Surface-Modified Talc Particles by Acetoxy Groups Grafting: Effects on Mechanical Properties of Polypropylene/Talc Composites

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This article deals with the effects of surface-modified talc particles on mechanical properties of polypropylene (PP)/talc composites. These materials were prepared by injection molding of PP blended with different concentrations of nontreated and treated talc, under the same processing conditions. Differential thermal calorimetry and scanning electron microscopy were used to assess thermal properties and morphology of the final composites. The reinforcing effect of talc, either treated or nontreated surface, on PP is analyzed through the tensile properties as a function of the mineral content (0–10 wt%). Morphological structure of composites revealed that the talc treatment improved the particle dispersion and distribution within the PP matrix and enhanced the interfacial PP-talc adhesion. The mechanical properties of these composites, especially the Young modulus, tensile strength and elongation at break, were found to be improved respect to PP-untreated talc ones. POLYM. ENG. SCI., 00:000–000, 2012. © 2012 Society of Plastics Engineers

INTRODUCTION

In the recent years, particle-filled composites have received considerable attention mostly because of their convenient relationship among cost, ease of high volume production and mechanical performance. For these reasons, they have attained great industrial interest, exhibiting remarkable improvement on properties when they are compared to the corresponding virgin polymer. Also, from the academic point of view, they represent a challenge when final properties improvement is desired. The best results are obtained when fillers of small particle size are added. However, when thermoplastic matrix is used, considerable problems occur during processing related to filler exfoliation, dispersion and distribution in the high viscosity molten polymer. Moreover, particle–polymer adhesion must be enhanced in order to increase the mechanical properties [1].

Polypropylene (PP) is one of the preferred commodity polymers for a wide range of modern technical applications. The incorporation of talc to PP is twofold attractive. On one side, the mineral particles play a strong nucleation effect on polymer crystallization; and on the other, talc has a relative high strength and stiffness, laminar particles with favorable aspect ratio (anisotropy), abundance in nature, and low cost [2]. The particle morphology, as well as the size, depends on talc genesis and mineral processing. Particles are the result of stacking hundreds or thousands elementary sheets, kept together by weak van der Waals forces; and their size can vary from approximately 1 μm to more than 100 μm.

The performance of PP/talc composites strongly depends on the talc delamination and adhesion with PP, as well as, particle dispersion and distribution within the matrix. The last two aspects are directly related with a good wetting of the filler by the matrix. Taking into account the nonpolar character of PP and the high surface energy of the talc particles, poor filler dispersion and low particle–matrix interfacial adhesion result, leading to a composite with poor mechanical properties [3–5]. In this sense, an increase in filler–polymer compatibility is needed. Many studies were performed to improve the filler-PP adhesion, looking for the optimization of mechanical and other physical properties. The compatibilization can be improved by modifications in the matrix, in the filler's surface or in both of them. Matrix modification is usually carried out by peroxide grafting of maleic anhydride or acrylic acid onto PP. Several commercial products are available in the market obtained by these reactions. However, the grafting reactions do not proceed to completion and can leave significant amount of unreacted monomer and, possibly, peroxide in the compositions. As a consequence, the final properties probably would be impaired due to the molecular weight degradation by chain scission [6–11]. Alternatively, the filler–matrix interphase can be enhanced by modification of the filler surface. To reduce the particle agglomeration and to increase the particle dispersion and distribution within the matrix, several authors proposed the use of coupling

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agents, including silanes, titanium esters, phosphates, aluminates, and zirconates [12, 13]. However, the disadvantage associated with this modification lies on the relatively high cost of the coupling agents; in particular when low cost fillers are used, as for example talc or carbonates. So, there is a need for developing low-cost treatments, applicable to filler-surfaces, that would allow a better filler-PP adhesion.

In our group, a methodology for talc treatment was developed. It was based on an acid treatment that allows delamination, particle size reduction and functionalization of the talc surface. Milled talc, having 10 μm-size particles, was treated with hot acetic acid in a closed stirring batch reactor. Hydroxyl groups react with the acid protons resulting in grafting of acetoxy groups ($-OCOCH_3$) to the talc surface and water as byproduct. Please note that talc is a layered silicate consisting of sheets of magnesium in octahedral coordination similar to brucite, $Mg(OH)_2$, sandwiched between sheets of silicon in tetrahedral coordination [7]. In this sense, talc particles present two kinds of surfaces, basal and edge ones with different composition. Acid attack is mainly performed on edge surfaces through hydroxyl groups, producing delamination by breaking down particle stacks into thin and individual plates. Acid also acts on basal surfaces and probably opens tetrahedral $-Si-O-Si-$ bonds, grafting acetoxy groups on them. The final particles preserve lamellar morphology with an important reduction in their particle size, mainly in thickness, and modified surface. In this sense, they present a strong hydrophobic character by the presence of acetoxy groups. More details of the procedure, results, and discussion are included in reference [14].

