Effect of Process Parameters and Composition on Mechanical, Thermal, and Morphological Properties of Polypropylene/Sawdust Composites

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ABSTRACT: Polypropylene/sawdust composites have been prepared according to a statistical experimental design, with varying sawdust and compatibilizer (maleic anhydride grafted PP) concentrations. To investigate process conditions, composites were first extruded in a twin screw extruder coupled to a Haake torque rheometer, without degassing, and then reextruded in a Werner Pfleiderer twin screw extruder, with two degassing zones. Process conditions were analyzed according to statistical techniques. Effect of the variables on mechanical properties was assessed through flexural modulus, tensile strength, and percent elongation at break and morphology was assessed by scanning electron microscopy. Comparison between the extruded and reextruded compounds indi-

INTRODUCTION

During the last years, a significant increase has been observed in research on polymers reinforced with natural fibers. Advantages of the use of these fibers in reinforced compounds include reduction in density and costs and increase in stiffness among others.^{1–4} One of these fibers is sawdust, a wood processing residue during consumer good manufacture. These residues are disposed of or very little used. The most common method of disposal is burning, which is economically little feasible as well as environmentally unfriendly. Some countries, mainly European, have therefore created laws that limit burning of cellulose residues.

Incorporation of sawdust in polymers has become a feasible possibility in obtaining products with suitable characteristics for use as a substitute for wood or in applications of important ecological appeal. Among the most widely used polymers is polypropylene (PP), because of its intrinsic properties and low melting temperature, which enable composite processing below 200°C and avoid degradation of the wood.⁵ However, polypropylene's nonpolar na-

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Key words: polypropylene; sawdust; composite; processing; compatibilizer

ture makes it incompatible with sawdust, whose main constituents are the polar cellulose, lignin, and hemicellulose. To overcome this difficulty compatibilizers may be used,^{6–10} which act at the interface and increase adhesion between the phases and transfer stresses. Another alternative is to modify the sawdust with coupling agents,^{11,12} to make it more compatible with PP and reduce moisture absorption by the sawdust, which besides impairing processing causes surface stains due to thermooxidative processes. Simultaneous utilization of the aforementioned techniques has also been proposed^{13,14} to obtain a more homogeneous mixture with improved adhesion at the interface. Plasma treatment in argon and in air has also been proposed to modify sawdust surface and hence increase compatibility with PP.¹⁵

In addition to the intrinsic properties of the composite constituents, which cause incompatibility, their final properties are strongly dependent on manufacturing processing. Because of wood's polar nature it absorbs significant amounts of water, which may impair processing. The hopper might get blocked up, leading to process discontinuity and porosity of the final parts. This will interfere in compound adhesion, as well as jeopardize parts appearance, as previously mentioned.^{16,17} Hence, the wood should be dried, before and/or during processing. In addition, it has been suggested to use twin screw



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extruders with length/diameter values, L/D, above 30 for the manufacture of this composite.^{4,18}

The objective of this investigation is to assess the compounding process of polypropylene with sawdust, through statistical analysis, and to analyze the effect of the independent variables, sawdust and compatibilizer, on mechanical, thermal, and morphological properties of the composite (dependent variables). It should be mentioned that in relation to other articles, this work analyzes high sawdust contents (between 40 and 60%).

EXPERIMENTAL

Materials

PP powder was supplied by Suzano Petroquímica (Mauá, Brazil), under code K, with melt flow index (MFI) of 3.0 g/10 min. Antioxidant and lubricant incorporated to the formulations were Irganox B215 and calcium stearate, respectively. Irganox B215 is a mixture of a phenolic antioxidant (Irganox 1010) and a phosphite (Irgafos 168) supplied by Ciba Especialidades Químicas (São Paulo, Brazil). The lubricant Struktol TPW 113, which is a blend of complex, modified fatty acid ester, supplied by Parabor (São Paulo, Brazil), was also incorporated to all formulations at fixed amounts of 2%.

The compatibilizer used was PP grafted with maleic anhydride (PP-g-MA), under code Polybond 3200 (MFI = 110 g/10 min at 190°C and 2.16 kg), supplied by Crompton-Uniroyal Chemical (São Paulo, Brazil).

