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REALIZATION OF POLYPROPYLENE/SISAL FIBER COMPOSITES BY REACTIVE EXTRUSION

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ABSTRACT

Sisal fibers are used for the reinforcement of a polypropylene matrix. The compatibilization between the two materials is assumed through treatment of cellulosic fibers with a maleated polypropylene (PP-*graft*-MA). The influence of different parameters is studied: fibers fraction, washing of raw fibers, chemical treatment of the fibers in a solvent or during extrusion, rate of maleic anhydride in PP-*graft*-MA and the rate of PP-*graft*-MA in the composites. The effects of these parameters are evaluated by determination of the mechanical properties of the composites (impact strength and breaking stress) and by the quantity of water sorbed by the composites. The elimination of pectic substances by washing of the fibers is favorable for the mechanical properties of the composites and also reduces their water sorption. The grafting of the fibers by PP-*graft*-MA enhances both the impact strength and the breaking stress.

INTRODUCTION

Low cost composites have been the subject of much research because of their possible application to transportation and other industries. The materials most widely used in this field are polypropylene as matrix, due to its low price and its chemical innocuity, and glass fibers as a reinforcement. However, these fibers, which may correspond to about 30–50% of the weight of the composites, present many inconveniences: they have a high density, lead to abrasion of the processing machines, and give rise to an important residue on combustion. Their replacement by vegetal fibers not only suppress or reduces these inconveniences but also offers the possibility of reusing waste materials of agricultural origin. This is particularly important in Third World Countries or at least in countries where the climate is favorable to high production of the biomass.

Many attempts have been made using the combination between different thermoplastic matrices and vegetal materials. Besides some exotic products (sugar cane bagasse, bamboo cane, nut shell flour, sisal), wood fibers and paper pulp are often used. The preparation of pure cellulosic fibers from raw material involves elimination of pectic substances by dissolution, generally in a basic aqueous medium.

Composites are then prepared by mixing the molten matrix with the fibers at a temperature around 200°C, but the hydrophobic matrix and the hydrophilic fibers generally lead to poor adhesion. In order to improve the wettability—and thus the adhesion—of the fibers by the matrix, a coupling or compatibilizing agent is deposited on the fiber or eventually reacted with the fiber, leading to an interface or interphase of high adhesion [1–4].

Idealized chemical agents which cause good transition between PP and cellulosic fibers include a PP chain on which functions able to react with the hydroxyl groups of cellulose have been grafted: maleic anhydride (MA) is efficient for this goal. The so-called PP-*graft*-MAs are then linked to the cellulose by ester functions and assume compatibilization and adhesion with the matrix by wettability, cocrystallization, and/or entanglements [5, 6].

In the present work, sisal fibers with or without PP-*graft*-MA treatment are used for the reinforcement of a polypropylene matrix. The influences of washing and treatment of the fibers are studied by testing the mechanical properties of the composites. The effect of aging in a moisture environment on these properties is also analyzed.

EXPERIMENTAL

Materials

Whitened sisal fibers (*Agave sisalana*) from Vietnam were used as furnished by the local market and referred as to R (Raw) fibers. Washed fibers (W) are obtained by soaking the raw fibers in an aqueous solution of sodium hydroxide (18% by weight) at 80°C for 30 minutes; this gives a sufficient defibrillation. The weight loss due to dissolution of mainly the pectic cements is 24%.

The washed fibers contain cellulose (84%), hemicellulose (5%), and lignine (11%). The natural presentation of sisal consists of fibers with a length reaching 0.60 m. It will be used as chopped fibers: length 3 to 8 mm and an aspect ratio L/d of 20 to 3.

The polypropylene matrix in pellet form has a melt flow index of 45 (230°C, 2.16 kg) and a crystallinity rate of 70%. Two different PP-*graft*-MAs have been used for the compatibilization:

PP-*graft*-MA *a* with an MFI of 65 contains 0.16% by weight of MA
PP-*graft*-MA *b* is Hostaprime HC5 (Hoechst) with a higher MA content ($\approx$ 4%)

Treatment of the Fibers

The PP-*graft*-MAs are activated by heating at 180°C for 1 hour to attain the total transformation of carboxylic acids into anhydrides [5]. The grafting of PP-*graft*-MA onto the sisal fibers is performed in two different ways as follows.

