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ORIGINAL PAPER

Supermolecular structure of wood/polypropylene composites: I. The influence of processing parameters and chemical treatment of the filler

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Abstract Wide-angle X-ray scattering (WAXS) and scanning electron microscopy (SEM) were used to investigate the effect of chemical modifications of wood as well as processing parameters on the supermolecular structure and morphology of wood/polypropylene (PP) composites. The surface of the filler was modified to enhance the adhesion between hydrophilic cellulose and hydrophobic polymer. Wood modification was performed by esterification with anhydrides (maleic, propionic, phthalic, crotonic and succinic) and by physical treatment with NaOH. The composites for structural characterization were prepared by the compression moulding method and injection technique. A new design of the compression mould, which ensured the wood pulling, was proposed. It was found that the polymorphs of PP matrix developed due to melt-shearing strongly depended on the pulling temperature as well as on the type chemical modification of wood. The modification of wood caused a significant decrease in the ability to generate the hexagonal phase of PP. Moreover, at a higher temperature of the mould, the amount of hexagonal phase of PP matrix slightly decreases. These investigations are very significant because characterize influence of real parameters processing as well as surface modification of filler on structure of composite materials.

Keywords Isotactic polypropylene · α-, β-polymorphs · Wood · Composites · WAXS

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Introduction

The composites of engineering polymers with natural components like wood, flax, hemp, ramie, etc. have recently been intensely studied because of the interesting properties of such materials [1–6]. Biodegradable natural components can act as reinforcing agents improving the tensile and flexural strengths, resistance and absorbing capacity of the product [7]. One of the essential problems in the technology of engineering polymers/natural filler composites is the poor adhesion between the components, because many matrixes (like polyethylene, polypropylene) are hydrophobic whereas the surface of wood is hydrophilic [8]. Improvement in the adhesion between the two components is a key to success in the area. Various strategies have been proposed to improve the compatibility between the lignocellulosic reinforcement and the polymer matrix: (1) the use of compatibilizer, (2) grafting of polymers onto the lignocellulosic components, (3) chemical modification of the lignocellulosic materials. Modification can be achieved by chemical pretreatment of wood with anhydrides [8, 9], silane coupling agents [10, 11] and isocyanates [12, 13]. Moreover, maleic anhydride modified polypropylene (PP) and such copolymers as styrene butadiene styrene (SBS) were applied as compatibilizing agents to improve adhesion [14, 15].

The mechanical properties of lignocellulosic materials/PP composites depend not only on the strength of adhesion, but also on the conditions of processing. An additional important problem, closely related to the processing of semicrystalline polymers like PP, is the supermolecular structure of polymer matrix, because the natural component can act as a nucleant for thermoplastic polymer causing changes in the morphology or even changes in the arrangement of chains in the crystal lattice of the matrix.

Moreover, analysis of the effect of processing conditions, during preparation of the composites, on the structure of polymer matrix is a focal issue because polymorphic changes in PP can be stimulated by the temperature of processing and the shearing forces applied. The isotactic PP can exist in several structural modifications [16, 17]: monoclinic (α), pseudo-hexagonal (β) and triclinic or orthorhombic (γ). The formation of polymorphs is controlled by the crystallization rate of the molten PP. The crystallization rate can be regulated by the processing conditions and shearing forces. The fact that shearing forces promote the hexagonal form of iPP was first observed by Leugering and Kirsch [18]. Polymer processing technologies (e.g. injection moulding, extrusion) impose intense shearing flows and have prompted investigation into the crystallization of polymer induced by shearing. Many authors [19–24] have observed that during shear treatment, crystalline polymer chains get oriented in the melt and can crystallize with both nucleation and growth different from those under quiescent conditions. Trotignon et al. [25] reported a detailed study of skin-core structures in injection-moulded PP, which showed that variations in the degree of crystallinity and the hexagonal phase were related to a complex layered morphology. Kalay and Bevis [26] noted that the difference in the relative proportions of α- and β-phases depends on processing conditions and the moulding method. They observed that the core region of iPP mouldings is composed mainly of small α-phase spherolites with sporadic β-phase spherolites.