The sawdust used was supplied by Uliana Lamber Company (Tietê, Brazil). Sawdust was collected from the storage silo containing a mix of several Brazilian timber types: pink cedar (*Cedrela* sp), arana cedar (*Cedrelinga cateniformis*), ipê (*Tabebuia* spp), angelim (*Hymenolobiem petroeum*), marupá (*Simarolba amara*), loro vermelho (*Nectanda rubra*), cumarú (*Dipterysa odorata*), jatobá (*Hymenoea* sp). The relative amounts of the timber types have not been determined. To adjust particle size, the sawdust was ground in a grinder equipment with a 0.8 mm screen. Particle size distribution is shown in Table I.

Methods

Preparation of PP/sawdust compounds

Preliminary processing to produce PP/sawdust compounds, with and without compatibilizer (PP-g-MA),

| TABLE I | | | | | | | | |
|---|--|--|--|--|--|--|--|--|
| Particle Size Distribution of the Sawdust Obtained in | | | | | | | | |
| the Grinder Equipment | | | | | | | | |

| ASTM Mesh | Percent retained material |
|-----------|---------------------------|
| No. 25 | 39 |
| No. 30 | 50 |
| No. 60 | 78 |

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TABLE II Preliminary Compositions of the Investigated PP/ Sawdust/PP-g-MA Compounds

| Formulation | Percent PP | Percent sawdust | PP-g-MA (phr) |
|-------------|------------|-----------------|---------------|
| T-01 | 70 | 30 | _ |
| T-02 | 70 | 30 | 3 |

were carried out in a corotating twin screw extruder coupled to a Haake torque rheometer (Karlsrube, Germany), with no degassing zone, at the following conditions:

Temperature profile: 160, 170, 180, 180, 180, 170, and 170°C.

Screw frequency: 120 rpm.

Prior to processing in the Haake torque rheometer, the sawdust was dried in an oven at 120°C for 15 h and mixed with the polypropylene and the remaining additives in a Henschel mixer (Rio Claro, Brazil).

The compositions investigated at this stage served to check whether the compatibilizer was necessary or not.

The compositions investigated in the preliminary tests are shown in Table II.

After verification of the effect of the compatibilizer (PP-g-MA) on the mechanical properties of the composite, the following stage of the work was commenced. To verify the effect of the independent variables on the mechanical properties of the composites and to construct response surfaces capable of describing their behavior in the experimental region, experiments were performed according to a Central Composite Experimental Design, where sawdust (C_s) and compatibilizer (C_c) concentrations were varied. Process conditions were the same as described previously. The samples extruded in the Haake rheometer were cooled in a water bath and chopped in a chopper. Next, they were injected and tested in tensile and bending, according to tests described as follows.

Table III shows the experimental design carried out. All formulations contained antioxidants and lubricants. The following reference formulation were

TABLE III Experimental Design

| | 1 0 | |
|------|-------------------------|-----------------------|
| Test | $C_s (\%)^{\mathrm{a}}$ | $C_c (\%)^{\epsilon}$ |
| 1 | 40 | 3 |
| 2 | 60 | 3 |
| 3 | 40 | 5 |
| 4 | 60 | 5 |
| 5 | 50 | 4 |
| 6 | 50 | 4 |
| 7 | 35.86 | 4 |
| 8 | 64.14 | 4 |
| 9 | 50 | 2.586 |
| 10 | 50 | 5.414 |
| | | |

^a C_s, sawdust content; C_c, compatibilizer content (PP-g-MA).

| | Tensile str | ength (MPa) | Percent elongation at break | | |
|---|--------------|--------------|-----------------------------|--------------|--|
| Sample | Average | Deviation | Average | Deviation | |
| PP/sawdust (70/30) PP/sawdust/compatibilizer (70/30/3) | 23.4 24.3 | 0.48 0.22 | 4.36 5.83 | 0.35 1.07 | |

TABLE IV Tensile Test Results of the Preliminary Processing Conducted in the Twin Screw Extruder Coupled to the Haake Torque Rheometer

also processed: Reference 1, consisting of PP, antioxidants, and calcium stearate, with no sawdust, no lubricant Struktol TPW 113 nor PP-g-MA, and Reference 2 containing the same components of Reference 1, however with lubricant Struktol TPW 113.

