Toughening of Wood Particle Composites—Effects of Sisal Fibers

Adrián J. Nuñez,[1] Mirta I. Aranguren,[1] Lars A. Berglund[2]

[1]*INTEMA-Universidad Nacional de Mar del Plata, Juan B. Justo 4302, Mar del Plata, Argentina*
[2]*Division of Lightweight Structures, Department of Aeronautical and Vehicle Engineering, Royal Institute of Technology, SE-100 44 Stockholm, Sweden*

Received 1 August 2005; accepted 3 November 2005
DOI 10.1002/app.23716
Published online in Wiley InterScience (www.interscience.wiley.com).

ABSTRACT: Sisal fibers were added to wood particle composites to enhance their toughness. The selected matrix was a commercial styrene diluted unsaturated polyester thermoset resin. Fracture tests were carried out using single-edge notched beam geometries. Stiffness, strength, critical stress intensity factor K_{IQ}, and work of fracture W_f of notched specimens were determined. The incorporation of sisal fibers into wood particle composites significantly changed the fracture mode of the resulting hybrid composite. For the neat matrix and the wood particle composites, once the maximum load was reached, the crack propagated in a catastrophic way. For hybrid composites, fiber bridging and pull-out were the mechanisms causing increased crack growth resistance. Addition of a 7% wt of sisal fibers almost doubled the K_{IQ} value of a composite containing 12% wt of woodflour. Moreover, the W_f increased almost 10-fold, for the same sample. In general, the two composite toughness parameters K_{IQ} and W_f increased when the fraction of sisal fibers was increased. © 2006 Wiley Periodicals, Inc. J Appl Polym Sci 101: 1982–1987, 2006

Key words: wood particle; sisal; mechanical properties; fracture toughness; fiber bridging

INTRODUCTION

The use of long fibers to enhance the toughness of brittle matrix composites is a well-established practice.[1–3] However, most of the previous studies were carried out on synthetic fiber-composites, rather than on natural fiber-reinforced composites.[4] The study of toughening composites by addition of natural fibers is justified because of the increasing interest in these materials as reinforcements. Fibers such as hemp, wood, sisal, jute, and others originate from renewable resources and often show low cost, low density, availability in large quantities, biodegradability, and cause little wear to processing equipment.[5] Composites reinforced with natural fibers often show lower cost in comparison to glass fibers-reinforced composites. Therefore, the use of natural fibers is economically and environmentally attractive in the manufacturing of composite materials.

Present applications of natural fiber composites are mainly in the automotive, packaging, and furniture industries,[5] but they are also used in building, flooring, decorative laminates, and general industrial applications. Such composites are frequently subjected to high mechanical loads. Therefore, the improvement of toughness is important in composites based on brittle thermoset matrices.

Wood particles are used in many commercially important composites. In the often termed "wood-plastic composites," woodflour particles or very short fibers are mixed with thermoplastics. The compounded material is extruded into, for instance, low-cost decking applications or injection molding components. Particle-board is another example of a low cost material based on wood particles. Compared with fiber composites, wood particle materials are brittle because of the lack of strong energy absorption mechanisms.

It is usual to think of the fiber as a reinforcing constituent in terms of Young's modulus and ultimate strength. However, in brittle matrices, fibers also contribute significantly to the work of fracture in the material. Kelly and Tyson demonstrated the importance of fiber pull-out as the major energy absorbing mechanism in fiber composites.[6] The argument is based on the critical fiber length concept l_c. For fiber lengths shorter than l_c , the fiber is pulled out of the matrix. The work of fracture per fiber is obtained from interfacial frictional shear stress during pull-out, pull-out length, and the fiber surface area of that length. The work of fracture increases with fiber length until l_c is reached. It then

Correspondence to: M. I. Aranguren (marangur@fi.mdp.edu.ar).
Contract grant sponsor: CONICET (National Research Council, Argentina).

decreases with fiber length above l_c since the amount of pull-out decreases.