The main objective of this work is to analyze the effect of this kind of talc modification on the mechanical properties of PP/talc composites, either for delamination, particle size reduction, or surface funcionalization. In this way, a comparative study of composites, prepared with both nontreated and treated talc particles, is performed.

EXPERIMENTAL

Materials

PP homopolymer 1102H, from Petroquímica Cuyo, was used as matrix. Its main physical characteristics are: MFI = 1.8 g/10 min, $M_w = 303{,}000$ g/mol and $M_w/M_n = 4.45$. To improve the composite compounding, PP pellets were reduced to fine particles of an average size of 1700 μm in a plastic disc shear grinder with a cool chamber. Talc (A10), supplied by Dolomita S.A.I.C, was used as filler. A10 is a microcrystalline talc sample having platy morphology and high purity, around 98%. Treated talc (A10A) was obtained by acid attack, following the methodology described in Castillo et al. [14]. This treatment favors particle size reduction and delamination, as well as, increases the hydrophobic character of talc due to the presence of acetoxy groups grafted on the surface. An accurate particle characterization, before and after treatment, is included in the same reference [14]. Table 1 summarizes the talc particle size of both samples.

Composite Compounding

PP and talc were mechanically premixed to prepare composites with different concentrations (1, 3, 5, and 10% w/w). These compounds were blended by melt extrusion in a Goettfert counter-rotating twin screw extruder ($D = 30$ mm, $L/D = 25$) with a cylindrical die (1 mm), at 30 rpm. The composite nomenclature was PP(Talc concentration)(Type of talc). Barrel temperature profile for extrusion was 170–190–200–210–220°C, from hopper to die, and it was kept constant during extrusion.

Molding

Composites were pelletized and then molded in a Fluidmec 60T injection-molding machine. The used mold has a cavity to produce tensile test specimens with nominal dimensions according to ASTM D638. The composite is injected in the cavity by a high thickness film gate from one of its ends with the following injection parameters: Screw speed: 100 rpm–Back pressure: 10^6 Pa–Barrel temperature profile: 165–190–195–210°C–Die temperature: 220°C–Injection Pressure: 6.6 10^6 Pa–Mould temperature: 40°C.

Characterization

Composite morphology and talc dispersion and distribution in test specimens were observed by scanning electron microscopy (SEM), using a Jeol 35 CF instrument. The SEM study was performed on cryofractured surfaces of the injected samples. All samples were coated with Au in a sputter coater PELCO 91000 to make them conductive, using an accelerating voltage of 5 kV. At least 100 particles, from around 20 micrographs of different specimen part and at different magnifications, were measured using Analysis PRO^{TM} software package. It is important to note that reference particle dimension values were obtained directly on talc particles previous compounding and following the methodology described in Castillo [14].

Thermal analyses under dynamic conditions were performed on PP and PP/talc composites with a Perkin Elmer Pyris 1 differential scanning calorimeter (DSC). Tests

TABLE 1. Mean particle length (L) and thickness (e) of talc samples.

Talc	L^a (μm)	e (μm)
A10	4.53 ± 1.65	0.139 ± 0.016
A10A	2.86 ± 1.20	0.079 ± 0.012

[a] L is the largest particle dimension.

were carried out in nitrogen atmosphere at a heating/cooling rate of 10°C/min, from 30 to 220°C. After the first heating step, each sample was kept in the molten state for 1 min. Then, the cooling scan begun, and this was followed by a second heating step. Temperature and enthalpy values were obtained using the Pyris software. The crystalline content of the matrix was calculated from the melting enthalpy ratio of the composite and the corresponding to 100% crystalline PP (209 J/g) [15]. Corrections were made to take into account the real amount of polymer in the composite.

Testing

Tensile mechanical properties were measured in an Instron 3369 universal testing machine at room temperature, according to ASTM D638-03 test procedure. Two routines were used. High precision modulus determinations were performed using an extensometer with cross head rate of 5 mm/min. Also, the overall stress–strain behavior was assessed at 70 mm/min, up to the sample breaks. For each composite, at least 10 samples were tested and the average results were reported.

