

A Review
on Natural Fibre-Based Composites—
Part II:
Application of Natural Reinforcements
in Composite Materials
for Automotive Industry

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ABSTRACT. Natural abundance, much higher strength per unit weight than most inorganic fillers, lower density and their biodegradable nature make natural fillers attractive as reinforcements of engineering polymer systems. However, certain drawbacks such as incompatibility with the hydrophobic polymer matrix, the tendency to form aggregates during processing and poor resistance to moisture greatly reduce the potential of natural fibres to be used as reinforcements in polymers. In this review, the main results presented in literature are summarized, focusing on the processing behaviour and final properties of natural fibres with polymeric matrices (thermoplastics, thermosets and biodegradables) and paying attention to the use of physical and chemical treatments for the improvement of fibre-matrix interaction and composite mechanical

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Part of the presented results have been obtained from the research work of the ECOFINA Project, in the framework of the 5th European Research Programme. The authors would like to thank all the partners of the Project for their useful research and the other colleagues who have kindly furnished contributions.

Journal of Natural Fibers, Vol. 1(3) 2004
<http://www.haworthpress.com/web/JNF>
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Digital Object Identifier: 10.1300/J395v01n03_03

properties. This work mainly focuses on the use of natural fibres for automotive applications. [Article copies available for a fee from The Haworth Document Delivery Service: 1-800-HAWORTH. E-mail address: <docdelivery@haworthpress.com> Website: <<http://www.HaworthPress.com>> © 2004 by The Haworth Press, Inc. All rights reserved.]

KEYWORDS. Natural fibres-based composites, chemical treatments, automotive application, ECOFINA

INTRODUCTION

The collapse in prices of engineering and standard plastics, the assumption of the future exhaustion of crude world-wide reserves and increasing environmental concerns have lead to the use of regenerable raw materials for the design and development of new components (Mohanty, Misra and Drzal 2002). After several years in which the development of synthetic fibres has dominated, it is remarkable how natural reinforcements have gained renewed interest, especially as a glass fibre substitute in automotive industries (Brouwer 2000). The main advantages of using natural fibers in composite materials can be summarized in the following points: they are process-friendly, they have lower specific weight and do not wear out tooling and they also have good thermal and acoustic insulating properties. However, some disadvantages, such as, variable quality depending on unpredictable influences such as weather and moisture absorption, limited maximum processing temperatures, lower strength properties, lower durability, poor fire resistance and the fluctuation of prices based on harvest results or agricultural politics can limit their industrial application. Most of the available information published over the last few years on natural fibre/wood composites concerns the following topics: identification and classification of fibres (volume, length and diameter), size of the crystalline fibrils and non-crystalline regions, crystal-structure (type of cellulose and defects) and void structure, treatments and adaptations of natural fibres, study of compatible fibre/matrix composites (Bledzki, Reihmane and Gassan 1996; Stamboulis, Baillie and Peijs 2001; Joseph, Thomas and Paul 1997; Valadez-Gonzalez, Cervantes et al. 1999). However, more extensive work is necessary to assess the standard quality of fibres (the function of raw material optimisation, growth-maturity-retting-decortication processes, intelligent preparation and processing of fibres) as well as to

improve the compatibility between the hydrophobic-thermoplastic or hydrophilic-thermoset matrix and the cellulose-based hydrophilic natural fibres. Fibre quality assessment together with the knowledge of the relationships concerning tetranomium mechanic-morphology-surface-chemistry, which is a function of the components used for composite realization and also of the manufacturing technology and conditions used, will lead to the development of competitive natural fibre composites as opposed to glass fibre-based ones. Therefore, a critical evaluation of the previous results obtained in the field of polymer NFCs has been made in this review, in order to suggest the right approach for the analysis of their mechanical and physical characteristics and to identify the best solution especially in the case of automotive part manufacturing.

THERMOPLASTIC MATRIX-NATURAL FIBRE COMPOSITES

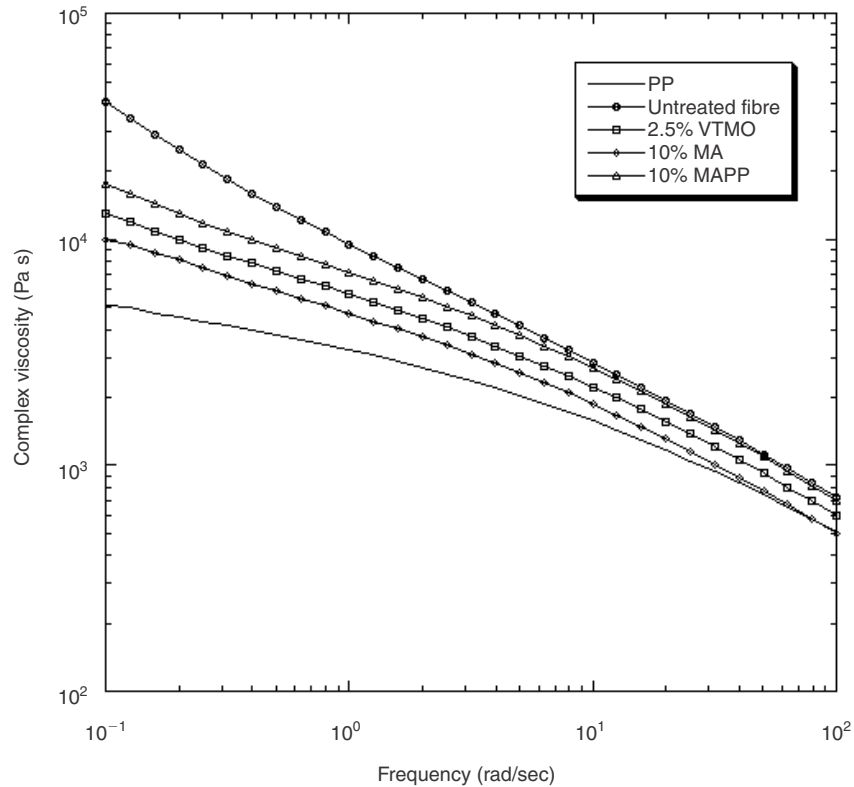
Lignocellulosic fibres were incorporated in a great assortment of thermoplastic matrices such as those reported in Table 1. The results reported in a large number of investigations have shown that natural fibres have the potential to be used as reinforcements in plastics (Lopez Manchado, Biagiotti and Kenny 2002; Oksman and Nilsson 1998; Rowell et al. 1999). One of the main advantages of natural fibre composites is the possibility of using the conventional processing equipment of thermoplastic-based systems with low maintenance costs as a consequence of the poor abrasiveness nature of natural systems. However, the maximum temperature values permissible before fibres begin to degrade or the water in hydrophilic fibres evaporate, represent the most important limitation to the use of these materials on an industrial scale. This processing factor limits the type of thermoplastic matrix, such as polyethylene (PE), polypropylene (PP) and polystyrene (PS) to be used with lignocellulosic-fibres. The final properties of the composite are generally strongly influenced by a number of parameters such as the mechanical properties and the geometrical characteristics of the reinforcement, the fibre-matrix and the fibre-fibre interactions and by the distribution and possible preferential orientation of fibres. The rheological behaviour of the material is also important for selecting the optimal processing condition for fabricating the product. Several research papers (Czarnecki and White 1980; Takase, and Shiraishi 1989; Joseph, Joseph and Thomas 1999; Hornsby, Hinrichsen and Tarverdi 1997) have studied the effects of processing parameters, such as mixing time, screw rate and temperature profiles, on the characteristics of the com-

TABLE 1. Thermoplastic matrix-natural fibre composites.

Fiber	Thermoplastic Matrix	References
Cellulose	PP	(Felix and Gatenholm 1991; Quillin, Caulfield and Koutsky 1993; Son, Lee and Im 2000; Biagiotti et al. 2002)
	PE	(Dong, Sapieha and Schreiber 1992; Sapieha, Allarda and Zang 1990)
	PA66	(Garcia Ramirez et al. 1994; Garcia Ramirez et al. 1995)
	PS	(Maldas, Kokta and Daneault 1989; Czarnecki and White 1980)
	PVC	(Matuana, Park and Balatinecz 1996)
Flax	PP	(Zafeiropoulos, Baillie and Matthews 2001; Wielage et al. 1999; Mieck, Nechwatal and Knobelsdorf 1995; Van de Velde and Kiekens 2002; Hornsby, Hinrichsen and Taverdi 1997)
	PE	(Stamboulis, Baillie and Schulz 1999; Van de Velde and Kiekens 2001)
Jute	PP	(Rana et al. 1998; Gassan and Bledzki 1997; Gassan 2002; Karmacker and Schneider 1996; Mitra, Basak and Sarkar 1998)
	PE	(Tripathy, Levita and Di Landro 2001)
Abaca	PHBV	(Shibata et al. 2002)
Sisal	PP	(López-Manchado M.A., Biagiotti and Kenny 2002; López-Manchado et al. 2000; Joseph, Joseph and Thomas 1999)
	PE	(Albano et al. 1999; Kuruvilla et al. 1996; Joseph, Thomas and Paul 1997; Kalaprasad et al. 1997)
	PS	(Nair et al. 2000; Nair, Thomas and Grueninckx 2001)
Kenaf	PP	(Caulfield et al. 1999; Rowell et al. 1999; Karnani, Krishnan and Narayan 1997)
Ramie	PP	(Angelini et al. 2000)
Broom	PP	(Avella et al. 1998; Contrafatto et al. 1998)
Henequén	PE	(Valadez-Gonzalez et al. 1999)
	PVC	(Ayora, Quijano and Marquez 1997)
	PMMA	(Canche-Escamilla et al. 1999)
Bagasse	PP	(Vázquez, Dominguez and Kenny 1999)
Bamboo	PP	(Thwe and Liao 2003)
Pineapple	PE	(George, Bhagawan and Thomas 1998a-b; George, Thomas and Bhagawan 1999)
Wood flour/fiber	PP	(Jana and Prieto 2002; Ichazo et al. 2001; Oksman and Clemons 1998; Balasuriya, Ye and Mai 2001; Coutinho, Costa and Carvalho 1997; Kazayawoko, Balatinecz and Matuana 1999; Raj et al. 1989; Belgacem, Bataille and Sapieha 1994; Sain and Kokta 1994; Czikovszky 1996; Sun and Hawke 1996)
	PE	(Oksman, Lindberg and Holmgren 1998; Raj, Kokta and Daneault 1990)
	PS	(Maldas and Kokta 1991; Simonsen and Rials 1996; Rials and Wolcott 1998)
	PVC	(Kokta et al. 1990)

posites obtained. A number of investigations on the rheological behaviour of natural fibre composites have also been reported (López-Manchado, Biagiotti and Kenny 2002; Nair et al. 2000). In general, natural fibre composites exhibit a pseudoplastic behaviour, manifesting higher non-Newtonian properties with respect to pure matrices (Figure 1). The overall increase in viscosity and non-Newtonian behaviour can be explained in terms of the flow hindrance produced by the fibres themselves. Clearly, the typology of flow influences the mastication of the fibre during processing, determining the final morphology in terms of

FIGURE 1. Complex viscosity of PP and of its flax fibre composites.

