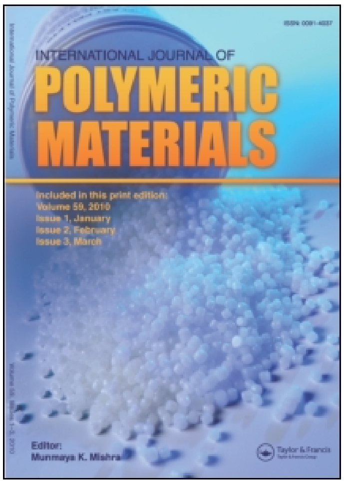


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# Water Absorption Behavior and Impact Properties of *Spartium Junceum* Fiber Composites

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# Water Absorption Behavior and Impact Properties of *Spartium Junceum* Fiber Composites

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*This paper presents the influence of* Spartium junceum *(SJ) fiber content, surface treatment and temperature on the water uptake of polypropylene*/Spartium junceum *fiber (PP/SJ) composites. Composites specimens were dried at 70°C to reach a constant weight and then were submerged in distilled water at different temperatures, 23°C and 85°C. Water uptake of PP/SJ fiber composites was found to increase with fiber content. Impact strength properties are dramatically affected by the water absorption. Water-saturated samples present poor impact strength. The SEM micrograph of* Spartium junceum *fiber untreated and treated with silane (Z-6020) illustrate the reduction of roughness via surface treatment of fiber.*

**Keywords:** Impact strength, polypropylene, *spartium junceum* fiber, surface treatment, water absorption



## INTRODUCTION

There has been a tremendous growth in polymer matrix composites containing vegetable fibers. Broom, hemp, sisal, jute, cotton and flax are the fibers most commonly used to reinforce polymers such as polyolefins, polystyrene, epoxy resins and unsatured polyesters. Most of the present applications are in the automotive sector and include composite parts produced by means of thermoforming or thermocompression molding techniques [1].

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The use of vegetable fibers in thermoplastics is beneficial because of their renewability, biodegradability, low density, high stiffness and relatively low price [2–8]. Vegetable fibers in general are hydrophilic in nature as they are derived from cellulose which contains strongly polar hydroxyl groups [9,10]. Due to the inherently poor compatibility between the hydrophilic vegetable fibers and hydrophobic thermoplastic, a treatment of the fiber surface is generally required [11,12]. The compatibility between composite components was improved using either physical or chemical modification of the fibers or polymer by using coupling agents [13–15].

Coupling agents based on alkoxysilanes are frequently used for vegetable fibers. Hydrolysis of the alkoxyl groups with subsequent reaction with hydroxyl groups from cellulose can provide chemical bonding with the fibers [15,16]. Nekkaa et al. [17] found that the addition of 30 wt% of *Spartium junceum* fibers treated with 1.5 wt% of silane N-[-3-Trimethoxysilylpropyl]ethylenediamine in PP results in increase of storage modulus.

Because of the presence of hydroxyl groups in various constituents of fibers, moisture regain is very high. The environmental performance of such composites is also poor due to delamination under humid conditions. The high moisture regain of the fibers leads to a reduction in the properties of the composites [18–20]. In some cases it was verified that the use of coupling agents also served to moderate and somewhat mitigate moisture movement though the composite, thus improving the mechanical properties of the materials [15]. Therefore, it is important to study in detail the water absorption behavior in order to estimate not only the consequences of water absorption, but also how this water uptake can be minimized in some way.

The aim of this study is to determine the influence of *Spartium junceum* fiber content, surface treatment by silane and temperature on the water uptake of PP/SJ fibers composites, and the effect of moisture content on the impact properties of PP/SJ fibers composites. Specimens were dried at 70°C to reach constant weight and then were submerged in distilled water at two temperatures, 23°C and 85°C. The effect of the addition of the silane coupling agents on the morphology of the composites was studied by scanning electron microscopy (SEM).

## EXPERIMENTAL

### Materials

The polymer matrix used in this study was polypropylene (PP) B-UP 123 (Exxon Mobil Chemical), having a density of 905 kg/m$^3$ and a melt flow index (MFI) experimentally determined (8.7 g/10 min at 230°C). Polypropylene was selected as the matrix because it is one of the major commodity plastics which may be processed below the decomposition temperature of cellulosic fibers (about 220°C).