Toughness is a problem if fibers are short, as in the woodflour case. Devi et al.[7] studied the impact behavior of pineapple leaf fiber-reinforced polyester composites as a function of fiber length. They determined that the fiber pull-out process was the main energy absorption mechanism. The authors observed a lower work of fracture for composites with fiber length shorter than a critical length. Ray et al.[8] investigated the fracture behavior of jute-reinforced vinylester composites. They improved the interfacial bonding by alkali-treating the jute fibers and observed that a stronger interface led to less fiber pull-out having shorter pull-out lengths, which decreased the impact fatigue resistance.

In the present study, long sisal fibers in the form of a mat were incorporated to enhance the toughness of wood particle-unsaturated polyester (UPE) composites. These hybrid composites were manufactured using the hand-lay-up technique followed by compression molding. Wood particles are generally incorporated to reduce the overall composite cost and, at the same time, improve certain mechanical properties, such as Young's modulus.[9] Furthermore, the presence of wood particles reduces matrix shrinkage and prevents resin cracking during the curing step.

The effect of long fibers on composites toughness, strength, and modulus was analyzed through the preparation of two series of hybrid composites. In one series, a fixed weight percentage of long fiber-mat was added to composites with different weight content of wood particles. In the second series, different weight percentages of long fiber-mat were added to a composite with a fixed wood particle content. The investigation of the mechanical properties emphasized toughness properties.

EXPERIMENTAL

Materials

A commercial styrene-diluted unsaturated polyester (UPE) matrix (Reichhold Norpol PO-4571, Norway) was selected for the work. The resin was mixed with 2 wt % of hardener (Peroxide Norpol 1, Norway). As filler, pine woodflour particle (Scandinavian Wood Fiber, Sweden) with a maximum particle size of 200 μm was used. The reinforcement consisted of nonwoven mats of sisal fibers (Mühlmeir Type SIMAT), with diameters in the range of 0.11–0.23 mm and lengths ranging from 10.0 to 18.5 cm.

The wood particles were oven-dried at 70°C for 1 day, until constant weight was achieved. The wood particles were mixed with the UPE resin, using a mechanical stirrer at high speed. The wood particle filled resin was then used to prepare both wood particle and hybrid composites. The wood particle composites were simply prepared by compression molding, whereas the hybrid composites were manufactured incorporating one or two layers of mats of long fibers, using a hand-lay-up technique followed by compression molding. The plaques were cured at room temperature under pressure for 6 h and post-cured at 150°C for 1.5 h. Final sample surface dimensions were 250 mm by 250 mm while thickness varied from 3 to 4 mm depending on the total weight of the material in the mold.

Different materials were prepared: neat crosslinked UPE, wood particle composites (WFC-XX, where XX stands for the volume fraction of wood particle), and hybrid composites (HC-XX-YY, where XX and YY stand for the wood particle and sisal volume fraction, respectively).

Hybrid composites with relatively high wood particle content were prepared by incorporating a single mat of sisal fibers to wood particle filled resins. The resulting sisal volume fractions were about (7 ± 0.4)% for all these composites (HC-XX-07). On the other hand, hybrid composites (HC-13-YY) were prepared varying the amount of sisal fibers and keeping the wood particle volume fraction approximately constant ((13 ± 1.2)%). This series of hybrid composites was prepared by incorporating two layers of mats and alternating layers of wood particle-filled resin, which were pressed to ensure impregnation of the filled polymer into the sisal mats.

Mechanical testing

Tensile tests

Tensile modulus and strength of the different materials were determined using a universal-testing machine, according to the procedures of the standard ASTM D3039. Specimens were cut and carefully polished to their final surface dimensions (120.0 × 15.0 mm^2). Samples were tested at room temperature at a crosshead speed of 2 mm/min.

In the study of the wood particle and hybrid composites with high wood particle content, an Instron 4411 universal-testing machine was used, while the study of the hybrid composites prepared with a moderate wood particle concentration (HC-13-YY) was realized in an Instron 8500 plus universal-testing machine.

