Effects of Hygrothermal Ageing on Mechanical Properties of Flax Pulps and their Polypropylene Matrix Composites

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ABSTRACT: The aim of this work was to study the kinetics of water uptake and its influence on mechanical behavior of both flax pulps and their composites with a maleic anhydride polypropylene copolymer (MAPP) modified polypropylene (PP) matrix by immersion in distilled water at 30, 50, 70, and 100°C. Both the influence of two different MAPP compatibilizers and the optimum doses of each ones were analyzed. The kinetics of water uptake was studied from weight measurements at regular interval times. The diffusion coefficient was dependent on the immersion temperature and MAPP content. Tensile modulus and strength of single flax fiber decreased by water immersion. Both flexural strength and modulus of composites decreased as a result of the combined effect of thermal ageing and moisture absorption. MAPP coupling agent increases moisture resistance and mechanical properties for MAPP-modified systems with respect to the unmodified ones. J Appl Polym Sci 102: 3438–3445, 2006

Key words: ageing; natural fiber; composites; moisture uptake; diffusion

INTRODUCTION

In recent years there has been a rapid growth in using renewable natural fibers as reinforcements in composite materials, aiming to produce an unique combination of high performance, great versatility, light weight, recyclability, biodegradability, and processing advantages at favorable cost.[1–5] Natural fibers are hydrophilic. On the contrary, most of the thermoplastic matrices used for composites such as polypropylene (PP) have a modest cohesive energy and a strong hydrophobic character. For a profitable use of lignocellulosic fibers as reinforcing agents for thermoplastic polymers, it is essential to lower the ensuing interfacial energy. For this purpose, several studies on the effect of different coupling agents, such as silanes and maleic anhydride-grafted PP, or modification by acetilation have been carried out.[5–9] Grafting a compatible matrix on the fiber surface has also been shown to improve both fiber–matrix adhesion and the dispersion of fibers within the matrix.[10]

One of the main disadvantages linked to the use of natural fibers is their moisture uptake when exposed to environmental conditions and its influence on the properties of composites. Data on the effects of moisture retention on mechanical properties of natural fiber-reinforced composites during long term service are crucial for their outdoor applications. Sorbed moisture can interact with both polymeric matrix and natural fibers but it can also affect the fiber–matrix interface, reducing the joint strength by breaking the bonds. Knowing how a property varies with environmental conditions can help to predict the service properties of the composites. Moisture diffuses into a polymer at varying degrees depending on molecular and microstructural aspects such as polarity and extent of crystallinity.

The present work deals with the water sorption effects on flax fiber-reinforced PP matrix composites through the analysis of the mechanical behavior of flax pulps and flax pulp-reinforced composites after water immersion. The influence of both temperature and maleic anhydride polypropylene copolymer (MAPP) modifier on water sorption has also been investigated.

Kinetics of moisture sorption

Moisture absorption into composite materials is addressed by three different mechanisms. The main process consists of diffusion of water molecules inside the microholes between polymer chains. Other common mechanisms are capillary transport into the gaps and flaws at the interfaces between fibers and polymer, due to the incomplete wettability and impregnation, and also transport through microcracks in the matrix, formed during the compounding process.[11]

The three diffusion mechanisms can be distinguished theoretically by the shape of the sorption curve represented by

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$$\frac{M_t}{M_\infty} = kt^n \quad (1)$$

where M_t is the moisture content at time t; M_∞ is the moisture content at the equilibrium; and k and n are diffusion kinetic parameters. The value of coefficient n presents different behavior; for Fickian diffusion, $n = 1/2$, non-Fickian, $n = 1$, and when it lies between 0.5 and 1, the diffusion is anomalous.[11–13] Moisture uptake in natural fiber-reinforced plastics usually follows Fickian behavior.[11–22]

Fick's law predicts that the sorbed mass of water increases linearly with the square root of time, and then gradually slows until an equilibrium plateau is reached. Diffusivity has been analyzed with the hypothesis of Fickian mechanism. One-dimensional approach was followed for the determination of the diffusion coefficient, D, which can be calculated as

$$D = \pi\left(\frac{d\theta}{4\Lambda m(\infty)}\right)^2 \quad (2)$$

where θ is the slope of the linear portion of the sorption curves, d the initial sample thickness, and Δm is the mass of sorbed water at infinite time.

MATERIALS AND METHODS

Materials

The flax pulps (CF05SU) were kindly supplied by Celesa (Spain). PP Eltex-P HV200 supplied by Solvay has been used as matrix. Two different maleic anhydride PPs have been used as coupling agents: E-43 ($MAPP_1$; M_n = 3900, M_w = 9100, and acid number 45) and G-3003 ($MAPP_2$; M_n = 27,200, M_w = 52,000, and acid number 8), both kindly supplied by Eastman.