*Spartium junceum* fiber was prepared in our laboratory. The *Spartium junceum* fibers were obtained from local sources, the shrub can be cultivated manually, and the fiber was cleaned and chopped into the desired length ranging from 2 to 4 mm.

## Pretreatment of *Spartium Junceum* Fibers

As pretreatment, the fibers were dewaxed by stirring in toluene/ethanol solution (2:1) for 24 h to remove the seizing (potato starch and waxes) followed by washing of the fibers in distilled water. After filtration, the fibers were dried at 105°C for 15 h.



## Surface Modification of *Spartium Junceum* Fibers

N-[-3 Trimethoxysilyl propyl] ethylene diamine was dissolved in distilled water. Then, the *Spartium junceum* fiber was immersed in the solution and kept there for 15 h with stirring at ambient temperature. The *Spartium junceum* fiber was filtered and then kept in the oven at 105°C for 15 h. The quantity of silane used was calculated at different concentrations: 0.5, 1, 1.5, and 2 wt% of the fiber content.

## Compounding and Processing

The composite materials were prepared by mixing the polymer matrix and the fibers in a two-roll mixer at 180°C. Different PP/SJ fibers composites were prepared; the *Spartium junceum* fiber amounts employed were 10, 20, 30, 40, and 50 wt%.

Pressed specimens of the composites were obtained by compression molding at 190°C under a pressure of 250 Kg/cm$^2$, followed by air cooling.

## Water Absorption

Water absorption studies were performed following the ASTM D 570-98 standard. Specimens were dried at 70°C to reach a constant weight and then were submerged in distilled water at the temperatures of 23°C and 85°C.

Mass uptakes of the samples were measured periodically by removing them from the water bath. The samples were wiped with tissue paper to remove the surface water before weighing. Water uptake was calculed using the equation below:

$$Water\ uptake(\%) = \frac{(Mt - Mo)}{Mo} \cdot 100$$

where, $Mt$ is the mass of sample at time $t$, Mo the mass of sample at $t = 0$.

## Impact Test

Impact testing was performed using the ASTM D256 Charpy impact method using a Ceast pendulum impact instrument. The capacity of the pendulum is 7.5 Kg.

## Microscopy Observations

The scanning electron microscope (SEM) Leica Sterosca 440 was used to investigate the morphology of untreated and treated fibers and the interface between the fiber and matrix of fractured surfaces of PP/SJ fiber composites.



# RESULTS AND DISCUSSION

## Effect of Content of Fibers on Water Absorption of PP/SJ Fiber Composites

Figure 1 shows the percentage of water absorption plotted against time for PP/untreated SJ fiber composites. In this figure, it is observed that the water absorption increases with increase in the SJ fiber content in the composite. This is completely logical, since PP is hydrophobic and fibers are hydrophilic. When the amount of fiber is increased in the composite, its polar character increases and hence the water retention increases.

The hydrophilic character of vegetable fibers is responsible for the water absorption in the composites, and therefore a higher content on fibers leads to a higher amount of water absorbed. These results are in agreement with those presented by Espert et al. [21] and Ichazo et al. [22].

## Effect of Temperature on the Water Absorption of PP/SJ Fiber Composites

The temperature of the absorption process also has an influence on the water absorption of composites. Figure 2 shows the variation of water

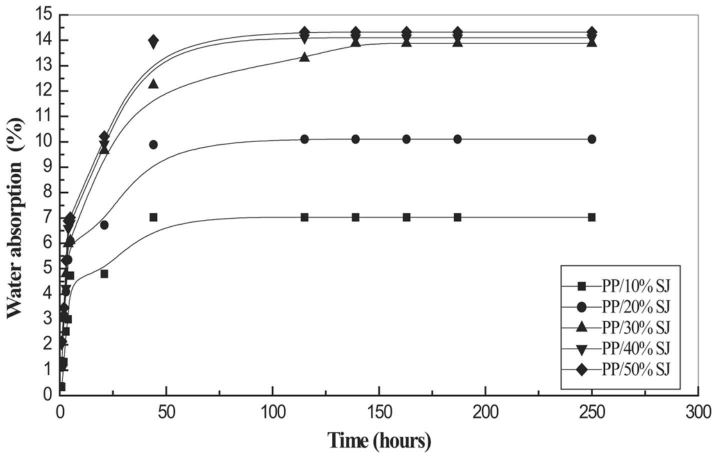



**FIGURE 1** Effect of content of fibers on the water absorption of PP/untreated SJ composites, at T = 23°C.



absorption with time for PP/untreated SJ fiber (70/30) composites at different temperatures (23°C and 85°C).

