JOURNAL OF MATERIALS SCIENCE **39** (2004) 3121–3128

Fique fiber-reinforced polyester composites: Effects of fiber surface treatments on mechanical behavior

P. GAÑÁN
Grupo de Nuevos Materiales, Universidad Pontificia Bolivariana, Circular 1 #70-01, Medellin, Colombia

I. MONDRAGON*
Grupo "Materiales + Tecnologias," Dpto. Ingenieria Quimica y M. Ambiente, Escuela Univ. Politecnica, Universidad del Pais Vasco/Euskal Herriko Unibertsitatea. Pza. Europa, 1. 20018 Donostia-San Sebastian, Spain
E-mail: iapmoegi@sc.ehu.es

Composites consisting of fique fibers (Colombian fibers) and unsaturated polyester (UP) matrix have been investigated. Fique fiber bundles were subjected to alkalization and/or treated with different chemical agents such as maleic anhydride, acrylic acid and a silane to provide increased compatibility between fiber and resin. The mechanical behavior of the composite materials was analyzed by flexural tests. Maximum mechanical properties were observed for composites with fibers subjected to alkalization and also when it was applied as previous process for the other treatments. Aspects of composite materials such as fiber bundle length, fiber content as well as two ways of preparing the material, lamination and BMC, have been evaluated. The influence of surface treatment of fiber on curing of the polyester resin was analyzed by differential scanning calorimetry (DSC). Dynamic mechanical properties were also evaluated to establish the influence of the interfacial interactions on the mechanical behavior of the laminates. © *2004 Kluwer Academic Publishers*

1. Introduction

In Colombia, the fique fiber is an important natural resource. The production is estimated at 30,000 tonnes/year [1], and this production extends to areas such as Ecuador and some regions of Central and South America. These fibers have medium thermal and mechanical properties. The fique fiber is employed in products such as ropes and sacks for seed, grain and coffee. The presence of synthetic fibers, such as polypropylene, in these markets has progressively substituted those hard fibers. This situation is common to other fibers such as jute and sisal.

In recent times, the composite materials approach allows for a good chance in a new area of applications for natural fibers. In the latter ten years increasing research for thermosetting matrix composites as jute/epoxy [2, 3], sisal/epoxy [4, 5], and sisal/unsaturated polyester [6], amongst others, has been performed.

However, there is agreement in the observations with jute and sisal fibers [2]. The fibers have problems for their incorporation within an organic matrix due to the presence of highly hydrophilic —OH groups in their chemical structure. The use of chemical agents applied to the fiber and also matrix modifications are some possibilities to overcome this difficulty.

For composites performed with unsaturated polyester (UP) matrix and natural reinforcements, the main chemicals used by other authors to enable the fiber/matrix adhesion are: alkaline treatments [7], silanization [8] and esterification [9].

To explore the different possibilities offered by fique fiber laminates in the composite field, both laminates and bulk molding compound (BMC) materials have been prepared in this investigation. The mechanical properties of composites formed by UP matrix and differently treated fique fiber bundles have been studied for several fiber lengths and contents.

In this study, the influence of untreated and treated fique fibers on the curing of the UP matrix has been evaluated using DSC. Finally, dynamic mechanical analysis has been used to establish the effect of the different treatments on the fiber/matrix adhesion.

2. Experimental

2.1. Materials

Compañía de Empaques S.A., Medellin, Colombia, kindly provided fique fiber bundles. The unsaturated polyester resin used was Cronolita 1.112 from Plastiform, containing 30 wt% styrene and no additives.

---

*Author to whom all correspondence should be addressed.

TABLE I Characteristics of the unsaturated polyester

Properties	
Viscosity (mPa·s)	600–700
Flexural strength (MPa)	95
Flexural modulus (MPa)	3820
Density (kg/m^3)	1230

TABLE II Fique fiber bundles characteristics

Properties	
Diameter (μm)	50–200
Ultimate stress (MPa)	200
Modulus (GPa)	8–12
Ultimate strain ($\times 10^2$ mm/mm)	4–6
Density (kg/m^3)	870

The initiator for polymerization of the UP resin was methylethylketone peroxide, also purchased from Plastiform, used in a 2 wt% with respect to the resin. For BMC compounds the initiator was terc-butyl perbenzoate. The most significant characteristics of the polymeric matrix [10] and the fique fiber [11] are reported in Tables I and II, respectively.

The chemical agents used were: sodium hydroxide for alkalization, maleic anhydride, MA (CEPSA), acrylic acid, AA (Fluka); and γ-methacryloxypropyl trimethoxy silane (Dynaslan MEMO), provided by Degussa-Hülls.

2.2. Surface modification of fiber bundles

The characteristics of the different treatments and reaction times and temperatures are shown in Table III. More detailed information has been previously published [12, 13]. After the different treatment processes, the fibers were dried for 24 h at 105 ± 5°C.

2.3. Composite processing

Laminates were produced using fiber bundles cut into lengths of 20, 30 and 40 mm. Then, to prepare the composites, the fibers were dried to 105 ± 5°C for 24 h.