Single fiber testing

Single fibers were carefully separated from the bundles manually and both fiber ends were glued on pieces of paper (paper tabs of size 5 × 10 mm^2) for handling purposes. Transparent two-component epoxy glue was used for this purpose. Fibers were tested after 2 and 7 days immersion in water. Fiber diameter was evaluated from optical microscopy images. An average of five apparent diameter measurements was taken at different locations along the fiber. Tensile tests were performed in a Minimat miniature mechanical test machine using a 20 N load cell and loading rate was 0.1 mm/min.

Processing

Test specimens containing 30 wt % carefully dried flax fibers (vacuum oven, 90°C for 24 h) were processed by extrusion mixing and further injection molding. For extrusion, a conical twin-screw extruder, Haake Rheomex CTW 100, was used. For both types of MAPP, 2, 4, 7, 10, and 15 wt % with respect to the flax/PP composite were added. The rotor speed was maintained constant at 30 rpm and temperature varied between 180 and 185°C. Samples for flexural testing were molded by injection molding using a Battenfeld Plus 250 injection machine, working at 175°C with 1057 bar injection pressure.

Hygrothermal ageing

To analyze the moisture content of flax pulp/PP composites, the maximum moisture content M_m and the diffusion coefficient have been calculated from Fickian representation. Prior to soaking them, composites were dried in a vacuum oven at 90°C until a constant weight was achieved. Flexural testing samples were then soaked in distilled water at different temperatures 30, 50, 70, and 100°C. Mass changes of the samples were recorded using an electronic balance at regular time intervals, immersing them again in water until equilibrium plateau was reached. Before each measurement the samples were blotted using a piece of filter paper.

Mechanical characterization of composites

Three-points bending tests were carried out using a universal mechanical testing machine Instron model 4206 with a 1 kN load cell. Rectangular samples of 80 × 10 × 4 mm^3 with a length span of 64 mm were tested at a test rate of 2.1 mm/min according to ASTM D-790M standard. The study on the effect of hygrothermal ageing on flexural properties was performed using samples subjected to water immersion up to 3 months.

RESULTS AND DISCUSSION

Kinetics of moisture sorption

As other natural fibers, flax pulps exhibit considerable moisture uptake due to its hydrophilic nature. Moisture is sorbed in their polymeric composites mainly by moisture sorption into the free volume present in the structure and by hydrogen bonding between hydrophilic groups of water and —OH groups of fibers.[11–14] Microcracks can also pave the way to moisture transport involving flow and storage of water within the cracks.[12–20,21]

The analysis of the diffusion kinetics was based on the Fick's theory. Values of the kinetic parameters n and k for moisture uptake data of flax pulp/PP composites were calculated using eq. (1). Figure 1 shows an example of the fitting of the experimental data to

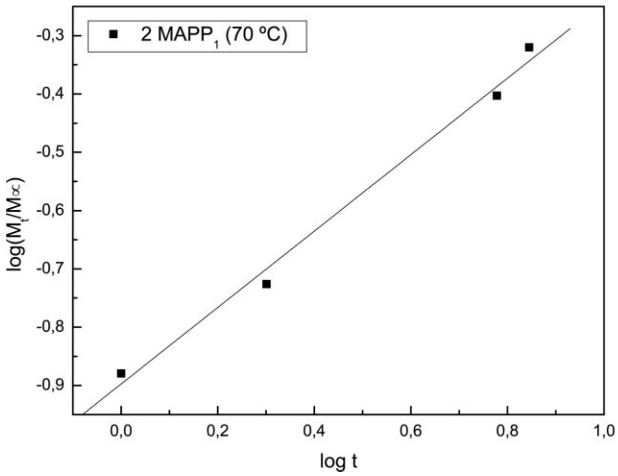

Figure 1 Diffusion case fitting plot for flax pulp/PP composites modified with 2 wt % $MAPP_1$.