Pretreatment of the Fibers in Solution (A)

Raw or washed fibers are immersed in a solution of 5% PP-*graft*-MA (calculated on the fibers) in toluene and heating at refluxing for 10 minutes. The fibers are then Soxhlet-extracted to remove all excess reagents. The grafting rate is around 0.08%. The fibers are then introduced with PP pellets into a twin-screw extruder to give granulates containing 20 or 40% by weight of fibers.

Reactive Extrusion (B)

Pellets of PP, the amount of fibers needed to reach a fiber content of 20 or 40%, and the powdered PP-*graft*-MA (3 or 5% calculated on the fibers) are granulated in a twin-screw extruder at 200°C.

Mechanical Tests on Composites

The prepared granulates are molded in the appropriate form of specimens for different mechanical tests:

Traction measurements on a DY 22 apparatus, following the norm NF-T 51-034 (5 samples)
Impact strength measurements following the norm NF-T 51-035 (20 samples)

Aging in Moisture Environment

The composite specimens are immersed in distilled water at room temperature. The weight gain is followed over time, and new mechanical tests are performed after 36 days of immersion.

The keys for codification of the composite specimens are

Fibers: Raw or washed		R	W
Fibers fraction (%)		20	40
Type of PP-*graft*-MA		*a*	*b*
PP-*graft*-MA added (%) (way B only)	0	3	5
Way of grafting: Solution (A) or extrusion (B)		A	B

RESULTS AND DISCUSSION

The factors analyzed in this work will be estimated on two measures:

The mechanical properties of composites: breaking stress and impact strength
The water sorption by the composites

Water Sorption by the Composites

Figure 1 shows that after 36 days of immersion, water sorption is still far from the equilibrium level. Figure 2 (water sorption versus the square root of time) shows that water diffusion in such composites is a non-Fickian phenomenon. The higher rate of sorption at the beginning of the experiment may be due to the emergence of fibers at the surface. This effect is more marked for a high fiber content.

Comparison between Raw and Washed Fibers

The elimination by washing of 24% of the pectic substances has consequences on the measures studied. Composites made of washed fibers compared with raw fibers show:

Higher impact strength and breaking stress (Figs. 3 and 4). It is clear that pectic cements act as a low cohesion layer
Lower water sorption (Fig. 1). Pectic cements are responsible of the high water retention. In the case of raw fibers composites, the sorption seems to have reached an equilibrium, but the apparent stabilization only reflects the