For laminates manufacturing, the fique fiber bundles were randomly arranged on the mould and subsequently the resin was added. The processing technique was compression in a hot press, SATIM, with a cure schedule of 2.5 h at 80°C at a pressure of 25 bars followed by a post-curing stage of 2 h at 120°C at the same pressure.

The BMC composite material was produced using the following typical recipe [14]:

	Relative weight
Unsaturated polyester	100.00
$CaCO_3$ filler	75.00
MgO	3.00
Zn-stearate	2.50
Terc-butyl perbenzoate	1.25
Chopped fiber reinforcement	Different contents respect to UP matrix

The BMC systems were prepared using fiber bundles cut off at mesh sizes of 4, 6 and 8 mm. The size of $CaCO_3$ particles is presented in Table IV. Compounding was made by mixing the components in a Haake mixing chamber during 10 min at 15 rpm at room temperature. The material was stored between polyethylene foils for 24 h. Compression press molding was carried out at 120°C and 25 bar for 1 h, afterwards cooling with water at the same pressure at a rate of approximately 4°C/min.

2.4. Test methods

Fiber bundles tensile properties were determinated using a Universal mechanical testing machine, Instron model 4026, equipped with a 10 N load cell. The deformation rate employed was 5 mm/min and gauge length was 50 mm. Thirty fibers were tested in each series. Individual fiber bundles were glued on small paper frames. Pneumatic clamps were employed, and prior to test the frame sides were cut to allow free straining of the sample. The transverse area of the fibers has been assumed to be circular in order to facilitate calculation of mechanical properties [12]. The diameter was measured using an optical microscope. Thirty fibers were studied for each type of treatment.

Composites mechanical properties were evaluated using a flexural test procedure, according to ASTM D-790, by using the same mechanical machine. The specimens employed were 60 mm long, 25 mm wide and around 3 mm thick. They were tested by using a loading span of 48 mm. Five specimens for each material have been tested.

For studying the influence of different treatments on curing of the UP resin, differential scanning calorimetry (Perkin-Elmer DSC-7) was used. About 15–20 mg of sample were placed in the sample pan. Each of samples

TABLE III Treatment conditions for fique fiber bundles

Treatment	Content[a] (wt%)	Solvent	Reaction time (h)	Reaction temperature (°C)
Alkalization (M)	20[b]	Distilled water	1	Room
Maleic anhydride (MA)	5, 10, 15	Acetone	25	55
Acrylic acid (AA)	5, 10, 15	Distilled water	1	Room
γ-methacryloxy propyl Trimethoxy silane (MEMO)	1, 2.5, 5	Distilled water	1	Room

[a] Amount calculated respect to fiber content.

[b] Solution concentration for treatment.

TABLE IV Filler size distribution for $CaCO_3$ particles

Mesh size (mm)	(%)
0.320	64
0.125	33
0.100	1
0.063	2

contained between 25–30 wt% fiber. The sample was scanned from 30 to 200°C at a rate of 10°C/min.

The determination of the acid values for the esterified fibers was performed following the methods described elsewhere [15].

A Metravib viscoanalyser was employed to obtain the storage modulus (E') and loss factor ($\tan\delta$) of the laminate specimens tested in form of small bars ($12 \times 60 \times 2$ mm^3). A bending device with 44 mm span was used. Temperature range from 20 to 200°C was analysed at a heating rate of 3°C/min. The frequency was fixed to 10 Hz.

A Jeol 6400 scanning electron microscope, SEM, was used to study the flexural fracture surfaces of the laminates.

3. Results and discussion

3.1. Laminate composites

3.1.1. Influence of fiber bundle length and content

Fig. 1 reports the effect of fiber bundle length on the flexural strength (σ_F) and modulus (E) for 30 wt% untreated fiber content. As seen, the flexural strength of UP matrix is higher than that for the composites whilst the stiffness of the composites slightly increases with respect to that for the matrix only for fiber lengths equal to or higher than 30 mm, though the changes are within the standard deviation. These results are similar to those obtained by other authors, such as Pothan *et al.* [16] using UP matrices containing other vegetable fibers. It is possible to appreciate the influence of the medium mechanical behavior of fique fibers on the modulus increase and also the low adhesion and/or lack of homogeneity in the interphase between fique fiber and UP matrix as strength was lower for composites than for neat matrix.

In composites with chopped fiber bundles, the stress along the fiber is not uniform; there exists a certain length where the transmission of stresses between matrix and fiber is better [16–19]. For fique laminates, this certain fiber length seems to be around 30 mm. Pothan *et al.* [16] reported a comparable value for unsaturated polyester matrix reinforced with banana fibers.

According to Fig. 2, both flexural strength and modulus increase with fiber content. However, there are not significant differences in strength between 30 and 40 wt% fiber laminates. These results suggest that for high fiber contents the interactions between fibers increase, so producing a lower stress transmission within the system. Besides, with the increment of fiber content, the disposition of the matrix for fiber wetting is reduced, and therefore the empty spaces and dry zones can increase, so affecting the interfacial bond strength [20].

