100, using a blade with an angle of 46° ± 15′ and a radius of 0.254 ± 0.025 mm. Single flame fire tests were performed according to EN ISO 11925-2 ''Reaction to fire tests-Ignitability of building products subjected to direct impingement of flame-Part 2: Single-flame source test'' in flexural bars, and durability tests were carried out in a QUV machine (QUV weathering tester, model QUV/basic) consisting of 4 h cycles of condensation at 50°C followed by 4 hours with UV-B light at 60°C. Vicat softening temperature (VST) was measured in an ATS Vicat machine, model MP/3, in accordance with EN ISO 306 ''Plastics-Thermoplastic materials-Determination of Vicat softening temperature (VST)'' following the method A50 (10N, 50°C/h).

Morphology of the fibers was determined in a Nikon Eclipse E400 optical microscopy adapted with a DS Camera Head DS-5M and a DS Camera Control Unit DS-L1 after blending in the high intensity mixer. The length and diameter values were obtained as mean values of at least 50 fibers.

HDPE and wood fiber composites

The polyethylene used was HDPE HMA 016 from ExxonMobil. The compatibilizer was maleinizated polyethylene (MA-PE) from Clariant. The lubricant was Loxamid OA (OA) from Clariant.

The wood fibers were conifer fibers from Rettenmaier & Söhne. Lignocel BK40-90 (Fiber A) has cubic structure and length of 300–500 μm. Lignocel S 150TR (Fiber B) has fibrillar structure with L/D ratio of 10/2 and length of 100 μm. Moisture content of

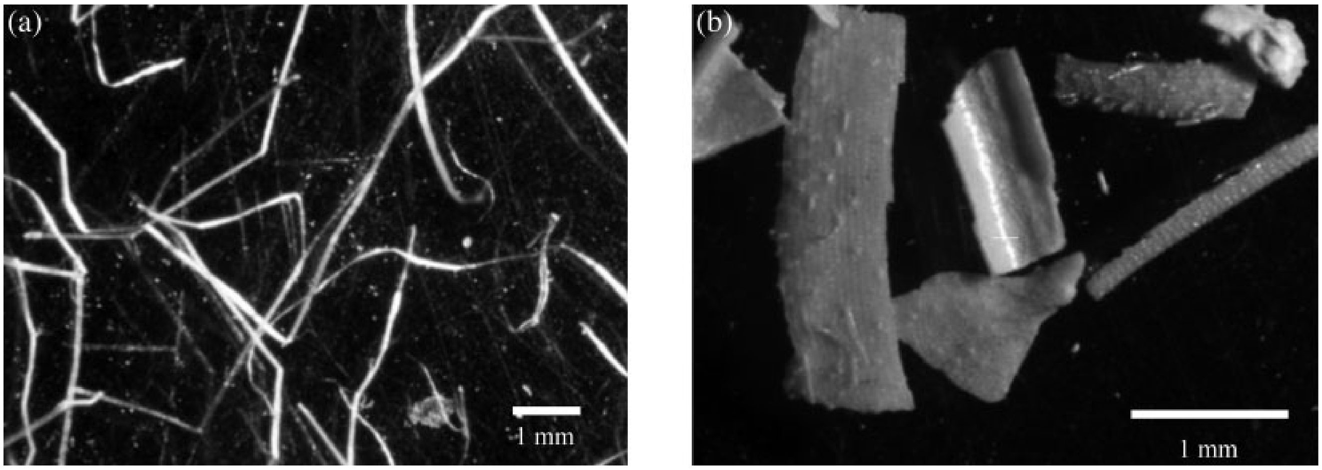

Figure 1 Kenaf fibers (a) and rice husks (b). A scale bar is reported in the pictures.

the fibers was not determined. Fibers were not chemically treated prior to processing.

Prior to injection, and after drying wood fibers at 80°C for 12 h, the components were hand blended. The injection process was performed in a Battenfeld PLUS 30/75 injection machine. ISO 527-2 ''Plastics-Determination of tensile properties-Part 2: Test conditions for molding and extrusion plastics'' Type 5 dog bone-shaped tensile specimens and 80 × 10 × 4 mm^3 flexural and impact specimens were obtained. Processing conditions were the following:

- Temperature profile: 175–180–185°C
- Injection speed: 35 cm^3/s
- Injection pressure: 1712, 2575, and 3038 bar

HDPE/MA-PE/OA/wood fibers composites with 0.5% lubricant to facilitate processability and different compatibilizer and fiber contents were obtained.

Tests were conducted according to the standards proposed by CEN/TC 249/WG 13 Plastics-Wood-plastic composites (WPC)-Part 1: Test methods. At least 5 specimens were tested in tensile and flexural tests and 8 specimens in impact tests. The data given are the arithmetic mean of the test values together with the typical standard deviation. Typical incertitude of the measurements is given in tables where mechanical properties are reported (Table IV). The typical incertitude is the incertitude of the measurement expressed as typical deviation. Type A evaluation method was used for the calculation of the typical incertitude. This method evaluates the incertitude of the measurement by means of statistical analysis of a number of observations.