TABLE II
Diffusion Kinetic Parameter *k* for Flax Pulp/PP Composites Immersed in Water at Several Temperatures

	k			
Composite	30°C	50°C	70°C	100°C
Unmodified	0.117	0.112	0.127	0.213
$MAPP_1$				
2	0.126	0.104	0.133	0.239
4	0.164	0.101	0.111	0.265
7	0.102	0.100	0.147	0.183
10	0.211	0.184	0.201	0.231
15	0.168	0.088	0.098	0.149
$MAPP_2$				
2	0.111	0.116	0.134	0.190
4	0.104	0.131	0.144	0.218
7	0.101	0.112	0.163	0.195
10	0.089	0.119	0.198	0.203
15	0.094	0.139	0.188	0.203

eq. (1). It is clear from Table I that in the case of flax pulp/PP composites, with a value of n about 0.5, the sorption process shows a tendency to approach Fickian behavior. In the case of $MAPP_1$, the Fickian diffusion case is more evident, but for $MAPP_2$, values deviate slightly from Fickian behavior. Similar observations were reported by other authors for other natural fiber-reinforced PP composites.[11,12,20] Table II lists the corresponding diffusion parameter k, which has increasing tendency observed with the temperature.

Effect of immersion temperature

The sorption of water by nonpolar polymers containing fibers depends mainly on the nature of the filler. For hydrophilic fibers, such as lignocellulosic fibers, an increase in water sorption can be expected due to their tendency to retain water in the interfibrillar space beside their hydrophility.[12,14]

Figure 2 shows the effect of temperature on moisture uptake of flax pulp/PP composites for four different temperatures. M_t increases in the initial stage, and then tends to a plateau when complete saturation is reached. This reveals that the moisture uptake kinetics follows Fickian law.[11,12]

The straight fitting against square root of time indicates that the porous structure of flax fiber leads to an initial capillary uptake.[11,20] A quantitative comparison between the results shown in Figure 2 reveals that increasing the temperature seems to reduce the time for achieving the equilibrium plateau. This supports the fact that temperature activates the diffusion process.

TABLE I
Diffusion Kinetic Parameter *n* for Flax Pulp/PP Composites Immersed in Water at Several Temperatures

	n			
Composite	30°C	50°C	70°C	100°C
Unmodified	0.542	0.594	0.655	0.409
$MAPP_1$				
2	0.566	0.514	0.547	0.539
4	0.430	0.591	0.519	0.484
7	0.524	0.585	0.469	0.433
10	0.235	0.601	0.498	0.436
15	0.414	0.552	0.633	0.517
$MAPP_2$				
2	0.452	0.444	0.526	0.675
4	0.362	0.449	0.529	0.641
7	0.512	0.468	0.487	0.724
10	0.353	0.428	0.430	0.652
15	0.418	0.452	0.521	0.652

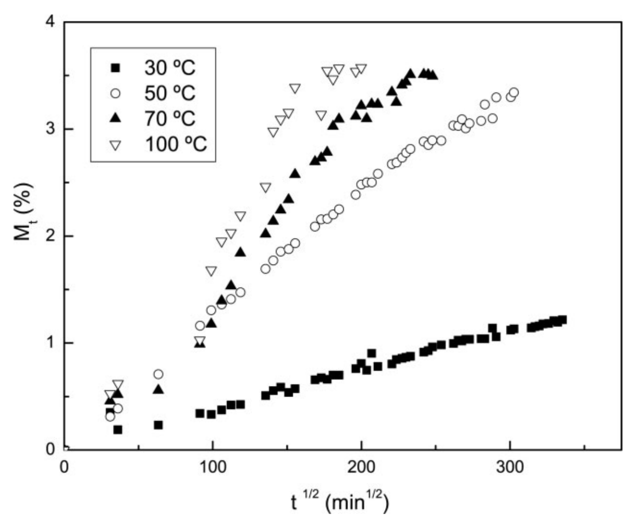

Figure 2 Effect of temperature on the moisture uptake behavior of flax pulp/PP composites.

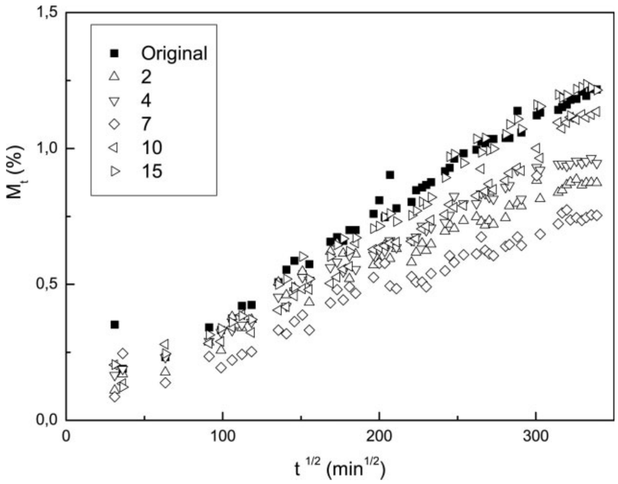

Figure 3 Effect of $MAPP_1$ coupling agent amount on the water uptake behavior of flax pulp/PP composites at 30°C.