3.1.2. Effects of fiber treatment on mechanical behavior

Table V shows the influence of fiber treatment on the mechanical properties of different laminates containing 30 wt% fibers with 30 mm length. The highest flexural properties were observed when alkaline treatment was applied as previous treatment, see Fig. 3. This behavior is because alkalization increases the fique fiber strength and modulus, a fact that can be associated with the changes introduced by this treatment on the orientation of molecular chains of cellulose [12] since it affects the molecular entanglements as well as its regularity. Additionally, increasing of the cellulose content in the fiber together with lowering of the other non-crystalline components of the fiber also reduces the fiber diameter [12], and it can lead to improved fiber wettability. Besides, as reported for other natural fibers, this treatment can generate more sites for mechanical interlocking [5] due to the formation of a rougher surface and to the increasing in the fiber fibrillation [20].

For esterification and silanization processes, the influence of the agent amount with respect to the weight

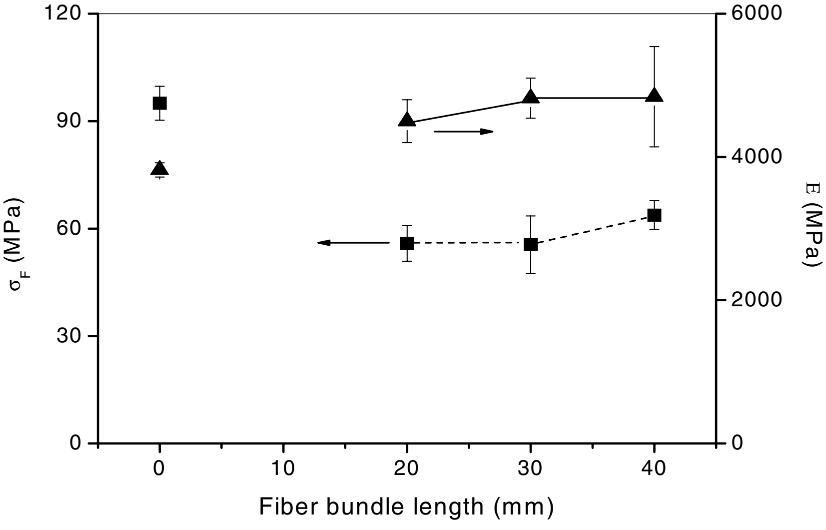


Figure 1 Variation of flexural properties vs. fiber bundle length for 30 wt% untreated fique fiber laminates.

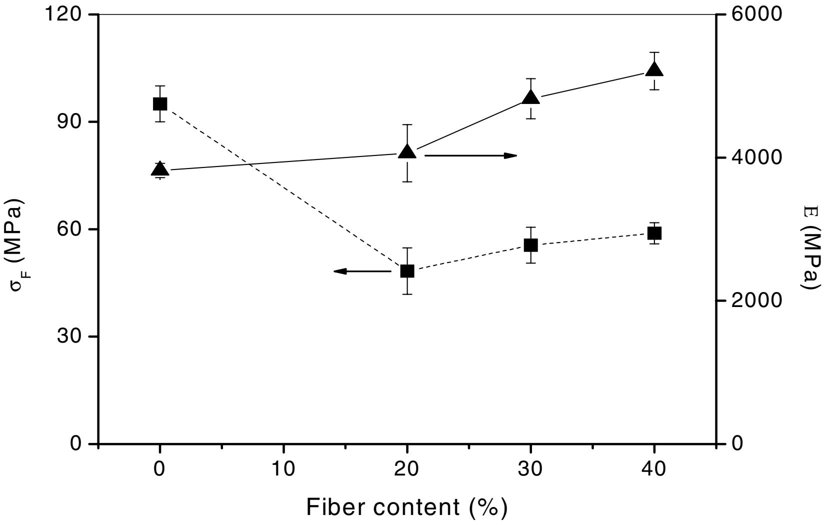


Figure 2 Variation of flexural properties vs. fiber content for untreated 30 mm length fiber bundle laminates.

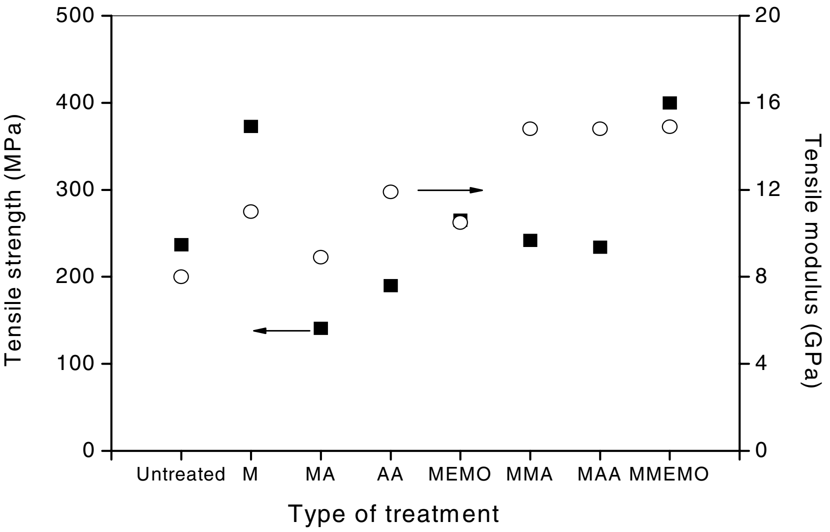


Figure 3 Tensile properties of untreated and treated fique fibers for the several coupling agents used.