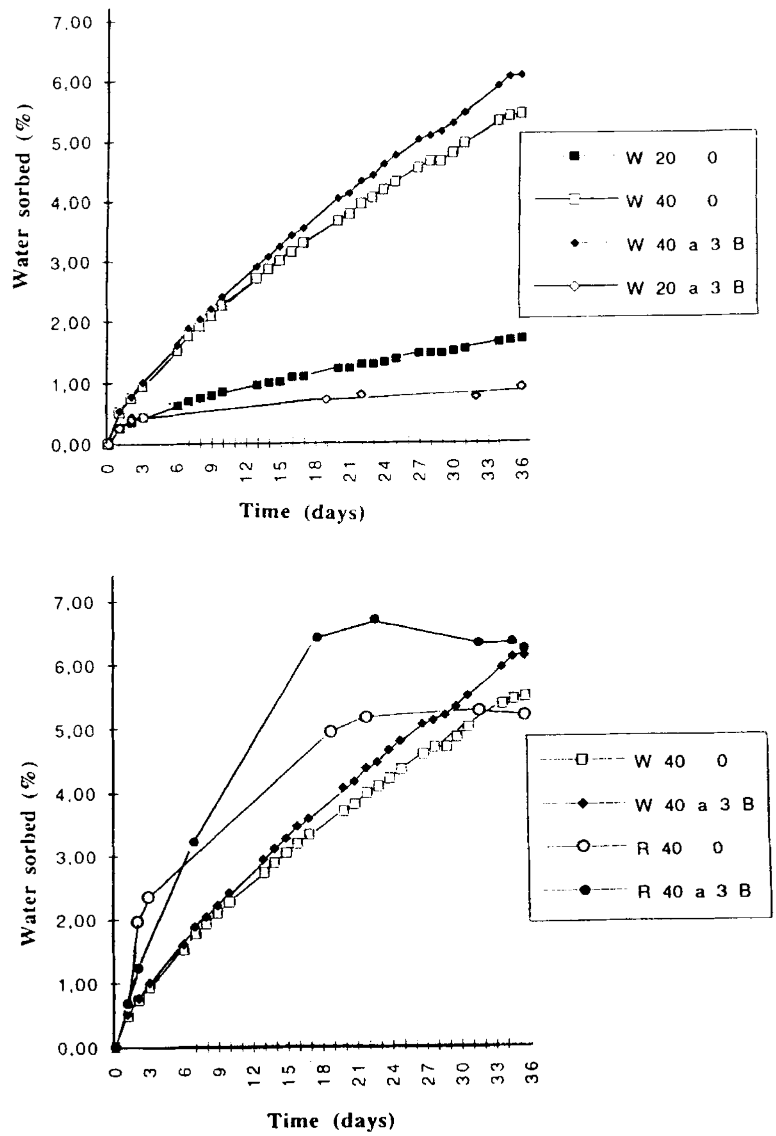

FIG. 1. Water sorption (% mass gain) versus time for PP-sisal composites immersed in water. Top: influence of fiber fraction and PPgMA treatment. Bottom: influence of washing and PPgMA treatment.

dissolution of pectic substances in the immersion bath, which takes on a brownish color

Influence of Fibers Fraction

The impact strength for composites is known to be much lower than for a pure matrix. That is also the case here, and the diminution is greater at a high fibers content (Fig. 3). Breaking stress increases with fibers content (Figs. 4 and 5) after

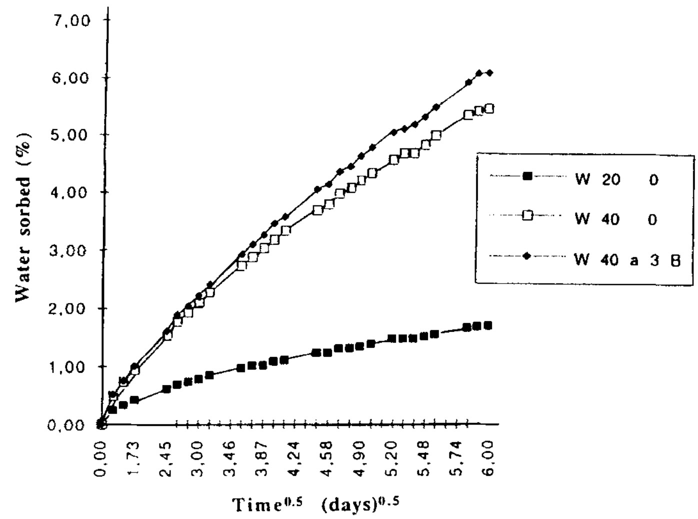

FIG. 2. Water sorption versus the square root of time for PP-sisal composites immersed in water.

washing, but with raw fibers often decreases slightly, as has been observed previously [7].

Since water sorption in composites is due to the fibers and not to the matrix, it will increase with the fibers content (Figs. 1 and 2). However, composites with 40% fibers sorb near 4 times the water sorbed by composites with 20% fibers. At too high fibers contents, voids in which water is accumulated seem to persist in the structure.

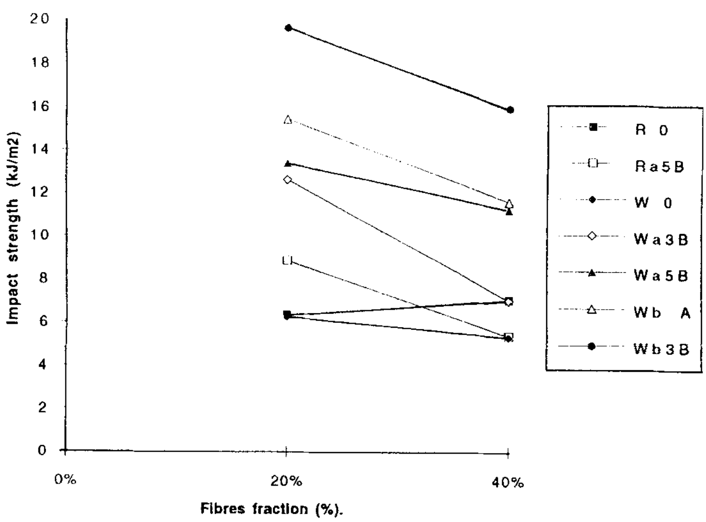

FIG. 3. Impact strength of PP-sisal composites versus fiber fractions.