The processing of composites with natural materials is difficult because of the temperature sensitivity of lignocellulosic component and the necessity of taking into regard additional shearing at the interface between the components. The problem of polymorphism in isotactic PP in the presence of fibres has been studied [27–32], but the mechanisms of formation of particular forms and their transformations in composite systems is not fully resolved yet. Varga and Karger-Kocsis [28] observed the presence of hexagonal phase of iPP after crystallization of molten PP with moving glass and carbon fibres. In our earlier model experiments by the WAXS method, the hexagonal form was found to appear when the flax or hemp fibres were moved [29, 30]. The content of the hexagonal form depends on the pulling temperature and on chemical modification of natural fibres, as well as on the speed of fibres in molten iPP. Recently, the composites of iPP with unmodified flax fibres as well as those modified by acetic anhydride obtained by extrusion have been analysed [33]. It was found that after extrusion the monoclinic (α-iPP) and hexagonal (β-iPP) crystal phases were present in PP matrix. The content of the β form depends on the temperature of the extruder die and the content of fibres.

Up to now, the composites obtained by compression moulding have been made of the monoclinic α-phase only, because the shear forces were not present. In this work, the shearing forces and pressure were applied simultaneously in the newly proposed compression process. Hence, the main aim of this study is the evaluation of the influence of pulling and chemical modification of wood on the supermolecular structure of PP matrix. In addition, the relationship between the conditions of the injection moulding and the polymorphic structure of polymer matrix was also studied for different composite systems.

Although many authors have studied PP and filler composites in recent years, the influence of chemical treatment of wood and processing parameters on the supermolecular structure of the composite materials has not yet been comprehensively analysed. Moreover, understanding of the influence of the shear fields applied and chemical treatment of the filler on the characteristic features of the crystalline phase, i.e. its polymorphic forms, is paramount for optimization of processing conditions of the composite materials.

Experimental

Materials

The isotactic PP matrix used in this study Malen F-401 was supplied by Basell Orlen Polyolefins (Plock, Poland) with a $\mathrm{MFI}_{230/2.16}$—2.4–3.2 g/10 min, isotacticity—95 %, $T_\mathrm{m} = 163$–164 °C. Scotch pine wood (*Pinus silvestris*) dust sieved through 1-mm mesh size was used as filling material. Moreover, pine wood plates (thickness—0.7 mm) were applied in press moulding.

Chemical modification of wood

To improve the adhesion between components, different types of chemical treatment of wood were realized. Before chemical modification the wood was dried at 70 °C for 24 h in a vacuum oven.

Mercerization process

Wood was treated with an aqueous solution of NaOH (17.5 %). The pine wood was immersed in the solution for 60 min at room temperature. After alkali treatment the wood was washed with distilled water to remove the excess of NaOH and then dried in the air at elevated temperature (ca. 110 °C). This process is the first step in modification of wood surface performed in order to remove waxes as well as fats and in consequence the fibres surface becomes more accessible to chemical reagents.

The process of mercerization has strong effect on the crystal structure of cellulose inducing the transformation of cellulose I (native form, Cell-I) to cellulose II (Cell-II). The efficiency of mercerization was estimated by the WAXS method [34].

Esterification reactions

In this study, to improve the interaction between wood and PP, the following anhydrides were used: maleic, propionic, crotonic, succinic and phthalic.

Part of the wood (after alkali treatment) was immersed in a one-molar solution of anhydride in xylene and then heated at reflux temperature for 8 h. The accurate procedure of chemical modification of wood, separation of wood from xylene solution and extraction process is described in our previous articles [35, 36]. The esterification process was confirmed by the FTIR spectra obtained and calculated as weight percent gain [35, 36].

Preparation of composite materials

Compression moulding

The PP and wood were compounded using compression moulding with the mould of a new design proposed by us, which enabled pulling of a wood plate in the molten polymer matrix.

The board composites of the size 100 × 100 × 3 were prepared in two steps. In the first step, the boards of PP of the size 100 × 100 × 1.2 mm were obtained.

The study was performed on the wood board of 0.7 mm in thickness and 80 mm in width. Such a board known as veneer was obtained on a veneer slicer. The principle of its work is as follows: a log of pine wood is mounted centrally in the lathe and turned against a razor sharp blade. Since this cut follows the log's annular growth rings, wood plate is produced. This method is widely used in furniture industry for the production of wood boards of small thickness.

The untreated or chemically modified wood plate was placed in the axial clearance of the press mould constructed according to the newly proposed design (clearance size 80 × 0.7 mm). Subsequently, the iPP plates were supplied to this compression mould and such a sandwich was heated. A scheme of the preparation of the composites in this process is presented in Fig. 1.