We observed a decrease in saturation time when the temperature of immersion increases. When the temperature is increased from 23°C to 85°C, the saturation is reduced by 70 h for the PP/untreated SJ fiber (70/30) composites.

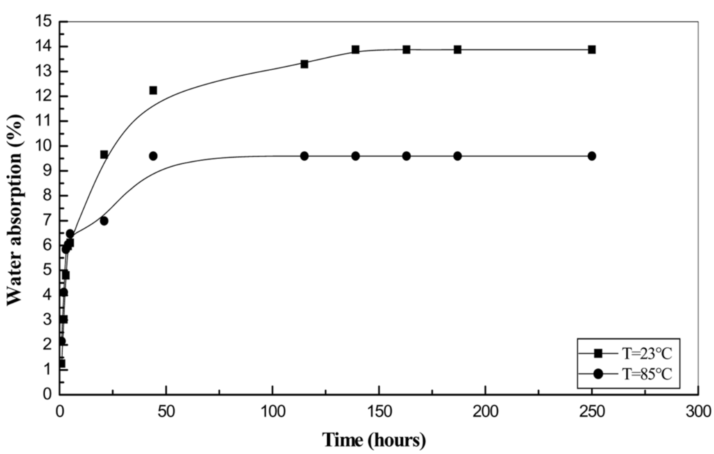



**FIGURE 2** Effect of temperature on the water absorption of PP/untreated SJ fiber (70/30) composites.

## Effect of Treatment on the Water Absorption of PP/SJ Fiber Composites

On the other hand, the fibers' surface treatment with N-[-3-Trimethoxysilyl propyl]ethylenediamine (Z-6020) at different concentrations influences the water absorption of PP/SJ fiber composites. This is presented in Figure 3. The results indicate that PP/SJ fiber composites exhibit low water absorption. This could be attributed to the lower amount of free OH in cellulose because some of them could interact with the coupling agent [22,23].

## Kinetics of the Water Absorption

In general, diffusion behavior in polymers can be classified according to the relative mobility of the penetrant and of the polymer segments. With this, there are three different categories of diffusion behavior [21].

1. *Case I, or Fickian diffusion, in which the rate of diffusion is much less than that of polymer segments' mobility*. The equilibrium inside the polymer is rapidly reached and it is maintained with independence of time.
2. *Case II, in which penetrant mobility is much greater than other relaxation processes*. This diffusion is characterized by the development of boundary between the swollen outer part and the inner

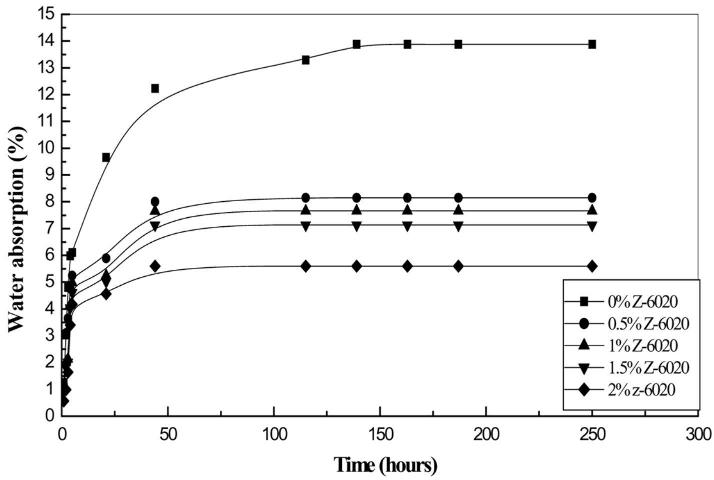



**FIGURE 3** Water absorption curves of PP/treated SJ fibers (70/30) at different concentrations of Z-6020, at $T = 23°C$.

