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Nanocomposites of isotactic polypropylene reinforced with rod-like cellulose whiskers

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Abstract

Nanocomposite films of isotactic polypropylene reinforced with cellulose whiskers highly dispersed with surfactant were prepared for the first time and compared with either bare or grafted aggregated whiskers. Films obtained by solvent casting from toluene were investigated by means of X-ray diffraction, differential scanning calorimetry, dynamic mechanical analysis and tensile testing. Evaluation of the crystallization behavior showed that the aggregated or surfactant-modified whiskers induced two crystalline forms (α and β) in the nanocomposites and also acted as nucleating agents for isotactic polypropylene. The linear mechanical properties above the glass–rubber transition were found to be drastically enhanced for all three of the nanocomposites as compared to the neat polypropylene matrix, and these effects were attributed to a mechanical coupling between the polypropylene crystallites and filler/filler interactions. For the mechanical experiments at large deformations, the quality of the whisker dispersion was found to play a major role. The nanocomposites obtained with the surfactant-modified whiskers exhibited enhanced ultimate properties when compared to the neat matrix or to the composites containing the other filler types.

Keywords: Cellulose whiskers; Polypropylene; Nanocomposites

1. Introduction

Since the pioneering work of the Toyota group on the incorporation of clays in thermoplastic matrixes [1,2], the addition of different kinds of anisometric nanofillers (either plates or needles of nanometric sizes) to various thermoplastic polymers has become widespread [3–11]. Among these matrixes, isotactic polypropylene (iPP) has been particularly studied because of its wide use. However, the hydrophobic character of iPP renders the dispersion of hydrophilic nanofillers particularly challenging. The goal is, for the most part, to obtain an increase in the Young's modulus, but in general this comes at the expense of the ultimate properties of the material – the elongation at break is particularly sensitive to the quality of the dispersion.

Cellulose whiskers hold a particular place among the various types of anisometric nanofillers. In natural structures, they already act as reinforcing elements. Cellulose whiskers consist of slender parallelepiped rods with nanometric lateral dimensions, high aspect ratios and important surface areas, and also have a renewable character. The tensile modulus of a single whisker has been found to be 143 GPa [12]. Cellulose whiskers have mainly been employed as fillers in several kinds of polymeric matrixes from aqueous suspension, giving rise to very strong and tough percolating networks of hydrogen bonded whiskers [13–18].

In a previous study, we have shown that the incorporation of surface modified cellulose whiskers from an organic solvent suspension in the apolar, amorphous matrix of atactic polypropylene [19] was feasible. A great enhancement of the Young's modulus was observed. The study also revealed that the

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ultimate properties were dependent on the quality of the dispersion, which could be preserved when modifying the whiskers with a surfactant. Although a model system, the atactic polypropylene matrix is not a representative of polyolefins which are, at most of the time, semicrystalline. A natural continuation of the work on atactic polypropylene would be to investigate the introduction of cellulose whiskers in isotactic polypropylene, iPP, in order to explore the role of crystallites on the reinforcing effect of the fillers.

Because of its wide use, numerous studies have already been devoted to the reinforcement of iPP with nanofillers, both with and without surface modifications [3,20–23]. The microstructure of this crystallizable polymer plays an important role on the macroscopic behavior of its resulting composite materials. For reinforced crystallizable matrixes, microstructural modifications due to the presence of fillers add to the classical phenomenon of reinforcement, and iPP is a typically crystallizable polymer for which the crystal morphology and structure may be greatly affected by the presence of fillers [24,25]. The polymer can crystallize into three crystalline forms, i.e. the α, β and γ forms. Usually, the α-phase crystallites represent the predominant part of the neat iPP, whereas the appearance of the other two phases (β or γ) may eventually be favored by the presence of fillers, under high pressure [26,27] or by thermal annealing [28,29]. The presence of the β-phase may have an important influence on the mechanical properties of the resulting composites, since its toughness is higher than that of the α-phase at temperatures both above and below the glass transition temperature of isotactic polypropylene [30].

It is also well known that some substrates may influence the way polypropylene crystallizes. For example, calcium carbonate, $CaCO_3$, treated with stearic acid may induce β-phase crystallization [31]. The occurrence of the γ form of polypropylene is extremely rare, and usually generated at high pressure. However, it was shown that under certain conditions, a mixture of α and γ forms can develop at atmospheric pressures in the presence of different fillers [32].