of fiber was also analyzed. Concerning esterification with maleic anhydride, a 10 wt% with respect to the fiber weight seems to be the appropriate value. At lower maleic anhydride concentration (5 wt%), probably less formation of ester groups and minor substitution of hydroxyl groups occur, and consequently the adhesion between fique fibers and UP matrix is lower. As shown in Table VI, these results agree with the acid values obtained after esterification. Nevertheless, at the optimal concentration of MA, the flexural strength is still smaller than for the laminates with untreated fique fiber. As shown in Fig. 3, this behavior is surely related to the changes on the fiber mechanical behavior by effect of the treatment. These variations are also reflected on the lower deformability and higher flexural modulus of the composites.

In the case of AA esterification, the best results are obtained for the 10 wt% content with respect to the

TABLE V Influence of fiber treatments on the mechanical behavior of laminates containing 30 wt% fique fiber (30 mm fiber bundle length)

Treatment	σ_F (MPa)	E (MPa)	Ultimate strain ($\times 10^2$ mm/mm)
Untreated	55.6 ± 7.1	4680 ± 230	3.00 ± 0.50
Alkaline (M)	102.6 ± 9.5	6190 ± 640	3.60 ± 0.20
5 wt% MA, 25 h	47.4 ± 3.9	5570 ± 330	1.20 ± 0.20
10 wt% MA, 25 h	51.4 ± 3.4	5670 ± 600	1.10 ± 0.10
15 wt% MA, 25 h	38.2 ± 1.3	5400 ± 370	0.80 ± 0.10
M + 10 wt% MA, 25 h	86.8 ± 5.5	6250 ± 810	2.10 ± 0.20
5 wt% AA	55.6 ± 5.7	5035 ± 354	1.80 ± 0.20
10 wt% AA	66.7 ± 3.7	530 ± 430	2.00 ± 0.30
15 wt% AA	64.3 ± 6.1	5150 ± 340	2.00 ± 0.20
M + 10 wt% AA	79.0 ± 4.8	5840 ± 200	2.10 ± 0.70
1 wt% MEMO	66.7 ± 8.0	4300 ± 380	3.09 ± 0.30
2.5 wt% MEMO	73.3 ± 8.8	4690 ± 410	3.00 ± 0.40
4.5 wt% MEMO	67.5 ± 6.2	4290 ± 410	2.90 ± 0.20
M + 2.5 wt% MEMO	103.2 ± 17.2	6060 ± 530	3.60 ± 0.10

TABLE VI Acid values for esterification treatments

Content[a] (wt%)	MA	AA
5	8.5	3.6
10	19.2	9.1
15	20.7	11.0

[a] Amount calculated with respect to fiber content.

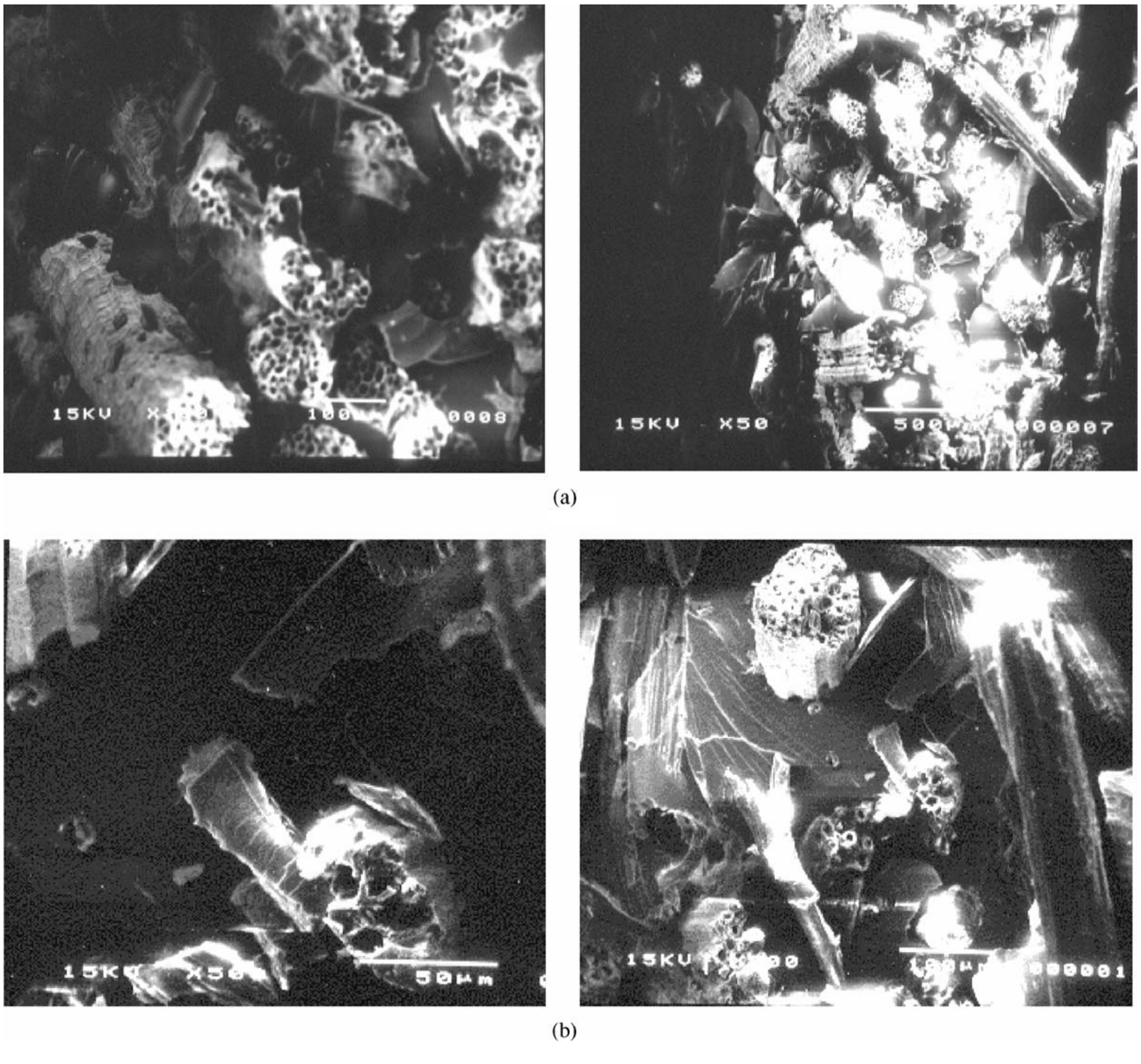
fiber weight. Accordingly, optimal conditions for esterification treatments seem to be around 10 wt% agent content. For a lower concentration of acrylic acid, such as 5 wt%, the progress of reaction is low such as observed in Table VI. At higher AA concentrations it is not possible to obtain higher flexural strength. Although less than for MA modification, flexural modulus also increases with respect to that for the untreated laminates. These variations possibly are a consequence of the changes introduced by esterification treatments on the mechanical behavior of the fiber, as shown in Fig. 3. On the other hand, the variations observed on mechanical properties caused by using coupling agents can also be a consequence of the increased homogeneity of the composites so obtained.

