Effect of the Talc Filler Content on the Mechanical Properties of Polypropylene Composites

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ABSTRACT: This research examines the effect of a microsize/nanosize talc filler on the physicochemical and mechanical properties of filled polypropylene (108MF10 and 33MBTU from Saudi Basic Industries Corp. and HE125MO grade from Borealis) composite matrices. A range of mechanical properties were measured [tensile properties, bending properties, fracture toughness, notched impact strength (at the ambient temperature and −20°C), strain at break, and impact strength] along with microhardness testing and thermal stability testing from 40 to 600°C as measured by differential thermal analysis and thermogravimetric analysis. Increasing filler content lead to an increase in the mechanical strength of the composite material with a simultaneous decrease in the fracture toughness. The observed increase in tensile strength ranged from 15 to 25% (the maximum tensile strength at break was found to be 22 MPa). The increase in mechanical strength simultaneously led to a higher brittleness, which was reflected in a decrease in the mean impact strength from the initial 18 kJ/m^2 (for the virgin polypropylene sample) to 14 kJ/m^2, that is, a 23% decrease. A similar dependency was also obtained for the samples conditioned at −20°C (a decrease of 12.5%). With increasing degree of filling of the talc–polypropylene composite matrix, the thermooxidative stability increased; the highest magnitude was obtained for the 20 wt % sample (decomposition temperature = 482°C, cf. 392°C for the virgin polymer). © 2008 Wiley Periodicals, Inc. J Appl Polym Sci 110: 2742–2747, 2008

Key words: composites; fracture; mechanical properties; poly(propylene) (PP); thermogravimetric analysis (TGA)

INTRODUCTION

In the beginning of the third millennium, polymeric materials represent the most important segment of all materials on the basis of production and usage volumes. In comparison with metals, polymers can offer better processing, a lower density, a higher strength-to-weight ratio, better resistance to corrosion, and often a better price/performance ratio.[1,2] A current increased demand for applications of synthetic polymers in the automotive and aeronautic industries is evident, mainly in the utilization of polyethylene, polypropylene (PP), polycarbonate, and polyamide components in the interior, exterior, and other functional parts of the vehicle.[3]

Minerals first served as additives to polymer systems as a cost-reducing technology: increased mineral addition led to a decreased polymer requirement.[4] The exploitation of mineral and polymer processing and chemistry has led to minerals being used as functional additives, bringing specific properties to the final composite.[4] Often these mechanical and physicochemical properties are optimized with the use of nanostructured or nanocomposite materials. Feynman[4] proposed in the late 1950s that materials whose structure would be possible to control on extremely small dimensions would exhibit new surprising properties. His forecast is now being realized.

In this study, we looked at the reinforcement of PP with the addition of the mineral talc. Talc is a phyllosilicate mineral with a trioctahedral layered structure [Mg$_3$Si$_4$O$_{10}$(OH)$_2$]. The idealized crystal structure contains a layered structure of a sandwich of magnesium oxide (brucite like) octahedra between tetrahedra of silica.[6] This leads to a neutrally charged system with all vacancies satisfied and with no net surface charge. As a result, the lamellar platelets are only held together by Van der Waals forces,

TABLE I
Processing Parameters Used for the Preparation of Samples A and B (SABIC; All Talc Compositions)
Through the Injection Molding of Pure and Filled PP

Injection pressure (bar)	Injection rate (mm/s)	Dye temperature (°C)	Screw resistance (bar)	rpm (1/min)	Melting temperature (°C)	Jet temperature (°C)
844	50	60	150	150	237	230

The parameters were kept constant for all samples.

which leads to talc being the softest mineral, defined as 1 on the Mohs scale. Furthermore, the main sites for chemical attack or amphiphilic reactions on the talc surface are mineralogical defects and platelet edges.[7]