Many studies have proved that macroscopic cellulose fibers act as nucleating agents for polypropylene [33–37]. In most cases the occurrence of a transcrystalline layer due to the high nucleation density has been observed. However, when nanoscopic cellulose whiskers are employed as fillers, their dimension prevents any experimental evidence of transcrystallinity by classical means, such as optical microscopy.

The aim of the present article was to identify the effects of the filler characteristics on the overall properties of the final iPP nanocomposites as well as the influence of the crystallinity on the reinforcing mechanism. The dispersion quality and surface activity of the whiskers were varied; either by maintaining the original whisker surfaces without modification, by grafting maleated polypropylene to the whisker surfaces [38] or by surface modification with a surfactant according to a process developed at CERMAV. This process enabled stable whisker suspensions in organic solvents where the whiskers were found to keep their fibrillar structure and their ability to form nematic phases [39]. According to a previous SANS study, the whisker cross sections could be described as homogeneously coated rectangles, and a thickness value of 15 Å could be extracted suggesting a folded conformation of the surfactant on the cellulose surface [40]. The crystallization behavior of the nanocomposites was examined by X-ray diffraction and differential scanning calorimetry and its effect on the mechanical properties of the materials was evaluated by dynamic mechanical measurements and tensile testing.

2. Experimental section

2.1. Polymer matrix

Isotactic polypropylene, iPP, with a number-average molecular weight of 22,000 g/mol and a weight-average molecular weight of 180,000 g/mol was supplied by Elf-Atochem. The iPP was used as received.

2.2. Cellulose whiskers from tunicin

Cellulose monocrystals, commonly called whiskers, were employed as the filler material. The whiskers were extracted from the sea animal tunicate and obtained as an aqueous suspension after an acid hydrolysis treatment [13]. The whiskers were 10–20 nm in diameter with lengths ranging from one to several micrometers, and thus had large aspect ratios of about 67 (estimated from transmission electron microscopy [13]) and significant interfacial areas of approximately 150 m^2/g.

Three types of whiskers with various surface characteristics were prepared in order to modify the filler/filler and filler/matrix interactions. The detailed description of their preparation has been reported elsewhere [19]. The first type was aggregated cellulose whiskers in toluene, denoted AGWH. The aggregated whiskers were obtained from individualized whiskers in water, which were freeze-dried and then redispersed in toluene. These whiskers had no surface modification, and the resulting dispersion was not stable and was found to flocculate at rest.

The second type was whiskers grafted with maleated polypropylene (PP*g*MA) according to an experimental process described elsewhere [38]. The grafted whiskers (GRWH) were dispersed in toluene by mechanical mixing with the Ultra-Turrax, and as for the aggregated whiskers the dispersion flocculated at rest.

The final type was novel as it consisted of whiskers suspended in a non-polar solvent. The suspension of cellulose in water was mixed with a phosphoric ester of polyoxyethylene-9-nonylphenyl ether (BNA, commercialized by CECA ATO Co.) at a weight ratio of 4:1 of BNA to cellulose. After adjustment of the pH to 8 with aqueous sodium hydroxide, the suspension was freeze-dried and then redispersed in toluene [39]. The final suspensions of the surface coated whiskers (SUWH) in toluene did not precipitate nor flocculate at rest.

2.3. Film preparation

Nanocomposites were prepared by mixing solubilized iPP in hot toluene (110 °C) with one of the three kinds of fillers

(AGWH, GRWH or SUWH) dispersed in toluene. The solvent was evaporated overnight in a ventilated oven at 80 °C to ensure the total solvent evaporation. The resulting iPP-composite materials were placed in a template frame consisting of folded aluminum foil sheets to ensure a constant film thickness and placed between press plates in a hot press heated to 200 °C. The assembly was left in the heated press for 14 min, without any pressure being applied, until the material was properly melted, and then pressed for 1 min at a pressure of 6.9×10^5 Pa. The samples were removed from the press plates and quenched in cold water after which the specimens were stored in sealed plastic bags in air awaiting analysis. The nanocomposite films reinforced with SUWH were practically transparent whereas the films reinforced with AGWH and GRWH were opaque, most likely because of the presence of aggregates of micrometric sizes and/or voids. The same fillers incorporated in an atactic polypropylene matrix have previously been shown [19] by SEM analysis to be properly dispersed in the case of SUWH and aggregated in the cases of AGWH and GRWH. It was thus assumed that equivalent states of dispersion were applied when isotactic polypropylene was used as the matrix.