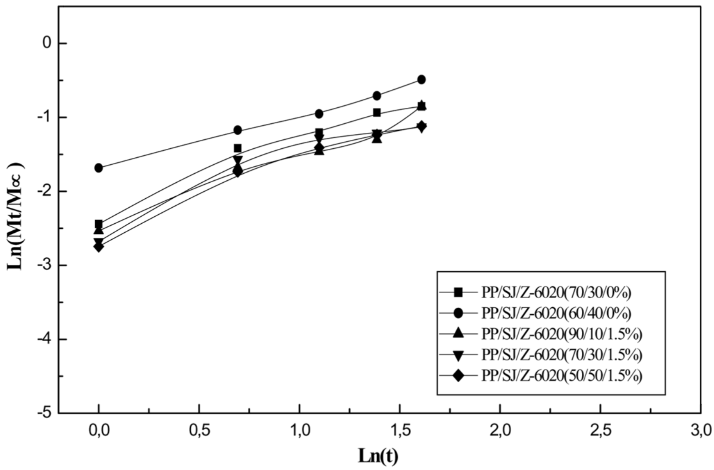



**FIGURE 4** Diffusion case fitting plots for PP/SJ fiber composites at T = 23°C.



core of the polymer. The boundary advances at a constant velocity and the core diminishes in size until an equilibrium penetrant concentration is reached in the whole polymer.

3. *Non-Fickian diffusion occurs when the penetrant mobility and the polymer segment relaxation are comparable*. It is then, an intermediate behavior between Case I and Case II diffusion.

These three cases of diffusion can be distinguished theoretically by the shape of the sorption curve represented by:

$$M_t/M_\infty = k \cdot t^n$$

**TABLE 1.** Values of the Parameters n and k for PP/SJ Fiber Composites at T = 23°C

| PP/SJ fiber Composites | k | n |
|---|---|---|
| 70/30/0% Z-6020 | 0.10 | 0.,52 |
| 60/40/0% Z-6020 | 0.18 | 0.53 |
| 80/20/1% Z-6020 | 0.05 | 0.60 |
| 60/40/1% Z-6020 | 0.97 | 0.50 |
| 90/10/1.5% Z-6020 | 0.08 | 0.43 |
| 70/30/1.5% Z-6020 | 0.07 | 0.60 |
| 50/50/1.5% Z-6020 | 0.06 | 0.57 |
| 80/20/2% Z-6020 | 0.07 | 0.60 |
| 70/30/2% Z-6020 | 0.10 | 0.57 |

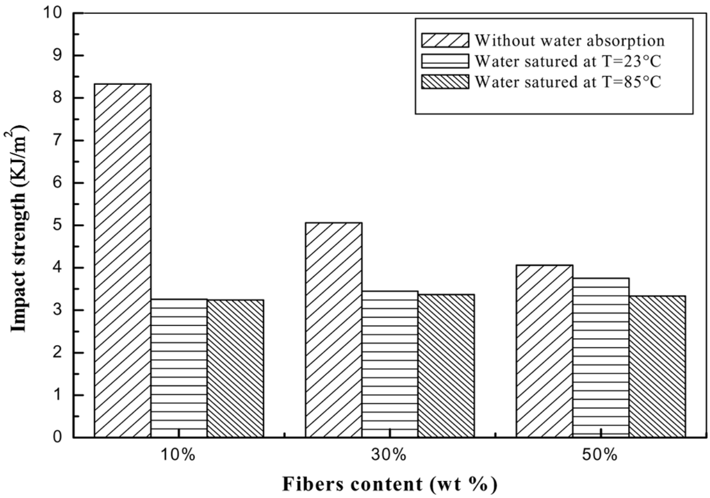



**FIGURE 5** Effect of the water absorption on the impact strength for PP/untreated SJ composites.



where $M_t$, is the moisture content at time; $M_\infty$ is the moisture content at the equilibrium; and k and n are constants. The value of the coefficient n shows different behavior between cases; for Fickian diffusion