Tensile and flexural tests were conducted in an Instron machine, model 5569 (Bucks, UK). Impact tests were carried out in an ATS15 Charpy impactometer. Tests were carried out at 23°C and 50% relative humidity. Tensile tests were performed according to ISO 527 ''Plastics-Determination of tensile properties'' (5.0 mm/min, type 5 specimens), flexural tests according to ISO 178 ''Plastics-Determination of flexural properties'' (2.0 mm/min, 80 mm × 10 mm 4 mm specimens) and notched and unnotched Charpy impact tests according to ISO 179 ''Plastics-Determination of Charpy impact strength-Part 1: Noninstrumented impact test'' (80 mm × 10 mm × 4 mm specimens). Test conditions were selected from the values recommended in ISO standards. Notches were performed in an ATS Charpy-Izod notcher, model 1013100, using a blade with an angle of 46° ± 15′ and a radius of 0.254 ± 0.025 mm.

RESULTS AND DISCUSSION

PLA with rice husks and kenaf

Biodegradable composites were produced by blending biodegradable PLA with kenaf and rice husks (RS). Kenaf structure was highly fibrillar ($L/D \cong 50$), while RS fibers were much shorter and wider ($L/D \cong 3.5$) as seen in Figure 1.

No characterization was carried out to determine the homogeneity of the fiber distribution after processing. It was assumed that the steps carried out during processing gave rise to homogeneous composites. This was validated by the homogeneity of the test results.

Figure 2(a,b) show the results of the tensile tests. Young's modulus increases dramatically after adding the fibers and increases at increasing the fiber content. On the contrary, tensile strength reduces after the addition of the fibers, especially with the RS. Thus, the fibers increase the fragility of the composites. These trends have also been reported for other extruded-injected natural fiber composites based on both biodegradable and nonbiodegradable polymers.[38] However, the results are very promising

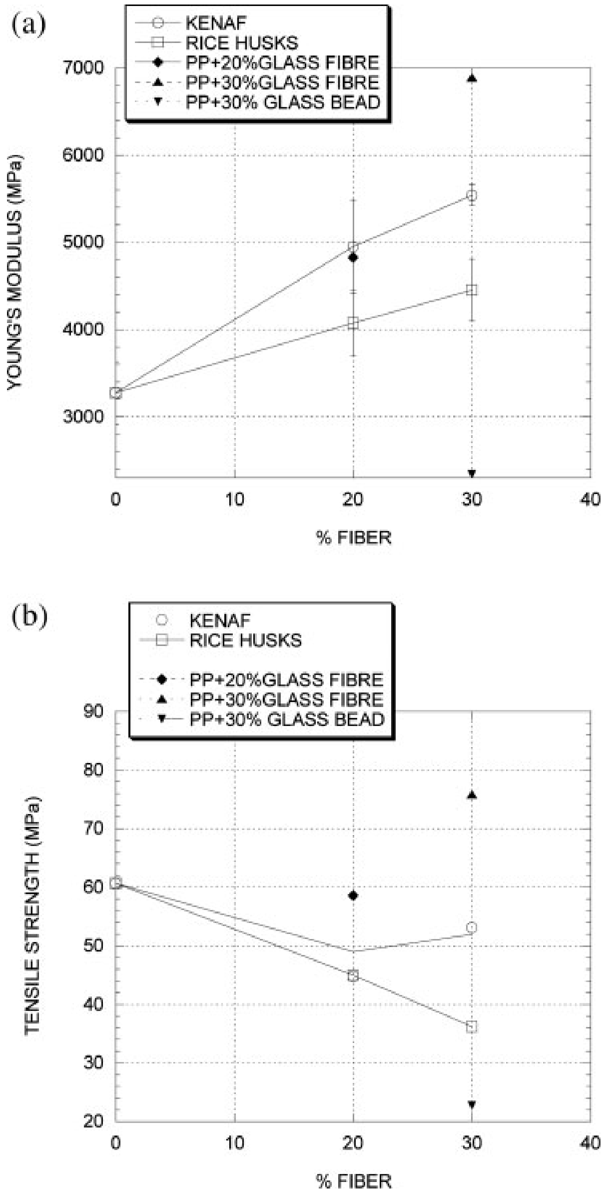

Figure 2 Young's modulus (a) and tensile strength (b) of the composites. PP composites are commercial materials. PP+20% glass fiber is RTP PP20GF; PP+30% glass fiber is RTP PP30GF and PP+30% glass beads is RTP100 GB30, all from RTP.

because, despite being lower than those of commercial 30% glass fiber-filled PP, kenaf composite results are similar to those of a commercial 20% glass fiber-filled PP and both kenaf and rice husks composites evidence much better performance than 30% glass bead-filled PP.

Figure 3(a,b) show the results of the flexural tests. As can be seen, increases are observed after the addition of kenaf in both modulus and strength. However, RS gives rise to decreases in both properties, indicating that it acts as a filler rather than a fiber, due to its low aspect ratio. As usual in composites, flexural and tensile deformation decreases after the addition of fibers. The results are very positive because in both kenaf and RS composites, modulus values are much better than that of glass bead-filled PP and even better than those of glass fiber-filled PP. On the contrary, strength values of natural fiber-filled composites are somewhat lower than those of glass fiber-filled PP, despite being much better than glass bead-filled PP. Thus, the new eco-composites can compete with typical plastics like glass fiber-reinforced PP. Serizawa et al.[29] also obtained properties similar to those of glass fiber composites in PLA/kenaf composites.