Assessment of the properties of these formulations, which are shown in the results section, indicated a great deal of retained water in the samples and inadequate compatibilization. It was therefore decided to reprocess the formulations in a ZSK25 Werner & Pfleiderer extruder (Tamm, Germany) containing two degassing zones. Composite extrudates were then cooled in air and ground in a P1001 FDR Primotécnica grinder (Mauá, São Paulo). The intention of this reprocessing was to eliminate the retained water through degassing as well as promote more intense mixing. The process conditions in the extruder were as follows:

Temperature profile: 150, 160, 170, 170, 170, 190, and 185°C.

Screw frequency: 400 rpm.

The composites obtained both in the preliminary processing and in the Haake experimental design processing, were dried in an oven prior to injection molding.

The compounds obtained in the ZSK25 Werner & Pfleiderer extruder, however, were not submitted to additional drying, as they were injected right after grinding.

Analyses of the preliminary compositions

At this stage, samples were analyzed by means of tensile tests and scanning electron microscopy.

Tensile tests were performed according to ASTM D638 in a Kratos Universal Testing Machine (Cotia, Brazil). Because of speed adjustment limitations of the machine, tests were conducted at 10 mm/min.

Fractured surfaces of the tensile tests were coated with platinum and analyzed in a LEO Stereoscan 440 scanning electron microscope, SEM, (Cambridge, UK.) under high vacuum (5 \times 10⁻⁶ torr).

Analyses of the compounds extruded in the twin screw extruder coupled to the Haake torque rheometer and in the Werner & Pfleiderer extruder

Tensile and bending tests were conducted in a 5565 Instron Universal Testing Machine (Norwood, US), according to ASTM D638 and ASTM D790, at test speeds of 5 and 1.3 mm/min, respectively.

Calorimetry analyses were carried out in a DSC 2920 TA Instruments Modulated DSC (New Castle, US). Samples weighing between 9 and 12 mg were heated at a rate of 10°C/min, from 30°C to 190°C. After 1 min at 190°C, samples were cooled to 30°C, at a rate of 10°C/min and then submitted to a second heating at the same conditions as the first to eliminate process history.

Samples analyzed by SEM were those fractured in the tensile tests, at the same conditions and in the same equipment described in the preliminary tests.

RESULTS AND DISCUSSIONS

The preliminary tests were performed to verify whether a compatibilizer was required or not for the composites. Tensile test results are shown in Table IV. From these results, it can be seen that incorporating the compatibilizer causes significant increase in tensile strength, but percent elongation at break did not show significant variations. Analyses of the differences were performed according to Tukey test at a significance level of 5%.

During extrusion, samples without compatibilizer were difficult to process as they broke easily on exiting the die. Furthermore, these samples presented white spots when injection molded.

Morphology of samples with and without compatibilizer (PP-g-MA), obtained by SEM, are shown in Figure 1. Figure 1(a,b) show fiber pull-out of the matrix, observed throughout almost the entire sample, indicating weak adhesion between the fibers and the polypropylene. The fractured surfaces in Figure 1(c,d) show pronounced deformation of the polymer matrix and significant reduction in fiber pull-out, indicating the compatibilizer likely affects interface, reducing interfacial tension and improving adhesion.

Analyses of mechanical properties and morphology obtained by SEM of the samples submitted to preliminary processing, in addition to visual assessment, indicate the need to incorporate polypropylene grafted with maleic anhydride (PP-g-MA) to the PP/ sawdust composites. PP-g-MA (Polybond[®] 3200) likely acts as a compatibilizer, reducing interfacial tension and increasing adhesion at the PP/ sawdust



Figure 1 SEM micrographs of the fractured surfaces of the PP/sawdust/PP-g-MA compounds: (a) and (b) 70/30/0, (c) and (d) 70/30/3.

interface. The need to compatibilize this system arises from the fact that PP is a nonpolar polymer whereas sawdust, consisting mainly of cellulose, lignin, and hemicellulose, has a polar nature, resulting in incompatibility between PP and sawdust.