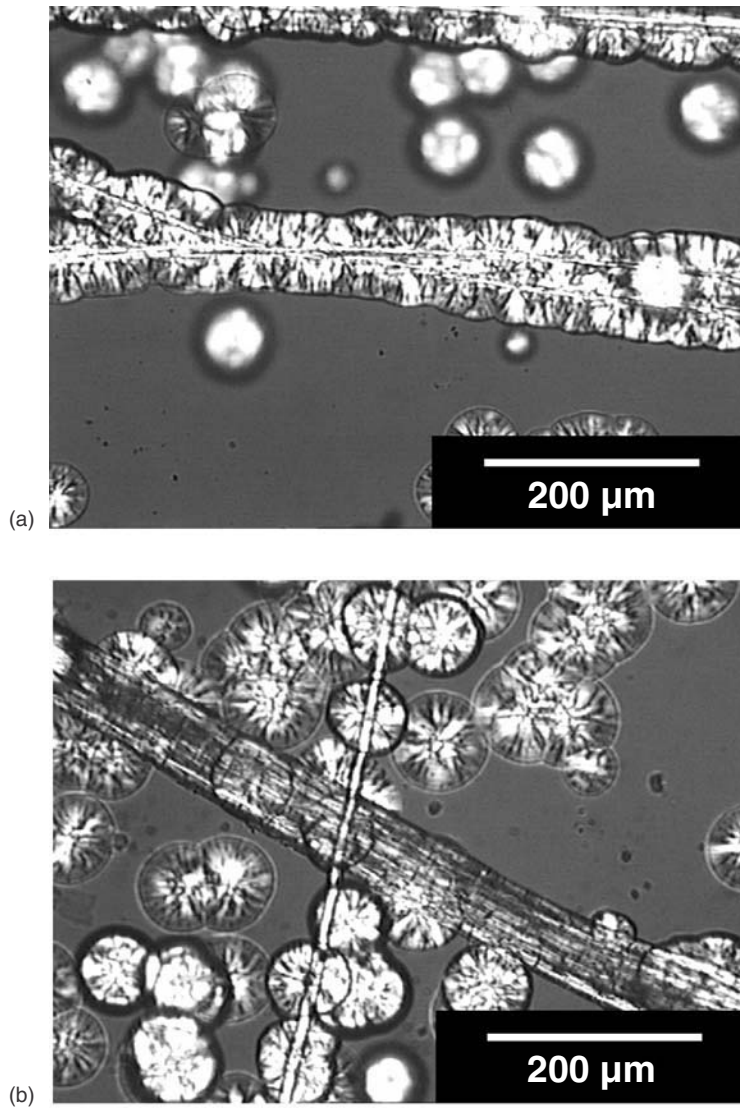


(Source: Biagiotti, J., D. Puglia, L. Torre, J.M. Kenny, A. Arbelaz, G. Cantero, C. Marieta, R. Llano-Ponte, and I. Mondragon. 2002, A Systematic Investigation on the Influence of Natural Fibre Treatments on the Final Behaviour of their Polymeric Matrix Composites. PART II: Analysis of Composite Properties. *Proceedings of the 23rd International SAMPE Europe Conference*, Paris, 299-322.)

length and orientation of the fibre in the matrix. The properties of the materials are strongly influenced by fibre loading, orientation and aspect ratio (ratio of length to diameter). There are several reports in literature about the correlation of the morphological and mechanical characteristics of fibres with the final composite by means of theoretical models (Kalaprasad et al. 1997; Hornsby, Hinrichsen and Tarverdi 1997).

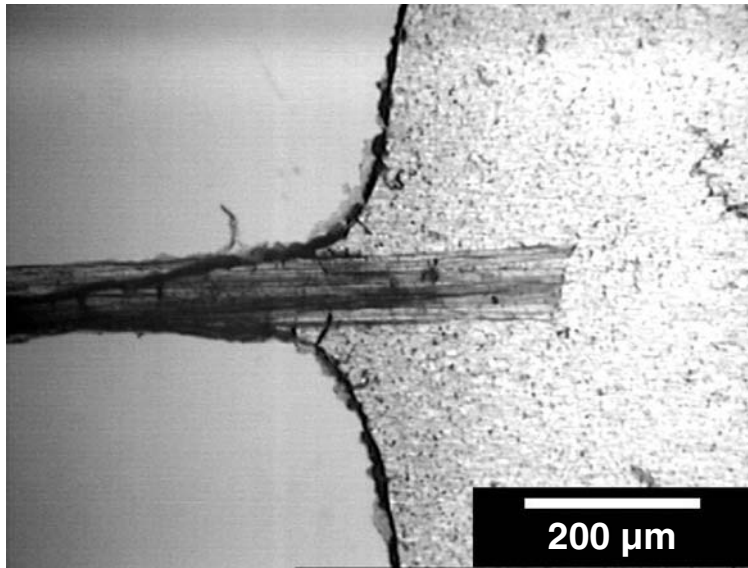
The modality of solidification of the melt during processing is another factor that can influence the properties of the final part. In particular, the study of the crystallization kinetics of the composite as a function of the processing conditions, from a macro-kinetic point of view, provides useful information for the analysis and the design of processing operations. There are several reports in literature about this effect (Lopez-Manchado et al. 2000; Nunez et al. 2002; Cyrus, Kenny and Vazquez 2001). In general, the accelerating effect of natural reinforcements on crystallization kinetics is evident as a consequence of the heterogeneous nucleation effect on natural fibers (Cantero, Arbelaiz et al. 2002). Moreover, the presence of transcrySTALLINE regions on the fibre surface seem to improve the quality of the fibre-matrix interfacial interaction (Figures 2a-2b), which plays a fundamental role in the upgrading of the mechanical properties of composite systems (Zafeiropoulos, Baillie and Matthews 2000). The evaluation of interfacial adhesion (Figure 3) can be carried out by means of micro-mechanical techniques already familiar in literature, such as, the single fibre fragmentation test (SFFT) (Kelly and Tyson 1965; DiBenedetto 1991), pull-out (López-Manchado, Arroyo et al. 2003; Li, and Netravali 1992) and microdebonding (Clark, Kander and Sauer 1999). Several authors have reported results obtained on interfacial adhesion between natural fibres and thermoplastic matrices by utilizing the above cited methods (Tripathy, Levita and Di Landro 2001; Valadez-Gonzalez et al. 1999; George, Bhagawan and Thomas 1998). In general, a poor adhesion level was measured due to the difference, in terms of polarity, of the constituents. Subsequently, stress transfer can be improved by pre-treating some of the composite constituents: the fibre, the matrix or both simultaneously. In this sense, the use of coupling agents or compatibilizing agents was largely studied and reported (Figure 4 and Figure 5). Interesting results were reported by Biagiotti et al. (2001, 2003a, 2003b), where the rheological and mechanical properties of ternary composites based on blends of isotactic polypropylene (iPP) and terpolymer ethylene-propylene diene rubber (EPDM) reinforced with flax fibres were studied (Figure 6). In order to evaluate the effect of the matrix composition and fibre content on the final properties of the composite, an experimental design based on a

FIGURE 2. Micrographs of crystalline growth after 260 sec at 130°C for PP with (a) untreated flax fibres and (b) 2.5% VTMO treated flax fibres.



(Source: Biagiotti, J., D. Puglia, L. Torre, J.M. Kenny, A. Arbelaz, G. Cantero, C. Marieta, R. Llano-Ponte, and I. Mondragon. 2002. A Systematic Investigation on the Influence of Natural Fibre Treatments on the Final Behaviour of their Polymeric Matrix Composites. PART II: Analysis of Composite Properties. *Proceedings of the 23rd International SAMPE Europe Conference*, Paris, 299-322.)

FIGURE 3. Optical polarizing microscopy of the pull-out sample for the PP/flax fibre system.