Fracture mechanics tests (single edge notched beam)

The tests were performed in a universal-testing machine, according to the procedures of ASTM D5045. Specimens were cut, carefully polished to the final dimension, and a prenotch was machined. A sharp crack was introduced in each of the precracked spec-

TABLE I
Mechanical Properties of Neat Unsaturated Polyester and Wood Particle Composites

	Tensile modulus (GPa)	Tensile strength (MPa)	K_{IQ} (Mpa × $m^{1/2}$)	W_f (kJ/m^2)
UPE	3.42 ± 0.05	53 ± 6	0.41 ± 0.14	0.04 ± 0.01
WFC-12	4.03 ± 0.25	35 ± 4	0.87 ± 0.33	0.17 ± 0.04
WFC-21	4.94 ± 0.23	28 ± 6	1.23 ± 0.30	0.25 ± 0.04

imens by sliding a fresh microtome blade. The fracture test was carried out using a single edge notched beam (SENB) configuration at room temperature at a crosshead speed of 1 mm/min.

Woodflour composites and hybrid materials with high wood particle content were cut to final surface dimensions of 56 × 12.7 mm^2. The specimens corresponding to the hybrid composites named HC-13-YY were manufactured from two layers of mats instead of one to achieve a higher concentration of the long sisal fibers. Larger samples were used in these fracture tests, 80 × 20 mm^2. Linear dimensions were measured with an accuracy of 0.01 mm.

The critical stress intensity factor, K_{IC} , is regularly used as a fracture toughness parameter. Since the validity of this parameter was not verified rigorously, it was denoted K_{IQ} and it was determined from the maximum load so as to characterize the resistance to crack initiation. The work of fracture (W_f) was calculated from the area under the load/displacement curve divided by twice the area of the fracture surface (since two new faces are created).[10–12] The work of fracture was defined simply as the total energy consumed to produce a unit area of fracture surface during the "complete" fracture process. The analysis requires no information on stress intensity at the crack tip, notch tip acuity, the elastic properties of the material, or its mechanical linearity.[12] This simple testing and data reduction procedure is motivated by the purpose to compare the performance of different material compositions.

RESULTS AND DISCUSSION

Wood particle composites

The addition of wood to the neat UPE polyester resin increased Young's modulus but reduced tensile strength (Table I). If the Halpin-Tsai equation for spherical particle composites is used, a particle with a modulus of 11.6 GPa predicts both experimental data with an accuracy of 95% or better. The reduction in strength is most likely due to early fracture initiation by particle-matrix debonding. The value of the modulus for the woodflour is in the range of values reported in the literature.[13,14] The longitudinal Young's modulus of clear Swedish pine-wood is typically in the range 10–15 GPa. The "effective" modulus of 11.6 GPa derived for a spherical Halpin-Tsai particle is, therefore, fairly high. Contributing reasons may include resin in the lumen and more favorable wood particle geometry than spherical. During the tensile tests of the wood particle composites, the load-displacement curves showed a slight deviation from linearity.

To characterize the material toughness in terms of K_{IQ}, the SENB testing geometry was used. For the neat UPE, the load increases linearly with displacement up to fracture. For the wood particle composites, a small deviation from linear elastic behavior was observed. In both cases, once the maximum load was reached, the crack instantaneously propagated through the material. The low K_{IQ} value measured for the neat matrix is in agreement with typical values K_{Ic}-values reported for thermoset polymers.[15,16] The wood particle addition increased the toughness of the starting material (Table I), an effect that has been previously observed in thermosets loaded with rigid particles.[15,17] For instance, the addition of 21% vol of wood particle increased the K_{IQ} value three times. Although the work of fracture W_f was low for the neat polyester, the wood particle composites could store or dissipate more energy prior to final failure so that W_f was increased (W_f for the 21% vol. sample is 6.25 times that of the neat thermoset). As load-displacement curves are analyzed, we see that the reason was that initiation of catastrophic crack growth required higher loads with increased wood particle addition (see also Fig. 1). One may speculate that subcritical crack growth was more difficult in the composites because of local irreversible deformation and crack stopping due to the wood particles.