After having determined the need for the compatibilizer, the next stage was to vary the concentrations of sawdust and PP-g-MA. The 10 formulations processed at this stage are listed in Table III.

Tensile and bending tests of these 10 formulations, of the two reference formulations processed in the twin screw extruder coupled to the Haake torque rheometer and of those reprocessed in the Werner Pfleiderer twin screw extruder are shown in Table V.

Table VI lists the coefficients of the equation fitted by multivariable analysis for the processing in the twin screw extruder coupled to the Haake torque rheometer. Multivariate analysis of the properties flexural modulus, tensile strength, and percent tensile elongation at break of Samples 1–10, processed in the Haake torque rheometer allow to verify that

TABLE V

Results of the Tensile and Bending Tests of the Samples Processed only in the Twin Screw Extruder Coupled to the Haake Torque Rheometer and Those Processed in Both Twin Screw Extruder Coupled to the Haake Torque Rheometer and in the Werner Pfleiderer Extruder

| | Tensile strength (MPa) | | Percen | t elongation at break | Flexural modulus (MPa) | | |
|--|------------------------|--------------------|--------------------|-----------------------|------------------------|--------------------|--|
| Sample Reference 1 Reference 2 H01 H02 H03 H04 | Haake | Haake and Extruder | Haake | Haake and Extruder | Haake | Haake and Extruder | |
| Reference 1 | 37.6 ^a | 38.4 ^a | 172.1 ^a | 120.7 ^a | 2049.6 | 2035.6 | |
| Reference 2 | 37.1 ^a | 36.3 ^a | 161.9 ^a | 319.8 ^a | 2009.1 | 1761.8 | |
| H01 | 28.5 | 28.0 | 5.19 | 6.20 | 3461.9 | 3726.3 | |
| H02 | 24.4 | 26.5 | 2.96 | 3.14 | 4995.4 | 5331.3 | |
| H03 | 29.2 | 28.5 | 5.01 | 5.71 | 3501.9 | 3834.6 | |
| H04 | 25.7 | 27.1 | 2.76 | 2.87 | 3491.3 | 5535.3 | |
| H05 | 27.0 | 27.9 | 3.90 | 3.98 | 4399.2 | 4664.4 | |
| H06 | 27.2 | 27.8 | 3.64 | 3.91 | 4419.5 | 4724.8 | |
| H07 | 28.7 | 28.8 | 6.03 | 7.50 | 3246.6 | 3440.1 | |
| H08 | 22.8 | 25.4 | 2.96 | 2.36 | 5320.1 | 5656.7 | |
| H09 | 27.6 | 26.9 | 4.20 | 4.25 | 4264.7 | 4333.5 | |
| H10 | 27.7 | 29.2 | 4.26 | 4.06 | 4143.3 | 4341.5 | |

^a Samples submitted to testing at 50 mm/min, different from the remaining at 5 mm/min.

| | | Linear parameters | | | | Quadratic paramete | Cubic parameters | | |
|-----------|---------|-----------------------|-----------|-----------------------|-----------------------|--------------------|-----------------------|-----------------------|----------------|
| Property | R^2 | <i>a</i> ₀ | a_1 | <i>a</i> ₂ | <i>a</i> ₃ | a_4 | <i>a</i> ₅ | <i>a</i> ₆ | a ₇ |
| TS | 0.93937 | 17.81043 | 0.587865 | _ | _ | -0.0078729 | _ | _ | _ |
| Percent E | 0.92570 | 9.6035 | -0.110268 | - | - | _ | _ | _ | _ |
| Modulus | 0.57772 | -42048.37 | 2770.365 | _ | - | -55.4073 | - | 0.369382 | - |

 TABLE VI

 Coefficients of the Fitted Equations to Construct Response Surface Plots of the Samples Processed Only in the Twin Screw Extruder Coupled to the Haake Torque Rheometer

Polynomial equation: $a_0 + a_1Cs + a_2C_c + a_3C_sC_c + a_4C_s^2 + a_5C_c^2 + a_6C_s^3 + a_7C_c^3$.