Alkalization can be considered as a pre-treatment when one wants to work with a follow-up process. For composites including fibers to which alkalization combined with MA and AA treatments were carried out, results reflected a high influence of alkaline process. Thus, the flexural properties of these laminates are higher than those for laminates containing MA or AA treated fibers but not previously alkalized.

For the silane treatment, the best flexural properties were obtained for a 2.5 wt% MEMO respect to fiber content. The flexural strength increases by around 30% with respect to that for the untreated fiber laminate. According to Fig. 3, silane treatment does not significantly affect the mechanical properties of the fique fibers. So, the constancy of modulus and deformability together with the higher resistance of these laminates with respect to the same properties in composites containing untreated fibers are possibly associated with a better stress transmission throughout the interface as well. The reported behavior is comparable to that shown by Singh *et al.* [21] with sisal fibers. The changes induced in the fique surface would also prevent the fiber–fiber interlocking contacts, which is a source of high stress concentration in the composites [22]. As for other treatments, prior alkalization leads to much better properties with respect to those for the laminates with untreated fibers.

According to previous work [12], with all treatments employed in this study a reduction on the hydrophilic behavior of the treated fiber with respect to the untreated fiber is obtained. Thus, it is possible that this reduction leads to an enhancement on the fiber/matrix adhesion and, consequently, on the mechanical properties of the fiber treated laminates.

For a better analysis of the effect of alkalization on the fiber/matrix adhesion, Fig. 4 presents SEM micrographs for untreated and mercerized fiber laminates.



(a)

(b)

Figure 4 SEM micrographs of UP laminates with: (a) untreated and (b) alkali treated fibers.

In Fig. 4a, fiber pull-out is present and many empty spaces between fibers can be observed, so suggesting poor adhesion between untreated fibers and UP matrix. On the contrary, for mercerized laminates, see Fig. 4b, the fibers are covered by UP matrix, and there are no empty spaces. This indicates a better wetting of the fiber. Besides, some regions present some level of deformation possibly due to the fibrillation of the fiber. Thus, these results confirm the enhancement on fiber/matrix adhesion by effect of alkalization.

3.1.3. Influence of the fique fiber treatment on the cure kinetics of UP resin

Another factor to be considered is the possibility that these fiber treatments can introduce variations on the curing of the UP matrix and also on its mechanical behavior. Some authors [23–25] reported variations on cure kinetics or T_g changes on such composites. Thermal results for different samples containing untreated or treated fique fibers are shown in Table VII. Each composite probe contained around 30 wt% fiber.

The total enthalpy of curing (ΔH) for mass unit UP resin in the untreated fiber laminate, 348.7 J/g, was comparable to that evaluated for the neat resin (353.4 J/g), and similar to that presented in the bibliography survey (around 340 J/g) [26]. These results indicate that the presence of the untreated fique fiber has no influence in the curing of UP resin. Lucas *et al.* [27] obtained a similar result working with UP and mineral fillers.