Hybrid composites of high wood particle content

During the tensile test of these hybrid composites, the matrix fractured catastrophically toward the end of the test, although most of the sisal fibers were undamaged. The fibers pulled out from the matrix and held the two pieces of the specimen together up to the final breakage (Fig. 2). As the volume fraction of wood particle was increased, the hybrid composite modulus increased (Table II). The distribution of wood flour and sisal fibers was uneven, and this may explain the unexpected modulus difference between materials

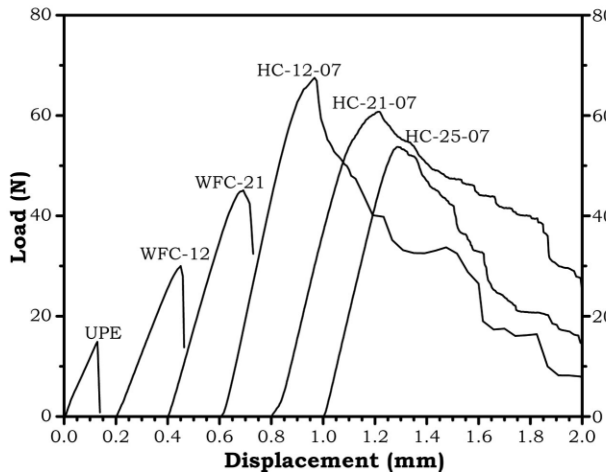

Figure 1 Representative load-displacement curves for neat polyester, wood particle composites, and hybrid composites with high wood particle content.

HC-21–07 and HC-25–07. The HC-25–07 also showed decreased tensile strength when compared with the other hybrid composites in Table II. The local volume fraction of wood particles in the nonfibrous layers of the laminate was very high whereas the impregnated sisal fiber layers showed low wood content. The present wood particle/resin mixture showed very high viscosity, and wood particles became concentrated in the regions outside the sisal mat (Fig. 3). In contrast, the sisal mat was impregnated by resin of low wood particle content. It is possible that high wood content layers show wood particle debonding at low strain, so that the overall laminate tensile strength is lowered.

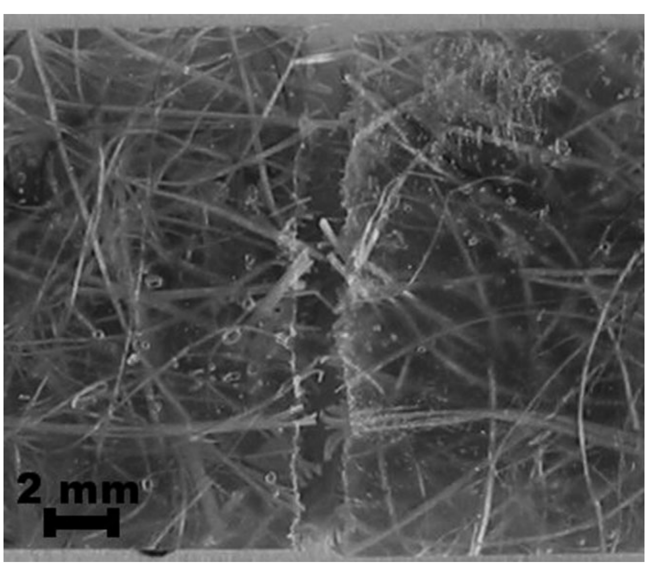

Figure 2 HC-12–07 specimen after being tested in tensile test.

TABLE II
Mechanical Properties of Hybrid Composites with a High Content of Woodflour

	Tensile Modulus (GPa)	Tensile Strength (MPa)	K_{IQ} (MPa×$m^{1/2}$)	W_f (kJ/m^2)
HC-12–07	4.32 ± 0.35	27 ± 3	2.18 ± 0.25	1.79 ± 0.32
HC-21–07	4.72 ± 0.17	26 ± 2	1.84 ± 0.31	1.73 ± 0.54
HC-25–07[a]	5.64 ± 0.28	22 ± 2	1.58 ± 0.64	1.00 ± 0.30

One mat of sisal fiber was used in the preparation.
[a] This sample showed a sandwich-like structure.