TS, tensile strength; percent E, percent elongation at break; modulus, flexural modulus.

incorporated sawdust content (independent variable) is the only variable affecting these.

All samples processed in the twin screw extruder coupled to the Haake torque rheometer were dried prior to injection molding. Nevertheless, injected samples presented white spots very similar to those of the preliminary studies without compatibilizer.

The fact that increasing PP-g-MA content did not affect the dependent variables indicates that PP-g-MA did not act as compatibilizer, contradicting the preliminary tests, as well as previous investigations.^{6–8}

This behavior might arise from two factors: the presence of retained moisture in the pellet, which may settle at the PP/sawdust interface, impairing compatibilizer effectiveness, and inadequate mixing of the compound. The first factor might be explained by the lack of a degassing zone of the extruder coupled to the Haake rheometer as well as the fact that the extrudate was cooled in a water bath. With regard to inadequate mixing, this might have occurred because of the low L/D ratio (about 28) of the Haake rheometer for composite processing.

Since the preliminary processing, which presented better results, were carried out at the same process conditions and in the same equipment, however samples were dried right after processing, it is very likely that the retained moisture is the factor that most affected the tests results.

It was therefore decided to reprocess the composite pellets of formulations H01–H10 as well as the reference formulations in a ZSK 25 Werner & Pfleiderer extruder with two degassing zones. To avoid contact with water, the reextruded composite was cooled in air and then ground and injected. Tensile and bending test results are shown in Table V (Haake and Extruder). These results were, then, submitted to multivariate analysis and the investigated dependent variables (tensile strength, percent elongation at break and flexural modulus) could be fitted in relation to the independent variables (sawdust and compatibilizer content). The coefficients of the equations fitted by multivariate analysis are shown in Table VII. The results shown in Table V together with the fitted equations show that both sawdust and compatibilizer content affect the investigated dependent variables.

In addition to the coefficients of the equations, that enable to analyze which variable affects the investigated response, the value of R^2 should also be analyzed. R^2 is the ratio between the sum of the squares of the model and the sum of the total squares and represents the correlation coefficient between the observed responses and the values predicted by the fitted model. The highest value of R^2 is 1 and only occurs when there is no residue and therefore, all variations around the average can be explained by the regression. The higher R^2 , the better the fit is of the model to the observed values.¹⁹ Comparing R^2 values of Table VI (processing in the Haake rheometer) and Table VII (processing in the Haake rheometer and Werner Pfleiderer Extruder), a better fit is seen of the curves obtained by multivariate regression, in the case where the compounds were reextruded.

 TABLE VII

 Coefficients of the Fitted Equations to Construct Response Surface Plots of the Samples Reprocessed in the Werner Pfleiderer Extruder (Haake and Extruder)

| | | Linear parameters | | | Quadratic par | Cubic parameters | | | |
|-----------|---------|-----------------------|-----------------------|-----------------------|-----------------------|------------------|-----------------------|-----------------------|----------------|
| Property | R^2 | <i>a</i> ₀ | <i>a</i> ₁ | <i>a</i> ₂ | <i>a</i> ₅ | a_4 | <i>a</i> ₅ | <i>a</i> ₆ | a ₇ |
| TS | 0.99723 | 43.55073 | -2.964265 | 24.59028 | _ | 0.062454 | -6.31184 | -0.000442 | 0.53431 |
| Percent E | 0.99292 | 63.91503 | -3.114068 | -0.12756 | _ | 0.055601 | _ | 0.000341 | _ |
| Modulus | 0.97827 | -1348.644 | 80.5068 | 990.6 | - | _ | -123.825 | _ | - |

Polynomial equation: $a_0 + a_1C_s + a_2C_c + a_3C_sC_c + a_4C_s^2 + a_5C_c^2 + a_6C_s^3 + a_7C_c^3$.

TS, tensile strength; percent E, percent elongation at break; modulus, flexural modulus.



Figure 2 SEM micrographs of the fractured surfaces of the PP/sawdust/PP-g-MA compounds (45.5, 50, 2.5): (a) and (b) processed in the Haake torque rheometer only; (c) and (d) processed in the Haake torque rheometer and in the Werner Pfleiderer Extruder.