Talc is a secondary mineral formed through the hydrothermal degradation of magnesium-rich metamorphic rocks.[8] For this reason, it is rarely found as a pure mineral, with chlorite groups {(Mg, Al, Fe)$_{12}$[(Si,Al)$_8$O$_{20}$](OH)$_{16}$}, magnesite (MgCO$_3$), and tremolite [Ca$_2$Mg$_5$Si$_8$O$_{22}$(OH)$_2$] being the most common impurities.[9]

The charge satisfaction of pure talc also leads to a different surface chemistry than many other clay minerals. There is a predominantly hydrophobic surface chemistry, which leads to poor wettability and dispersion in aqueous systems but a decreased dispersant demand when used in the polymer matrix. This surface chemistry, combined with the high aspect ratio of the particles (breadth/thickness), leads to the reinforcing properties that the addition of talc lends to commodity polymers.

The application of talc to PP is known to increase the modulus while also leading to a decreased impact strength.[10–13] PP has the highest stiffness and melting point and the best thermal resistance of all of the polyolefins. The modulus increase can be concomitant with an increase in the aspect ratio of platy minerals, but PP shows greater performance than, say, an uncoated kaolin, because of the greater compatibility of surface chemistries.[14] Because of the wide complexity of the PP molecular architecture and morphology, the synergistic interplay between several structural levels that starts from the atomic scale through the molecular architecture and supramolecular arrangements, which include crystallites and spherulites, up to the phase structure of blends and composites is vital for the final matrix mechanical properties.

The aim of this study was to better understand and characterize the basic physicochemical and material data of the highly filled microcomposite/nanocomposite matrix of PP–talc bases.

EXPERIMENTAL

Materials

PP [108MF10 natural copolymer (sample A); lot no. 6091218L23] and a talc-filled PP copolymer with ethylene–propylene–diene elastomer [33MBTU (sample B); lot no. 0060271436], both from Saudi Basic Industries Corp. (SABIC), were used in this study. For comparison, another PP [HE125MO grade (sample C)] from Borealis was used. Filled samples with different talc contents (5, 10, 15, 20, 25, and 30 wt %) were prepared by the homogenization of pure and filled PP in a Brabender microblender at 180, 185, and 195°C. The samples were then injection-molded in a Demag NC4 Ergotech 50-200 system. The processing parameters for samples A, B, and C are summarized in Tables I–III.

Methods

Tensile strength testing

Tensile strength tests were performed on a Tiratest 2160 universal tensile test machine according to standards CSN EN ISO 527-1 and 527-2. Fixing grips with a swinging pivot eliminated possible nonaxial loading. The applied deformation rates were 50, 200, and 500 mm/min. Measurements were performed

TABLE II
Processing Parameters Used for the Preparation of Samples A and B (SABIC) Through the Injection
Molding of Pure and Filled PP

	Sample/talc concentration (wt %)						
Parameter	A/0	B/5	B/10	B/15	B/20	B/25	B/30
Postpressing time (s)	25	25	25	24	23	23	21
Cooling time (s)	40	40	38	37	36	34	29

Variable parameters were used for different samples.

Master batch	Baker Perkins MP2030 corotating, twin-screw extruder with a temperature profile of 160, 150, 160, 165, 170, and 180°C (die), a screw speed of 300 rpm, and torque ranging from 25 to 40% (depending on the mineral)
Compound	Baker Perkins MP2030 corotating, twin-screw extruder with a temperature profile of 140, 150, 160, 165, 170, and 180°C (die), a screw speed of 300 rpm, and 50% torque
Injection molding	Arburg Allrounder 320M injection molder with barrel temperatures of 200–230°C, a mold temperature of 30°C, an injection rate of 35.5 cm^3/s, a holding pressure of 1000 bar, and cooling time as required

under ambient laboratory conditions (25°C and 50% relative humidity). Samples (dogbone coupons 150 mm in length) were conditioned before testing for 72 h at 25°C and 50% relative humidity. Acquired data were processed with SigmaStat Version 2.03 (SPSS, Inc.) software, and the maximum tensile load and elongation at break were determined. Each measurement was repeated five times.