RESULTS AND DISCUSSION

In filled polymer composites, a good reinforcing degree is obtained when particles are well dispersed and distributed within the matrix. This also contributes to the uniformity of mechanical properties throughout the whole specimen. SEM observations of cryofractured surfaces, from tensile specimens, were performed in order to check the filler dispersion and distribution. SEM micrographs were taken at different points of the cross section for all the prepared composites. Particularly, in Fig. 1, micrographs of PP/A10A composites with 10 wt% of talc are presented and related to the spatial distribution within the specimen. Good particle distribution along the cross section is observed in all the samples. However, the dispersion was better in PP/A10A composites than in PP/A10 ones. Initially, it is attributed to the surface treatment. In the same figure, the injection point and the indication of particle preferred orientation are also indicated. The particle orientation plays a vital role in strengthening the polymer matrix, being optimal when particles present their basal planes parallel to the tensile strain direction. This particle organization is a consequence of the platelike structure of talc and its motion in a viscous medium during the injection-molding process. The film gate assures that the flow into the mold is plug like. Then, during the mold filling, talc/molten PP suspension flows, and the laminar particles align straight along the flow direction [16, 17].

A further confirmation of talc dispersion in PP matrix was achieved at higher SEM magnifications, as shown in Fig. 2 for composites with 1 wt% of A10 and A10A. Small agglomerates appear in PP/A10 composites (Fig. 2a and b), but the dispersion is clearly improved when A10A talc is used. This figure also shows a lower particle thickness in PP/A10A composites, as compared to the A10A talc values from Table 1, revealing further particle delamination during the composite processing. This behavior was similar for all the studied talc concentrations. A comparative examination between the fractured surface of PP/A10 and PP/A10A composites (Fig. 2) also shows differences in talc-PP adhesion. In the last ones, remaining particle-matrix polymer links after cryogenic fracture are observed. This is the typical aspect of well-adhered particles to a matrix, where the adhesion strength is higher than fracture forces. The behavior agrees with the more hydrophobic character of the talc surface, giving by the grafted acetoxy groups [14]. This result is consistent with the changes observed in mechanical properties, where PP/A10A composites exhibited higher elongation at break than PP/A10 ones, as shown later.

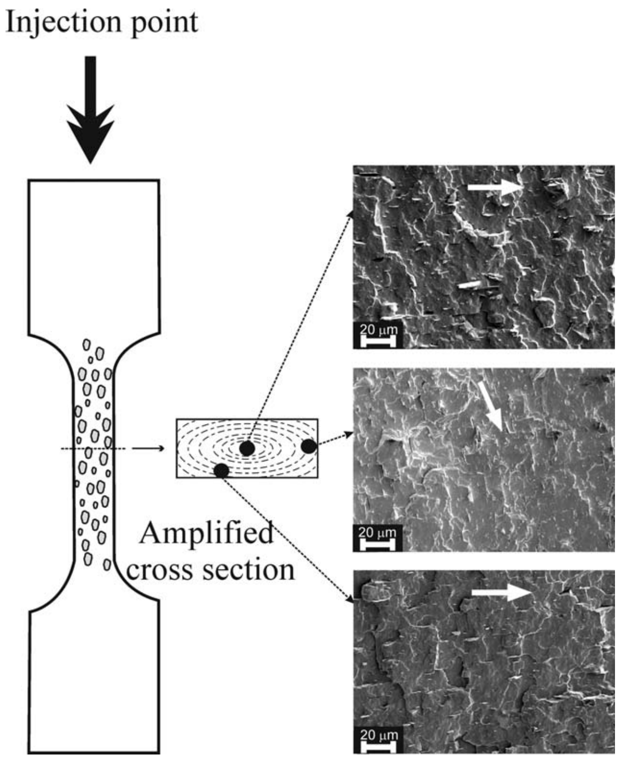

FIG. 1. Orientation of talc particles in the injected composite specimen including SEM micrographs (×2,000) of PP10A10A at different points of the cross section.