TABLE III
Diffusivity Coefficient for Flax Pulp/PP Composites After Water Immersion at 30, 50, 70, and 100°C ($D \times 10^{13}$ m^2/s)

Composite	*T* (°C)			
	30	50	70	100
Unmodified	0.548	5.90	11.23	30.03
$MAPP_1$				
2	0.516	5.90	6.37	18.86
4	0.354	5.39	4.25	15.50
7	0.239	5.45	4.28	16.43
10	0.263	1.95	8.51	19.18
15	0.291	5.87	7.88	19.99
$MAPP_2$				
2	0.479	4.59	7.65	23.12
4	0.408	5.60	7.71	25.97
7	0.351	4.27	7.34	28.88
10	0.352	6.52	8.93	25.29
15	0.355	6.92	7.12	25.98

Effect of coupling agent

The effect of coupling agent on water sorption can be understood from the respective sorption curves. As shown in Figure 3, the improved moisture resistance at each analyzed time caused by addition of $MAPP_1$ can be explained by an improved fiber/matrix adhesion. The reduction in moisture level can be attributed to improved interfacial adhesion that reduces water accumulation in the interfacial voids and decreases volume fraction of fiber in contact with water, thus preventing water from entering flax fiber.[11,15,21] Some hydroxyl groups of cellulose and other fiber constituents become chemically bonded with MAPP coupling agent; therefore, the hydrophilic character of the fiber is reduced.[12,16,19] However, increasing $MAPP_1$ content up to 7 wt % leads to increase in water diffusion inside the micrograps between polymer chains.[12] Figure 4 shows the sorption behavior of $MAPP_2$-treated composites. It is worth noting that the water uptake also decreases with increasing $MAPP_2$ coupling agent content. Both coupling agents sorption tendencies as a function of their content are slightly different due to their different acid number and molecular weight.[10,16] Acid number measures the functionality present in the coupling agent, being dependent on MA units grafted per polymeric chain.[10,16] $MAPP_2$ has lower amount of maleic groups per chain length, and so at low modifier content, it does not have enough maleic groups to produce an optimal coupling efficiency. So, the probability to block fiber matrix linkages is low and the optimum dose is achieved at 7 wt % $MAPP_2$. On the contrary, as MA content in $MAPP_1$ is higher, after adding only 2 wt % $MAPP_1$, the chance for fiber–matrix linkages increases. Compatibilizer content beyond 7 wt %, the optimum concentration for $MAPP_1$, decreases moisture resistance since the chance for the

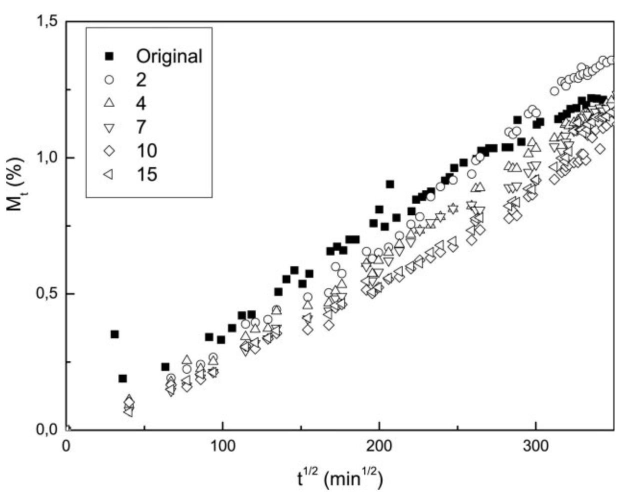

Figure 4 Effect of $MAPP_2$ coupling agent amount on the water uptake behavior of flax pulp/PP composites at 30°C.

TABLE IV
Arrhenius Parameters for Flax Pulp/PP Composites

Composite	Arrhenius parameters	
	E_a (kJ/mol)	D_0 ($\times 10^{10}$ m^2/s)
Unmodified	2.32	9.85
$MAPP_1$		
2	1.68	1.37
4	1.93	3.69
7	2.34	9.16
10	2.16	6.60
15	2.81	37.8
$MAPP_2$		
2	2.86	51.3
4	2.86	44.0
7	3.05	68.4
10	3.21	129
15	3.25	153