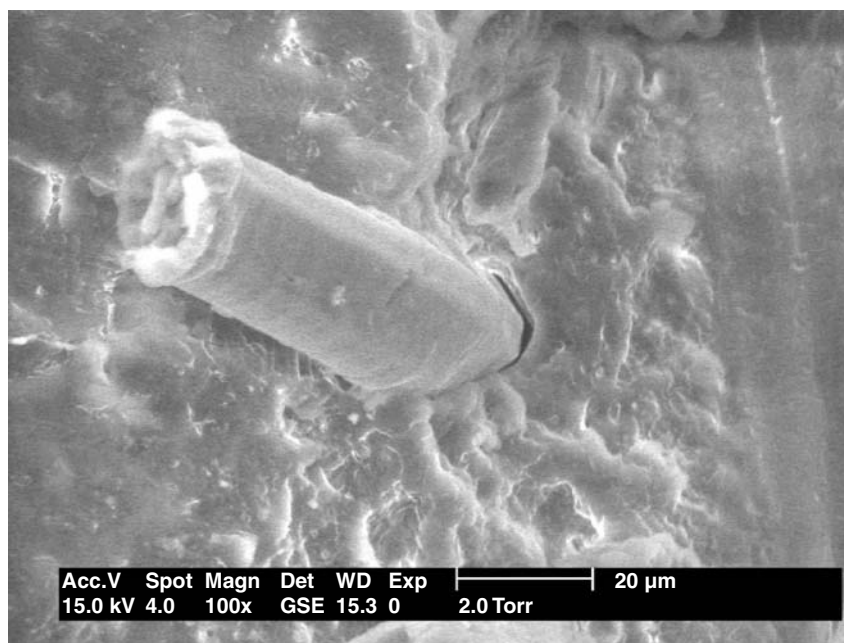


(Source: Biagiotti, J., M.A. Lopez-Manchado and J.M. Kenny. 2002. Studio della Modificazione Chimica di Elastomeri termoplastici Poliiolefinici Rinforzati con Fibra di Lino. 5 "CONGRESSO NAZIONALE AIMAT" Modena, Italy, September 8-11.)

Doehlert Uniform Net was employed. The results showed that flax fibres behave as an effective reinforcing agent, with a marked improvement of the composites' mechanical properties. It is, in fact, the incorporation of the fibres that gives rise to a more rigid material with a great increase in both the strength and modulus of the composite material. The higher the EPDM percentage in the blend, the more this effect was evident.

Moreover, a systematic and statistical approach to evaluate and predict the properties of random discontinuous natural fibre reinforced composites was also presented (Biagiotti et al. 2003). Different composites based on polypropylene and reinforced with natural fibres were produced and their mechanical properties were measured together with the distribution of the fibre size and the fibre diameter. The values obtained were compared with theoretical predictions, using a combination of the Griffith theory for the effective properties of the natural fibres and

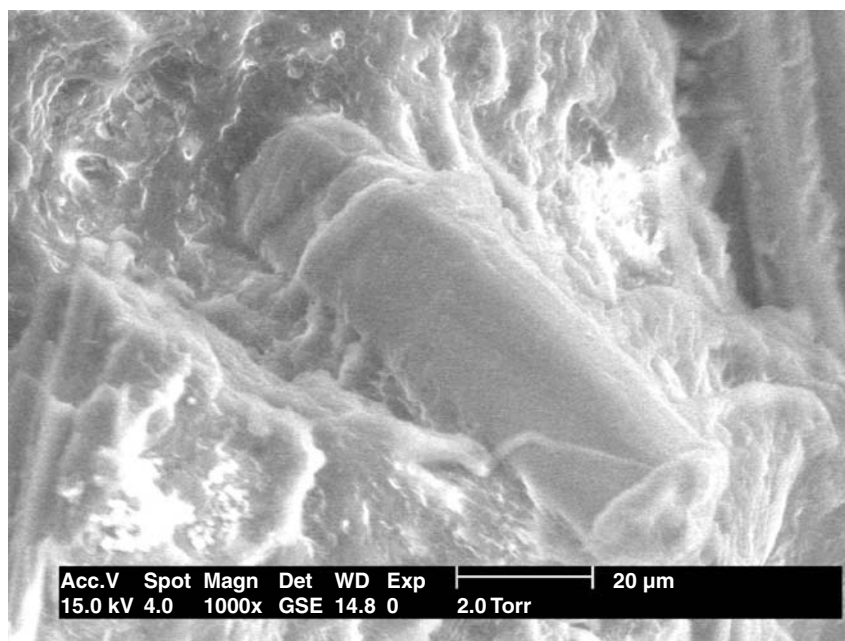
FIGURE 4. Fracture surface of flax fibre reinforced PP-EPDM composites.



(Reprinted with permission of John Wiley & Sons, Inc. from López-Manchado, M.A., M. Arroyo, J. Biagiotti and J.M. Kenny. 2003. Enhancement of the mechanical properties and interfacial adhesion of PP/EPDM/flax fiber composites by using maleic anhydride as a compatibiliser. *Journal of Applied Polymer Science* 90(8): 2170-2178.)

the Halpin-Tsai equation for the elastic modulus of the composites. The relationship between experimental results and theoretical predictions were statistically analysed using a probability density function estimation approach based on neural networks. The results obtained show a more accurate expected value with respect to the traditional statistical function estimation approach. In order to point out the particular features of natural fibres, the same proposed method was also applied to PP-glass fibre composites. It was observed that the theoretical elastic modulus predicted was close to the experimental value. The relatively small differences between the expected values of the moduli could be attributed to imperfections, in terms of fibre/matrix adhesion and voids, in the analysed composites. With the proposed method, the dispersion of properties could be approached utilizing a more accurate semi empirical method, which could be useful in the design and the optimisation of the processing of natural fibre reinforced composites.

FIGURE 5. Fracture surface of flax fibre reinforced PP-EPDM composites with maleic-anhydride modified EPDM.

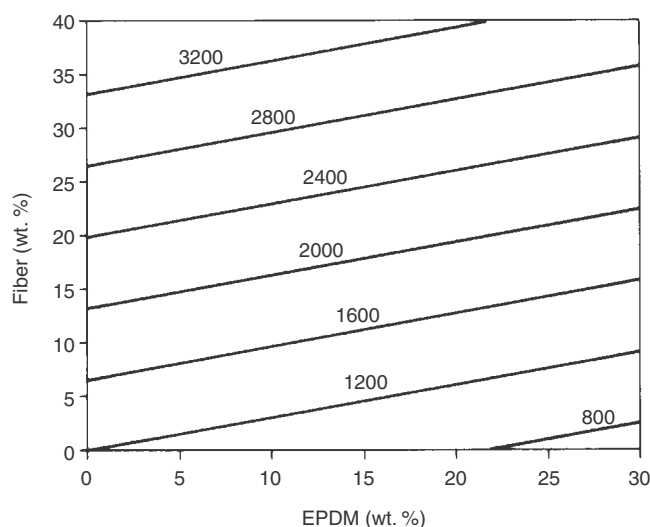


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THERMOSET MATRIX-NATURAL FIBRE COMPOSITES

In thermoset matrix composites, the fibres are impregnated with thermosetting resins and then exposed to high temperatures for curing. Usually lignocellulosic reinforcements are combined with phenolic, unsaturated polyester, phenol-formaldehyde, novolac type phenol-formaldehyde and epoxy resins to form composite materials such as those reported in Table 2. Reinforced thermosetting resin composites are structurally excellent materials but under some conditions can suffer environmental degradation, with a loss in mechanical performance (Mishra, Naik and Patil 2000). In order to minimize the effect of external agents, an insight on developing optimal hydrophobic morphology at the fibre/matrix interface is necessary prior to their use under wet/dry environments.

FIGURE 6. Iso-level curves of tensile Young's modulus (MPa) as a function of the composite composition.



(Reprinted with permission of John Wiley & Sons, Inc. from Biagiotti, J., M.A. López-Manchado, M. Arroyo and J.M. Kenny. 2003. Ternary composites based on PP-EPDM blends reinforced with flax fibers. Part II: Mechanical properties/morphology relationship. *Polymer Engineering and Science* 43(5): 1031.)

Taking this backdrop into account, a concerted effort is still necessary to establish and maintain a suitable level of quality and long term performance with respect to humidity, alternate wetting and drying, weathering, biological attack, fire and user habit conditions. Regarding the manufacturing technologies, natural fibre composites with thermosetting matrices have been manufactured using hand lay up, modified lay-up/press moulding, pultrusion, vacuum infusion and Resin Transfer Moulding (RTM), to achieve high performance components with equipment presently available and in order to eliminate large investments which could have been necessary for their application with natural fibres. However, a study and optimisation of these moulding technologies are required (for using of these new types of reinforcements). In addition to Sheet Moulding Compound (SMC) moulded plastics, Bulk Moulding Compound (BMC) composites with natural fibres with good mechanical properties can be produced (Saheb and Jog 1999; Bledzki and Gassan 1999). In the materials group of natural fibre reinforced thermosetting plastics, especially in the group of natural fibre rein-

TABLE 2. Thermosetting matrix-natural fibre composites.