To verify the effect of processing on composite morphology, SEM analysis was performed on the surface of tensile fractured Samples 9 (PP/Sawdust/ PP-g-MA, 45.5/50/2.5) and 10 (PP/Sawdust/PP-g-MA, 42.5/50/5.5), processed in the twin screw extruder coupled to the Haake torque rheometer (H)



Figure 3 SEM micrographs of the fractured surfaces of the PP/sawdust/PP-*g*-MA compounds (42.5, 50, 5.5): (a) and (b) processed in the Haake torque rheometer only; (c) and (d) processed in the Haake torque rheometer and in the Werner Pfleiderer Extruder.

and extruded in the Haake and reextruded in the Werner Pfleiderer Extruder (HE). Figure 2 shows analyses of Samples 09H and 09HE and Figure 3 shows analyses of Samples 10H and 10HE.

All samples presented improved adhesion at the polymer/sawdust interface, as seen by minimized fiber pull-out and near-surface fiber rupture. However, comparison between Samples H and HE, shown in Figures 2 and 3, shows increased deformation of the matrix of the samples reprocessed in the extruder, as well as more adhered fibers. The samples processed in the Haake torque rheometer only, showed a less deformed matrix, a greater amount of voids, as well as less adhered fibers. This behavior is an indication of the improved adhesion brought about by reprocessing in the extruder due to more adequate mixing and moisture removal.

Statistical analysis of the mechanical behavior and morphology analyses indicate that water (moisture) retained in the composite pellets likely takes place at the interface, impairing compatibilization of PP with the sawdust. Because of the polar nature of sawdust the water is likely retained at its surface. On reprocessing in the Werner extruder, equipped with two degassing zones, moisture could be removed, enabling action of the compatibilizer at the interface. Moreover, the high L/D ratio of ~ 40 of the Werner Pfleiderer Extruder allowed improved mixing of the compound.

The previous analysis shows the importance of process control of PP/sawdust compounds, the need for reducing sawdust moisture, before and during processing, as well as the importance of avoiding compound cooling in water bath.



Figure 4 Effect of the variables sawdust and compatibilizer (PP-*g*-MA) concentrations on tensile strength. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]



Figure 5 Effect of variables sawdust and compatibilizer (PP-*g*-MA) on percent elongation at break. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

After having solved the problems originating from sawdust processing, response surface graphs were plotted from the equations of which the coefficients are presented in Table VII (Haake and Extruder). Graphs of tensile strength, percent tensile elongation at break, and flexural modulus are presented in Figures 4, 5 and 6, respectively.

Figures 4 and 5 show that increase in sawdust concentration causes a reduction in tensile strength and percent elongation at break. This indicates that the sawdust likely acts as a stress concentrator. However, increase in compatibilizer (PP-g-MA)



Figure 6 Effect of the variables sawdust and compatibilizer (PP-*g*-MA) concentrations on flexural modulus. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

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TABLE VIII Thermal Test Results Obtained by Differential Scanning Calorimetry

| Sample | T_m (°C) | Percent crystallinity |
|-------------|------------|-----------------------|
| Reference 1 | 165.36 | 42.51 |
| Reference 2 | 164.74 | 40.98 |
| H01 | 163.86 | 26.9 |
| H02 | 163.24 | 16.5 |
| H03 | 164.32 | 25.0 |
| H04 | 163.39 | 16.1 |
| H05 | 163.65 | 20.5 |
| H06 | 164.02 | 21.7 |
| H07 | 164.39 | 25.7 |
| H08 | 163.15 | 15.0 |
| H09 | 164.34 | 20.7 |
| H10 | 164.48 | 21.7 |
| | | |

content in the composite results in a slight, yet significant, increase in tensile strength and reduction in percent elongation at break, suggesting a more effective compatibilization at the interface when PP-g-MA content is increased, minimizing interfacial stresses and increasing adhesion between components.

The highest tensile strengths occur at lower sawdust contents and higher compatibilizer contents, indicating that at higher sawdust contents, the amount of incorporated compatibilizer should exceed the levels investigated.