The main variations obtained for composite reaction enthalpies are associated with samples containing silanized and AA treated fibers. However, these values can be affected by the fiber content. The introduction of treated fibers slightly delayed the curing process, this behavior being more evident for AA treatment.

For analysis of these results, Table VII also includes the reaction enthalpy for the neat UP resin mixed with each chemical agent employed, with the appropriate concentration according to flexural behavior. For samples containing AA or MEMO chemicals a reduction of total enthalpy with respect to the neat UP matrix is also observed. Besides, these samples present a slight increment in maximum temperature of curing. This situation is possibly associated with the interference of the double bonds of the agents on curing of the UP matrix. Shieth *et al.* [28] and Ishida *et al.* [29] reported a similar behavior with a MEMO silane applied on glass beads and E-glass fibers, respectively.

According to these results, it is possible to suggest the changes observed on the curing process of UP matrix due to the fiber treatments can also affect the ultimate properties of the analyzed composites.

TABLE VII Influence of fiber treatments on curing of UP matrix

Treatment	ΔH/Composite 30 wt% fiber (J/g)	ΔH/UP in composite (J/g)	ΔH neat UP (J/g)	Maximum temperature (°C)
Neat UP	–	–	353.7	115.4
Untreated fiber	248.4	348.7	–	116.1
Alkaline	240.4	341.0	–	116.0
MA	248.0	332.3	367.7	117.6
AA	220.3	299.1	342.4	118.7
MEMO	244.0	322.8	328.1	118.4

3.2. BMC

Tables VIII and IX show the results for BMCs prepared with untreated and alkalized fique fiber bundles. As can be seen, the flexural behavior is affected by different factors as amount of fiber, fiber length and also alkalization applied onto the fiber. The best mechanical behavior for composites containing untreated fibers is achieved using 60 phr of fibers with respect to UP matrix. At higher weight contents, it is possible that interactions between fibers increase, so affecting the mechanical behavior of the material. For all BMCs, inhomogeneties in the bulk exist due to the possible interference between the mineral fillers and fique fibers. This can be attributed to the increased incompatibility between fique fibers and fillers due to the difference in shape and size that reduces the stress transmission through the material. Rozman *et al.* [30] reported a similar behavior for thermoplastic composites reinforced with glass and coconut fibers.

The variation of the mechanical behavior with fiber length is shown in Table IX for 60 phr fiber composites. Flexural strength presents a slight increase with the average fiber length in the 2.9 to 6.5 mm range, though the mechanical properties do not show important changes in the fiber length range used. As reported in Fig. 5, that is possibly due to the wide size distribution that each mesh size employed presents.

Similarly to the laminates, for BMC materials, alkalization appears to be an adequate treatment in order to enhance the mechanical behavior. A comparison of results shown in Table VIII indicates that both flexural strength and modulus are higher for all fiber contents than those corresponding to composites with untreated fibers. This increase is related to better mechanical properties of the fibers because of changes introduced by the alkaline treatment on the fiber structure and also to the enhancement on the wettability of the fiber.

TABLE VIII Mechanical behavior of several BMCs. Mesh size: 4 mm

	Untreated fibers		Alkaline fibers	
Fiber content (%)	σ_F (MPa)	E (MPa)	σ_F (MPa)	E (MPa)
0	43.7 ± 2.1	6310 ± 290	43.7 ± 2.1	6310 ± 290
40	42.6 ± 6.5	7320 ± 170	59.9 ± 4.9	8690 ± 170
60	52.1 ± 4.8	8690 ± 310	70.8 ± 2.6	9170 ± 130
80	48.4 ± 1.3	8300 ± 80	62.8 ± 3.4	9610 ± 780
100	50.1 ± 4.0	8700 ± 290	–	–

TABLE IX Variation of mechanical behavior with fiber length for BMCs containing 60 phr untreated fique fiber

Mesh size (mm)	Average fiber length (mm)	σ_F (MPa)	E (MPa)
4	2.9	52.1 ± 4.8	8690 ± 310
6	3.7	60.0 ± 3.7	8700 ± 420
8	6.5	59.3 ± 5.3	9010 ± 320

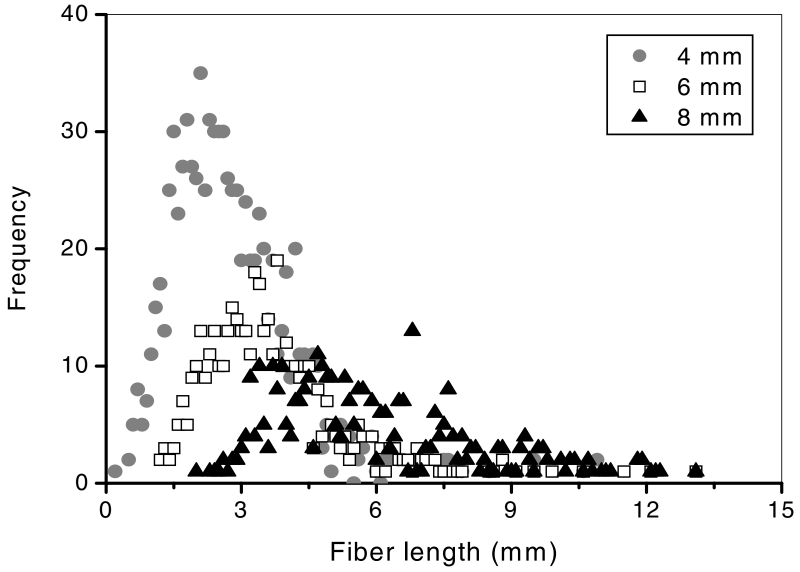


Figure 5 Distribution of fiber length with respect to mesh sizes used for BMCs.