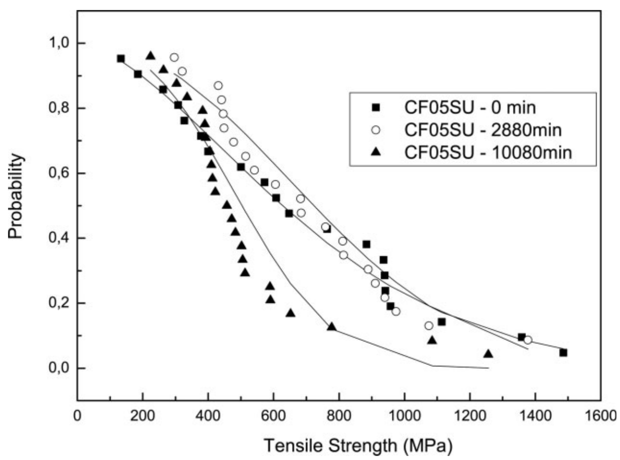

Figure 5 Weibull statistical distribution of tensile strength of flax pulp after immersion at different times: (■), 0 min; (○), 2 days; and (▲), 7 days.

coupling agent to react with the fibers is lower than the one that react with themselves.[16]

Diffusion coefficients

The diffusion coefficient characterizes the ability of water molecules to diffuse through the composite. The values of D coefficient obtained in the present study are summarized in Table III. Values are similar to the values reported by other authors[13–16] being minimal at 30°C. The presence of MAPP in flax pulp/PP composites results in a slight reduction in the values of D as compared to neat flax pulp/PP composites. The improvement in the interfacial bonding between PP matrix and flax pulp is believed to give further hindrance on the moisture absorption into the composites. Joseph et al.[13] found that the use of coupling agents lowers the diffusivity for sisal fiber-reinforced PP. The lowest values appear for 2–7 wt % $MAPP_1$ and about 4–7 wt % for $MAPP_2$ at all times that, as shown below, corresponds to the better mechanical performance. This can be due to the rate of the diffusion processes decreases since there are less gaps in the interfacial region and also more hydrophilic groups are blocked by the coupling agent.[11]

The Fickian diffusion coefficient is related to temperature by the Arrhenius relationship:

$$D = D_0 \exp\left(\frac{-E_a}{RT}\right) \tag{3}$$

where D_0 is the diffusivity index, E_a is the activation energy of the diffusion process, R is the universal gas constant, and T is the absolute temperature. Table IV presents the results of the Arrhenius parameters.

The values of the activation energy vary from 1.68 to 3.25 kJ/mol, being similar to those reported by other authors for PP and different types of natural fibers.[11,12]

Flax pulp tensile testing

Amorphous cellulose, as well as other fiber constituents, play a cementing role in the three-dimensional network of the fiber structure, also accounting for their high elongation. During the sorption process as was published by Sreekala and Thomas,[20] the microporous nature of the flax pulp helps the retention of moisture on the fibers by thus entering water molecules into the spaces between the cellulose fibrils. The presence of water molecules destroys the network cementing ac-

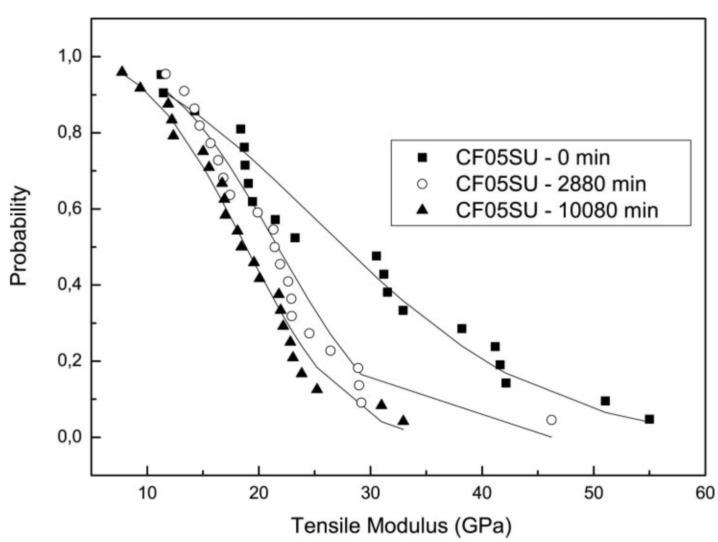

Figure 6 Weibull statistical distribution of tensile modulus of flax pulp after immersion at different times: (■), 0 min; (○), 2 days; and (▲), 7 days.