The composite materials were heated to 200 °C at the rate of 10 °C/min. and held at this temperature for 5 min, in order to eliminate the effects of their thermal history. The pressure used in the experiments was 0.2 MPa. Subsequently, the samples were cooled to 60 °C at the rate of 10 °C/min. During such non-isothermal crystallisation, the wood plate was pulled mechanically in one direction at the rate of 0.66 mm/s. for 15 s at different temperatures (T_{pull}). The pulling temperatures were in range from 130 to 180 °C. A scheme of the temperature program is presented in Fig. 2.

Injection moulding

In first step, the isotactic PP/wood composites were obtained by the extrusion method using a "Fairex" (Mc Nell Akron Repiquet, France) single-screw extruder, L/D = 25. During the extrusion, the temperatures of the three processing zones were chosen as: 140, 180, 195, respectively, and the extrusion die temperature of 190 °C. The extrusion speed was 25 to 30 rpm. Extrusion temperature was kept below 200 °C to avoid decomposition and degradation of wood. The product obtained contained 50% of wood.

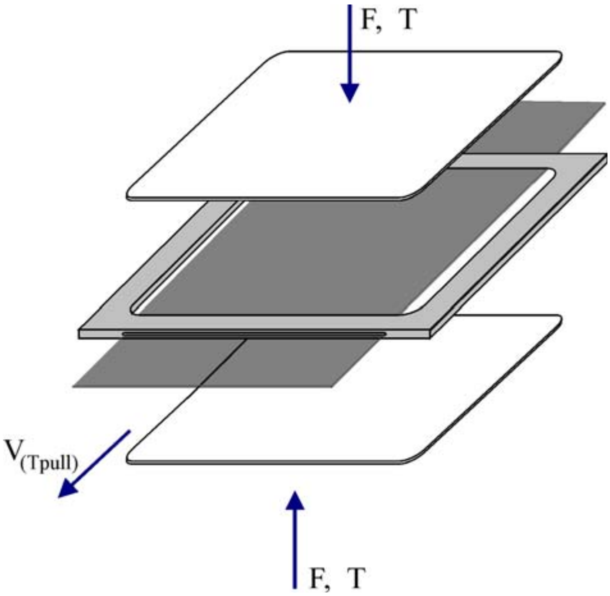

Fig. 1 Scheme of the new design of compression mould enabling the pulling of wood plate ($V_{(T_{pull})}$ rate of pulling, *F* pressure force, *T* processing temperature)

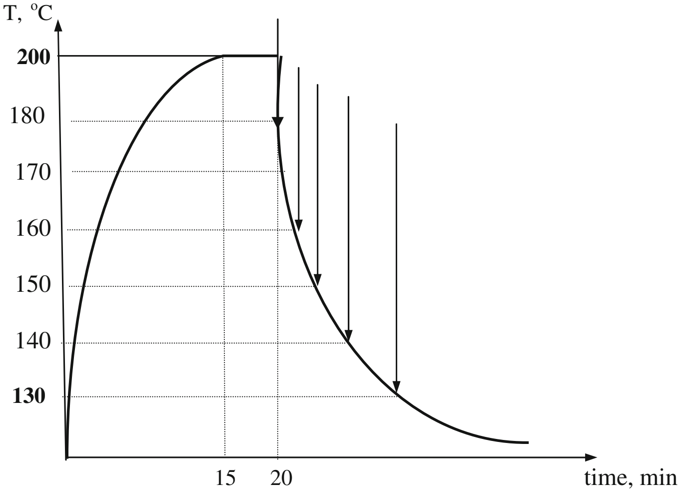

Fig. 2 Scheme of the temperature program of compression. The *arrows* indicate the temperatures at which the wood plate was pulled

The samples used for the structure investigation were prepared using Engel injection moulding machine. The composite materials were obtained using a barrel temperature of 210 °C, injection pressure of 60 bar, cooling time of samples in injection mould of 30 s. Moreover, two different mould temperatures of 30 and 45 °C were applied. After moulding, the specimens were immediately sealed in a polyethylene bag, and placed in a vacuum desiccator for minimum of 24 h prior to structural testing.