2.4. X-ray diffraction

X-ray diffractograms were recorded at ambient temperature using a Siemens diffractometer D500, operating at 40 kV and 20 mA, equipped with a scintillation detector. Samples were mounted on a support and exposed to Cu Kα_1 radiation with a wavelength of 1.5406 Å. The angle of incidence, 2Θ, was varied from 10° to 25° by steps of 0.04°.

2.5. Differential scanning calorimetry (DSC)

Differential scanning calorimetry was conducted on a Setaram DSC 131 on the nanocomposite films. All DSC scans were carried out under nitrogen atmosphere on approximately 10 mg of material. The runs were performed from 60 °C to 200 °C (10 °C/min) and, after a 3 min isothermal, back down to 60 °C (10 °C/min) in order to observe the melting and crystallization behaviors of the film materials.

2.6. Dynamic mechanical analysis (DMA)

Dynamic mechanical analysis (DMA) was performed on an RSAII from Rheometrics. The experiments were conducted in tensile mode under isochronal conditions at a frequency of 1 Hz. The value of the strain magnitude was set at 0.05% in order to be in the domain of the linear viscoelasticity of the materials. Curves displaying the storage (E') modulus were recorded as a function of temperature between −100 °C and 200 °C at a heating rate of 3 °C/min. The shape of the film samples was rectangular, approximately 15 mm × 5 mm × 0.40 mm.

2.7. Tensile testing

The mechanical behavior at large deformations for the neat fillers, the unfilled matrix and the nanocomposites was analyzed with an INSTRON 4300 tensile tester with a load cell of 100 N. Experiments were performed with a cross head speed of 10 mm/min at room temperature, 25 °C. The sample dimensions were 15 mm × 5 mm × 0.40 mm, and the results were the averages of five measurements. The force, F, was recorded as a function of the sample elongation, $L-L_0$, and the stress, σ, and strain, ε, of the material could be calculated as:

$$\sigma = \frac{F}{A}$$

and

$$\varepsilon = \frac{L-L_0}{L_0}$$

respectively, where A was the cross-sectional area of the sample and L_0 was the initial sample length.

3. Results and discussion

3.1. Characterization of the nanocomposite films

Films of the nanocomposites as well as that of the neat iPP were characterized by X-ray diffraction and differential scanning calorimetry in order to identify the crystal morphology, the crystallization behavior and the thermal properties of the materials. Fig. 1 shows the X-ray diffractograms with the intensity as a function of the scattering angle, 2Θ. Five peaks

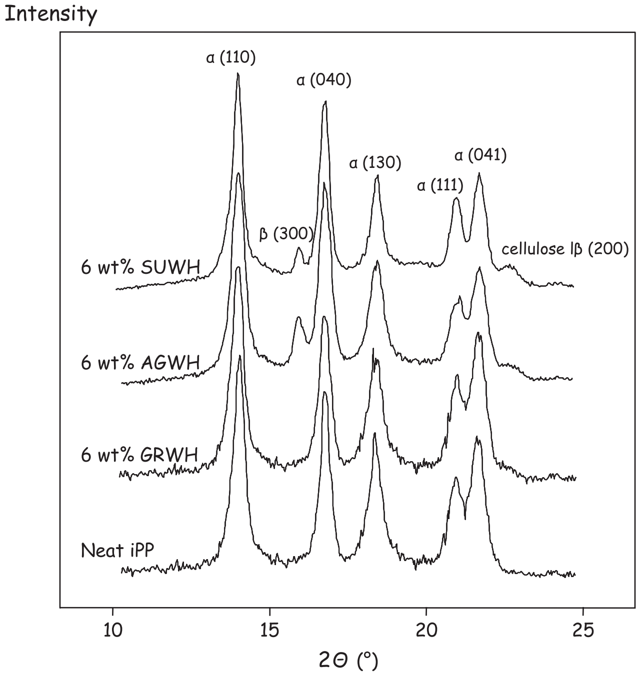

Fig. 1. X-ray diffractograms of films of neat iPP and composites reinforced with 6 wt% SUWH, AGWH and GRWH.

portraying the α-phase of iPP, given by the planes (110), (040), (130), (111) and (041), were expected at the scattering angles 14°, 17°, 18.5°, 21° and 22°, respectively. The most representative peak of the β-phase of iPP, the (300) plane, was expected at 16°, and the main cellulose peak corresponding to the Iβ plane (200) was expected at 23°.