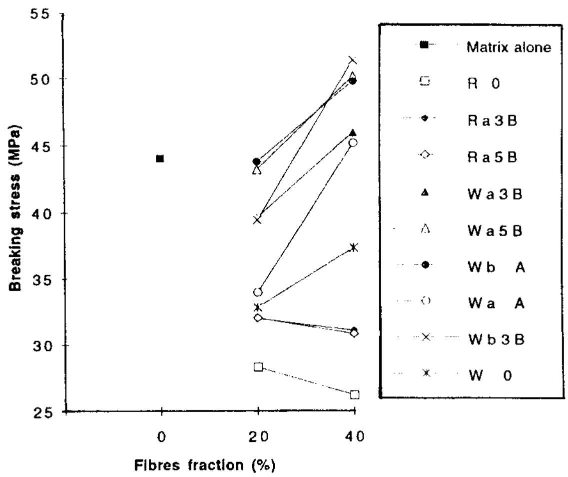

FIG. 4. Breaking stress of PP–sisal composites versus fiber fractions.

Influence of the PP-*graft*-MA Content

Treatment of the fibers with 3% PP-*graft*-MA increases the breaking stress and the impact strength in comparison with nontreated fibers. When the rate of PP-*graft*-MA is increased from 3 to 5%, a further increase is observed for washed fibers only (Fig. 5). For raw fibers, the maximum is obtained at 3% of PP-*graft*-MA.

The *b* type of PP-*graft*-MA, with a higher MA content, has a higher breaking stress than type *a* (Fig. 4).

The influence of the PPgMA treatment on the water sorption is generally small (Fig. 1) and not always in the same direction.

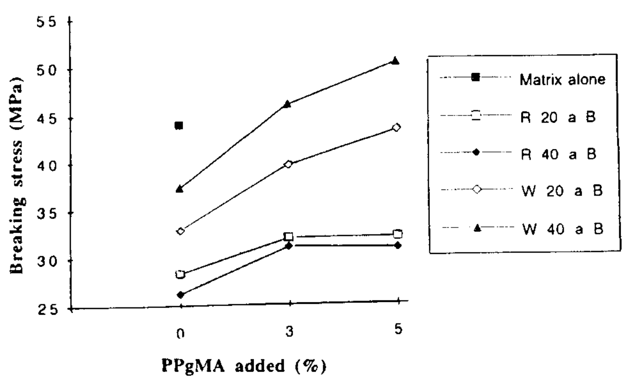

FIG. 5. Breaking stress of PP–sisal composites versus PP-*graft*-MA content.

Influence of the Method of Grafting

Direct comparisons are difficult because the quantities of PP-*graft*-MA present in the composites are different due to the two methods used:

Treatment in solution (A) give around 0.1% grafted PP-*graft*-MA onto the sisal fibers

In the extrusion method (B), only a small part of the 5% added PP-*graft*-MA is actually grafted (also at around 0.1%). The remaining amount is dispersed in the matrix and modifies the mechanical properties

The results obtained by reactive extrusion are not systematically unfavorable to mechanical properties. However, the time savings of this method are an advantage.

CONCLUSION

The grafting of sisal fibers by PP-*graft*-MA occurs normally on cellulose for washed fibers and leads to composites with higher impact strength and breaking stress than for nontreated fibers. These effects increase with the rate of PP-*graft*-MA added and with the maleic anhydride content of the PP- *graft*-MA. The water sorption by such composites is not influenced by any eventual treatment. For composites made with raw fibers, the results on the mechanical properties show an unfavorable effect of the pectic substances (low cohesion layer). Water sorption is enhanced by these substances which are slowly extracted during immersion.

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