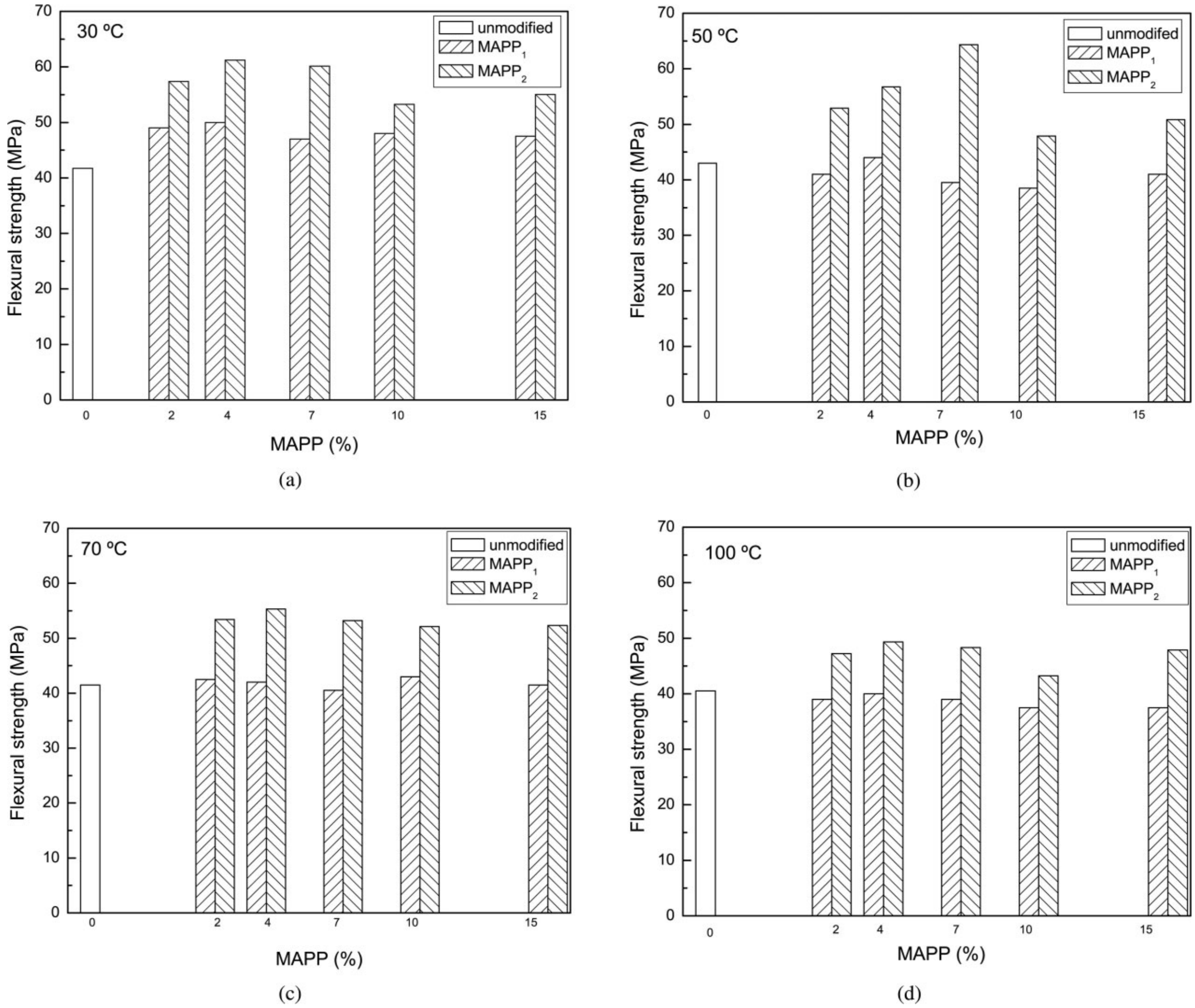

Figure 7 Flexural strength for flax pulp/PP composites after water immersion during 3 months: (a) 30, (b) 50, (c) 70, and (d) 100°C.

tion, thereby decreasing mechanical properties of the fibers.[11,14]

Micromechanical properties of flax pulps after water immersion at 30°C are shown in Figures 5 and 6. Some studies[23,24] have reported that environmental temperature in the range of −80 to 70°C has a limited effect on tensile strength, with a small decrease of strength at higher temperatures related to the loss of water. The tensile properties of flax fiber depend on the direction of the fiber and exhibit considerable scatter, thus needing a statistical treatment. The tensile strength variation of the analyzed flax fibers was obtained by application of the Weibull distribution. The two parameter Weibull distribution is represented by

$$F(\sigma) = \exp\left[-\frac{\sigma}{\beta}\right]^{a} \quad (4)$$

Equation (4) is found to provide good agreement for single flax fiber strength data, where $F(\sigma)$ is the probability of survival at stress, α the shape parameter, and β the scale parameter of the Weibull distribution.[23,24] Tensile strength decreases about a 20% after a week of water immersion from 786 to 580 MPa. Tensile modulus decreases from 32 to 21 MPa. Under the conditions of the experiment, the reason for this decrement in mechanical properties can be due to the destruction of amorphous region by water penetration through the interface and subsequent diffusion through the porous structure.[11,13,20] Similar results were founded by Sreekala and Thomas[20] for oil palm fiber.