As it is known, composite mechanical performance depends on matrix and filler mechanical properties, their relative concentration and adhesion, filler dispersion and distribution, particle morphology, aspect ratio and orientation [18]. In this kind of composites, it is important to note that matrix morphology depends on the filler presence since it usually behaves as a nucleating agent. Particularly, the PP crystal perfection, as well as its degree of crystallinity is highly sensitive to the processing conditions and the presence of talc particles. To assess the inci-

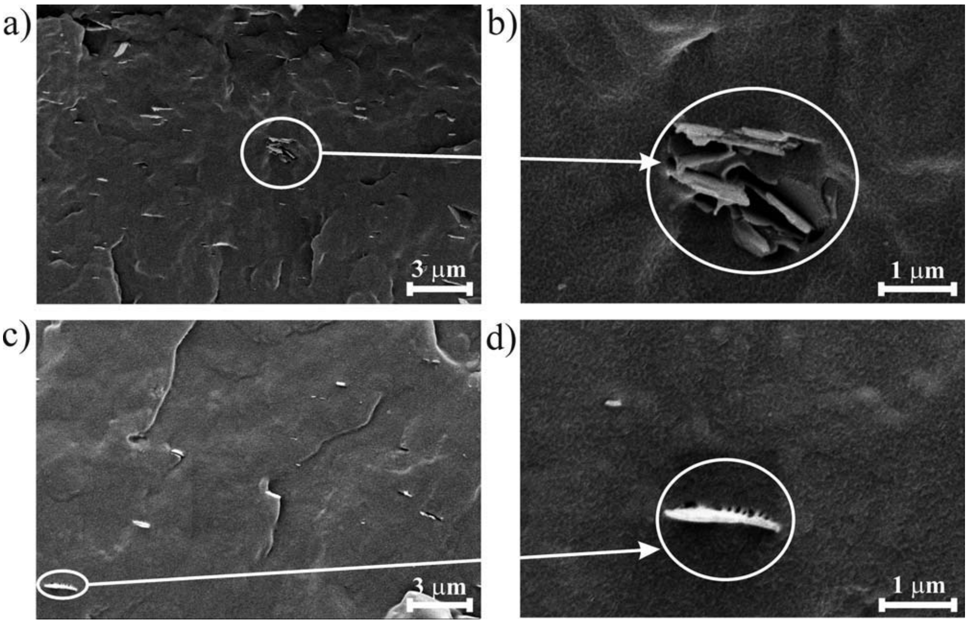

FIG. 2. SEM micrographs of: PP1A10 at (a) ×15,000, (b) ×50,000, and PP1A10A at (c) ×15,000, and (d) ×50,000.

dence of talc (treated and nontreated) on PP crystallization and composite properties, a systematic DSC study was carried out. Thermograms were recorded with the same thermal history and cooling/heating rate for PP and the composites. Table 2 shows the temperatures and enthalpies of crystallization and melting (named T_c, ΔH_c, T_m, and ΔH_m, respectively) of PP, PP/A10, and PP/A10A composites. In this table, the degree of crystallinity is also included (X_c %). There is a significant change in these parameters when PP and composites with 1 wt% of talc are compared, which is expected since this filler is a good nucleating agent of PP [2]. However, T_c and ΔH_c present a small variation with talc concentration. It can be explained from the fact that the nucleating effect is dominant during PP crystallization, but saturation is reached at about 1 wt% of talc [19]. Indeed, the addition of A10A talc leads to composites with T_c slightly lower than A10 ones: T_c reaches up to 125.6°C with 10 wt% of A10A and 126.5°C with the same concentration of A10. This may be due to the effect of grafted groups on the talc surface, which could hinder the PP crystallization.

The melting temperature of the composites shows an increment of about 2°C from PP value, for all talc concentrations and treatment, indicating that the crystalline perfection induced by this filler is similar among them. Moreover, ΔH_m increases slightly with talc content. A10A talc induces a relative higher degree of crystallinity in PP than A10 one, especially at low concentrations. Two opposite phenomena contribute to different crystallization between PP/A10 and PP/A10A composites, the increment in the number of nuclei and the possible hindering of organic groups attached to particles surface in A10A, respect to A10. In this sense, the increment of the nuclei number seems to be the responsible of the slight increment in crystallization degree, although saturation was reached. To investigate this phenomenon, a more detailed work, involving isothermal crystallization studies of these composites, show results that agree with these observations [20].

The stress–strain curves of PP and PP/talc composites are shown in Fig. 3. All the samples form neck before breaking, being their propagation the major difference between PP/A10 and PP/A10A composites. For PP specimens, within the neck, chains become aligned in the tensile direction, rendering localized strengthening. A similar mechanical behavior is observed for treated talc compo-

TABLE 2. Thermal parameters of PP and PP/talc composites.