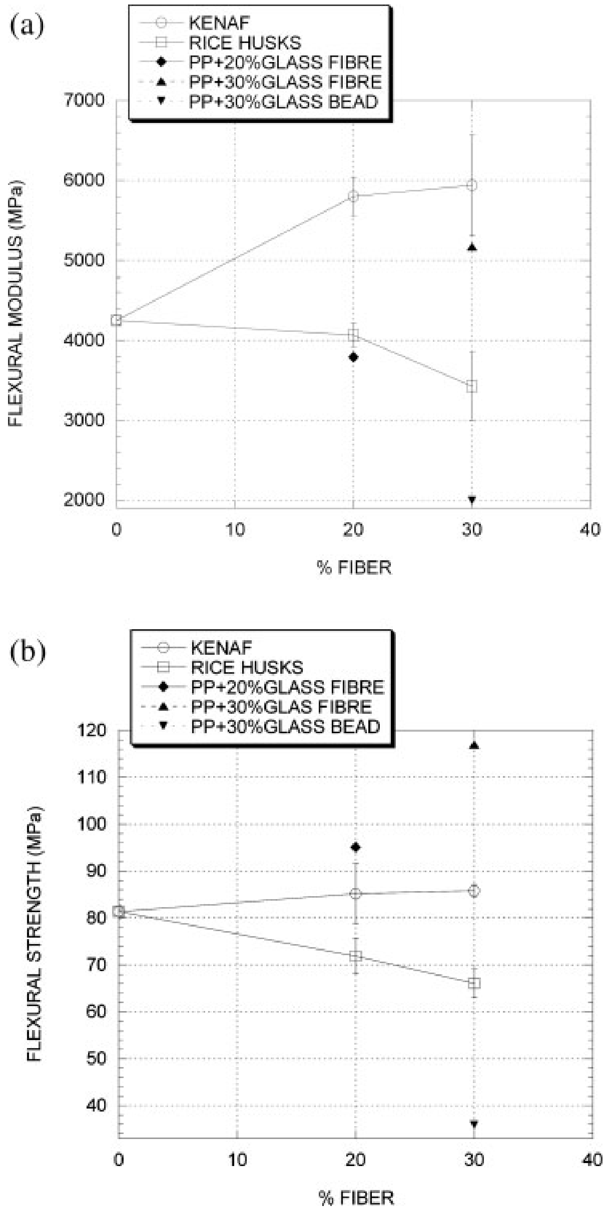

Figure 3 Flexural modulus (a) and flexural strength (b) of the composites. PP composites are commercial materials. PP+20% glass fiber is RTP PP20GF; PP+30% glass fiber is RTP PP30GF and PP+30% glass beads is RTP100 GB30, all from RTP.

TABLE II
Vicat Softening Temperature of the Composites

Composition	VST (°C)	
	Kenaf	Rice husks
PLA	58	58
20% Fiber	56	57
20% Fiber, 4% PIG, 10% FR	64	66

Charpy impact strength of the composites is shown in Table I. While RS hardly changes the notched impact strength of the matrix, kenaf gives rise to increases of more than 50%. Depending on the nature and the size of the fibers, the impact results show great differences. It was reported[38] that cut flax, hemp, sisal or cotton fibers gave rise to higher impact resistance than wood chips or ground straw due to their higher factor in terms of form. As usual in composites, unnotched impact strength decreases after the addition of the fibers. However, kenaf composites display better results than RS composites, once again due to their higher aspect ratio.

On the whole, taking into account the standard deviation of the data shown by the error bars and except for Young's modulus, results did not increase at increasing fiber content. Moreover, processing was much easier with 20% fiber for both kenaf and RS. This is because 20% in weight of fiber means 84% in volume of kenaf and 75% in volume of RS. In the case of higher fiber contents, the plastic does not embed the fiber properly, hindering the improvement in mechanical performance. Thus, composites with 20% fiber were considered the most suitable as regards their performance.

To improve the durability and fire resistance of the composites, fire retardant ($Al(OH)_3$) and pigment (iron oxide) was added to composites with 20% fiber. After some preliminary tests, 4% of pigment and 10% of fire retardant were selected as the most suitable contents. The addition of natural fibers did not increase the VST of the materials. However, as seen in Table II, the inorganic additives increased the VST remarkably. The addition of talc also increased the temperature resistance of HDPE/wood composites.[39]

Fire tests showed remarkable enhancements after the addition of the fire retardant as shown in Figure 4. PLA did not evidence any value because it lost integrity immediately when coming into contact with the flame. Burning time of both kenaf and RS composites was similar prior to the addition of the fire retardant. However, the time required for burning the specimens increased by 30% in kenaf composites and by more than 50% in RS composites when the aluminium hydroxide was added. The effectiveness of the fire retardant was higher in the RS composites due to the lower specific area of the fibers to be covered by the fire retardant. Kenaf composites with 20% fire retardant showed similar burning times to those of RS composites with 10% fire retardant. A higher fire retardant content did not lead to greater increases in the burning time, indicating that the selected fire retardant content is the minimum required for the maximum effectiveness in fire performance.