The five α-peaks could be seen in all four materials at their corresponding diffracting angles (Fig. 1). The cellulose peak could also be determined from the diffractograms for the three nanocomposites, even though the low percentage of whiskers (6 wt%) gave rise to a peak of relatively low intensity. The neat iPP film did not display the presence of the β-phase and neither did the nanocomposite with GRWH. For the two nanocomposites reinforced with SUWH and AGWH, on the other hand, Fig. 1 clearly shows peaks corresponding to the β-phase. It could also be seen in the diffractograms that the intensity of the β-phase was larger for the AGWH composite as compared to the one reinforced with SUWH. These results indicate that the more hydrophilic the whisker surface, the more it appeared to favor the appearance of the β-phase. A comparison can be made to results of iPP reinforced with stearate-coated carbonate [31]. In this case, the appearance of a β-phase was also observed, suggesting that the presence of a surfactant induced a nucleating effect of this phase within the polymer.

The thermal properties of the four materials were investigated by means of DSC and the characteristics from the runs are summarized in Table 1. Fig. 2a shows the DSC traces displaying the endothermic melting peak for neat iPP and the three nanocomposites. The four materials displayed similar thermograms and the small differences in their melting temperatures were within the experimental precision of the equipment. Fig. 2b displays the exothermic peak of the crystallization, obtained when the melted materials were cooled at 10 °C/min. The SUWH and AGWH nanocomposites displayed double crystallization peaks whereas the neat iPP and the material containing GRWH showed single peaks. These results indicate that the unmodified whiskers and those modified with a surfactant acted as nucleating agents for the iPP matrix; the unmodified whiskers having the largest nucleating effect. The whiskers grafted with maleated polypropylene on the other hand did not act as a nucleating agent. This leads to the conclusion that the nucleation of iPP is influenced by the surface characteristics of the whiskers.

It is highly probable that there existed a correlation between the appearance of β-phase in the X-ray diffractograms and the early onset of crystallization upon cooling for the AGWH and SUWH nanocomposites. However, such a correlation would certainly not be straightforward. One must take into account that while the results from the X-ray diffraction and DSC measurements did support each other qualitatively, their quantitative values could not be compared. This is most likely due to the differing crystallization conditions experienced in the DSC capsules and in the bulk material, especially in terms of temperature gradients. It is, however, apparent that only AGWH and SUWH seem to be β-nucleating, as compared to the neat iPP and GRWH systems.

In terms of melting and crystallization enthalpies, the values of neat iPP are portrayed in Table 1. When taking into account the fact that the nanocomposites contained only 94 wt% of the matrix material it could be stated that the films of 6 wt% AGWH and 6 wt% SUWH displayed equivalent enthalpy values to the neat iPP. The composite containing 6 wt% GRWH on the other hand showed a significant decrease in these values. Judging from these results in crystallinity as well as the fact that AGWH and SUWH were believed to be β-nucleating, variations were expected in the individual microstructures of the nanocomposites.

3.2. Dynamic mechanical analysis (DMA)

Films of the nanocomposites with 6 wt% of the three whisker types as well as the neat iPP were analyzed by DMA. Fig. 3 gives the evolution of the storage modulus, E', at 1 Hz vs temperature of the four materials. The unfilled matrix displayed the typical behavior of a semicrystalline polymer. For temperatures below T_g the polymer was in the glassy state and the modulus decreased slightly with temperature, but remained roughly constant, around 3.5 GPa. The onset of a significant drop of the tensile modulus, corresponding to the glass–rubber transition, was then observed. The crystalline regions of the iPP, which can be considered as physical crosslinks, enabled the material to maintain a significant modulus after the amorphous relaxation. As the temperature was increased the modulus decreased only slowly as a result of the crystalline relaxation. Above 150 °C, the modulus dropped irremediably due to the melting of the crystalline zones of iPP.