Sample	T_m (°C)	ΔH_m (J/g)	T_c (°C)	ΔH_c (J/g)	X_c (%)	Sample	T_m (°C)	ΔH_m (J/g)	T_c (°C)	ΔH_c (J/g)	X_c (%)
PP	163.8	88.5	115.0	92.2	42.7						
PP1A10	165.8	92.4	126.0	92.7	44.6	PP1A10A	164.7	94.0	124.8	95.7	45.4
PP3A10	165.7	91.2	126.2	92.2	44.0	PP3A10A	165.8	94.4	123.8	96.5	45.6
PP5A10	166.0	92.1	126.2	92.5	44.5	PP5A10A	165.5	92.4	125.2	93.6	44.6
PP10A10	165.8	93.6	126.5	94.5	45.2	PP10A10A	166.0	94.1	125.6	94.0	45.4

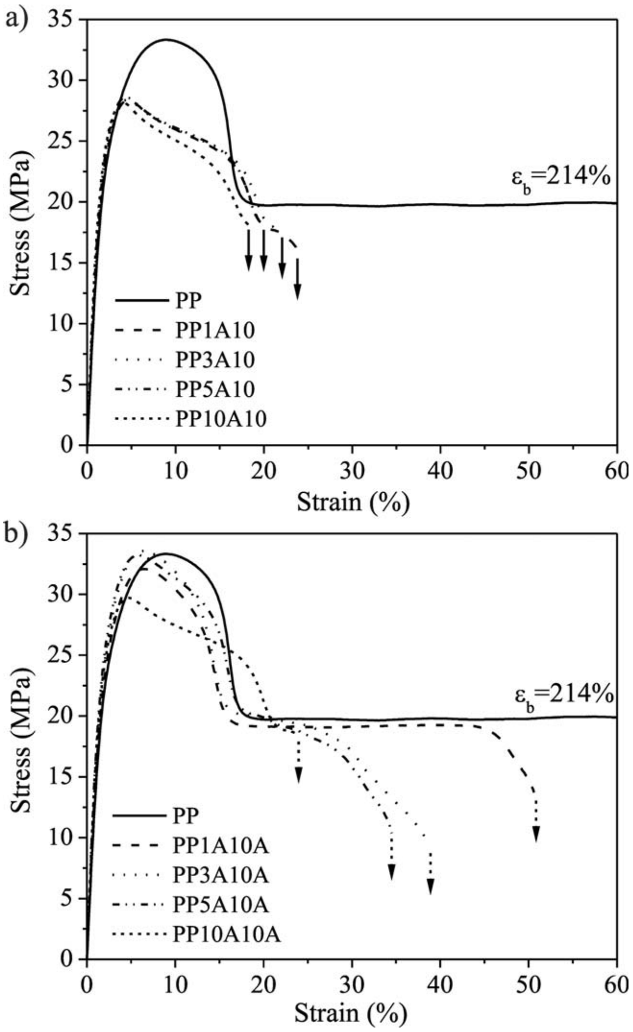

FIG. 3. Stress–strain curves of PP and PP/talc composites: (a) PP/A10 and (b) PP/A10A.

sites either for strength or for elongation at break, mainly at low talc concentration. This indicates that A10A-PP interphase resists enough, allowing the alignment of the chains and the neck propagation, in order to confine the deformation. However, and depending on the concentration, the strength and neck propagation are lower than in PP. As a consequence, the composite breaks at low deformation, as it is expected. This observation is in agreement with the presence of a large number of particles, due to particle delamination, upon treatment. At 10% of A10A talc, small agglomerations could occur and the interphase becomes weaker, lowering the strength and elongation at break, even so this sample elongates up around 20%.

The mechanical performance of untreated talc composites (PP/A10) differs considerably from PP/A10A. For untreated talc filled PP, the strength is about 20% lower. Also, in these samples, the neck forms but it does not propagate. This behavior can be attributed to the poor talc-PP adhesion and non homogeneous filler dispersion, which allows particle debonding at very low strain, impairing the elongation at break. It proceeds from the interphase failure, impeding the confination of the deformation within the neck region. The differences in mechanical behavior, mainly in the neck propagation, are clear evidence that the talc treatment improves the talc/PP adhesion, according to the observed in Fig. 2.

In addition, the overall mechanical behavior showed in stress–strain curves agrees with the macroscopical behavior presented in the photographs of PP and both composites specimens after mechanical testing (Fig. 4). Also, a fair amount of stress whitening is observed at the neck region of the composites, characteristic of the PP response [21].

In order to quantify the general behavior observed in Figs. 3 and 4, the tensile mechanical properties of PP and PP/talc composites are summarized in Table 3. It is noted that these properties exhibit a low standard deviation, indicative of a repetitive type of morphology for the tested specimens. This data confirms the results of structural analysis showed above, where a good talc dispersion and distribution within the matrix was observed (Fig. 1).