Durability tests showed that composites without pigment evidenced bleaching, cracking and chalking after keeping them for 500 h in the ageing machine. After 1500 h, the test bars were completely broken. However, the composites with pigment took on a much better appearance. Pigmented composites did not evidence any bleaching nor chalking—just some cracking in very few bars after 1500 h.

As regards retaining the properties, it was observed that the addition of the pigment and the fire retardant did not change the flexural properties of the neat composite before ageing. However, as seen in Table III, flexural modulus, strength and strain markedly decreased after maintaining the composites under severe conditions. The decrease in the modulus is not so noteworthy, indicating that the rigidity of the materials at low deformation levels is maintained after ageing. As stated by Stark,[40] this could be due to an increase in crystallinity of the polymer rich surface. However, as usual, ageing gives rise to a decrease in the elasticity of the materials, and, thus, ductility diminishes. As a consequence, a strong decrease in the flexural strength takes place. This is claimed[41,42] to be because moisture penetration into the composite degrades the fiber-polymer interface, which in turn causes loss of strength as the stress transfer from matrix to fiber becomes less efficient.

The overall conclusion is that kenaf composites give rise to much better mechanical performance than RS composites. However, an improvement in

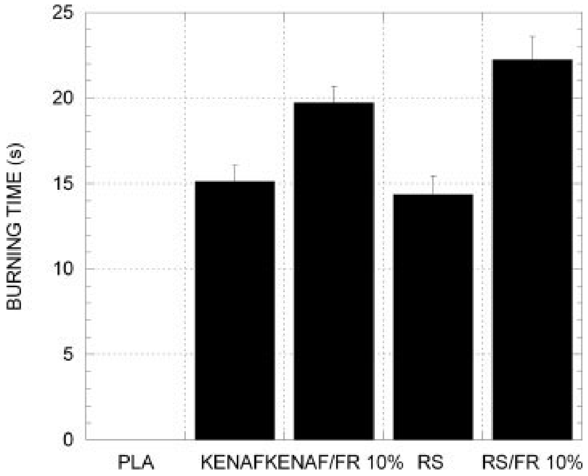

Figure 4 Fire tests of the composites.

TABLE III
Flexural Properties of Aged Composites

Ageing time (h)	Kenaf			Rice husks		
	Modulus (MPa)	Strength (MPa)	Strain (%)	Modulus (MPa)	Strength (MPa)	Strain (%)
0	5797 ± 239 (1300)	82 ± 9 (19)	2.4 ± 0.3 (0.5)	4067 ± 147 (946)	77 ± 10 (17)	2.9 ± 0.6 (0.6)
500	5249 ± 306 (1175)	32 ± 6 (7)	1.0 ± 0.2 (0.2)	4164 ± 40 (940)	29 ± 9 (12)	1.0 ± 0.5 (0.5)
1500	4877 ± 503 (1117)	29 ± 7 (7)	1.1 ± 0.3 (0.3)	4039 ± 179 (1648)	12 ± 1 (6)	1.0 ± 0.5 (0.5)

Figures in brackets indicate incertitude of the measurements.

the fire resistance of PLA/rice husks composites is even higher than that of kenaf composites. For applications where high performances are not required, such as housing or decorative appliances, PLA/rice husks composites, which show better performance than glass bead-filled PP, are a good choice in order to obtain a completely biodegradable material which is cheaper than usual biopolymers because of the low price of RS. On the other hand, the addition of kenaf led to high increases in the tensile and flexural modulus without noticeable changes in the strength. Moreover, notched impact strength and fire resistance also increased remarkably. To enhance durability of the materials, it is necessary to add a pigment to the composites. Four percentage iron oxide is enough to maintain the correct appearance of the composites without high loss in terms of rigidity.

HDPE and wood fiber composites

Fillers and glass fibers are often added to polyethylene in order to enhance its performance. If wood fibers are added instead of inorganic materials, degradability of the composites is greatly enhanced, giving rise to more ecological materials. Furthermore, the abrasion generated by the wood fibers is much less than that of the glass fibers—thus, they reduce the wear in the processing machines. Price is also a key factor because wood fibers are cheaper than glass fibers and even cheaper than HDPE, and so composites are cheaper than the neat polymer.

Using the direct injection processing method, the homogeneity of the fiber distribution could be deficient. However, the aim of this research was to analyze whether direct injection was an efficient method for the development of composites with homogeneous properties. If so, the processing method would be useful for obtaining composites despite some heterogeneities in the fiber distribution. However, no special measurement was taken.