Figure 1 shows representative load-displacement curves for the materials in SENB fracture mechanics configurations. Considerable differences are observed between wood particle and hybrid composites. Wood particle composites showed catastrophic crack propagation at lower loads, whereas cracks in hybrid composites propagated in a more stable manner. An illustrative example is the comparison between WFC-12 and HC-12–07. Addition of 7% by volume of sisal fibers more than doubled the peak load where macroscopic crack growth started. During subsequent crack growth, much more energy was dissipated as apparent from the gradual rather than dramatic decrease in load-displacement data, which resulted in an increase of the W_f of 10 times over the value of the neat thermoset.

For the hybrid composites with constant fiber content, increased wood content was accompanied by decreased load at crack initiation. For this reason, K_{IQ} was decreased with increasing wood particle content. Crack growth apparently initiated at lower load as the concentration of wood particles increased. In addition, W_f decreased with increasing wood content. The improved crack growth toughness was not sufficient to compensate for the lowered load at crack growth initiation.

The hybrid composites showed improved ability to carry load also after maximum load was reached. The long sisal fibers increased the energy required for crack growth, primarily by fiber pull-out, see Figure 4.

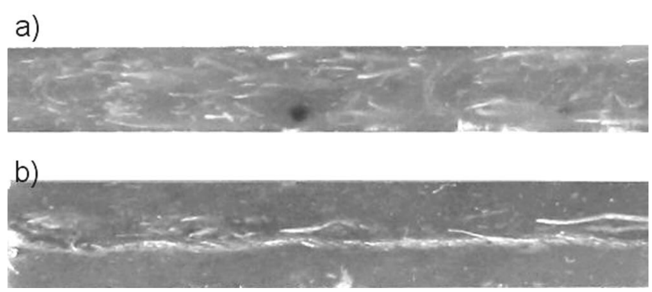

Figure 3 Photograph of hybrid samples, thickness side: (a) HC-12–07 (b) HC-25–07.

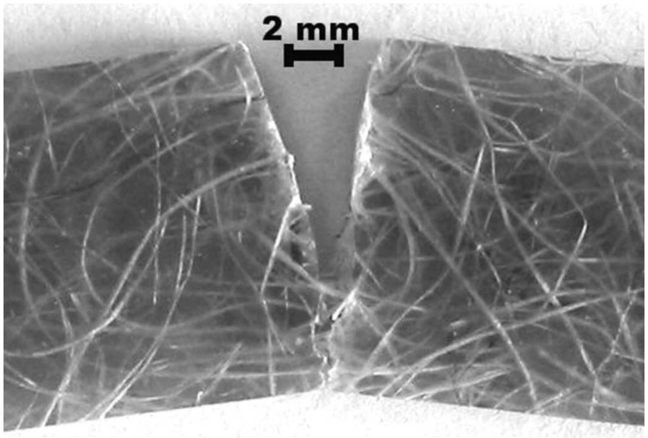

Figure 4 HC-12–07 specimen after the 3-point bending test.

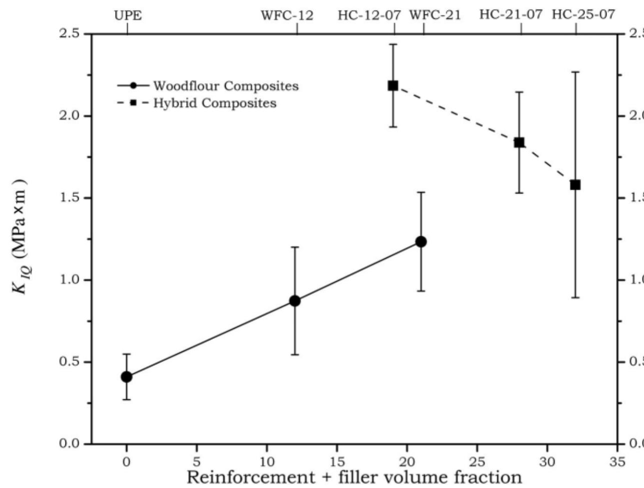

Figure 5 Stress intensity factor for neat polyester, wood particle composites, and hybrid composites with high wood particle content.