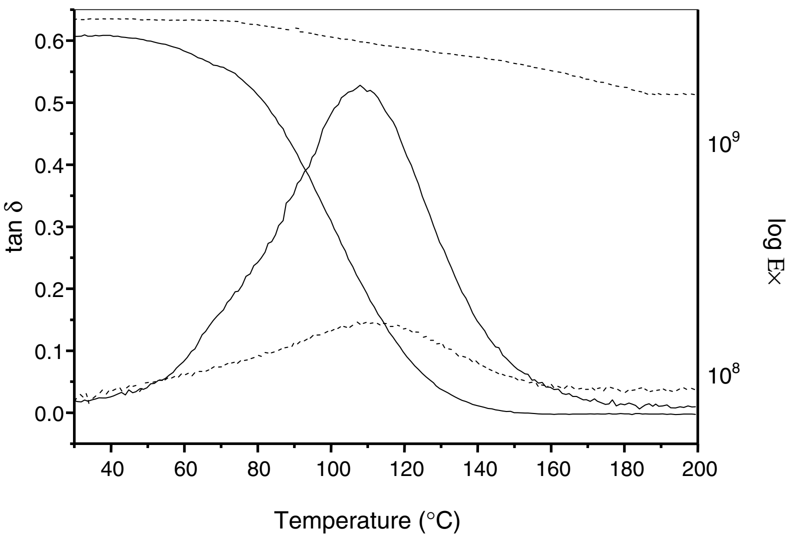


Figure 6 Variation of log E' and tan δ with temperature for neat resin (——) and 30 wt% untreated fiber laminate (- - -).

3.3. Dynamic mechanical testing

Fig. 6 presents the variation of storage modulus (E') and loss tangent (tan δ) with temperature for neat both matrix and untreated 30 wt% fiber laminate. The glass transition temperature (T_g) of the laminate is similar to that for the neat matrix. Marcovich *et al.* [31] and Mwaikambo *et al.* [32] reported similar results. The height of the tan δ peak is lower for the laminate than that for the UP resin. This change is produced by the fiber presence since it leads to hindering of the molecular movement of the UP matrix. The E' of the untreated laminate is higher than that for the UP matrix as the incorporation of fibers into the polymer increases the stiffness of the material. It can be further noted that the difference in storage modulus between laminates and UP matrix increases at higher temperatures.

Fig. 7 shows the variations of tan δ upon temperature for different laminates. Results correspond to the second scan for reducing the plastifying effect of the moisture observed by others authors [33]. In a similar way to that reported by Saha *et al.* [34] for cyanoethylated jute fibers, for all treatments only a slight variation of T_g and small variations of shape of damping peak are observed. Some authors [35] attribute the variations on shape of damping peak respect to temperature with changes in the orientation of fibers as well as with

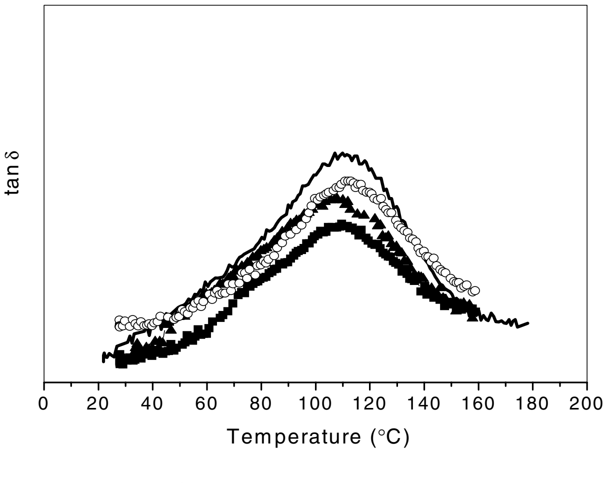


Figure 7 Variation of tan δ versus temperature for laminates containing 30 wt% fiber: untreated (——), MA (■) AA (□), and silane treated (▲).

variations in fiber—matrix adhesion. In these composites, the untreated fiber laminate has a higher damping than those for the treated fiber laminates, which could be associated with some wettability increment of the treated fibers by the UP matrix.

4. Conclusions

The importance of choosing an adequate chemical agent and its amount has been proven for composites containing fique fiber bundles. Flexural properties of fique fiber laminates and BMC systems can be increased with fiber content and fiber length. However, in BMCs the effect of the size distribution has to be considered.

For the mechanical behavior of the treated materials, the best results are achieved when the fiber is subjected to treatments such as alkalization. These variations are associated with the enhancement of the fiber—matrix adhesion observed through microscopy techniques but also with the changes introduced by treatments on the structure and mechanical properties of fique fibers and also on the homogeneity of the composites. This behavior has been observed for both laminates and BMCs.