Fig. 3 also displays the temperature dependence of the storage moduli of the three nanocomposites. As a consequence of the differences in surface modification for the three filler types, they should give rise to different dispersion qualities, various interfaces with the matrix and different filler/filler

Table 1
Thermal data obtained by DSC measurements for films of neat iPP and composites reinforced with 6 wt% AGWH, GRWH and SUWH

Sample	Melting temperature (°C)	Enthalpy of melting (J/g)	Crystallization temperature (°C)	Enthalpy of crystallization (J/g)	Degree of crystallization[a] (%)
iPP	163	101	115	109	52
6 wt% AGWH	162	91	115/127	94	46
6 wt% GRWH	163	75	115	74	38
6 wt% SUWH	164	94	115/122	103	49

[a] Calculations are based on ΔH_m for 100% crystalline iPP being equal to 209 J/g [44]. Consideration was taken to the nanocomposites containing only 94 wt% iPP.

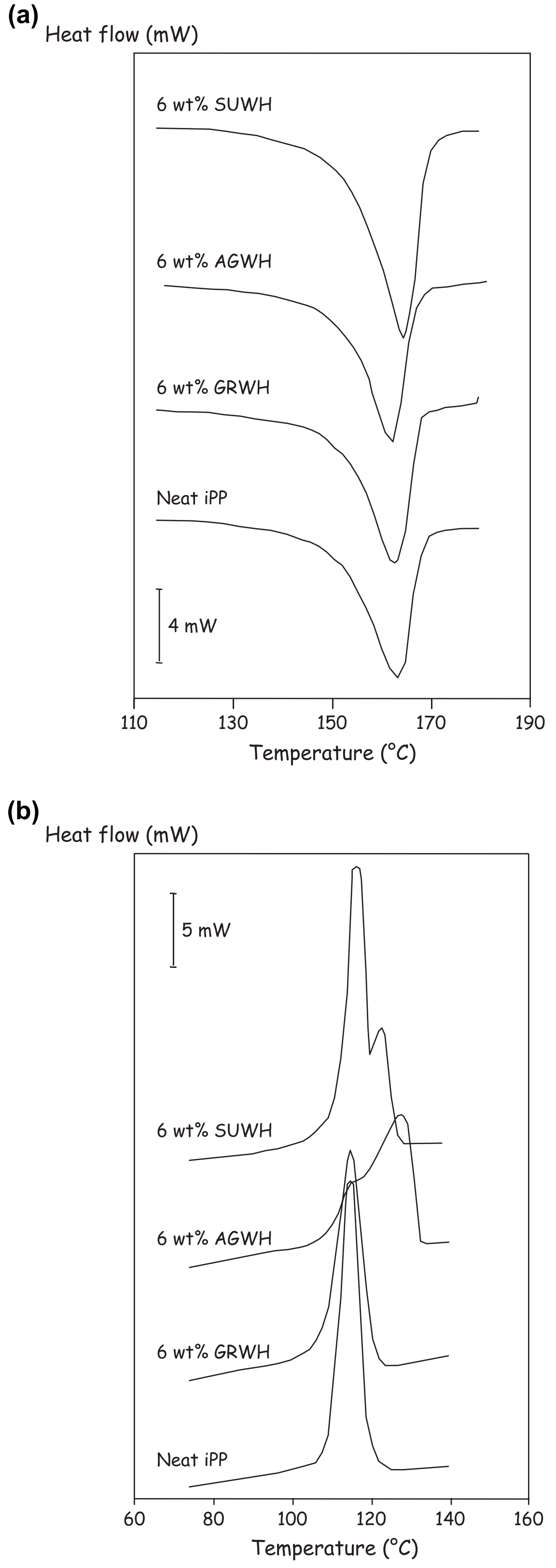

Fig. 2. DSC thermograms portraying (a) the melting and (b) the crystallization of neat iPP and composites reinforced with 6 wt% SUWH, AGWH and GRWH.

interactions [19]. Below T_g, the moduli of the nanocomposite materials differed somewhat from the neat iPP matrix as usually observed for a configuration of rigid fillers in a matrix below its T_g [13]. In the present experimental results, the SUWH composite displayed a higher modulus whereas the other two materials showed slightly lower moduli. A method that accounts for possible experimental errors in the modulus, induced by for instance the sample dimension, is the normalization of all the curves to a given value [41–43]. The results in the present study could, however, be explained by the GRWH and AGWH nanocomposites having lower densities than expected as a consequence of poor dispersability and the formation of aggregates (and thus air pores) in the matrix giving rise to the lower storage modulus values. The SUWH material, on the other hand, displayed a better dispersion leading to a material without aggregates or pores, and thus the reinforcing effect of the whiskers became visible.