Fiber	Thermosetting Matrix	References
Cellulose	Epoxy	(Low, Schmidt and Lane 1995)
Flax	Epoxy	(George, Ivens and Verpoest 1999; Hepworth et al. 2000; Lamy and Baley 2000; Gassan, Mildner and Bledzki 1999)
	MF	(Hagstrand and Oksman 2001)
Jute	Epoxy	(Gassan and Gutowski 2000; Datta, Basu and Banerjee 2002; Mishra et al. 2000; Tripathy et al. 2000; Costa and D'Almeida 1999; Gassan and Bledzki 1997; Gassan and Bledzki 1999)
	Polyester	(De Albuquerque et al. 2000; Dash et al. 1999; Saha et al. 1999; Mohanty and Misra 1995)
	Vinylester	(Ray, Sarkar and Bose 2002)
	Phenolic	(Singh, Gupta and Verma 2000)
Sisal	Epoxy	(Oksman et al. 2002; Rong et al. 2002)
	Polyester	(Singh, Gupta and Verma 1998; Fernandes et al. 2002; Aquino, D'almeida and Monteiro 2001)
	Novolac	(Mishra, Naik and Patil 2000)
Kenaf	Epoxy	(Zimmerman and Losure 1998)
	Phenolic	(Dansiri et al. 2002)
Hemp	Phenolic	(Richardson and Zhang 2000)
	Epoxy	(Hughes et al. 2002; Hepworth et al. 2000)
	Polyester	(Hughes, Hill and Hague 2002; Sebe et al. 2002)
	Vinylester	(Thielemans et al. 2002)
Cotton	Polyester	(Mwaikambo and Bisanda 1999)
Coir	Polyester	(Khalil et al. 2000; Samal et al. 1995; Hill and Khalil 2000)
Banana	Polyester	(Pothan, George and Thomas 2002; Zhu, Tobias and Coutts 1995)
	Phenolic	(Joseph et al. 2002)
Bagasse	Phenolic	(Zarate, Aranguren and Reboredo 2000; Patil et al. 2000)
Bamboo	Epoxy	(Rajulu, Baksh and Reddy 1998)
	Polyester	(Deshpande et al. 2000; Jain, Kumar and Jindal 1992)
Pineapple	Polyester	(Uma Devi, Bhagawan and Thomas 1997)
	Phenolic	(Mangal et al. 2003)
Wood flour	Polyester	(Marcovich, Reboredo and Aranguren 1998; Doss et al. 1991; Marcovich, Reboredo and Aranguren 2001)
Oil palm	Polyester	(Sreekala, Kumaran and Thomas 1997; Khalil et al. 2001)
	Phenolic	(Agarwal et al. 2000; Sreekala et al. 2002)

forced epoxies, there is little knowledge about the influence of suitable coupling agents on the parameters of composites in comparison to natural fibres and reinforced thermoplastics. George, Ivens and Verpoest (1999) investigated the influence of various fibre parameters such as lignin content, pectin content and degree of polymerisation on the properties of flax reinforced epoxy. Fibre surface modifications such as alkali, silane and isocyanate treatment were also studied, with significant results in terms of the improved mechanical properties of the composites. In this area, Gassan and Bledzki (1999) reported the possibility of improving the mechanical properties of jute/epoxy composites by means of treating fibres with alkali. Results in terms of better tensile and fracture behaviour were collected, reporting an optimisation of the NaOH treatment with different alkali concentrations and shrinkages. Ray, Sarkar and Bose (2002) also studied the alkali treatment of jute fibre in vinylester resin matrix composites, in particular, the impact fatigue behaviour with the final conclusion that in addition to fibre reinforcement, the defect concentration of the fibres treated for less time played an important role in the improvement of the fatigue behaviour of the composites under impact loading. Detailed research on the effects of various chemical treatments designed to allow epoxy resin to penetrate the plant fibre cell walls was performed by D.G. Hepworth et al. (2000), where a treatment that involved swelling the plant cell walls with urea solution, washing out the excess urea and then replacing the water with alcohol in a graded series was developed. The improved adhesion between the resin and the fibre facilitated by the urea-alcohol treatment suggests that this treatment could be a useful method for controlling the variability of natural fibre composites while retaining their desired mechanical properties. Gassan and Gutowski (2000) also studied the effect of corona discharge and UV treatment on the properties of jute/epoxy composites. The results confirmed that both the treatments lead to a significant increase in the polar component of the free surface energy but in the case of UV oxidation, a balance between the increased polarity of the fibre surface and a decrease in fibre strength is necessary.

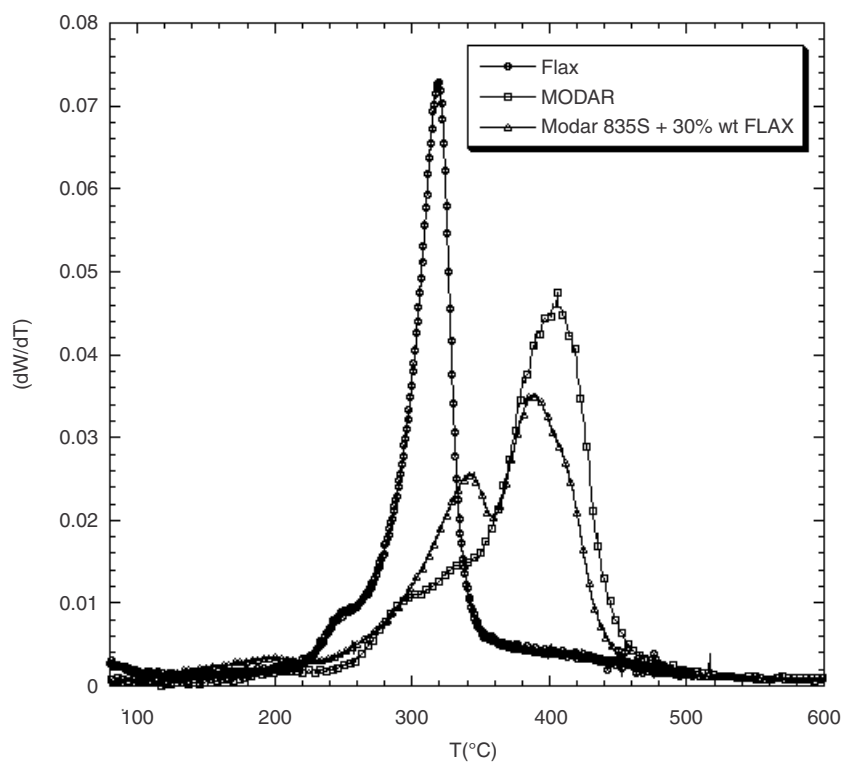
Other investigations were performed with melanine-formaldehyde resins, successfully used together with cellulose containing materials such as wood flour, paper pulp, cotton or α -cellulose in a variety of applications. Hagstrand and Oksman (2001) compared the mechanical performance of non-woven flax fibre mat reinforced and particle filled melanine-formaldehyde (MF) composites, processed by means of compression moulding with similar MF composites reinforced with glass fi-

bre. Even if there was a negative effect on tensile performance, the difference was relatively small and an advantage compared to the commercial glass fibre reinforced grades is lower damage on straining. Even if the more costly epoxy resins could be used to obtain higher quality in the product, unsaturated polyesters seem to be the most popular resins, for their low cost and adaptability for large composite structures. There are many articles currently in literature regarding this type of thermosetting matrix. The effect of the incorporation of sisal treated with organotitanate, zirconate, silane and methacrylamide (Singh, Gupta and Verma 1996), coir with alkalization and graft polymerization with methylmethacrylate (MMA) (Rout et al. 2001) and pineapple leaf fibre (Uma Devi, Bhagawan and Thomas 1997) have been studied and the results confirmed the useful selection of fibres and treatment for improved strength in composites. The effect of acetylation and coupling agent treatments on the biological degradation of plant fibre reinforced polyester was investigated and the superior bio-resistance of acetylated fibre with respect to silane treated fibre was found. Marcovich, Reboredo and Aranguren (1998, 2001) extensively investigated the mechanical behaviour of unsaturated polyester resins reinforced with untreated and maleic anhydride wood-flour as well as the role of moisture content in the final results. With the same intention in mind, studies on the thermodegradative behaviour and moisture absorption of acrylic resin reinforced both with flax and glass fibre were performed (Puglia 2001) and the results have shown good behaviour of hybrid composites with respect to glass reinforced samples. By means of the deconvolution of the degradation peaks of the complex TGA thermograms, obtained for degradable composites, with a powerful tool for the kinetic analysis of their thermal stability, the thermal stability of the acrylic matrix slightly affected by the presence of the flax fibres was observed and it is a good requirement for a fire retardant acrylic resin usually used for fire protection elements (Figure 7).

BIODEGRADABLE MATRIX-NATURAL FIBRE COMPOSITES

Increasing environmental awareness, social concern and decreasing landfill space has led to the search for biodegradable plastics as an alternative to traditional plastics. Biodegradable plastics such as poly(lactic acid) cellulose esters, starch plastics, poly(ϵ -caprolactone) and aliphatic polyesters/co-polyesters are now emerging as potential replacements

FIGURE 7. DTG curve ($\mu\text{g}^\circ\text{C}^{-1}$), at $10^\circ\text{C}/\text{min}$, of Modar[®] acrylic resin and 30% wt flax composite.



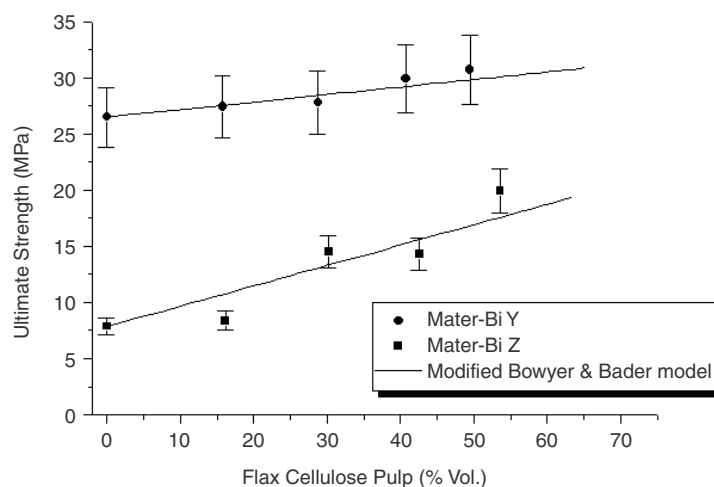
(Source: Puglia, D. 2001. A Comparison of the Effects of Flax and Glass Fibres on the Mechanical Properties of Thermosetting Matrix Composites. *Proceedings of 3rd National Conference INSTM*. Trento, June 18-20.)