Effect of water on mechanical properties of composites

The flexural strength of composites is higher than that for neat PP because the fibers have a higher stiffness than the polymer.[25,26] Moreover, there are many studies concerning the action of MAPP, and most of them show high increases on strength of MAPP-treated composites.[10,16]

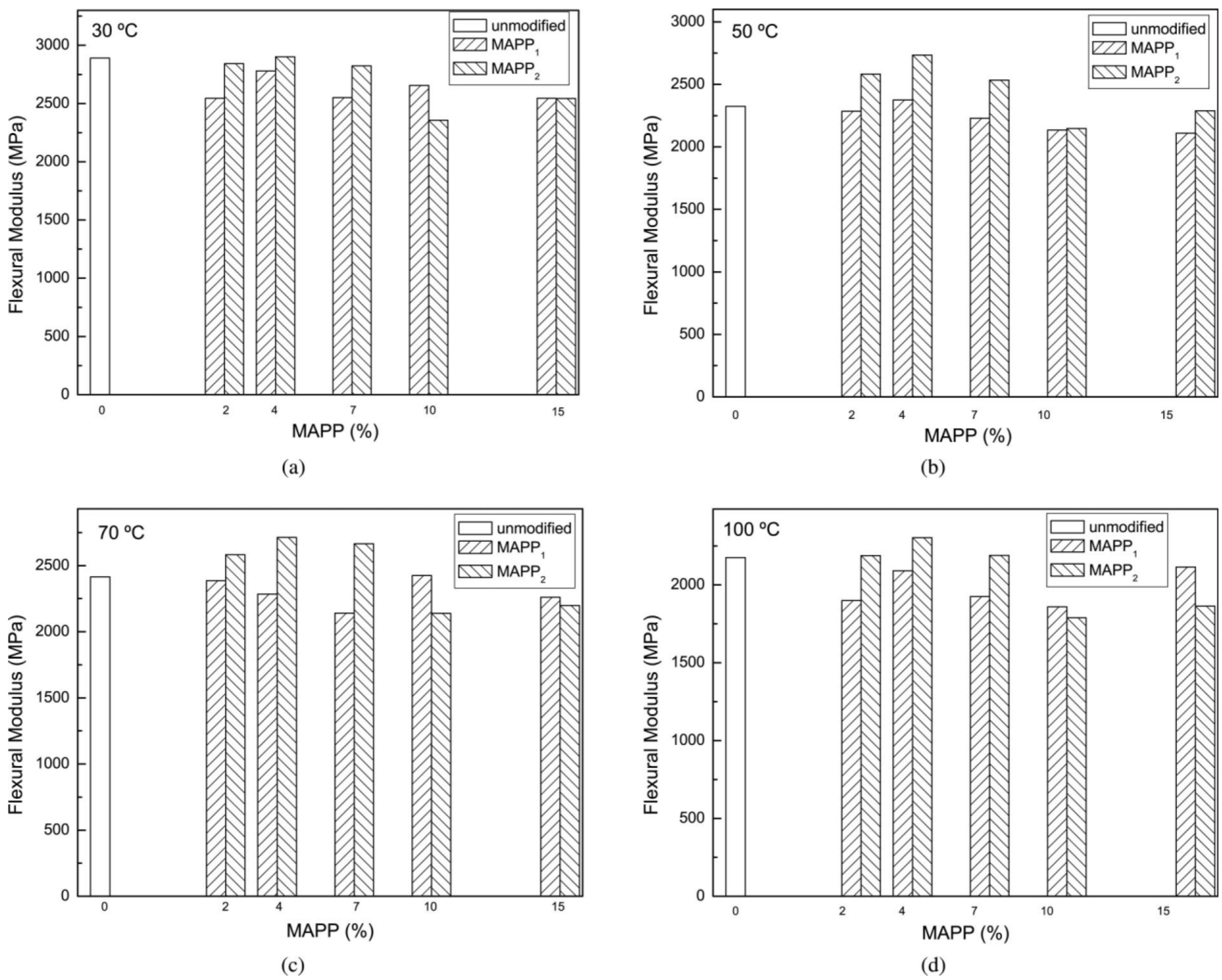

Figure 8 Flexural modulus for flax pulp/PP composites after water immersion during 3 months: (a) 30, (b) 50, (c) 70, (d) and 100°C.