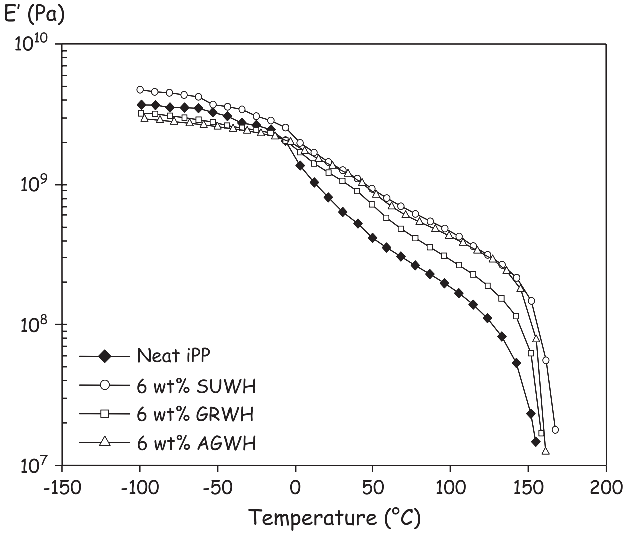

Fig. 3. The temperature dependence of storage modulus curves from DMA runs comparing experimental data for films of neat iPP and iPP reinforced with 6 wt% SUWH, GRWH and AGWH.

Above T_g, there was a significant mechanical reinforcement for the composite materials (Fig. 3). The drop in storage modulus associated with the glass transition and the crystalline relaxation was dramatically reduced as compared to neat iPP; for instance, at 100 °C the composite containing 6 wt% SUWH presented a modulus almost three times that of the neat matrix. When comparing these results with those obtained for composites containing mineral fillers [3], cellulose whiskers appear to be equally efficient with the advantage of having much lower densities. The augmentation in storage modulus at high temperatures was the highest for the composites containing SUWH and AGWH, and somewhat lower for the GRWH material. These results suggested that the larger degree of crystallinity in the SUWH and AGWH nanocomposites, as compared to the GRWH material, enhanced their mechanical properties. It should also be noted that the nanocomposite materials displayed their irremediable modulus reductions at higher temperatures than the neat matrix. Particularly, the film reinforced with 6 wt% SUWH showed an improved thermal stability as it was able to maintain a significant modulus up to 170 °C.

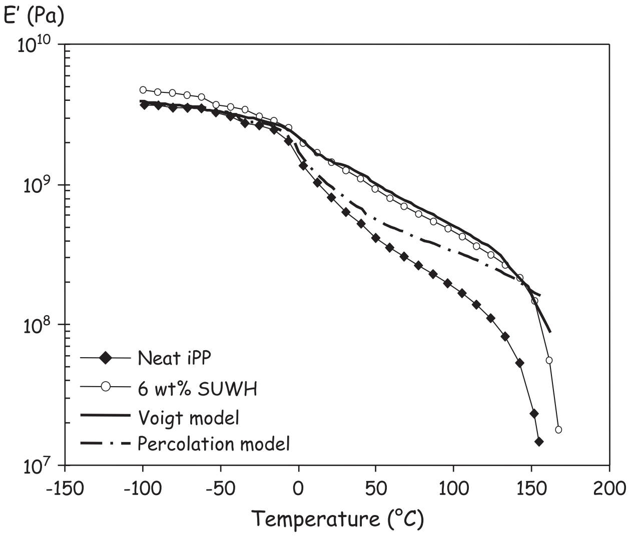

Fig. 4. The temperature dependence of storage modulus curves from DMA runs comparing experimental data for films of neat iPP and iPP reinforced with 6 wt% SUWH, with predicted data from the theoretical Voigt and percolation models.