for non-biodegradable traditional plastics in certain applications. The realization of a biocomposite, a fully biodegradable material, is actually possible by combining a biodegradable matrix with natural fibres. The so-called biopolymers, which can be used as matrices in this new kind of composite, are normally used in the packaging industry and for other applications with minor strength requirements. Some of the modern applications of this class of material involve the realization of tubes, car doors, interior panelling, sandwich plates, etc. (Herrmann, Nickel and Riedel 1998). Although these materials can offer very interesting properties, only a few studies have been reported (Cyras et al. 2001; Iannace, Nocilla and Nicolais 1999). The latter could probably be attributed to

the ratio performance/cost of the biocomposites which is still low in comparison to traditional polymer based composites. However, some interesting results were presented by Gatenholm, Kubat and Mathiasson (1992) that observed good properties for cellulose-polyhydroxybutyrate systems. In fact, the incorporation of cellulose seems to greatly improve the mechanical properties of the bacteria-produced polyester. The same kind of matrix blended with jute fibres was studied by Mohanty, Khan and Hinrichsen (2000a) who analysed the influence of the chemical treatments of the reinforcement on the final characteristics of the biocomposite. The suitability of existing biodegradable plastics as matrix polymers in biocomposites has been studied, in order to find emerging applications for such biodegradable composites. Extrusion processing followed by compression moulding of biocomposites from various surface-modified natural fibres (Jute, Kenaf, Hemp, Sisal, Henequen) and commercial biodegradable plastics has been investigated (Mohanty, Khan and Hinrichsen 2000b). Studies on the mechanical performance of biodegradable systems were conducted by Puglia, Tomassucci and Kenny (2002). The mechanical and thermal properties of composites based on different grades of Mater-Bi and flax cellulose pulp were studied. The results showed that good mechanical properties of pulp fibre biodegradable composites with Mater-Bi matrices can be obtained with strength values higher than the equivalent glass fibre composites, suggesting that natural fibres are more compatible with the matrix (Figure 8). Furthermore, water absorption had an important influence on the mechanical behaviour of the composites and the main effect of water has been associated to the negative influence of moisture on fibre-matrix adhesion (Figure 9). The thermal stability of the Mater-Bi composites was also investigated and the effect of the cellulose fibres on the shift of the degradation peaks to higher temperatures and a slight reduction in the activation energy of the peak associated to cellulose decomposition (Figure 10), were demonstrated.

Cyras, Kenny and Vazquez (2001) and Cyras, Martucci, Iannace and Vazquez (2002) investigated the processing and properties of biodegradable composites based on Mater-Bi and sisal fibres. In particular, calorimetric studies were performed to develop a kinetic model for the crystallisation behaviour and the influence of processing conditions on fragmentation and disgregation of the fibre during composite extrusion. Correlating the reduction of fibre size and mixing time, an alkaline treatment was performed in order to improve the adhesion and the com-

FIGURE 8. Fitting and experimental values of ultimate strength for Mater-Bi Z and Mater-Bi Y with flax cellulose pulp.



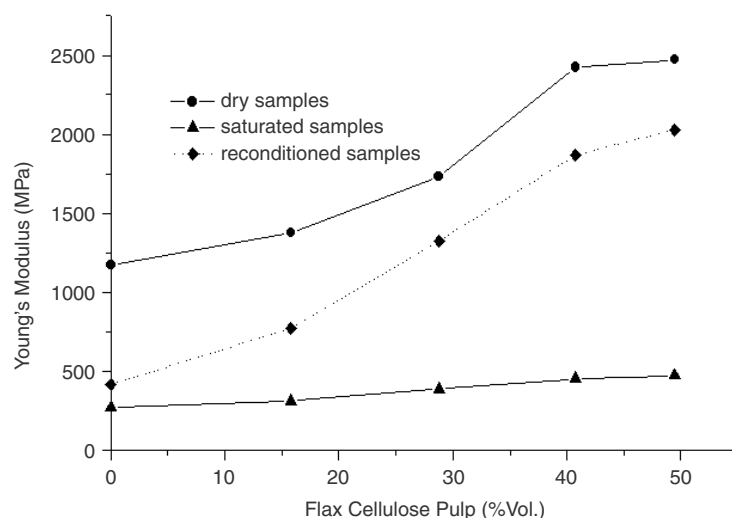
(Reprinted with permission of John Wiley & Sons, Ltd. from Puglia, D., A. Tomassucci, J.M. Kenny. 2003. Processing, properties and stability of biodegradable composites based on Mater-Bi® and cellulose fiber. *Polymers for Advanced Technologies* 14(11-12): 752.)

patibility of the fibre with the matrix, without affecting the final dimensions of the fibre after enough mixing time.

TREATMENTS FOR NATURAL FIBRES

It is important to understand the properties of the components of the cell wall and their contributions to fibre properties, in order to appreciate how lignocellulosic fibre can be used in property-enhanced applications. Dimensional instability, flammability, biodegradability and degradation caused by acids, bases and ultraviolet radiation are a result of the environmental attempt to convert natural composites back to their basic building blocks (carbon dioxide and water). Fibres biologically degrade because organisms recognize the carbohydrate polymers (mainly hemicelluloses) in the cell wall and have very specific enzyme systems capable of hydrolysing these polymers into digestible units. Moreover, fibres change dimensions with a change in the moisture content because the cell wall polymers contain hydroxyl and other oxygen-containing groups that attract moisture through hydrogen bonding. Hemicelluloses

FIGURE 9. Comparison of Young's Modulus values of Mater-Bi Y-flax cellulose pulp composites for dry, saturated and reconditioned samples.

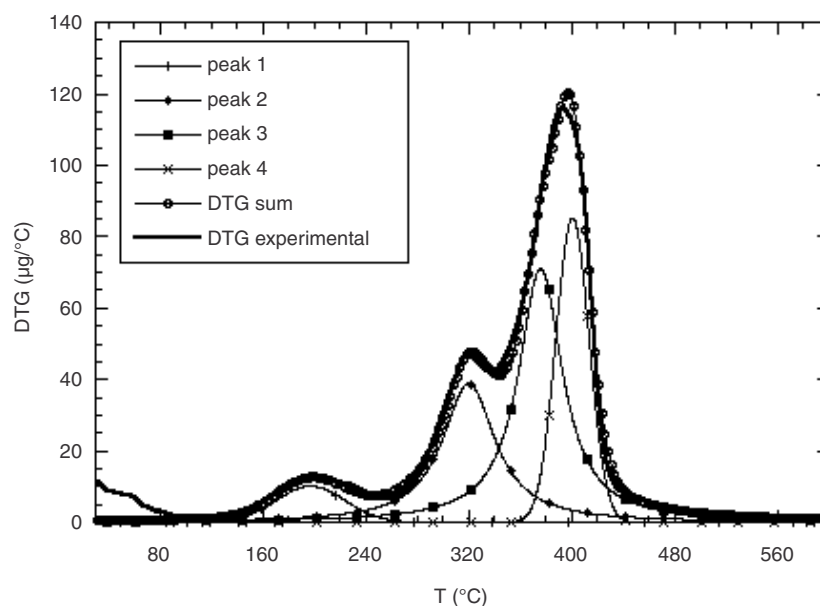


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are mainly responsible for moisture absorption, but the accessible cellulose, non-crystalline cellulose, lignin and surface of the crystalline cellulose also play major roles. Strength is also lost as the cellulose polymer undergoes degradation through oxidation, hydrolysis and dehydration reactions. Photochemical degradation that take place primarily in the lignin component should be considered (cellulose is much less susceptible to ultraviolet light degradation). After the lignin has degraded, the poorly bonded carbohydrate-rich fibres erode easily from the surface, which exposes new lignin to further degradative reactions. The lignin component contributes to char formation and the charred layer helps insulate the composite from further thermal degradation.

Another important factor that affects processing and the ultimate properties of NFCs is the interfacial adhesion between the lignocellulosic and the matrix. Due to the lack of adhesion between the components, the composites exhibit poor mechanical properties. Good interfacial adhesion for composites containing natural fibres and fillers is achieved in general by means of one of the following ways: fibre modification, the use of interface-active additives or matrix modification. Fibre modi-

FIGURE 10. Deconvolution peaks for the DTG ($\mu\text{g}^\circ\text{C}^{-1}$), at $10^\circ\text{C}/\text{min}$, of Mater-Bi Z with 30% of flax cellulose pulp.



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fication involves grafting functional groups on the lignocellulosic fibres or coating fibres with additives that carry suitable functional groups, in order to make the fibre surface more compatible/reactive with the matrix material. The various reactive species that can be used for fibre modification include one or more of the following—acetic anhydride, n-alkyl isocyanates, styrene, maleic anhydride and silanes.

The second method for promoting interfacial adhesion involves the use of additives that act as coupling agents. Polyesteramide polyol, titanates, chemicals based on trichloro-s-triazine, poly[methylene(poly phenyl isocyanate)], maleic anhydride and silanes can be employed for this purpose. The use of interface-active additives is the least effective method of improving the adhesion characteristics, while matrix modification is a quick, effective method. In contrast, fibre modification, which is also an effective method, mostly involves solvent-based processes, which are slower and not very attractive environmentally and from a commercial point of view.

Chemical Treatments of Natural Fibres

Since the properties of the fibres result from the chemistry of the cell wall components, the basic properties of a fibre can be changed by modifying the basic chemistry of cell wall polymers. Dimensional stability can be greatly improved by bulking the fibre cell wall either with simple bonded chemicals or by impregnation with water-soluble polymers. For example, acetylation of cell wall polymers using acetic anhydride produces a fibre composite with greatly improved dimensional stability and biological resistance. The same level of stabilization can also be achieved by using water-soluble phenol formaldehyde polymers followed by curing.