The debonded fibers presented a clean surface, which indicated a weak interface between the sisal and the polyester matrix.

The addition of sisal fibers affected the toughness of the composites strongly (Fig. 5). For instance, as 12% by volume of wood particle was added (WFC-12), the toughness K_{IQ} increased to 2.1 times the value measured for the neat matrix (UPE). Adding also a mat of long fibers at 7% by volume (HC-12–07), the K_{IQ} value became 5.3 times that of the matrix. All hybrid composites showed higher toughness values than the matrix and wood particle composites because of the development of fiber bridging and fiber pull-out in the process zone.

The lowered toughness of hybrid composites due to increased wood content deserves further discussion. It was related to the inhomogeneous dispersion of wood particles at high concentrations (HC-25–07). The microscopic details of crack growth were also different in the hybrid composites (sequence of events, interaction between mechanisms, etc.) so that simple additive toughness contributions from sisal and wood particle were not expected. A more homogeneous microstructure is most likely a desirable feature of hybrid composites with improved toughness.

Figure 6 shows the large differences in the work of fracture W_f between the hybrid and wood particle composites. This was due to the fact that the hybrids were capable of bearing load after reaching the maximum load, while the wood particle composites broke catastrophically at that point. The load at crack initiation was increased when long fibers were added. In addition, more energy was required to propagate the crack in the hybrid composites.

Hybrid composites of moderate wood particle content (HC-13-YY)

These materials showed linear and brittle stress–strain behavior during tensile tests. Strength and modulus increased as the sisal fiber content was increased (Table III). As previously observed in hybrid composites with higher wood particle content, the tensile tests ended by fracture of the matrix, with the long fibers pulled out of the matrix, but still joining the two broken specimen parts.

A relatively large data scatter was observed in the fracture parameters (Table III), which was due to the inhomogeneous distribution of the reinforcing fibers in front of the crack path. The load-displacement behavior is sensitive to local fiber volume fraction, spatial distribution, strength, and interfacial friction stress of the fibers in the path of the crack. Differences between the data corresponding to HC-12–07 (Table II) and HC-13–09 (Table III) are most likely due to the different preparation processes, only one sisal mat for

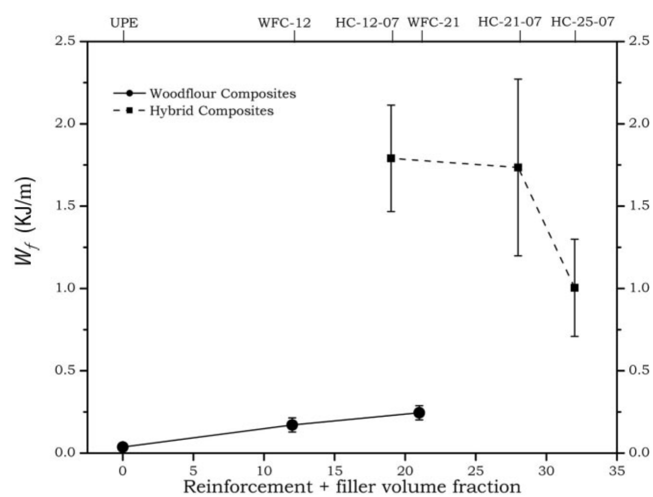

Figure 6 Work of fracture for neat polyester, wood particle composites, and hybrid composites with high wood particle content.