Fig. 4 gives the experimental tensile modulus curves for neat iPP and the nanocomposite containing 6 wt% SUWH, and compares them with theoretical predictions. The Voigt and percolation models were calculated for 6 wt% of SUWH by taking into account that the modulus of an SUWH sheet was 1.4 GPa as determined in a previous study [19]. Detailed calculations can be found elsewhere [13].

Within the experimental precision of the measurements in the glassy state, both models appeared to slightly underestimate the modulus of the composite. At higher temperatures, the percolation model clearly underestimated the measured data, whereas the Voigt model predicted a modulus that better described the experimental data. In the case of atactic polypropylene (aPP) reinforced with 6 wt% of the same surfactant-modified whiskers, the nanocomposite displayed a storage modulus below that of the Voigt model and much closer to the percolation model, suggesting a percolating network with dead branches [19]. On the one hand, the nature of the two matrixes is not comparable because aPP is completely amorphous and iPP is semicrystalline. Though the Voigt model represents an upper boundary in terms of modulus for a composite, it does not take into account the possible ultrastructural changes in the crystallinity (e.g. type of phase, overall crystallinity, etc.). On the other hand, as was clearly shown from the X-ray diffractograms (Fig. 1) and the DSC measurements (Table 1), the crystallinity in the iPP/SUWH composite decreased and changed form (went from α to β to a certain degree), and such a behavior would in normal circumstances have led to a decrease in storage modulus. However, nothing is known on the precise mechanism of crystallization, and it is possible that transcrystallization occurred around the whiskers. Thus, the large increase in modulus that was observed suggests that in the semicrystalline iPP there existed a mechanical coupling between the crystallites in the matrix and the reinforcing whiskers. This phenomenon can be interpreted in terms of a continuous network of reinforcing fillers and matrix crystallites without dead branches, as usually described by the percolation model [13]. The existence of such a percolating network is further supported by the fact that it was the two systems that showed the highest reinforcing effect in the DMA measurements (Fig. 3), i.e. the SUWH and AGWH composites, that also displayed a whisker-induced nucleating effect upon cooling according to the DSC thermograms (Fig. 2b).

3.3. Tensile testing

The mechanical behavior at large deformations for films of the neat iPP matrix as well as the composites reinforced with 6 wt% of the three filler types was performed by tensile testing at room temperature. Fig. 5 shows the stress–strain curves of the four film materials. The tensile strength, σ, and elongation at break, ε, were determined from the curves and the results are presented in Table 2.

It can be observed that the unfilled matrix displayed a nonlinear elastic behavior with a tensile strength of 19 MPa and an elongation at yield of 0.08. As a result of its semicrystalline nature, the tensile behavior for neat iPP, was characterized by extensive deformation after yielding. By incorporating 6 wt% of one of the fillers, the tensile strength was drastically changed. Contrary to the neat matrix, none of the three composite materials showed signs of stress-whitening or yielding. This led to believe that the mechanism behind the elongation and rupture of the composites was quite different from that of the neat iPP, and that the values of stress at break for the composites in Table 2 should be compared with that of the stress at yield for the iPP sample.

The nanocomposite reinforced with 6 wt% AGWH displayed reduced mechanical properties as compared to the neat iPP for both tensile strength (15 MPa) and elongation at break (0.04). As a consequence of the aggregates in the AGWH composites, zones with accentuated fragility were created, which lead to a brittleness in this material. These tensile

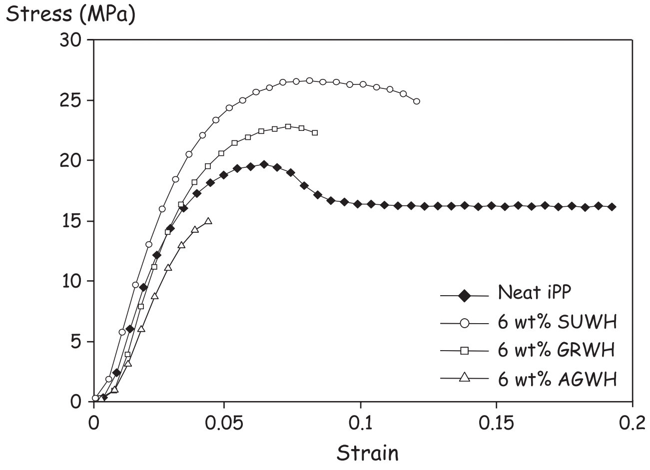

Fig. 5. Stress–strain curves for films of neat iPP and composites reinforced with 6 wt% SUWH, GRWH and AGWH.