The biological resistance of fibre-based materials can be improved using several methods, for example, bonding chemicals to cell wall polymers increases resistance due to the lowering of the equilibrium moisture content and restraining the points needed for microorganism attack and by changing the conformation and configuration requirements of the enzyme-substrate reactions. Toxic chemicals can also be added to the composite to stop biological attack. This is the basis for the wood-preservation industry. Resistance to ultraviolet radiation can be improved by adding polymers to the cell matrix, in order to prevent the water-leaching of undegraded carbohydrate polymers. Fire retardants can be bonded to the fibre cell wall to greatly improve fire performance. Soluble inorganic salts or polymers containing nitrogen and phosphorus can also be used. These chemicals are the basis of the fire-retardant wood-treating industry.

The strength properties of fibre-based composites can be greatly improved in several ways. The finished composites can be impregnated with a monomer and polymerised in situ or impregnated with a pre-formed polymer. In most cases, the polymer does not enter the cell wall and is located in the cell lumen. By using this technology, their mechanical properties can be greatly enhanced. For example, composites impregnated with acrylates, methacrylate, epoxy or melamine monomers and polymerised to weight gain levels of 60% to 100%, show increases (compared to untreated controls) in density of 60% to 150%, in compression strength of 60% to 250%, and in tangential hardness of 120% to 400%. Static bending tests show increases in the modulus of elasticity of 25%, modulus of rupture of 80%, fibre stress at a proportional limit of 80%, work to proportional limit of 150% and work to maximum load of 80% and at the same time a decrease in permeability of 200% to 1200%.

Many chemical-reaction systems have been published for the modification of agro-fibres.

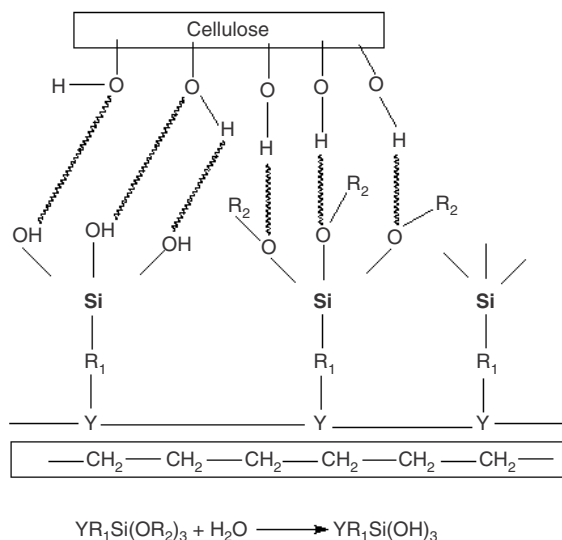
Silane: Organosilanes are the main group of coupling agents developed to couple virtually any polymer to mineral fibres. The organofunctional group in the coupling agent causes the reaction with the polymer. This could be copolymerisation and/or the formation of an interpenetrating network (Zafeiropoulos et al. 2002; Gassan 2002; Bisanda and Ansell 1991; Gassan and Bledzki 1999). This curing reaction of a silane treated substrate enhances the wetting of the resin. Analogous to glass fibres, silanes are used as coupling agents for natural fibre polymer composites. The theories used for silane treatment of natural fibres are contradictory and therefore further studies are necessary (Figure 11).

Isocyanate: The mechanical properties of composites reinforced with wood-fibres can be improved by an isocyanate treatment of those cellulose fibres or the polymer matrix. In the comparison between both methods, that is, the treatment with silanes and the treatment with isocyanates, the latter is more effective. The method consists of washing and drying fibre which has previously been treated with NaOH, whereafter the fibres are soaked with CCl_4 in a glass vessel (Figure 12). Toluendiisocyanate (TDIC) with 0.1% of a dibutyl tin dilaurate (DBTDL) catalyst is added to the vessel and stirred well, the reaction continues for 1 h at 50°C with continuous stirring. Fibres are then purified by refluxing with acetone for 5 h and finally are washed with distilled water and oven dried at 100°C (Vazquez, Dominguez and Kenny 1999; Bledzki, Rehmane and Gassan 1996; Raj, Kokta, and Daneault 1989).

Acetylation: This process involves the soaking of the plant fibre in acetic anhydride with or without an acid catalyst (Figure 13). Since acetic acid does not react sufficiently with cellulose, acetic anhydride is preferred. However, because acetic anhydride is not a good swelling agent for cellulose and in order to accelerate the reaction, cellulose materials are first soaked in acetic acid and subsequently treated with anhydride at higher temperatures for a period of 1 and 3 h. In natural fibre reinforced composites, acetylation of the hydroxy group will swell the plant fibre cell wall, greatly reducing the hygroscopic nature of the cellulose fibre. This will consequently result in the dimensional stability of the composites, as any absorbed water will not cause further swelling or shrinkage of the composite material (Rana et al. 1997).

Benzoylation: The enhanced strength of composites treated with benzoyl peroxide may arise from crosslinking or grafting reactions (Evans et al. 2002; Kumar et al. 2000). The presence of peroxides also introduces polarity in the host polymer, thereby increasing adhesion at the

FIGURE 11. Scheme of the silanization process.



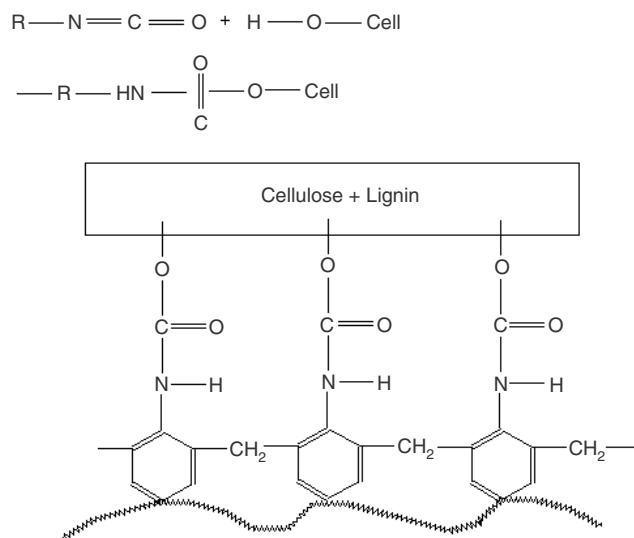
(Reprinted with permission of John Wiley & Sons, Inc. from Bledzki, A.K., S. Reihmane, J. Gassan. 1996. Properties and modification methods for vegetable fibres for natural fibre composites. *Journal of Applied Polymer Science* 59(8): 1329-1336.)

polymer/fibre interface. The degree of dispersion of the peroxide or its preferential localization at the fibre/matrix interface is a factor in the fibre/matrix modification (Joseph, Thomas and Paul 1997). Benzoyl-peroxide-initiated radicals may attack the cellulose backbone to generate cellulose radicals with subsequent promotion of the grafting of cellulose on the polymeric matrix. It is also possible that the presence of polar groups on the cellulose fibres modifies the reactive species generated, thus facilitating the cross-linking of the polymeric matrix (Figure 14).

Stearic acid: The aim of the stearic acid treatment is the reaction of the hydroxyl group of the fibre with the stearic acid group, to hydrophobise the fibre's surface, yielding better compatibility. This kind of treatment does not significantly affect fibre strength for either green or dew retted flax at low reaction times (Zafeiropoulos, Baillie and Matthews 2001; Eichhorn et al. 2001).

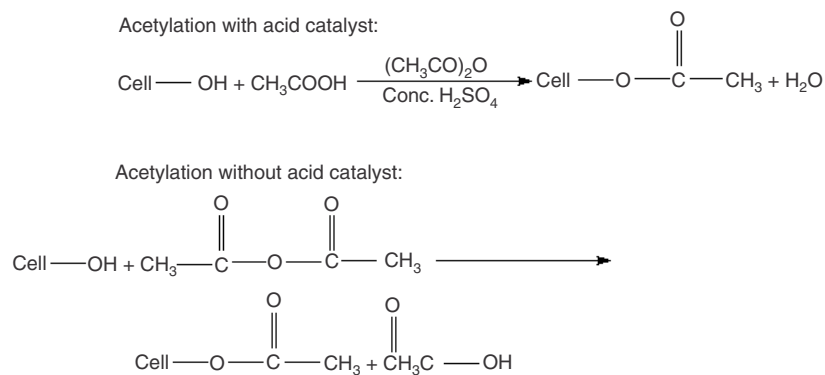
Graft copolymerisation: Initiation by free radicals is one of the most common methods used for the grafting of vinylic monomers onto cellulose. These free radicals are produced as a result of a reaction of the cellulosic chain in a redox system. In this reaction, the oxidation of the

FIGURE 12. Scheme of the isocyanate process.



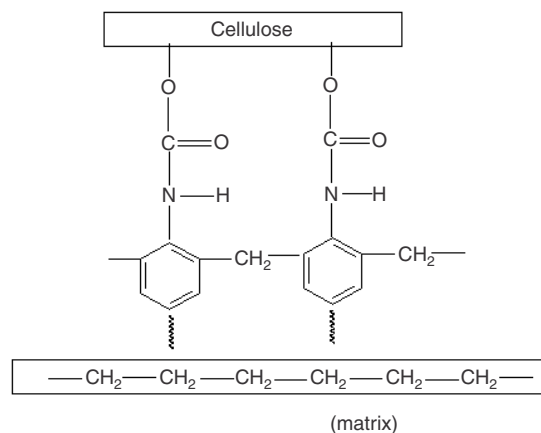
(Reprinted with permission of John Wiley & Sons, Inc. from Maldas, D., B.V. Kokta and C. Daneault. 1989. Influence of coupling agents and treatment on the mechanical properties of cellulose fibre/polystyrene composites. *Journal of Applied Polymer Science* 37: 751-775.)

FIGURE 13. Scheme of the acetylation process.