In general, mechanical properties of composite materials decrease after moisture uptake, due to the effect of water molecules, which change the structure and properties of the fibers, matrix, and the interface between them.[11] The presence of water induces to a plasticization effect so that the mechanical behavior of the composites is reduced. It is known that when plant fibers begin to swell in the composites exposed to humidity, interfacial shear stress can be developed owing to the increasing osmotic pressure, leading to adhesive debonding. Water diffuses through the matrix and reaches the interphase region on the fiber. As the efficiency of stress transfer is closely associated with the fiber–matrix interfacial bonding, there is a great possibility that hygrothermal ageing at higher temperatures disrupts the bonding quality between fiber and matrix.[12–14] Similar observations have been found by Ishak et al.[15] for rice husk PP composites. Espert et al.[11] investigated the effect of different humidity conditions on crops reinforced PP composites finding that as humidity and temperature increased mechanical properties decreased, since hygrothermal ageing may also lead to the degradation of natural fibers by a hydrolysis mechanism.

The effect of immersion temperatures on the flexural strength and modulus of composites containing both types of MAPP are shown in Figures 7 and 8, respectively. Although water immersion reduces flexural properties, reduction is less marked for MAPP-modified composites above all low temperatures, and so the use of compatibilizer reduces the extent of degradation during ageing. For $MAPP_1$-modified samples reduction in mechanical properties with temperature is more evident, and at 100°C, flexural properties of $MAPP_1$-modified samples are lower than for unmodified ones. Similar observations were reported by Thwe et al.[22] for bamboo–glass fiber-reinforced PP composites and MAPP coupling agent where mechanical property retention was better with PP-MAPP matrix. Ishak et al.[15] found that the presence of 3-APE (3-aminopropyltriethoxylane) coupling agent in short glass fiber/poly(butylenes terephtalate) composites enhanced the mechanical performance of the composite especially in hygrothermal ageing conditions. The reduction in mechanical properties for $MAPP_2$-modified composites is not so evident after hygrothermal ageing, which can be attributed to more entanglement between fiber and matrix due to its higher molecular weight of $MAPP_2$. Felix et al.[27] found that for cellulose/PP composites modified with two different MAPP coupling agents, with low and high molecular weight, the longer the PP chain of the modifier the better tensile properties were. They suggested that high molecular weight MAPP has more flexible PP chains able to diffuse into the matrix. Thus, MAPP chains become more involved in interchain entanglements and thereby they contribute to the mechanical continuity of the system.

In comparison with the strength plots given in Figure 7, the decline of modulus is more obvious. The stiffness of cellulose fibers drops considerably after being aged in boiling water due to the softening of the desorbed zones of microfibrils.[11,26] Substantial water uptake would lead to the formation of hydrogen bonding between cellulose and water molecules so that the amount of intermolecular hydrogen bonding in cellulose fibers would be reduced accordingly.

CONCLUSIONS

Water uptake on flax fiber/PP composites containing two different MAPP coupling agents has been investigated at 30, 50, 70, and 100°C. Under the studied experimental conditions, the diffusion mechanism for all systems has been found to follow the predictions of Fick's law, where the amount of sorbed moisture increases linearly with the square root of time and then gradually slows down until an equilibrium moisture content is reached. MAPP-modified composites exhibit a reduction in water uptake; however, each coupling agent shows different tendency. $MAPP_1$-modified composites present an optimum dose between 2 and 7 wt % where the water uptake is the lowest. Nevertheless, a continuous decrease in water uptake with the coupling agent content has been observed for the $MAPP_2$-modified composites not finding significant differences after 7 wt % of $MAPP_2$ addition. The apparent diffusion coefficient has been found to be strongly dependent on the immersion temperatures and MAPP coupling agent. Increasing immersion temperature increases diffusivity. Diffusion coefficient is lower with optimum dose as a result of less water uptake. Micromechanical properties of flax fiber are also reduced by water. Increasing immersion time decreases fiber tensile strength. Hygrothermal ageing also reduces flexural properties. Mechanical properties are dependent on MAPP coupling agent content and immersion temperature. As MAPP coupling agents decrease water uptake, subsequently the reduction of mechanical properties by water increase is not so evident. Increasing immersion temperature the decline in mechanical properties is drastic.

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