For some treatments, such as silanization or AA, some slight changes on curing behavior of UP matrix can take place.

The fiber presence produces a reduction on the damping properties of composites with respect to the neat matrix, due to the restrictions of chain movements of the UP matrix.

Acknowledgments

The authors would like to thank Compañía de Empaques in Medellin (Colombia), Universidad del Pais Vasco/Euskal Herriko Unibertsitatea and Universidad Pontificia Bolivariana for the financial and logistics support that has made this research work possible.

References

1. C. ALVÁREZ, "III Congreso Internacional de Fibras Naturales," Rionegro (1998) p. 20.
2. J. GASSAN and A. BLEDZKI, *Polym. Comp.* **18** (1997) 179.
3. *Idem., ibid.* **20** (1999) 604.
4. A. K. MOHANTY and M. MISRA, *Polym. Plast. Techn. Eng.* **34** (1995) 729.
5. E. BISANDA and M. ANSELL, *Comp. Sci. Tech.* **41** (1991) 165.
6. K. JOSEPH, S. VARGHESE, G. KALAPRASAD, S. THOMAS, L. PRASANNAKUMARI, P. KOSHY and C. PAVITHRAM, *Eur. Polym. J.* **32** (1996) 1243.
7. J. GASSAN and A. BLEDZKI, *Comp. Sci. Tech.* **59** (1999) 1303.
8. E. BISANDA and M. P. ANSELL, *J. Mater. Sci.* **27** (1992) 1690.
9. N. MARCOVICH, M. M. REBOREDO and M. I. ARANGUREN, *J. Appl. Polym. Sci.* **70** (1998) 2121.
10. K. DE LA CABA, P. GUERRERO, J. GAVALDA and I. MONDRAGON, *J. Polym. Sci.: Part B: Polym. Phys.* **37** (1999) 1677.
11. P. GAÑÁN, "COMAT 2001, Proceedings," Mar del Plata (2001) p. 39.
12. P. GAÑÁN and I. MONDRAGON, *Polym. Comp.* **23** (2002) 383.
13. *Idem.*, "Materiales Compuestos 99," Málaga (1999) p. 81.
14. O. OWOLABI, T. CZVIKOVSZKY and I. KOVACS, *J. Appl. Polym. Sci.* **30** (1985) 1827.
15. M. GEIER, in "Quality Handbook for Composite Materials" (Chapman & Hall, London, 1994).
16. L. POTHAN, S. THOMAS and N. NEELAKANTAN, *J. Reinf. Plast. Comp.* **16** (1997) 744.
17. W. ZHU, B. TOBIAS and R. COUTS, *J. Mater. Sci. Lett.* **14** (1998) 508.
18. S. FU and B. LAUKE, *Comp. Sci. Tech.* **56** (1996) 1179.
19. A. SANADI, R. YOUNG, C. CLEMONS and M. ROWELL, *J. Reinf. Plast. Comp.* **13** (1994) 54.
20. A. DE ALBURQUERQUE, K. JOSEPH, L. DE CARVALHO and J. D'ALMEIDA, *Comp. Sci. Tech.* **60** (2000) 833.
21. B. SINGH, M. GUPTA and A. VERMA, *Polym. Comp.* **17** (1996) 910.
22. J. KARDOS in "Molecular Characterization of Composite Interfase," edited by H. Ishida and G. Kumar (Plenum Press, New York, 1985).
23. A. GARTON, W. STEVENSON and S. WANG, *J. Polym. Sci.: Part A: Polym. Chem.* **26** (1988) 1377.
24. S. WANG, A. GARTON and W. STEVENSON, *J. Appl. Polym. Sci.* **40** (1990) 99.
25. Z. PETROVIC and N. STOZAKOVIC, *Polym. Comp.* **9** (1988) 227.
26. J. KENNY and M. OPALICKI, *Composites* **27A** (1996) 229.
27. J. LUCAS, J. BORRAJO and R. J. J. WILLIAMS, *Polymer* **34** (1993) 1886.
28. J. SHIETH and T. HSU, *Polym. Eng. Sci.* **32** (1992) 335.
29. I. ISHIDA and J. KOENIG, *J. Appl. Polym. Sci.* **17** (1979) 615.
30. H. ROZMAN, G. TAY, R. KUMAR, A. ABUBAKAR, H. ISMAIL and Z. MOHD, *Polym. Plas. Techn. Eng.* **38** (1999) 997.
31. N. MARCOVICH, M. REBOREDO and M. ARANGUREN, *J. Appl. Polym. Sci.* **68** (1998) 2069.
32. L. MWAIKAMBO and E. BISANDA, *Polym. Test.* **18** (1999) 181.
33. N. MARCOVICH, M. REBOREDO and M. ARANGUREN, *J. Appl. Polym. Sci.* **61**, (1996) 119.
34. A. SAHA, S. DAS, D. BATTA and B. MITRA, *ibid.* **71** (1999) 1505.
35. L. NIELSEN and R. LANDEL, "Mechanical Properties of Polymer and Composites," 2nd ed. (Marcel Dekker Inc., New York, 1994).

Received 10 February
and accepted 17 December 2003