Three, 5, and 7% of compatibilizer was added to the composites with 40% of fiber and the tensile and flexural modulus, strength and strain being studied. The results showed that unlike strain, where no differences were observed, modulus and strength increased at increasing the compatibilizer content from 3 to 5%, but no improvement was noted in the case of higher compatibilizer contents. Similar results were obtained for PP based WPC with different compatibilizers,[43] showing a maximum tensile, flex-

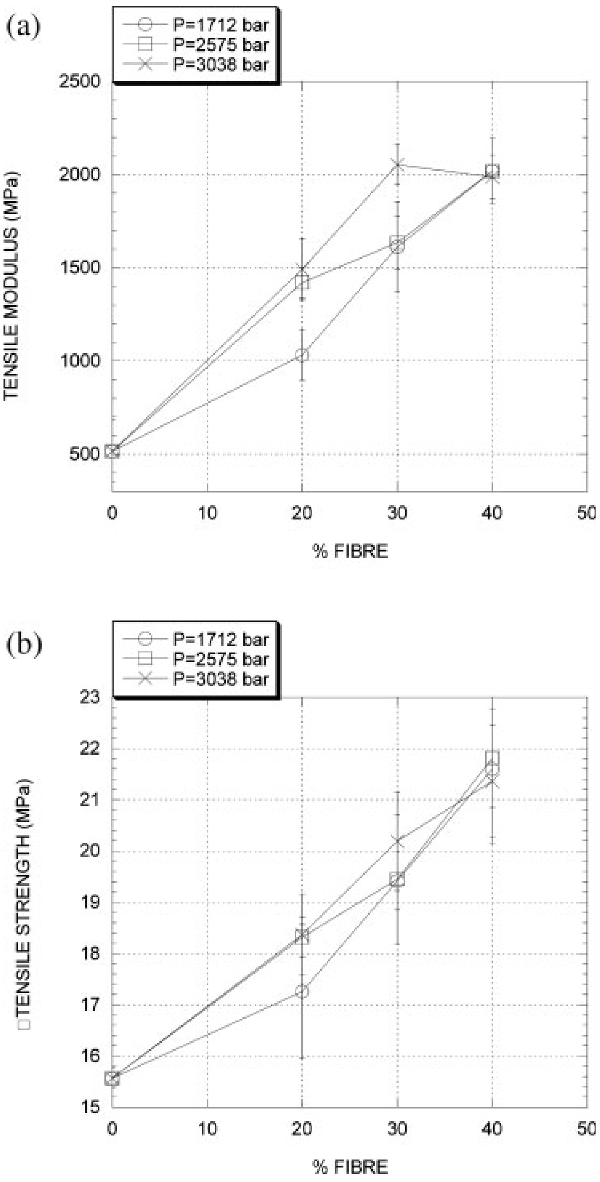

Figure 5 Young's modulus (a) and tensile strength (b) of the composites obtained at various injection pressures.

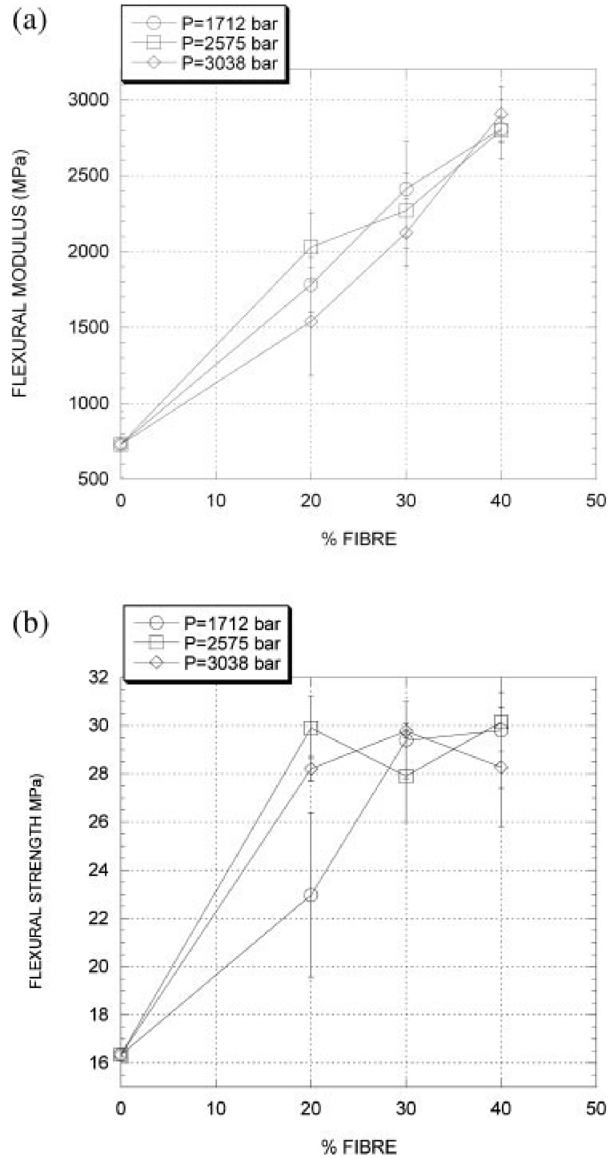

Figure 6 Flexural modulus (a) and strength (b) of the composites obtained at various injection pressures.

ural and impact strength values at around 4–5% in all cases. Thus, the optimum compatibilizer content was found to be 5%.