Young moduli (*E*) of composites, having both non-treated and treated talcs, resulted higher than of virgin PP value (Table 3); as would be expected. The PP modulus increases by talc addition, obeying to different reasons. First, talc has a significantly higher stiffness (about 170 GPa) than 1.7 GPa corresponding to PP [22, 23]. Also,

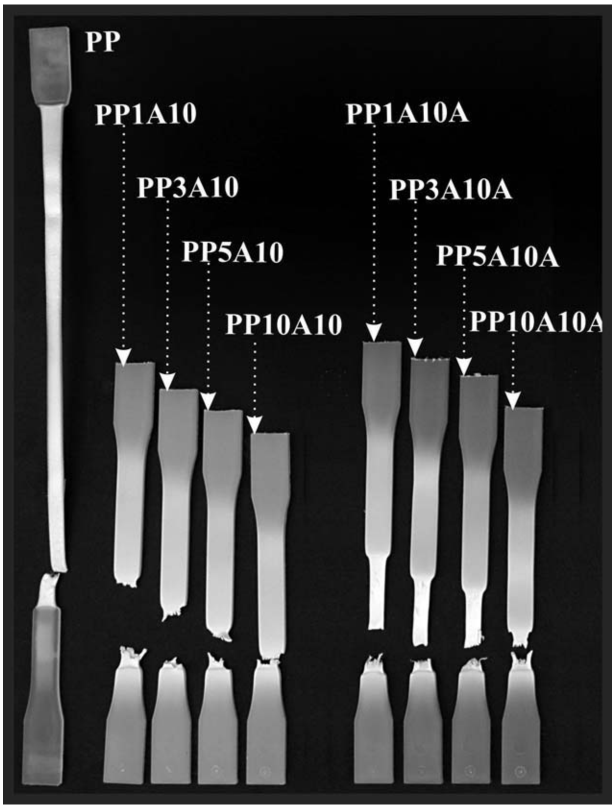

FIG. 4. PP and PP/talc composites specimens after mechanical testing.

TABLE 3. Mechanical properties of PP and PP/talc composites.

Sample	E (MPa)	σ_y (MPa)	ε_b (%)	Toughness (J/m^3)
PP	1623 ± 13	33.4 ± 0.2	214.5 ± 7.2	111.4 ± 4.0
PP/A10				
1%	1694 ± 20	28.6 ± 0.3	22.0 ± 3.2	12.4 ± 1.6
3%	1793 ± 18	28.2 ± 0.3	22.1 ± 3.1	12.5 ± 1.8
5%	1886 ± 24	28.5 ± 0.2	20.8 ± 2.2	12.1 ± 1.3
10%	2042 ± 24	28.1 ± 0.3	18.8 ± 2.2	10.9 ± 1.2
PP/A10A				
1%	1712 ± 20	32.0 ± 0.3	48.6 ± 7.8	25.4 ± 3.9
3%	1893 ± 25	34.2 ± 0.4	38.4 ± 4.9	21.2 ± 3.5
5%	1940 ± 8	33.1 ± 0.1	34.5 ± 2.3	19.4 ± 1.1
10%	2059 ± 32	30.0 ± 0.5	25.4 ± 5.0	15.0 ± 2.9

talc has a nucleating effect on the PP crystallization, leading to composites with more uniform and smaller sized PP crystals, which are often associated with an increased stiffness [24–27]. Finally, the aspect ratio and orientation of talc particles in PP matrix can increase the composite stiffness. These contributions are more important for higher filler concentrations, except for the nucleating effect of talc that reaches saturation at relatively low content (less than 1% w/w). Particularly, the increment in modulus, showed in Table 3, for 1 wt% talc can be assigned to the nucleation ability. The other contributions are less important in this case since, at low concentration, the modulus will not increase significantly by additivity [25]. As can be seen in Table 3, the modulus of PP/A10A composites are slightly higher than the measured for PP/A10 ones. This modulus variation can be as high as about 6% for 3 wt% talc. The observed increment could be explained in terms of the extremely small talc particle size, as a result of the acid treatment (see Table 1). It is important to note, that the crystallinity degree does not vary so much between PP/A10 and PP/A10A composites. Also, such treatment makes filler more hydrophobic that favors better talc dispersion in PP and allows a modulus increase by the formation of a percolation network, as reported by Svehlova and Poloucek'e [28]. A percolation theory, where each particle is surrounded by a matrix zone affected by a stress concentration, was formulated by He and Jiang [29]. In it, if the particles are dispersed close enough, these zones join together and form a percolation network, resulting in an increased modulus. Nevertheless, the increase in rigidity could also be accounted by modification of the matrix fraction near the treated particle surface. PP crystal phase close to the interphase, as nucleated by treated talc, has a more regular and fine morphology than the bulk PP matrix [25, 30].