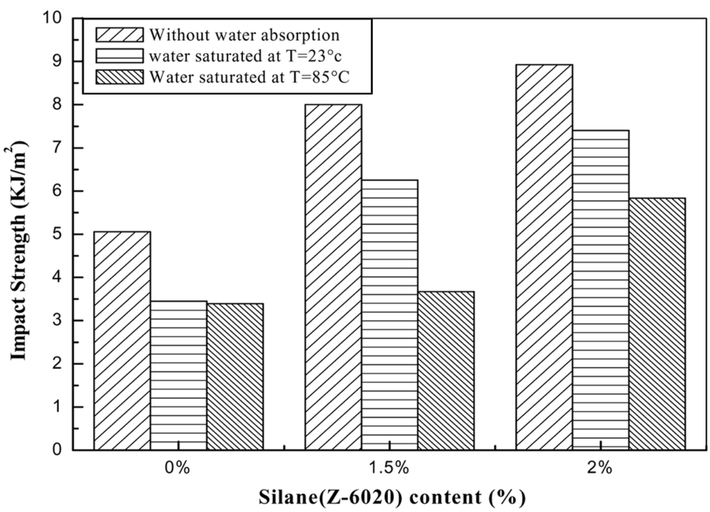
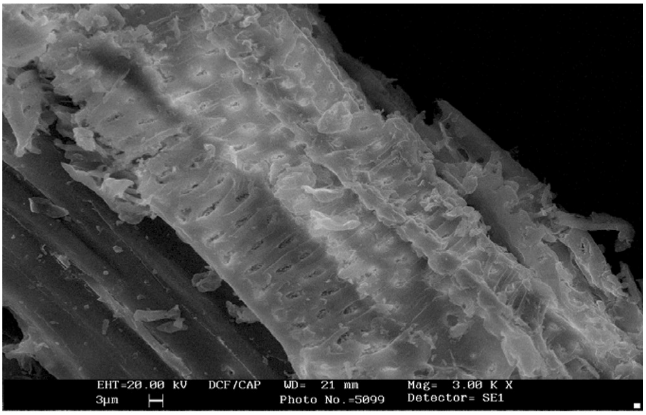


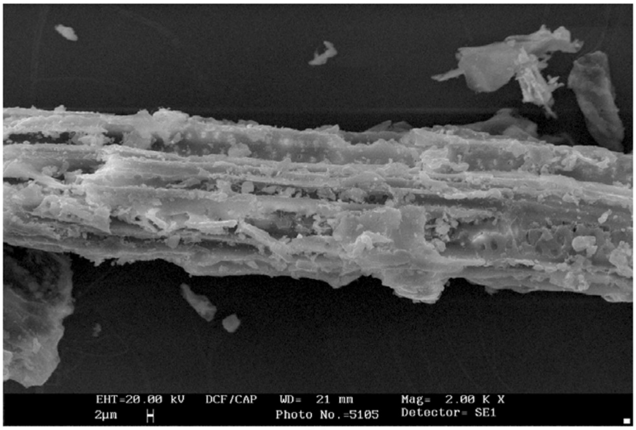

**FIGURE 6** Effect of the water absorption on the impact strength for PP/treated SJ fiber (70/30) composites.

$n = 0.5$, while for Case II $n = 1$. For anomalous diffusion, n shows an intermediate value ($0.5 < n < 1$). Moisture absorption in natural fibers reinforced plastics usually follows Fickian diffusion case I [21].

Figure 4 shows an example of the fitting of the experimental data. In Table 1, the values of the parameters n and k resulting from the fitting are shown for the samples. The absorption of water in PP/SJ fiber composites approaches towards the Fickian diffusion case I, as the values of n are very similar for the samples and they are very close to the value of $n = 0.5$.




(a)