The results of composites with fiber A for tensile tests are shown in Figure 5. As can be seen, modulus and strength increase as fiber content increases regardless of injection pressure, especially in fiber contents above 20%. Improvements in tensile properties when increasing wood content have also been observed in other WPCs.[44,45]

Flexural modulus and strength are shown in Figure 6. As in the case of tensile results, the modulus increases as fiber content increases regardless of injection pressure. Nevertheless, the high increase in the flexural strength after the addition of fiber, especially at higher injection pressures, indicates that the composites achieve a high stiffness level with low fiber contents. The ductility decrease compensates for the increase in the strength and gives rise to similar strength values for all the composites. However, at low deformation values where the modulus is measured, the increase in the stiffness with the fiber content is easily observed.

As usual in WPC composites,[45,46] the unnotched impact strength of the composites decreased as fiber content increased. Notched impact strength remained constant with composition. Results are shown in Table IV.

Results revealed that the best mechanical performance was obtained with 40% A fiber. However, the processing of the composite in the case of this composition was very difficult because the matrix could hardly wet all the fiber, giving rise to constant variations during the processing. Thus, it was considered that the composite with 30% fiber gave rise to the overall best mechanical and processability performance.

When the same tests were performed with the composites composed of fiber B, unexpected results were obtained. After the addition of 20% B fiber, the tensile and flexural modulus increased to similar values achieved after the addition of 30% A fiber, but with higher fiber contents the modulus was maintained constant or even decreased because of the inability of the matrix to wet all the fibers. The tensile and flexural strength showed the same trend as the modulus, even with higher values than those of A composites for the flexural strength due to the larger aspect ratio of the fibers. The higher aspect ratio of hemp compared with wood also gave rise to higher properties in PVC composites.[46] However,

TABLE IV
Notched and Unnotched Charpy Impact Strength of the Composites With Fiber A

Fiber content (%)	Unnotched impact strength (J/m)			Notched impact strength (J/m)		
	1712 bar	2575 bar	3038 bar	1712 bar	2585 bar	3038 bar
0	Not broken	Not broken	Not broken	14.0 ± 1.5 (1)	14.0 ± 1.5 (1)	14.0 ± 1.5 (1)
20	35 ± 3 (8)	42 ± 4 (9)	41 ± 1 (9)	13.9 ± 0.8 (1)	13.8 ± 0.5 (1)	13.9 ± 0.6 (1)
30	30 ± 3 (7)	31 ± 2 (7)	30 ± 4 (8)	13.8 ± 1.1 (1)	13.4 ± 1.0 (1)	14.1 ± 1.3 (1)
40	27 ± 3 (6)	22 ± 3 (5)	22 ± 3 (5)	14.1 ± 1.5 (1)	13.0 ± 1.2 (1)	13.1 ± 1.2 (1)

Figures in brackets indicate incertitude of the measurements.

the notched impact strength decreased after the addition of the fibers because they act as weaknesses in the composites. These results indicate that the composites with 20% B fiber have the best mechanical performance, together with a good processability.

The difference in the mechanical performance of both fibers A and B might be due to their density. Fiber B has a density of 100–135 g/L, while that of A is 170–230 g/L. Taking into account that the density of HDPE is approximately 950 g/L, both 20% B and 30% A by weight means 67% fiber in volume. So, the mechanical results are very similar when the same volume of wood fibers is blended with HDPE, despite their characteristics. Thus, this results in a critical wood fiber volume above which the polymer cannot wet the fiber properly, giving rise to decreases in terms of mechanical properties. On the other hand, appearance is more plastic-like when smaller fibers are employed.

Finally, in order to apply these composites in proper applications, their properties must be compared with those of typical commercialized composites based on HDPE. The results are shown in Table V. As can be seen, tensile modulus is similar to that of commercial WPCs with 20% wood. Moreover, flexural modulus is comparable to talc, mica and 20% glass fiber composites and even higher than commercial 20% wood fiber composites. Tensile and flexural strengths are also comparable to those of wood fiber composites and just slightly lower than those of mica, talc or glass fiber composites. Thus, the properties of directly injected WPCs are comparable to commercially available wood fiber-filled and inorganic filler and fiber-filled composites.

Comparison between PLA and HDPE composites

It is difficult to compare the results for HDPE/wood fiber composites with those for PLA/natural fiber composites, due to the use of both different matrix and fiber. HDPE has lower performance than PLA, so the effect of reinforcement is more pronounced in HDPE composites. In fact, the addition of 30% wood fiber to HDPE enhances the tensile and flexural moduli by over 200%, while the increases after fiber addition are below 50% in PLA composites. However, the flexural modulus of PLA/20%Kenaf and PLA/20%RS composites are 2.5 and 1.8 times that of HDPE/WPC. Pouteau et al.[38] stated that when comparing different matrixes (polycaprolactone and HDPE) with the same natural fibers, the biodegradable matrix gave rise to higher increases. The differences with this study could be because of the effect of the different nature and *L/D* ratio of the fibers used in each type of composite.