Table 2
Tensile strength, σ, and elongation at break, ε, for films of neat iPP and composites reinforced with 6 wt% AGWH, GRWH and SUWH

Sample	σ (MPa)	ε
Neat iPP	19	0.08[a]
6 wt% AGWH	15	0.04
6 wt% GRWH	23	0.09
6 wt% SUWH	27	0.12

[a] Elongation at yield.

testing results and the lack of compatibility between the whiskers and the matrix in the AGWH composites thus led to the conclusion that there were very little or no stress-transfer properties in this material [12]. The GRWH composite, on the other hand showed enhanced mechanical properties in comparison with the neat matrix. The tensile strength was larger (reaching values of 23 MPa), and the elongation at break was slightly longer (0.09) than the elongation at yield for iPP. Thus, by grafting PP*g*MA to the cellulose whiskers, a better compatibility as well as better stress-transfer properties between the fillers and the matrix was enabled.

The film displaying the best results in terms of mechanical properties was the SUWH composite. Significant increases were observed in tensile strength (27 MPa as opposed to 19 MPa for iPP) and the elongation at break was 50% larger than the elongation at yield for the neat matrix (see Table 2). These features did not follow the classic trend in polymer composites where any factor that improves stiffness is detrimental to elongation. However, the simultaneous enhancement of both tensile strength and elongation at break in the SUWH/iPP nanocomposite confirmed what was already observed for atactic polypropylene reinforced with the same whiskers [19]. The incorporation of the whiskers enabled the increase in stiffness while due to the presence of the surfactant the decrease in plasticity could be avoided.

Thus, by modifying the surface characteristics of the cellulose whiskers through addition of a surfactant, and thereby obtaining a better dispersability, the mechanical and stress-transfer properties of the whisker-reinforced nanocomposite could be significantly enhanced. At temperatures well above ambient, the surfactant-modified whiskers are potentially even more efficient, as can be concluded when taking into account the higher reinforcing effect observed in the DMA experiments at such temperatures (Fig. 3).

4. Conclusions

Cellulose whiskers with three types of surface characteristics were prepared: aggregated whiskers without surface modification, AGWH, aggregated whiskers grafted with maleated polypropylene, GRWH, and novel surfactant-modified whiskers, SUWH. The whiskers were incorporated as nanometric fillers in isotactic polypropylene, iPP, by solvent casting from toluene followed by film pressing. The crystallization behavior of the films, as evaluated by X-ray diffraction, displayed two crystalline forms (α and β) in the nanocomposites containing AGWH and SUWH, whereas the neat matrix and the material reinforced with GRWH only crystallized in the α-form. Differential scanning calorimetry experiments also indicated that the aggregated and surfactant-modified whiskers acted as nucleating agents for the iPP.

The mechanical properties of the nanocomposite films were evaluated in the linear range by dynamic mechanical analysis, and were found to be significantly enhanced by the incorporation of the cellulose whiskers. Particularly, the materials with SUWH and AGWH displayed increased storage moduli as compared to the neat matrix and the GRWH composite. The experimental data above the glass transition temperature from the DMA measurements were accurately predicted by the theoretical Voigt model. It was thus concluded that a mechanical coupling most likely existed between the reinforcing effect of the fillers and the crystallites in the matrix.

At large deformations, the mechanical properties were found to depend strongly on the dispersion quality of the whiskers. The AGWH composite had mechanical properties inferior to the neat iPP as a result of the whisker aggregates rendering the material fragile. The polypropylene grafts in the GRWH composite enabled a better dispersion quality between the whiskers and the matrix as compared to the AGWH composite, but the most efficient filler dispersion was obtained in the SUWH nanocomposites as a result of the surface modification with surfactant. The high dispersion quality induced enhanced mechanical properties at large deformations at room temperature and the SUWH composite displayed an increased tensile strength and strain at break as compared to the neat matrix. There is also reason to believe that the tensile properties of the nanocomposite would be even more superior to those of the neat matrix at elevated temperature, making a polypropylene composite containing surfactant-modified cellulose whiskers an interesting candidate for high-temperature applications.

Acknowledgements

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