Structural investigations

For the WAXS measurements, samples obtained from the single-screw extruder were injection moulded into disks of 35 mm diameter and 3 mm thickness. Moreover, the composites were compression moulded with the nominal dimension of 100 × 100 × 3 mm. These composites were cut into specimens (disks) of 35 mm in diameter and 3 mm in thickness. To eliminate possible orientation effects, samples were analysed in the rotation mode. Measurements were performed with the X-ray beam perpendicular to the flow direction in the moulded piece as well as to the wood plate pulling direction in the compression moulding.

The structure of iPP matrix was analysed by means of wide angle X-ray scattering (WAXS) using CuKα radiation at 30 kV and 25 mA anode excitation. The X-ray diffraction pattern was recorded for the angle range of $2\,\Theta = 10\text{–}30°$ in the step of $0.04^{\circ}/3$ s. Deconvolution of peaks was performed by the method proposed by Hindeleh and Johnson [37], improved and programmed by Rabiej [38]. After separation of X-ray diffraction lines, the contents of β-phase (k) was calculated by using the Turner–Jones formula [39]. The changes in the supermolecular structure of PP were analysed as a function of pulling temperature of the wood fillers and of the mould temperature.

Surface morphology of different wood surfaces was revealed by scanning electron microscopy (SEM).

The abbreviations of the composite materials studied in this article are the following: PP/cW—polypropylene and unmodified wood; PP/prW—polypropylene and wood modified with propionic anhydride, PP/maW—polypropylene and wood modified with maleic anhydride, PP/crW—polypropylene and wood modified with crotonic anhydride, PP/suW—polypropylene and wood modified with succinic anhydride, PP/phW—polypropylene and wood modified with phthalic anhydride, PP/merW—polypropylene and mercerized wood.

Results and discussion

The supermolecular structure of composite materials was analysed as a function of: (a) chemical modification of wood and (b) processing parameters like pulling temperature of wood in polypropylene matrix and temperature of injection mould. The polymorphic changes in PP taking place on applying the extrusion or injection method were studied in many works [33, 40–43]. The presence of shearing forces causes induction of hexagonal form. However, during press moulding the shear between wood and polymer matrix does not occur and consequently the monoclinic phase only exist.

In this study, the influence of pulling of a wood plate in polymer matrix on the supermolecular structure of iPP was analysed. The X-ray diffraction patterns show the crystallisation of iPP in the presence of pulling of a wood plate (Fig. 3).

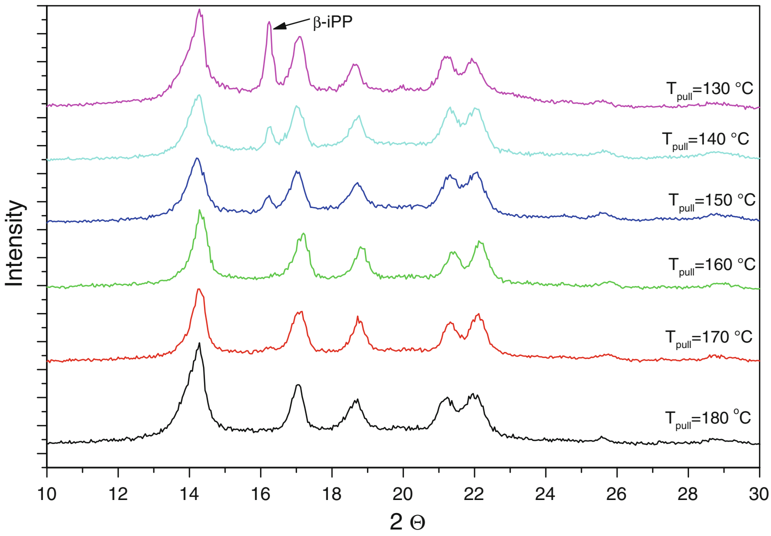

Fig. 3 WAXS patterns of PP/suW during crystallization at different pulling temperatures

After crystallisation with moving wood, the presence of β-phase of iPP besides the α-one is observed (diffraction maximum of β-phase at 16.12° of 2Θ). It can also be observed that on increasing the pulling temperature of wood, the intensity of the reflection corresponding to β-phase decreases, disappearing at around 160 °C. WAXS patterns indicate that the β-phase was predominant in the samples containing wood pulled at 130 °C. When the wood was pulled in PP matrix at above 160 °C, only the α-phase was found.