(Reprinted with permission of Wiley-VCH from Mwaikambo, L.Y., and M.P. Ansell. 1999. The effect of chemical treatment on the properties of hemp, sisal, jute and kapok for composite reinforcement. *Die Angewandte Makromolekulare Chemie* 272(1999): 108-116.)

FIGURE 14. Scheme of the benzoylation process.

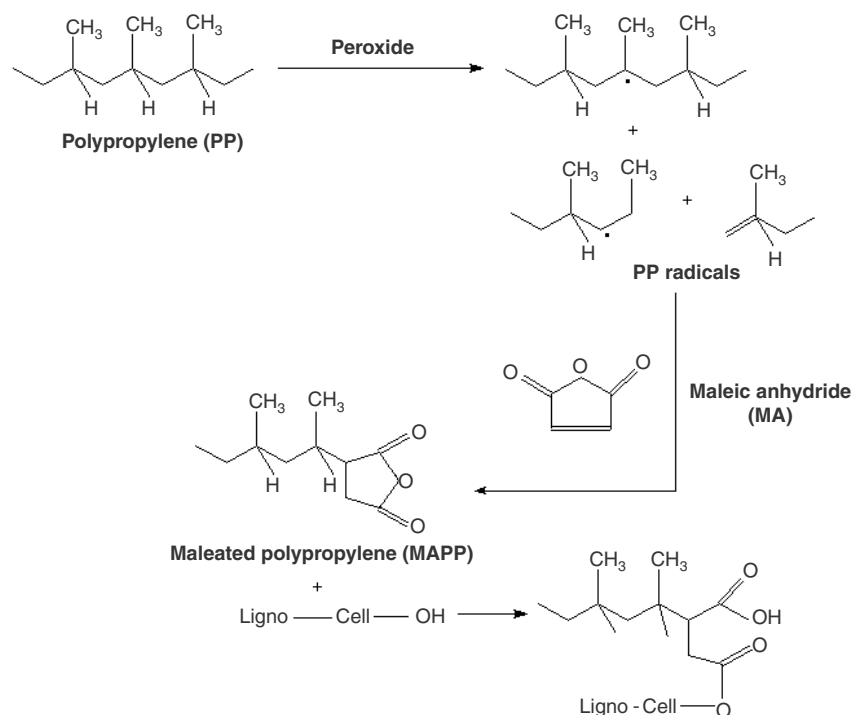


(Reprinted with permission of John Wiley & Sons, Inc. from Maldas, D., B.V. Kokta and C. Daneault. 1989. Influence of coupling agents and treatment on the mechanical properties of cellulose fibre/polystyrene composites. *Journal of Applied Polymer Science* 37:751-775.)

anhydroglucose units occurs along the cellulosic chains and macro-cellulosic radicals are generated on the surface of the fibre (Figure 15). When the reaction is carried out in the presence of a monomer, the oxidation or depolymerisation is lower than in the case of cellulose alone. In this case, the macro-cellulose radicals generated by the initiator are used to carry out the graft copolymerisation of the polymer and the degradation of the cellulose is reduced. When the cellulose molecule cracks and radicals are formed, the radicals' sites are treated with a suitable solution (compatible with the polymeric matrix), e.g., vinyl monomer, acrylonitrile, methyl methacrylate and polystyrene. The resulting copolymer possesses properties that are characteristic of both a fibrous cellulose and a grafted polymer (Pavlock 1992).

Preimpregnation: A better combination of fibre and polymer is achieved by the impregnation of reinforcing fabrics with polymer matrices compatible with the polymer. For this purpose, polymer solutions or dispersions of low viscosity are used. The most promising methods for improving the specific properties of wood cell material is chemical impregnation under vacuum or pressure. Compounds that are highly reactive to the hydroxyl groups of cellulose, hemicellulose and lignin components of wood include epoxides, isocyanates, anhydrides, lactones and diols. Chemical impregnation also has the potential of reduc-

FIGURE 15. Scheme of the Graft copolymerisation.



(Reprinted with permission of John Wiley & Sons, Inc. from Karnani, R., M. Krishnan, and R. Narayan. 1997. Biofibre-reinforced polypropylene composites. *Polymer Engineering and Science* 37(2): 476-483.)

ing the susceptibility of wood to biological degradation. Another widely studied system is the crosslinking of wood via impregnation with formaldehyde in the presence of an acid catalyst (Mathias et al. 1991).

Duralin: A proven treatment that replaces retting and also removes hemicellulose and lignin to a certain extent is the Duralin method developed by Ceres b.v. (Wageningen, Netherlands). This novel treatment of vegetable fibres reduces moisture sensitivity in a simple process that only uses energy and water and involves three steps, viz. hydrothermolysis, drying and curing. The raw material for the Duralin process applied to flax is green rippled flax straw. This eliminates the need for traditional dew-retting, where the freshly harvested flax stems lay on the field for about four weeks. The Duralin process reduces moisture absorption and biological degradation, the fibre yield is higher than af-

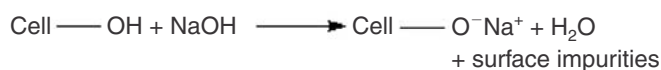
ter dew-retting and the shives can be used for making a water-proof particle board. The three main reasons for the reduced water uptake after the Duralin treatment are the extraction of hemicellulose, the network formation through crosslinking of the degradation products of hemicellulose and lignin and the increased crystallinity of cellulose (Quillin, Caulfield and Koutsky 1992).

Other minor treatments, such as, the use of potassium permanganate (Paul, Joseph and Thomas 1997) and acrylic acid (Karlsson et al. 1998) can be used to improve the properties of the fibre, but little research has been done to collect data.

Physical Treatment of Natural Fibres

Mergerisation: The standard definition for mergerisation proposed by ASTM D1695 (Standard Terminology of Cellulose and Cellulose Derivatives) is “the process of subjecting a vegetable fibre to the action of a fairly concentrated aqueous solution of a strong base so as to produce great swelling with resultant changes in the fine structure, dimension, morphology and mechanical properties.” Sodium hydroxide (NaOH) is the most commonly used chemical for bleaching and/or cleaning the surface of plant fibres and it also changes the fine structure of native cellulose I to cellulose II, with de-polymerisation and the production of short length crystallites (Figure 16). The influence of parameters such as alkali concentration, temperature, time, fibre pre-treatment and fibre tension were studied. It has been demonstrated that the basic fibre properties (strength, modulus, elongation at break) could be varied over a wide range by a suitable choice of mergerisation parameters. The influence of mergerisation on the static and dynamic parameters of a unidirectionally reinforced epoxy resin test composite is much stronger than the influence on the measured fibre characteristics. It has been found that the flexural modulus of the specimen containing mergerised fibres was doubled and its fatigue resistance significantly improved (Bledzki et al. 1999).

FIGURE 16. Scheme of the mergerisation process.



(Reprinted with permission of John Wiley & Sons, Inc. from Mwaikambo, L.Y., and M.P. Ansell. 2002. Chemical modification of hemp, sisal, jute, and kapok fibres by alkalization. *Journal of Applied Polymer Science* 84(12): 2222-2234.)

Corona, cold plasma: Corona treatment is one of the most interesting techniques for surface oxidation activation. This process changes the surface energy of the cellulose fibres and in the case of wood, surface activation increases the amount of aldehyde groups. The same effects are obtained with cold plasma treatment. Depending on the type and nature of the gases used, a variety of surface modifications can be achieved. Surface crosslinking could be introduced, surface energy could be increased or decreased and reactive free radicals and groups could be produced. Electrical discharge methods are known to be very effective for “non-active” polymer substrates such as PS, PE and PP (Sakata et al. 1993).

APPLICATIONS OF NATURAL FIBRE COMPOSITES

Whereas wood-fibre composites compete largely with talc- or mica-filled PP in automotive parts, natural long-fibre composites are aiming to compete with glass-fibre composites. Leading contenders for wood and glass replacement are bast fibres from flax, hemp, kenaf and jute. These fibres are carded or air laid and then usually needle-punched into mat form. Leaf fibres from sisal as well as abaca and banana leaves are also being explored. Natural fibres, which traditionally were relegated to the world of thermosets, are now rapidly becoming one of the fastest-growing additives for thermoplastics as well. For centuries they have been made into baskets, clothing, sacks, ropes and rugs. In recent years, the market has moved towards thermoplastics and demand has taken off for such products as decking, window and door profiles, fencing, siding, railings, furniture, flooring, marine components and automotive interior parts. Now plant-derived natural fibres of kenaf, hemp, flax, jute and sisal are making their way into components of cars. In the last decade, natural-fibre composites of thermoplastics and thermosets have been embraced by European carmakers for door panels, seat backs, headliners, package trays, dashboards and trunk liners. Technology for using natural-fibre composites in interior trim is being cultivated by Tier I and II automotive suppliers, typically in partnership with producers of natural-fibre-based mat materials. Most developmental work is focused on polypropylene-based composites produced by compression moulding or thermoforming extruded sheets or commingled mats of PP and plant fibres (Kafus Bio-Composites produced non-woven mats of kenaf and PP, Cambridge Industries made flax/PP composites for heavy trucks and flax/PP composites for rear-shelf trim panels).

Visteon aims to use post-industrial and post-consumer recycled PP and recycled nylon 66. It is also working mostly with kenaf and hemp blends and PP or TPO. The blending of different natural fibres has been studied as well and the results confirmed the optimal physical properties obtained.