Bending strength testing

Bending strength tests were performed on the Tiratest 2160 universal test machine according to standard CSN EN ISO 178-64 0607 (a three-point flexural test). Testing was based on the continuous loading of the test specimens up to their breaking point or up to a previously determined magnitude of deformation. During the testing process, the applied force on the specimen was recorded. The applied deformation rate was 5 mm/min. The support span length was adjusted to 60 mm. The determined maximum deformation was set to be 6 mm (i.e., 1.5 mm sample thicknesses), as the samples were not broken during testing. Each measurement was repeated five times.

Microhardness measurements

Surface microhardness was measured according to the Czech standard ČSN EN ISO 2039-1 ("Plastics—Determination of the Hardness—Part 1: Sphere Impressing Method") by use of a Rockwell-Rapid instrument. Measurements were performed at 25°C and 42% relative humidity. The sphere was impressed into six different positions of the test surface at a defined preload (132.4 N) applied for 10, 30, 60, 120, and 240 s. The results, shown later in Figure 2, are readings after 10 s. The surface of the imprint was calculated from the depth measured.

Microhardness is defined as the ratio of applied loading to the obtained surface of the imprint. Each measurement was repeated five times.

Fracture toughness

Fracture toughness was determined on a Charpy's hammer (0.4-kPm hammer) at 25 and −20°C for each material under study. *Charpy fracture toughness* is defined as the kinetic energy of the hammer used for the fracture of the tested specimen normalized to the specimen original cross section. Each measurement was repeated five times.

Notched impact toughness

Notched impact toughness was determined according to standard TL 526 31 on the Charpy's hammer (0.4-kPm hammer) at 25 and −20°C for each material under study. *Charpy notched impact toughness* is defined as the kinetic energy of the hammer used for the fracture of the notched tested specimen recalculated to the specimen original cross section. Each measurement was repeated five times.

Thermal stability

The thermal stability of the samples was characterized by means of thermogravimetry and differential thermal analysis. Measurements were performed on a DTG 60/60H differential thermogravimetric analyzer (Shimadzu) under an air atmosphere. Samples were in the form of granules (2 mm in diameter). The measurement parameters were as follows: heating rate = 10°C/min, starting temperature = 40°C/min, ending temperature = 600°C/min. The instrument temperature scale was calibrated by In and Zn. Mass decrease was measured with 2–3% error.

RESULTS AND DISCUSSION

The results of the mechanical tensile tests of samples A, B, and C (for various filler concentrations) are given in Table IV and are shown in Figure 1. The obtained results confirm the expected behavior, that is, the gradual increase in tensile strength with increasing filler concentration accompanied by a respective decrease in the elongation at break. Similar trends were obtained for all measured deformation rates. The observed behavior suggested that, with increasing filler content, samples did not preserve their original polymer chain rearrangement mobility, most probably because of the creation of specific particle–polymer interactions. Talc is a strong reinforcing filler because of its thin platy or flaky nature and possesses the capability of orienting to the polymer flow during processing.[10] Because the talc layers

TABLE IV
Summarized Results of the Tensile Tests of Samples A and B (SABIC) and Sample C (Borealis) with Different Talc Filler Contents