TABLE III
Mechanical Properties of Hybrid Composites

	Tensile Modulus (GPa)	Tensile Strength (MPa)	K_{IQ} (MPa × $m^{1/2}$)	W_f (kJ/m^2)
HC-13–09	4.73 ± 0.25	23 ± 3	1.86 ± 0.17	0.84 ± 0.12
HC-13–13	5.21 ± 0.23	22 ± 3	2.78 ± 0.20	2.43 ± 0.15
HC-13–14	5.64 ± 0.33	27 ± 2	3.67 ± 0.68	2.51 ± 0.33

Two mats of sisal fiber were used in the preparation of these materials.

the first sample and two sisal mats for the second one. The method of impregnation of one or two layers of mats certainly affected the final properties, because of the sensitivity of the parameters (especially fracture parameters) to the distribution of the reinforcing fibers.

In general, the toughness parameters increased when the fraction of long fibers was increased (Table III). Increased fiber content caused increased energy dissipation through fiber bridging and pull-out.

CONCLUSIONS

A significant toughening effect was demonstrated in wood particle composites as long sisal fibers were added. In the reference material, composites based on wood particles and polymer only, the modulus was increased whereas tensile strength decreased with wood particle content. A Halpin-Tsai analysis assuming spherical particles resulted in an effective wood particle modulus of 11.6 GPa. The decreased tensile strength was most likely due to fracture initiation from interfacial debonding of wood particles at low strain. Increased K_{IQ} toughness was measured with increased wood particle content.

Hybrid composites based on wood particle and sisal fiber mat reinforcement showed a large increase in K_{IQ} toughness and work of fracture. For instance, a composite with 12% by volume of wood particles shows a 10-fold increase in work of fracture by addition of just a 7% by volume of sisal. The addition of fibers completely changed the brittle mode of fracture in the wood particle composites. After peak load, the load-displacement curve for fracture mechanics specimens showed a much more gradual decrease. The reason was crack bridging from sisal fibers and the associated fiber pull-out process. This increased the energy required to break the specimen. As expected, a higher fiber content increased composite toughness.

The authors thank CONICET (National Research Council, Argentina) for the fellowship awarded to one of the authors (A. J. Nuñez).

References

1. Banthia, N.; Sheng, J. Cem Concr Compos 1996, 18, 251.
2. Savastano, H., Jr.; Warden, P. G.; Coutts, R. S. P. Cem Concr Compos 2003, 25, 311.
3. Kagawa, Y.; Sekine, K. Mater Sci Eng A 1996, 221, 163.
4. Kim, J. K.; Mai, Y. W. Engineered Interfaces in Fibre Reinforced Composites; Elsevier: Oxford, UK, 1998.
5. Bledzki, A. K.; Gassan, J. Prog Polym Sci 1999, 24, 221.
6. Kelly, A.; Tyson, W. R. J Mech Phys Solids 1965, 13, 329.
7. Devi, L. U.; Bhagawan, S. S.; Thomas, S. J Appl Polym Sci 1997, 64, 1739.
8. Ray, D.; Sarkar, B. K.; Rana, A. K. J Appl Polym Sci 2002, 85, 2588.
9. Marcovich, N. E.; Reboredo, M. M.; Aranguren, M. I. J Appl Polym Sci 1998, 70, 2121.
10. Shannag, M. J.; Brincker, R.; Hansen, W. Cem Concr Res 1997, 27, 925.
11. Aksel, C.; Warren, P. D. Compos Sci Technol 2003, 63, 1433.
12. Sakai, M.; Ichikawa, H. Int J Fract 1992, 55, 65.
13. Woodhams, R. T.; Thomas, G.; Rodgers, D. K. Polym Eng Sci 1984, 24, 1166.
14. Buttrey, D. N. In Polymer Engineering Composites; Richardson, M. O. W., Ed.; Applied Science: London, 1977; Chapter 12.
15. Singh, R. P.; Zhang, M.; Chan, D. J Mater Sci 2002, 37, 781.
16. Auad, M. L.; Frontini, P. M.; Borrajo, J.; Aranguren, M. I. Polymer 2001, 42, 3723.
17. Spanoudakis, J.; Young, R. J. J Mater Sci 1984, 19, 473.