The effects of talc surface modification on yield stress (σ_y) of PP/talc composites are also depicted in Table 3. The yield stress of composites depends on several factors: yield stress of polymer matrix, specific nature of the talc surface, filler–matrix adhesion and talc particles anisotropy. The tensile yield stress, a property determined under deformation effects, is a good indicator for interfacial interactions in heterogeneous polymer systems. In particular, any disruption or weakness at the interphase, that could reduce the stress transfer, will be magnified by such large deformation. The yield stress is strongly related to the particle size. The smaller the particle size, the greater the surfaces, consequently the more uniform the stress transfer throughout the composite which favors the yield stress. Also, the increment in σ_y may be associated with the finer crystalline morphology (smaller and thinner lamellar spherulites) by talc nucleated PP crystallization [19]. In this sense, a smaller spherulitic size is usually related to a greater yield stress [25]. The effect of talc treatment on the yield stress results in an increment up to 21%, as seen in Table 3, by comparing PP/A10A and PP/A10 composites at 3 wt% of filler. This result is consistent with works reported by other authors and it shows the incidence of stress transfer at the interphase on the yield stress [23, 31].

Considering the elongation at break (ε_y) of PP/A10 and PP/A10A composites, it is observed that both show a decreasing tendency with talc concentration, as it is expected. However, there is an increase up to 44%, when 3 wt% of treated talc is incorporated, compared to PP/A10. The reason of this improvement resides in the talc treatment, especially by the particle size reduction and the matrix–filler adhesion increment. In the same direction, the toughness increases when treated talc is used, as expected, being this effect more important at low concentrations where the increment is about 105%, respect to PP/A10. Then, this mechanical property decreases gradually with talc concentration up to 38% for 10 wt% of talc. The treated talc incorporation to PP allows the absorption of more energy than PP/A10.

The proposed simple talc treatment, by acid attack, allows the grafting of acetoxy groups onto the particle surface, rendering the talc more compatible with PP. The increment in talc/PP adhesion is evident from direct observation and it was corroborated by the increment of the mechanical properties.

CONCLUSIONS

The effect of talc modification on mechanical properties of PP/talc composites is analyzed. Taking into account that the studied talc treatment allows delamination, particle size reduction, and surface funcionalization; a comparative study of composite properties, with both nontreated and treated talc, is performed. Better particle dispersion and distribution, as well as an improvement in talc-PP interfacial adhesion, were obtained by using treated talc particles. These results are attributed to the hydrophobic character increment in the treated talc surface, respect to untreated ones, due to the presence of the grafted acetoxy groups.

The nucleation effect of talc in PP was corroborated by comparing the values of the temperature and enthalpy of crystallization between composites and pure PP. Moreover, the same parameters show slight changes between PP/A10 and PP/A10A composites, mainly at low concentrations. Although, treated talc has low particle size and better dispersion and distribution in PP composites, the crystallization temperature in PP/A10A is lower than in PP/A10. It can be attributed to a possible crystallization hindered by acetoxy groups which are grafted on A10A particle surface. On the other side, the crystallization reaches saturation at low talc concentration (1 wt%) as thermal properties present a slight variation with talc concentration for each composite set.

The overall mechanical performance of the composites is improved when treated talc is used. The modulus of PP/treated talc composites is slightly higher than the measured for PP/nontreated ones. This variation is within the experimental errors. Taking into account that, at the same talc concentration, the induced crystallization is similar; the slight modulus increment could be explained in terms of the particle size reduction and the more hydrophobic talc character in A10A talc. Both aspects favor the particle dispersion and allow the formation of a percolation network.

The tensile behavior of the two set of composites is different, evidencing the talc treatment effects on the matrix–filler adhesion. The results reveal that there is a notable increase in necking propagation length in PP/A10A composites respect to PP/A10 ones. In addition, the incorporation of treated talc leads to higher values of strength in comparison with PP/nontreated talc composites. This observation agrees with the major number of particles due to the particle delamination, favoring the stress transfer at the interphase. The elongation at break, which is the main parameter that indicates the improvement in the adhesion, strongly increases in PP/A10A composites respect to PP/A10 ones. In the same direction, the toughness increases when treated talc is incorporated to PP, absorbing more energy than PP/nontreated composites.