It has been established that the development of polymorphs of PP matrix due to melt-shearing (caused by wood pulling) strongly depends on the pulling temperature as well as the type of chemical modification of wood. The modification process of wood caused a significant decrease in the ability to induce the hexagonal phase of iPP. In the presence of unmodified wood the β phase (28% in $T_{\text{pull}} = 130$ °C) forms in greater amount than that of esterificated wood (about 20% in $T_{\text{pull}} = 130$ °C) (Fig. 4).

The results prove that the modification of wood with the anhydrides does not influence the contribution of hexagonal phase. However, the composite systems containing wood modified by acid anhydrides were characterized by greater contributions of this phase than the materials subjected to mercerization process.

The amount of β-iPP polymorph also depends on the temperature of the compression mould. The formation of hexagonal phase has been found particularly profound at low pulling temperature. With increasing pulling temperature, the amount of β-form decreases rapidly and reaches a minimum when the pulling temperature is 160–180 °C. In the composites with unmodified wood, the amount of

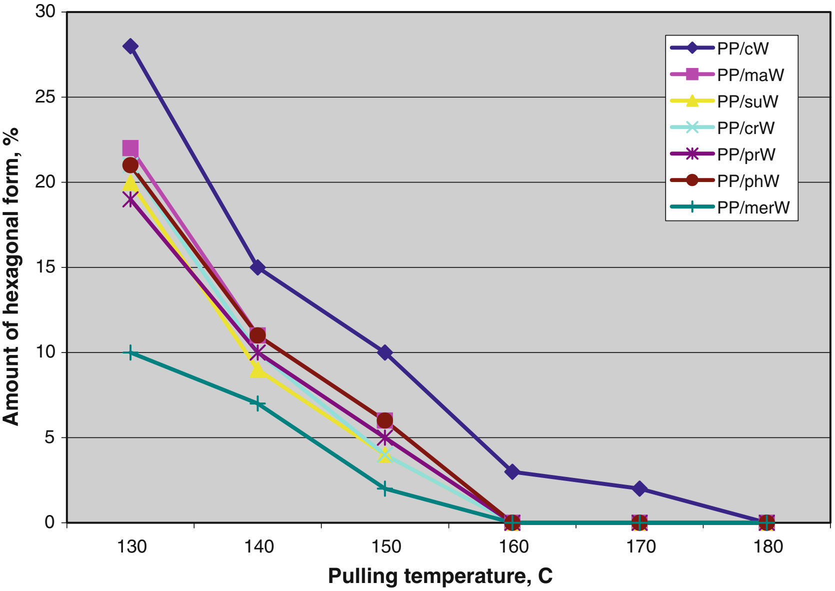

Fig. 4 The amount of the hexagonal phase versus pulling temperature for the composites with unmodified and chemically modified wood

hexagonal form rapidly decreases up to 160 °C and for higher temperatures remains at the same level (up to 180 °C). On the basis of the WAXS data, it was possible to determine the critical temperatures (T_{cr}) of wood pulling above which the β-iPP phase did not form. Interestingly, the T_{cr} values were observed to decrease significantly as a result of chemical modification of wood with anhydrides or alkalies. The critical temperature in the composites of PP and unmodified wood was 180 °C, while in those of PP and modified wood it was 160 °C.

A similar character of the dependence of polymorphic structures of iPP versus temperature processing during extrusion was found in recent works [33]. However, in the case of extrusion of crude and also chemically modified cellulosic fibres, the presence of the hexagonal form up to 180 °C was noted. Moreover, the composites obtained by extrusion process showed significantly greater content of β-iPP phase (in $T_{extrusion} = 140$ °C, β-iPP 28%; in $T_{extrusion} = 150$ °C, β-iPP 26%; and in $T_{extrusion} = 160$ °C, β-iPP 20% [33]) in comparison to those obtained by compression moulding. This indicates that the formation of the hexagonal form of iPP in the composite systems depends on the cooling rate. In the method of compression moulding, the cooling rate was lower (10 K/min) in comparison to that in the extrusion process (>100 K/min). Our observations suggest that the phase $\beta \rightarrow \alpha$ phase transition is blocked in the PP-unmodified fibre system due to a faster rate of crystallization.

Our experiments correspond to the results reported by Varga and Karger-Kocsis [28], who analysed the pulling of glass and carbon fibres in PP matrix in model systems. They have established that the polymorphism of iPP strongly depends on the pulling temperature. According to these authors, the shearing of fibres yields α-row nuclei and the surfaces of these α-row nuclei may induce formation of β-form of iPP.