| Sample/talc concentration (wt %) | Deformation rate (mm/min) | | | | | |
| | 50 | | 200 | | 500 | |
	σ (MPa)	ε (%)	σ (MPa)	ε (%)	σ (MPa)	ε (%)
A/0	16.68 ± 0.12	265.25 ± 90.86	17.94 ± 1.83	130.82 ± 15.13	17.70 ± 0.37	130.50 ± 17.06
B/5	16.61 ± 0.41	95.82 ± 11.61	17.72 ± 0.52	67.57 ± 11.41	17.87 ± 1.98	66.54 ± 11.85
B/10	17.83 ± 0.45	83.30 ± 23.76	18.31 ± 0.28	58.21 ± 8.43	19.21 ± 0.96	66.54 ± 11.86
B/15	18.52 ± 0.35	47.51 ± 9.59	19.26 ± 0.24	37.53 ± 5.88	20.45 ± 0.61	50.74 ± 4.07
B/20	20.46 ± 0.83	24.35 ± 5.27	20.05 ± 0.34	28.78 ± 4.80	19.93 ± 0.98	28.92 ± 8.02
B/25	21.20 ± 0.43	13.21 ± 1.39	21.43 ± 0.11	15.08 ± 2.60	22.32 ± 1.50	6.94 ± 1.27
B/30	21.93 ± 0.39	5.22 ± 0.55	22.51 ± 0.24	7.29 ± 0.97	22.55 ± 0.19	18.68 ± 3.36
C/0	33.95 ± 0.52	15.05 ± 2.03	—	—	—	—
C/10	36.35 ± 0.80	16.80 ± 8.2	—	—	—	—
C/30	36.1 ± 0.82	5.80 ± 1.52	—	—	—	—

ε = elongation at break; σ = tensile strength.

in the talc/PP composites were aligned along the flow direction during injection molding, this could have also induced a preferential orientation of the PP macromolecular chains, as confirmed by wide-angle X-ray diffraction measurements.[15] Because of shearing during compounding, some talc layers might have been delaminated. Talc has a high aspect ratio, which increases the wettability of the filler by the polymer matrix, which thus creates fewer microvoids and increases the interaction between the filler and the matrix. The increase in tensile strength for the 50-mm/min deformation rate ranged from 16.68 to 21.93 MPa, a 31.5% increase. Corresponding to the latter increase of the tensile strength, a 98% decrease in the elongation at break was detected (a decrease from the original 265% to 5%). With increasing de-

formation rate, the loss of elongation was not so great for the 200-mm/min deformation rate (94%) and was 86% for the 500-mm/min deformation rate. The obtained increase in tensile strength for the virgin sample A for increasing deformation rates suggested successful pseudoplastic deformation of the amorphous polymer chain part. With increasing filler content, higher crystal-like morphologies were created, which were reflected in the latter mentioned increase in the tensile strength and which suggested substitution of PP by the largely more rigid filler. The filler could thus restrict the mobility and deformability of the matrix by the introduction of a mechanical restraint.[10] This effects could also be attributed to the fact that talc is a strong crystallization agent that affected the nucleation processes in the PP matrix, which might have changed the crystalline structure from weaker β spherulites to stronger α spherulites. As confirmed by thermal analysis data (see Table V), our samples' melting
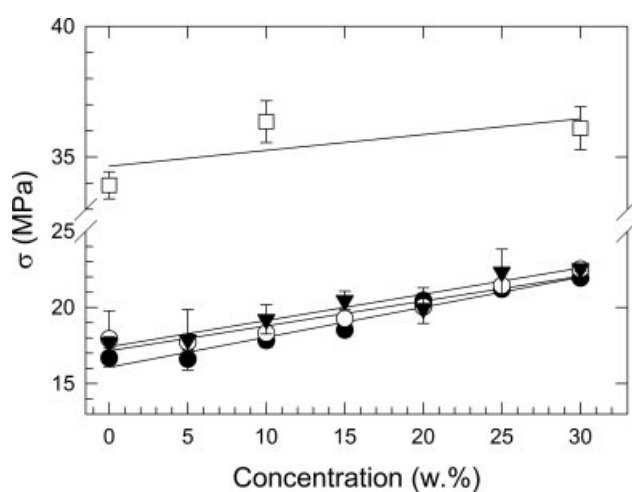

Figure 1 Talc filler concentration dependence of the tensile strength (σ) for samples A, B, and C. The applied deformation rates were (●) 50 and (○) 200 mm/min for samples A and B (SABIC) and (□) 50 mm/min for sample C (Borealis).