In this scenario, most industries in the field of natural composites have developed active programmes with auto-parts makers, in particular, as technologies installed mat-making lines at its plant in Brussels, using hemp, kenaf, flax, sisal and abaca. Cargill Ltd. makes Durafibre from the bast of the flax plant and also makes fine-particle-size Durafill from the shive (inner core) of flax, suitable for compounding into thermoset BMC or SMC, as well as thermoplastics for extrusion or moulding. With the same intention, Dexter Corp.'s Nonwoven Materials Div. makes sisal-based nonwoven mats in roll form for compression moulding or thermoforming (a wet-form nonwoven process similar to papermaking is used for these mats, which also contain some wood pulp, polyester fibre and a bit of EVA binder) and Georgia Composites' developed sisal/PP composite that can be used to boost the performance of "Woodstock"-type materials (50/50 PP and wood flour). Furthermore, Global Resource Technologies has compounded virgin and recycled PP and HDPE with kenaf, jute, hemp, sisal, flax, coconut fibre and wood pulp since 1996. Also Kenex Hemp Ltd. has produced mats from hemp or blends of hemp with flax or jute and virgin or recycled PE or PP. Pinnacle Technology teamed up with the U.S. Dept. of Agriculture's Forest Products Laboratory to develop and commercialise an "agro-plastic" manufacturing process using abundant wheat straw. The product is ground to a fine powder with an aspect ratio near 1. Pinnacle compounded 50% wheat straw with PP or HDPE for injection moulding and extrusion. Wheat straw reportedly provides much lower density and significantly higher tensile and flexural strengths, flex modulus and higher distortion temperature (HDT) than mineral-filled polyolefins.

From the point of view of processing, in the case of non-woven mats combining PP fibres and natural fibres in a needle-punched sandwich, the mat is generally heated in an oven until soft and then transferred to a cold press where it is compression moulded. However, new technologies such as low-pressure compression moulding technology, considered for applications like instrument-panel topper pads or door-trim panels in which a preheated natural-fibre/PP mat is in-mould laminated with a TPO coverstock have been considered. CPI has offered to license its patented process for in-line compounding and compression mould-

ing of glass- or natural-fibre composites working with PP composites of flax from Durafibre or kenaf from Kafus.

Automotive Use and Market Orientation of Natural Fibre Composites

Natural fibre composites are used at different levels in the European automotive market than the U.S. automotive market. Although they are not considered mature materials, most of the natural fibre composites manufactured in Europe are being used by the automotive industry. In Europe, the driving force for using natural fibres in the automotive industry is environmentally related. This is different from the issue of material recyclability. There are some projects in Germany and Italy (Contrafatto et al. 1998) that look at recyclability, but the consumer would not notice a recycled part in an automobile, although the average European consumer is concerned about environmental issues. As a result, the environmental driving force stems partly from European regulations. The effect is two-fold. Natural fibre composites in the automotive industry both reduce material waste and increase fuel efficiency. For example, there are regulations in Italy that require the reduction of dumping waste materials. Only materials that cannot be recycled in any other way by the end of 2002 will be land-filled. This means that no organic materials, plastics, wood or metals will be allowed in landfills resulting in recycling the majority of waste. The main question for the automotive industry is what to do with the glass fibres of a glass fibre-plastic composite after its life cycle. This leads to a clear advantage for using natural fibre composites, which can be recycled. The second environmental benefit is to reduce fuel emissions. Europe is committed to following the Kyoto concepts. By 2005 there is a commitment to reduce fuel consumption in Europe by 50 percent. If lighter materials are used in the automotive industry, fuel efficiencies rise, making it easier to meet this goal. In the current situation, it is estimated that no more than 50 kg (110 lb) of natural fibres can be used in a car. This corresponds to a reduction of about 10 kg (22 lb) if glass fibre composites are replaced with natural fibre composites in an automobile. If the weight of a car can be reduced by 10 to 20 kg (22 to 44 lb), the effect on the environment will be significant. European car production is 55 million cars annually. This results in approximately 1 million metric tons of natural fibre materials targeted to be consumed by the automotive industry. However, there are also disadvantages of using natural fibre composites in the automotive industry. Fibres are produced all over the world and

may have different histories, geographic production, pre-treatment and providers. The prices of natural fibres have increased since 1999, while the price of glass fibres has remained stable. A cost analysis should be carefully undertaken using accurate fibre prices. Flax is typically more expensive than glass fibres due to competition between the use of flax in the textile industry and flax in the reinforced composite industry. There are composites used in the automotive industry with wood flour used as a filler as well. Woodstock and Polywood are two familiar materials that are widely used. These materials are good for automotive panels where properties are less of a concern, but the industry needs to distinguish between applications where mechanical properties are not so important and applications where properties are more important so that natural fibre composites can replace some glass fibres composites. The properties of natural fibre composites will not reach those of glass fibre composites, but will probably reach properties on the order of 50 to 70 percent of glass fibre composites. Although compression moulding has been the main process in automotive applications, we have the possibility of refining processing. Compression moulding is simple and can take advantage of non-woven mats that can be impregnated with thermoplastics or thermosets. The automotive industry has shown a lot of interest in injection mould natural fibre composites. Many automotive companies are looking at the possibility of injection moulding for producing lower cost interior or exterior panels. Based on general experience, the most difficult processing method is compounding natural fibres. Germany is a leader in the use of natural fibre composites. The German auto-manufacturers, Mercedes, BMW, Audi and Volkswagen have taken the initiative to introduce natural fibre composites for interior and exterior applications. The first commercial example is the inner door panel of the 1999 S-Class Mercedes-Benz, made in Germany, of 35% Baypreg F semi-rigid (PUR) elastomer from Bayer and 65% of a blend of flax, hemp and sisal. The 2-mm-thick door panel was made by the new NafpurTec process at Bayeros Hennecke Machinery Unit, whereby a robot placed a natural-fibre mat in an open mould and a second robot poured PUR over it before the mould was closed. The interest therein started almost 10 years ago, with actual development beginning 5 years ago. It should be emphasized that luxury automotive manufacturers are on board which could be seen as evidence that natural fibre composites are being used for environmental needs and not to lower costs. Other auto-manufacturers in Europe, such as, Fiat are following this trend. Fiat has developed a prototype car, Ecobasic, a small car that is expected to use 3 L (0.79 gal.) of fuel to travel 100 km (62 miles). Fiat

has been rethinking the meaning of a Class-A finish for aesthetical appearance. Achieving a Class-A finish has long been a concern of automotive makers, but the question why cars need a Class-A finish has been raised. In this sense, Fiat is working at eliminating the paint shop, with the production of Ecobasic, by manufacturing a car with pigmented plastic panels that do not need painting. If a scratch were to occur through the thickness, the difference of colour would not show. The plastic panels can be replaced quickly and cheaply following an accident. The Ecobasic prototype has many natural fibre composites incorporated in it, mainly to save weight. It is expected to weigh just 750 kg (1,650 lb). Another innovation is that the engine compartment is fully enclosed. The consumer need only worry about filling fluids through designated holes in the car, while a mechanic can work on the engine if needed.

ECOFINA EUROPEAN PROJECT

In order to develop high performance natural fibre composites, the automotive industry and research institutions in Europe are collaborating in European Commission FP5 GROWTH Projects. In particular, the 'ECOFINA' Project addresses the substitution of mineral fillers and fibres presently used in automotive parts made with organic matrices, with polymeric matrix composites based on annually renewable natural fibres. This substitution will allow the production of vehicle components with potentially complete recyclability. Moreover, the lower weight attainable due to the low density of the natural fibres will allow a lowering of gas emission in vehicles, enhancing quality of life. Complementarily, the use of European natural fibres, extracted from related-plants, has become a very interesting alternative for the agriculture sector in which, an increase in economic activities and employment could be expected, when these natural fibre composite materials are applied in mass production sectors, like in the processing of vehicle parts. The final objective of this Project includes the standardization of at least one processing technology to produce natural fibres of constant quality for natural fibre composites and the development of procedures to increase the compatibility of resins and fibres. Long-term objectives are the development of specific recycling routes for natural fibre composites, the creation of a database which includes the properties of natural fibres, thermosetting and thermoplastic resins and natural fibre-based compos-

ites and technical, economical and life cycle assessment of the production and recycling of parts of natural fibre-based composites (Kenny 2001).

The actual final goal of the Project is the production of prototypes of automotive parts, illustrated in Figures 17a-b, in thermoplastic and thermosetting matrix NFCs, for the end users of the Project: C.R.F. (Torino,

FIGURE 17. ECOFINA Project prototypes: (a) inner door pocket, and (b) spare wheel well.

a.



b.



(Source: Puglia, D., J. Biagiotti and J.M. Kenny. 2002. ECOFINA: Ecoefficient Technologies and Products Based on Natural Fibre Composites. *Proceedings of 10th European Conference on Composite Materials–ECCM10*. Old St. Jan Conference Centre–Brugge, Belgium June 3-7, 2002.)

Italia) (Research Centre in the FIAT Automotive Sector) and SAAB (Vehicle Manufacturer) (Puglia, Biagiotti and Kenny 2002).

CONCLUSIONS

Natural fibre polymer composites have recently had a great renewed interest for a variety of reasons. Among these, are the quest for increased fuel efficiency in cars, cheaper and better building materials and a growing public interest in environmental preservation.

An increasing amount of interest has developed over the past few years in natural fibre reinforced composites (NFRCs) because of their ease of production, subsequent increase in productivity, cost reduction, lower density and weight and use of renewable resources. However, there are many problems associated with the production of bio-based composite materials, such as the compatibility of natural materials with synthetic polymers, lack of dimensional stability and problems with processability. In any good composite it is necessary to create a good interface (or interphase) whereby applied stress can be transferred between the two different materials. Due to the significant weight savings and the low cost of the raw constituent materials, attractive alternative to glass and carbon fibre reinforced polymer composites, the automobile industry has been approached to overcome these problems and has begun to apply NFRCs in a variety of exterior and interior panel applications. However, further research needs to address significant material and production obstacles before commercially available NFRCs can be widely used in the automotive sector.

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RECEIVED: May 6, 2003

REVISED: January 23, 2004

ACCEPTED: February 9, 2004