The complete mechanical behavior of PP/A10A composites and their enhancement comparing to PP/A10 ones is a corroboration of the effectiveness of the proposed talc treatment. This simple, effective and economic method seems to be a promissory alternative for surface modifications of fillers in order to improve their performance in polymer composite field.

REFERENCES

1. S.Y. Fu, X.Q. Feng, B. Lauke, and Y.W. Mai, *Compos.: Part B*, **39**, 933 (2008).
2. B. Pukánsky, “Particulate Filled Polypropylene: Structure and Properties,” in *Polypropylene. Structure, Blends and Composites*, Chapman & Hall, UK (1995).
3. M.Y. Boluk and H.P. Schreiber, *Polym. Compos.*, **7**, 295 (1986).
4. F. Candau, *Polymerization in Organized Media*; Gordon and Breach: Reading, PA, (1992).
5. E. Ruckenstein and J.S. Park, *Polymer*, **33**, 405 (1992).
6. M. Xanthos, *Functional Fillers for Plastics*, Wiley-VCH Verlag, GmbH&Co KGaA, Weinheim (2005).
7. H. Karian, *Handbook of Polypropylene and Polypropylene Composites, Headquarters*, Marcel Dekker, New York (2003).
8. E. Borsig, A. Fiedlerová, and L. Hrčková, *J. Macromol. Sci. Part A Pure Appl. Chem.*, **32**, 2017 (1995)
9. S.H.P. Bettini and J.A.M. Agnelli. *Polym. Test.*, **19**, 3 (2000).
10. S.B. Brown, in *Reactive Extrusion: Principles and Practice*, Oxford University Press, New York (1992)
11. F.P. La Mantia and J.L. Gardette, *Polym. Degrad. Stab.*, **75**, 1 (2002).
12. M. Alonso, J.I. Velasco, and J.A. de Saja, *Eur. Polym. J.*, **33**, 255 (1997).
13. A.W. Chuah, Y.C. Leong, and S.N. Gan, *Eur. Polym. J.*, **36**, 789 (2000).
14. L.A. Castillo, S.E. Barbosa, P. Maiza, and N.J. Capiati, *J. Mater. Sci.*, **46**(8), 2578 (2011).
15. A.L. Marinelli and R.E. Bretas, *J. Appl. Polym. Sci.*, **87**, 916 (2003).
16. B. Fisa, “Injection Molding of Thermoplastic Composites,” in *Composite Materials Technology*, Hanser Publishers, Munich (1990).
17. S. Barbosa and J.M. Kenny, *Polym. Eng. Sci.*, **39**, 1880 (1999).
18. L.E. Nielsen and R.F. Landel, *Mechanical Properties of Polymers and Composites*, Marcel Dekker, New York (1994).
19. J.I. Velasco, J.A. de Saja, and A.B. Martínez, *Revista de los Plásticos Modernos*, **477**, 271 (1996).
20. L.A. Castillo, S.E. Barbosa, and N.J. Capiati, *J. Appl. Polym. Sci.* DOI: 36846. Accepted for publication (2012).
21. J. Song, M. Prox, A. Weber, and G.W. Ehrenstein. *Self-Reinforcement of Polypropylene, in Polypropylene Structure, Blends and Composites. I. Structure and Morphology*, Chapman and Hall, London (1995).
22. F. Martinatti and T. Ricco, *J. Mater. Sci.*, **29**, 442 (1994).
23. M. Denac, V. Musil, and M. Makarovie, *Metals Alloys Technologies*, **32**, 69 (1998).
24. J. Menczel and J. Varga, *J. Thermal Anal.*, **28**, 161 (1983).
25. S.F. Xavier, “Development of Fine Morphology in Polypropylene Composites,” in *Two-Phase Polymer Systems*, Hanser Publishers, Munich (1991).
26. J. Kalfus and J. Jancar, *Polym. Compos.*, **28**, 365 (2007).
27. R. Popli and L. Mandelkern, *J. Polym. Sci., Part B: Polym. Phys.*, **25**, 441 (1987).
28. V. Svehlova and E. Poloucek, *Angew. Makromol. Chem.*, **214**, 91 (1994).
29. D. He and B. Jiang, *J. Appl. Polym. Sci.*, **49**, 617 (1993).
30. P.H.T. Vollenger and D. Heikens, *Polymer*, **30**, 1656 (1989).
31. M. Denac, V. Musil, I. Smit, and F. Ranogajec, *Polym. Degrad. Stab.*, **82**, 263 (2003).