TABLE V
Results of the Thermal Analysis of Samples A and B (SABIC) by Thermogravimetry (TG) and Differential Thermal Analysis (DTA) Measurements

| Sample/talc concentration (wt %) | T_m (°C) by DTA | TG | |
		T_{start} (°C)	Weight loss (%)
A/0	172.6	260.5	100 ± 2
B/5	171.4	298.5	93.5 ± 1.9
B/10	174.9	280.1	88.8 ± 1.8
B/15	170.4	288.7	84.2 ± 1.7
B/20	170.6	303.6	78.2 ± 1.6
B/25	170.2	283.1	73.6 ± 1.5
B/30	171.5	275.4	67.6 ± 1.4

T_m = melting temperature; T_{start} = starting temperature of decomposition.
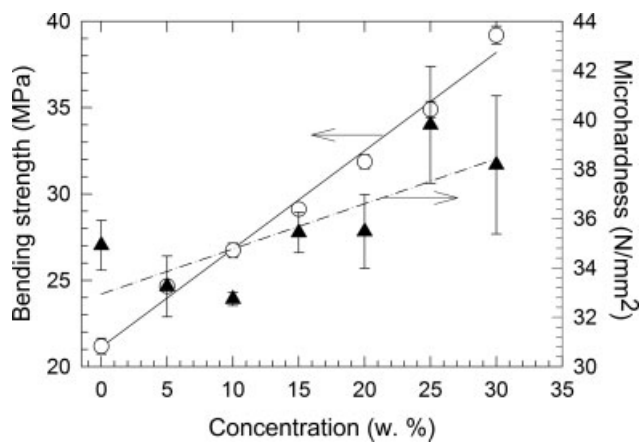
Figure 2 Talc filler concentration dependence of (○) the bending strength and (▲) the microhardness of samples A and B (SABIC).

temperatures were in the range of 170°C, which is typical for α modification (the melting temperature for β modification is approximately 152°C).[16,17] This conclusion was also supported by the very low observed β-phase indices (0.02) for talc-filled PP, whereas for unfilled PP, the index was approximately 0.06.[18] A similar trend was also confirmed in the corresponding increase of the bending strength from the original 21.18 to 39.20 MPa for the 30 wt % talc concentration, that is, an increase of 85% (see Fig. 2). As shown in Figure 2, with increasing filler concentration, the microhardness gradually increased by approximately 9% (from the original 34.94 to 38.18 N/mm^2). With increasing hardness, a corresponding increase in brittleness may be expected; thus, measurements of both the fracture toughness and notched impact toughness were per-

formed at ambient temperature and −20°C. The results are summarized in Figures 3 and 4. In the case of fracture toughness, dependencies at both ambient temperature and −20°C followed the same gradual linear decrease pattern with a gradient of −0.1 kJ/m^2 and abscissas at 16.7 kJ/m^2 (for 25°C) and 23.7 kJ/m^2 (for −20°C). Different dependencies were obtained for notched impact toughness measurements for SABIC PP, where at 15 wt % (for 25°C) and at 5 wt % talc concentration (for −20°C), a vigorous decrease in impact toughness was found, which suggested a step change in the sample brittleness. This phenomena corresponded to void formation and microcrazing. Of course, these processes might have been accompanied by the filler actively suppressing the polymer matrix's ability to undergo plastic deformation. This triggered a change in the failure mode of the PP matrix from ductile to brittle. Such a strong increase in brittleness corresponds to the change in the spatial arrangement of filler particles in the composite matrix, which was reflected in thermal gravimetric data. Here, a strong increase in the decomposition starting temperature was found in the case of 20 wt % talc concentration, from the original 260.5 to 303.6°C, which suggested the presence of strong intermolecular or interparticle bonds. Because of the fact that the studied injection-molded samples of which the mechanical properties were measured underwent a highly complex thermal and rheological history, one must bear in mind the fact that this will have had as profound an effect on the microstructure as the filler particles themselves.[11–13] As confirmed earlier by Rybnikar[19] by means of X-ray diffraction and by electron microscopy,[18] the talc platelets were arranged in parallel with each other in the matrix, which triggered
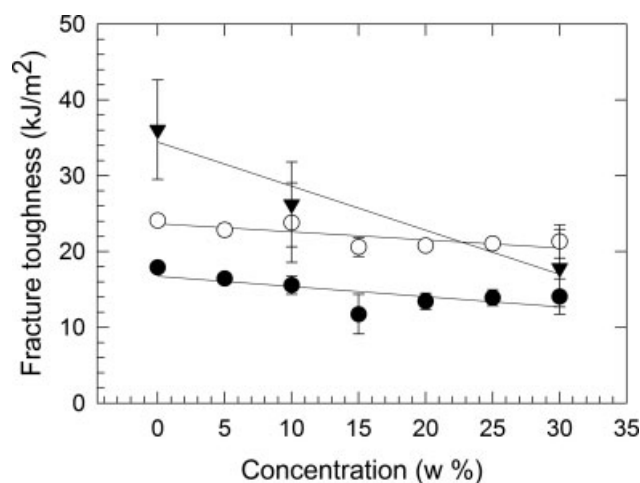