Nevertheless, both composites showed some common advantages and disadvantages. Among the

TABLE V
Comparison of Mechanical Properties of Various Composites

	Filler							
	Mica 25%	Talc 30%	Talc 40%	Glass fiber 20%	Wood fiber 20%	Wood fiber 40%	Fiber A 30%	Fiber B 20%
Property	Code PE4325 from Spartech Polycom	Code PE4130 from Spartech Polycom	Code RTP727 from RTP	Code HD0234620UVL from Aclo Accutech	Injection molding grade from North Wood	Injection molding grade from North Wood	From this study	From this study
Tensile modulus (MPa)	–	–	3100	3000	1700	2700	1635	1700
Tensile strength (MPa)	34.5	32.4	20.7	35 (yield)	17	18	19.4	26.6
Flexural modulus (MPa)	2000	2137	2410	2200	1400	2400	2270	2309
Flexural strength (MPa)	44.8	46.9	37	39	31	35	27.9	33.7

TABLE VI
Forces, Weaknesses, and Industrial Uses of Eco-Composites

<table>
<tr><th></th><th>PLA composites</th><th>HDPE composites</th></tr>
<tr><td>Forces</td><td>Biodegradability
Lower price than pure biopolymer
Mechanical properties comparable to glass fiber filled composites susceptible of substitution (PP composites)
Improved fire performance</td><td>Low price
Easier degradation
Mechanical properties comparable to glass fiber filled HDPE composites
Possibility of wood-like appearance
Easy processing at low fiber contents</td></tr>
<tr><td>Weaknesses</td><td colspan="2">Lower impact strength
Need of agents against ageing</td></tr>
<tr><td>Industrial uses</td><td>Structural applications
Civil engineering applications
Automotive applications
Aerospace
Sports</td><td>Building applications
Containers
Consumer goods</td></tr>
</table>

advantages, easier degradation and an increase in the tensile and flexural properties is displayed. The major disadvantage is the decrease in the unnotched impact strength. Table VI exhibits the forces, weaknesses and possible industrial uses of these composites. It should be highlighted that the main differences between both composites are the following:

- Lower price of HDPE composites compared with PLA composites
- Lower mechanical properties of HDPE composites compared with PLA composites
- Easier processing of wood fiber filled composites than other natural fiber filled composites

CONCLUSIONS

Eco-composites were obtained by blending natural fibers with petroleum-derived polymers and biopolymers. The addition of natural fibers such as kenaf, rice husks or wood to polymeric matrixes improves the mechanical performance of the composites remarkably, regardless of the nature of the polymer. Mechanical performance similar to or even better than typical inorganic filler and fiber-filled composites can be achieved with lower environmental impact, extending the application spectrum of the eco-composites.

In the case of fibers obtained as products and subproducts coming from annually grown plants such as kenaf and rice husks, the aspect ratio of the fibers plays a key role in the performance of the composites—the higher the aspect ratio, the higher the mechanical properties. However, for applications where high mechanical performance is not necessary like housing or decorative appliances, the price of the biodegradable materials can be easily reduced by adding agricultural subproducts to bioplastics. Durability and fire tests also reveal good results when fire retardants and pigments are added to the composites.

The study performed in wood fiber composites reveals that composites can be easily obtained by direct injection molding by up to almost 70% in volume of fiber. Furthermore, injection parameters do not seem to affect the mechanical performance of the composites, which is only fiber content dependant. Different types of wood affect more the appearance of the composite rather than their mechanical properties.

This work was supported by the EU within the framework of an INCO Project entitled ''Eco-houses based on eco-friendly polymer composite construction materials''. CIDEMCO thank the EU for the financial support and the consortium for allowing the publication of technical data as examples.

References

1. Fifth International Conference on Woodfiber-Plastic Composites, Forest Products Society, Madison (Wisconsin), USA, 1999.
2. Leveque, J. Natural Fibres Forum Proceedings, JEC Composites Forums, Paris, 2006.
3. Principia Partners. Wood Plastic Composites 2003 Study, Principia Partners, Exton, USA, 2003.
4. Karus, M.; Kaup, M.; Lohmeyer, D. Markets and Prices for Natural Fibers Study, Nova Institute, Germany, 2000.
5. Optimat Ltd., MERL Ltd. Wood plastic composites study-Technologies and UK market opportunities, The waste and resources action programme, Banbury (Oxon), UK, 2003.
6. Hackwell Group. The European Wood Plastics Composites Market in Europe 2003: Construction, Furniture, Automotive and other Applications, Hackwell Group, UK, 2003.
7. Hackwell Group. Wood Plastics Composites in Europe (2006): Analysis–Technology–Market Opportunities, Hackwell Group, UK, 2006.
8. Pritchard, G.; Hackwell, B. Second Wood Fibre Polymer Composites Symposium: Application and Perspective, CTBA, Bordeaux, France, 2005.
9. Schut, J. H. Wood-plastic composites weathering quality issues, Plastic Technology, Sept, 2005.
10. Morrell, J. J.; Stark, N. M.; Pendleton, D. E.; McDonald, A. G. Wood Design Focus 2006, 16, 7.
11. Wang, W.; Morrell, J. J. Forest Product J 2004, 54, 209.
12. Pascal, K. D.; Jiang, H.; Cui, W.; Freed, J.; Matuana, L. M. Compos Part A: Appl Sci Manuf 2004, 35, 347.