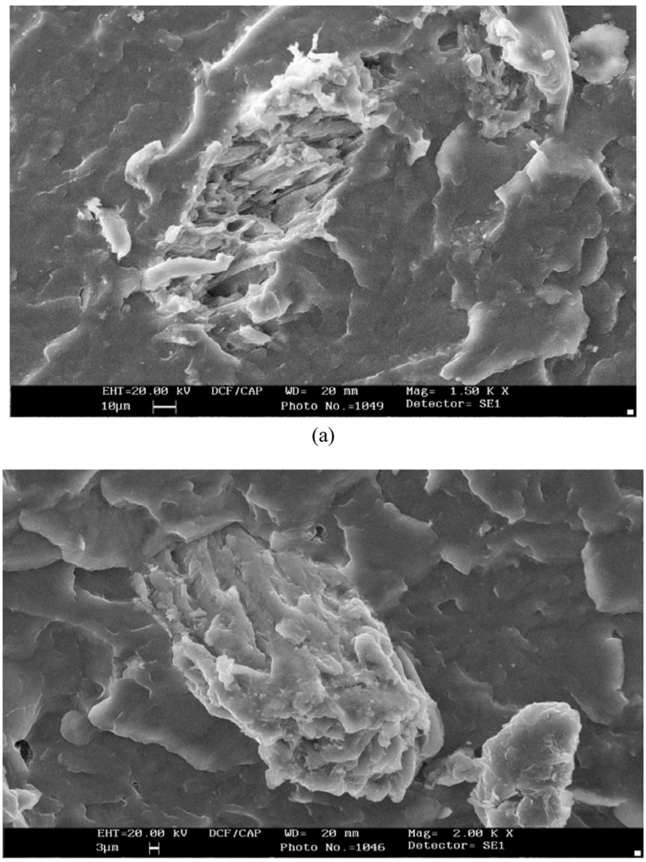



(b)

**FIGURE 7** SEM micrographs of Spartium junceum fiber (a) untreated fibers, (b) 1.5 wt% Z-6020 silane treated.

## Effect of Water Absorption on Impact Strength of PP/SP Fiber Composites

The results of Charpy impact strength are shown in Figures 5 and 6. Figure 5 shows the comparison of experimental data of the impact strength values of PP/untreated SJ fiber composites before and after water absorption at different temperatures. We observed that the impact strength of PP/untreated SJ fiber composites decreased after moisture uptake. Cellulosic fibers contain numerous hydroxyl groups available for interaction with water molecules by hydrogen bonding. So, cellulosic fiber-reinforced polymers can take up high amounts of water, which generally causes a reduction in mechanical properties.



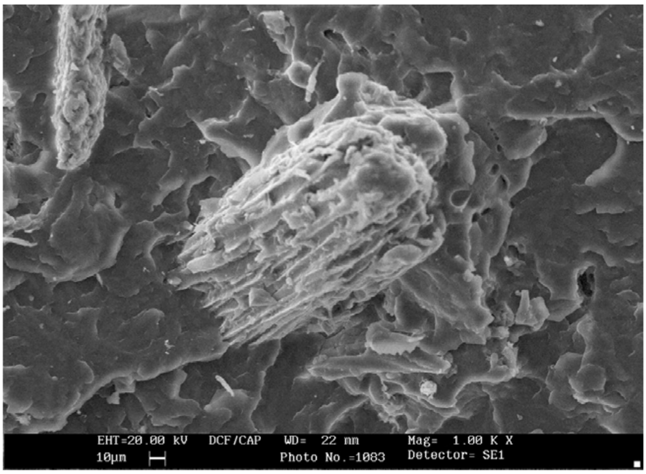

(a)

(b)

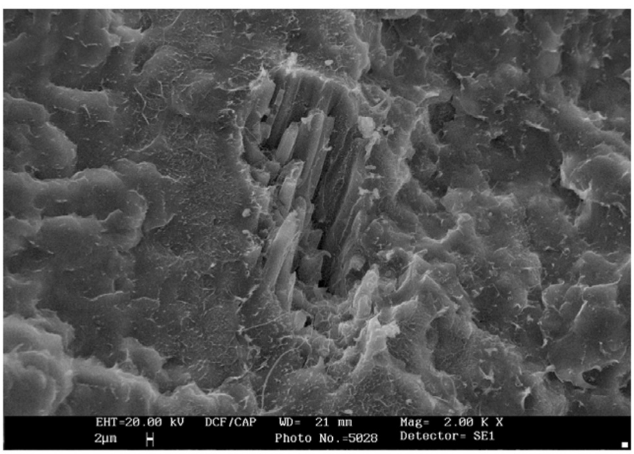
**FIGURE 8** SEM micrographs of PP/untreated SJ fiber composites (90/10), at different magnitudes (a) 1500×, (b) 2000×.

As polypropylene is an apolar polymer matrix, penetration into pits, cracks, and other voids in fibers tends to be limited. Under moisture conditions, spaces can be filled with water [16].