The above-mentioned statistical analysis could be corroborated by morphology observations of the compounds shown in Figures 2 and 3, where increase in PP-*g*-MA content in Samples 9 and 10 caused improved adhesion of the fibers to polypropylene and increased deformation of the polymer matrix.

Figure 6 shows the effect of C_s and C_c on the dependent variable flexural modulus. As can be seen from the response surface graph the flexural modulus increases with increasing sawdust concentration, indicating a significant increase in composite stiffness.

Increase in compatibilizer content results in an increase in elastic modulus up to a certain value after which this property reduces. Increase in this property indicates that as PP-g-MA is incorporated, compatibilization becomes more effective, by stress transfer from the sawdust to the matrix. However, reduction is likely due to the plasticizing effect of

the compatibilizer at high incorporated percent, since this copolymer has a very low molecular weight compared to PP.

Thermal analyses

The results of crystalline melt temperature and percent crystallinity obtained during the second heating in the DSC runs are listed in Table VIII. Analyses were performed during the second heating to eliminate processing history and analyze the effect of the independent variables (sawdust and compatibilizer content) on the aforementioned responses. Comparison of percent crystallinity and crystalline melt temperature (T_m) of the reference samples and of those containing sawdust (H01–H10), shows that the presence of the sawdust and of the compatibilizer significantly reduce the two investigated variables. This result might be explained by the fact that the presence of the sawdust hinders approximation of the polypropylene chains and this way impairs crystallization and renders less perfect crystals.

When Samples H01–H10 are submitted to multivariate analysis, the only variable seen to affect the investigated responses was sawdust content. Table IX presents the coefficients of the fitted equations.

Increase in the variable sawdust in the compound caused a significant reduction in percent crystallinity and a slight reduction in melt temperature. However, the low value of R^2 (0.26748), shown by the variable T_m , indicates that analysis of the effect of increasing sawdust content on this variable is not very reliable. On analyzing the effect of the independent variables, sawdust and compatibilizer content, on percent crystallinity the only variable seen to affect percent crystallinity is sawdust content. Its increase leads to reduction in percent crystallinity, due to the increasing difficulty of the polymer chains to approach one another.

CONCLUSIONS

The conclusion arrived at is that incorporation of elevated amounts of sawdust in polypropylene (above 40%) is possible. However, the difficulties in process-

| TABLE IX |
|---|
| Coefficients of the Fitted Equations to Construct Response Surface Plots of the Thermal Analyses of the |
| Samples Reprocessed in the Werner Pfleiderer Extruder (Haake and Extruder) |

| | | Lin | Linear parameters | | | Quadratic parameters | | | Cubic parameters | |
|--------------------|--------------------|-----------------------|-------------------------|-----------------------|-----------------------|----------------------|-----------------------|-------|---------------------|--|
| Property | R^2 | <i>a</i> ₀ | a_1 | <i>a</i> ₂ | <i>a</i> ₃ | a_4 | <i>a</i> ₅ | a_6 | a ₇ | |
| Percent C T_m | 0.95233 0.26748 | 42.48605 165.957 | $-0.430341 \\ -0.04151$ | _ | _ | _ | _ | _ | | |

Polynomial equation: $a_0 + a_1C_s + a_2C_c + a_3C_sC_c + a_4C_s^2 + a_5C_c^2 + a_6C_s^3 + a_7C_c^3$.

 T_m , melt temperature; percent C, percent crystallinity.

ing are pronounced. One of the main factors that should be controlled, during processing, is retained moisture, which may affect compatibilization between polypropylene and sawdust and in addition impair processing.

Incorporation of polypropylene grafted with maleic anhydride showed to be necessary for system compatibility, however, added amounts were low for the high sawdust contents, indicating the need to increase PP-g-MA content for improved compatibilization.

Increase in sawdust content led to increase in polymer stiffness and reduction in tensile strength, indicating sawdust acted as stress concentrator. Crystallinity degree was severely reduced by incorporation and increase in sawdust content in the polypropylene as the presence of sawdust hinders approximation of the polymer chains.

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