13. Dányádi, L.; Renner, K.; Szabó, Z.; Nagy, G.; Móczó J.; Pukánszky B. Polym Adv Technol 2006, 17, 967.
14. Dash, B. N.; Rana, A. K.; Mishra, H. K.; Nayak, S. K.; Mishra, S. C.; Tripathy, S. S. Polym Comp 1999, 20, 62.
15. Kamel, S. Polym Adv Technol 2004, 15, 612.
16. Andersons, J.; Sparnins, E.; Joffe, R. Polym Comp 2006, 27, 221.
17. Avella, M.; Bozzi, C.; Dell, ER.; Focher, B.; Marzzetti, A.; Martuscelli, E. Die Angewandte Makromolekulare Chemie 1995, 233, 149.
18. Yang, H. S.; Wolcott, M. P.; Kim, H. S.; Kim, S.; Kim, H. J. Comp Struct 2007, 79, 369.
19. Cao, Y.; Shibata, S.; Fukumoto, I. Compos Part A: Appl Sci Manuf 2006, 37, 423.
20. Bledzki, A. K.; Faruk, O. Cellular Polym 2002, 21, 417.
21. Yeh, S. K.; Al-Mulla, A.; Gupta, R. K. J Polym Eng 2006, 26, 783.
22. Najafi, S. K.; Tajvidi, M.; Chaharmahli, M. J Appl Polym Sci 2006, 102, 3907.
23. Yang, H. S.; Wolcott, M. P.; Kim, H. S. Polym Testing 2006, 25, 668.
24. Bledzki, A. K.; Faruk, O. Int Polym Proc 2006, 21, 256.
25. Ochi, S. Compos Part A: Appl Sci Manuf 2006, 37, 1879.
26. Kurek, B.; Mougin, G. Third Wood Fibre Polymer Composites International Symposium: Innovative Sustainable Materials Applied to Building and Furniture, CTBA, Bordeaux, France, 2007.
27. Deb, P. C.; Rajput, L. D.; Hande, V. R.; Sasane, S.; Kumar, A. Polym Adv Tech 2004, 14, 749.
28. Nishino, T.; Hirao, K.; Kotera, M.; Nakamae, K.; Inagaki, H. Sci Tech 2003, 63, 1281.
29. Serizawa, S.; Inoue, K.; Iji, M. J Appl Polym Sci 2006, 100, 618.
30. Bodros, E.; Pillin, I.; Montrelay, N. Comp Sci Tech 2007, 67, 462.
31. Oksman, K.; Skrifvars, M.; Selin, J. F. Comp Sci Tech 2006, 63, 1317.
32. Khondker, O. A.; Ishiaku, U. S., Nakai, A. Compos Part A: Appl Sci Manuf 2006, 37, 2274.
33. Ganster, J.; Fink, H. P. Cellulose 2006, 13, 271.
34. Avella, M.; Bogoeva-Gaceva, G.; Buzarovska, A. J Appl Polym Sci 2007, 104, 3192.
35. Tajvidi, M.; Shekaraby, M. M.; Motiee, N. Polym Comp 2006, 27, 563.
36. Nishino, T.; Hirao, K.; Kotera, M. Compos Part A: Appl Sci Manuf 2006, 37, 2269.
37. Yew, G. H.; Yusof, A. M. M.; Ishak, Z. A. M. Polym Deg Stab 2005, 90, 488.
38. Pouteau, C.; Lakhdar-Chaouch, H.; Barq, P. Mechanical properties of natural fibre composites, processed by injection moulding. Structure/properties relationship, Second Wood Fibre Polymer Composites Symposium, Bordeaux, 2005.
39. Crépin-Leblond, J.; Jouffret, F.; Clark, R.; Noel, O. Talc reinforced wood plastic composites, Second Wood Fibre Polymer Composites Symposium, Bordeaux, 2005.
40. Stark, N. Changes in wood flour/HDPE composites after accelerated weathering with and without water spray, Second Wood Fibre Polymer Composites Symposium, Bordeaux, 2005.
41. Stark, N. J Thermoplast Comp Mat 2001, 14, 421.
42. Rangaraj, S. V.; Smith, L. V. J Thermoplast Comp Mat 2000, 13, 140.
43. Putz, R. F.; Stadlbauer, W.; Burgstaller, C.; Gösweiner, A.; Rittberger, R.; Weichslberger, G. Influence of wood type and fibre geometry, polymer type and additive type on the mechanical properties of directly extruded wood-plastic-composites, Second Wood Fibre Polymer Composites Symposium, Bordeaux, 2005.
44. Pouteaux, C.; Grimaud, X. Wood and natural fibre composites processed by hybrid injection moulding: Evaluation and testing of the performances of the material, Third Wood Fibre Polymer Composites Symposium, Bordeaux, 2007.
45. Hajji, P.; Cygan, Z. Wood/PVC composites formulation optimisation, Third Wood Fibre Polymer Composites Symposium, Bordeaux, 2007.
46. Bloyaert, B. PVC and natural fibres, a successful association, Second Wood Fibre Polymer Composites Symposium, Bordeaux, 2005.