Figure 3 Talc filler concentration dependence of the unnotched fracture toughness: (●) +25 and (○) −20°C for samples A and B (SABIC) and (▼) 25°C for sample C (Borealis).
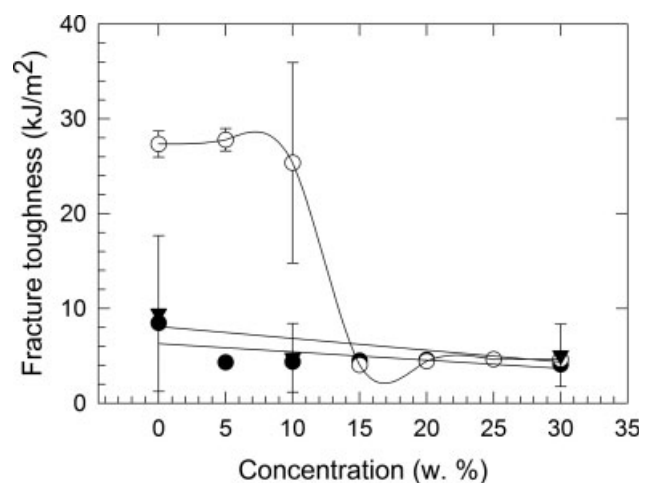

Figure 4 Talc filler concentration dependence of the notched fracture toughness: (●) 25 and (○) −20°C for samples A and B (SABIC) and (▼) 25°C for sample C (Borealis).

epitaxial growth of PP on the talc surface. This allowed anisotropic crack propagation that was rather easier parallel to the talc plates than perpendicular to them, which was most probably because of a reduction in crack pinning or blocking. The obtained decrease in fracture toughness could in part be attributed to the high stress concentration on talc sharp edges during composite deformation.[18]

CONCLUSIONS

In this study, with increasing degree of filler content, an increased mechanical strength of the PP–talc composite matrix was found. This effect was ascribed to the influence of the excellent dispersion and high order (crystal-like lattice) of microparticles (their spatial configuration) in the three-dimensional PP matrix and to their mutual synergistic effect, which resulted in the creation of specific bonds between individual particles and the polymer matrix. The observed increase in the tensile strength ranged between 15 to 25% [the maximum tensile strengths at break were found to be 22 MPa (SABIC) and 36 MPa (Borealis), respectively]. Simultaneously, an increased brittleness was obtained, which was reflected by a decrease in the mean impact toughness from the initial 18 kJ/m^2 (for the virgin PP sample) to 14 kJ/m^2, that is, a 23% decrease. A similar dependency was obtained also for the samples conditioned at $-20°C$ (a decrease of 12.5%). With increasing degree of filling of the talc–PP composite matrix, the thermooxidative stability increased; the highest magnitude was obtained for the 20 wt %

sample (the virgin unfilled sample showed a decomposition temperature at 392°C; the 20 wt % sample showed a decomposition temperature at 482°C).

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