The effect of water absorption on the impact properties for the composites containing 30 wt% SJ fiber without Z-6020 and for similar composites containing 1.5 and 2 wt% of Z-6020 is show in Figure 6. It was observed that the treatment of the fibers with silane Z-6020 improves the impact properties of the composites. Silane coupling agents form




(a)




(b)

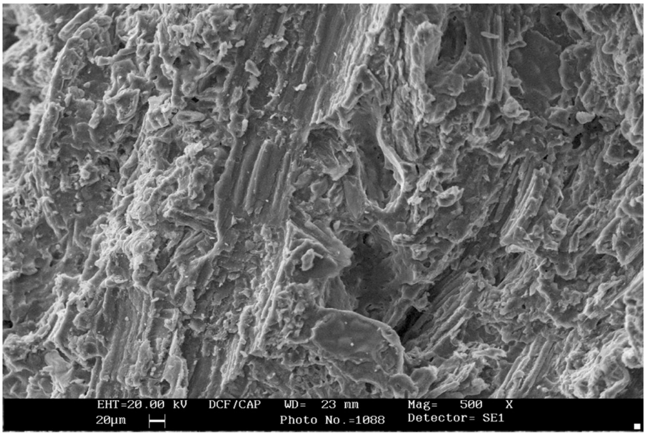
**FIGURE 9** SEM micrographs of PP/SJ/Z-6020 composites (90/10/1.5), at different magnitudes (a) 1000×, (b) 2000×.

hydrogen bonds with some free hydroxyl groups of cellulose, which reduce the water absorption capacity of cellulose [3]. On the other hand, we observe that the impact strength of composites decreases with increased immersion temperature.

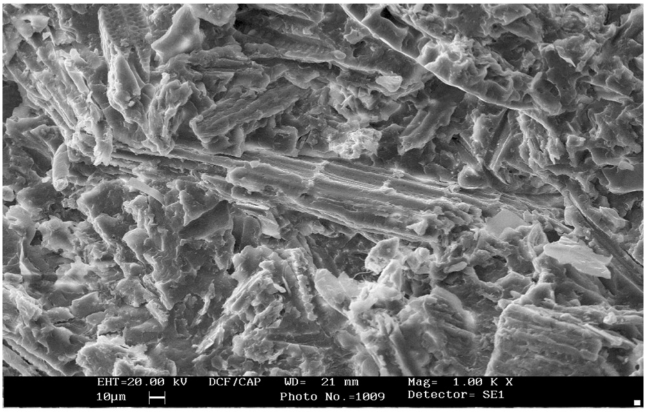
## MORPHOLOGY

### Morphology of Spartium Junceum Fibers

The SEM micrograph of *Spartium junceum* fibers untreated and treated with 1.5% of silane (Z-6020) is shown in Figure 7. Micrographs




(a)




(b)

**FIGURE 10** SEM micrographs of PP/SJ/Z-6020 composites (50/50/1.5), at different magnitudes (a) 500×, (b) 1000×.

illustrate the reduction of roughness via surface treatment of fibers. The results can be accepted as proof for the surface coverage of the fibers with a siloxane layer resulting in a decrease of the surface roughness.

## Morphology of PP/SJ Fiber Composites

Examination of the fracture surface of PP/untreated SJ composites is presented in Figure 8. The micrographs indicate that there are voids between fibers and matrix which is evidence of poor adhesion. Poor adhesion seems to facilitate debonding of the fiber. Fracture surface of these composites shows holes and fiber ends, indicating that most of the fibers have pulled out without breaking during the fracture of the composite.

SEM micrographs of PP/treated SJ composites are presented in Figures 9 and 10. These figures clearly indicate that the treatment facilitates good adhesion between fibers and matrix.



# CONCLUSIONS

The effect of *Spartium junceum* fiber content, surface treatment and temperature on the water absorption and the influence of moisture content on the impact behavior of PP/SJ fiber composites were investigated in this study.

The following points can be drawn from these studies.

The moisture content increases in composites with the increase of *Spartium junceum* fiber content.

A decrease in saturation time when the temperature of immersion increases for the PP/ SJ fiber composites.

Water absorption results showed that silane treatment reduced the water absorption capacity compared to untreated composites.

The absorption of water in PP/SJ fiber composites approaches the kinetics of a Fickian diffusion case I at ambient temperature.

Impact strength properties are dramatically affected by the water absorption. Water–saturated samples show poor impact strength.

Scanning electron microscopy of *Spartium junceum* fibers untreated and treated with silane (Z-6020) illustrates the reduction of roughness via surface treatment of SJ fibers and facilitates good adhesion between fibers and polypropylene matrix.

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