Analysis of the kinetic aspect of spherolitic growth has shown that the formation of the β-phase of iPP, induced by shearing is preferred when the growth of β-phase ($G\beta$) is greater than that of α-form ($G\alpha$) [44]. The kinetic differences in the spherolitic growth ($G\beta > G\alpha$) were observed only over a certain range of temperatures.

To check the influence of processing parameters on the polymorphous structure of PP matrix, the composites obtained by the injection method were cooled at mould temperatures. Figure 5 shows the contribution of the hexagonal phase in all the composite materials analysed.

The composite systems containing wood modified by acid anhydrides or by alkaline chemicals were characterized by the lower contents of β-form than the systems without any modifications (PP–cW). It should be stressed that the PP matrix was characterized by distinctly the lowest amount of hexagonal form (below 10%). As follows, the type of the acetic anhydrides applied for modification of pine wood has no influence on the content of β-form. The development of iPP polymorphs in the process of injection moulding is determined by the temperature and shear flow. In addition to the control parameters related to the processing conditions of injection technique, the important structure-related parameters to consider include the temperature gradient of moulding and mould temperature. At the higher temperature of moulding (45 °C), the amount of hexagonal form in the composites slightly

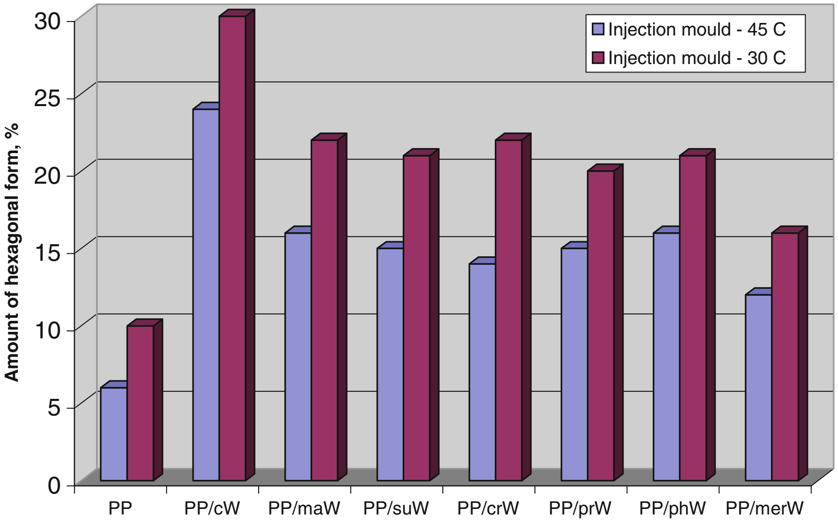

Fig. 5 The amount of hexagonal phase versus different temperatures of injection mould for the composites with unmodified and chemically modified wood

decreases (about 20–30%). It is also worth noting that the content of hexagonal phase in PP matrix is the higher the lower the mould temperature. The investigation is of interest as it provides information on the effect of real parameters of processing and the filler surface modification on the composite structure.

According to our results, high rate of cooling (at a low mould temperature) favours formation of β-form. This result agrees well with literature [45] claiming that for PP, on application of shear stress, part of the PP macromolecules would orient forming row nuclei. These row nuclei would begin to grow at a higher temperature than the growth of the homogeneous system. At a high degree of undercooling, β-nuclei might also be formed on the α-row nuclei or on the growing α-cylindritic front [45].

Another explanation of the variation in the amount of the hexagonal form versus mould temperature may be related to the kinetic aspect of formation of both polymorphic structures of isotactic PP. The phase transition from the metastable β-phase to the stable α-phase has been extensively studied [44, 46–49]. The authors of this concept maintain that the polymorphic transition is a characteristic feature of β-isotactic PP, which transforms from the metastable β-phase to the thermodynamically more stable α-form via a recrystallisation process. Garbarczyk [50] suggested that one of the factors determining the formation of a given form are perturbations in mobility of the chains during crystallization, which would make it difficult for polymer chains to reach the arrangement with the lowest potential energy (α form). The phase transformation process consists of a number of intermediate stages based on rotations and transitions of the iPP chains. Also, Varga [45, 51, 52], on the basis of a DSC study, showed that the formation of α-phase from β one is preceded by an endothermic effect identified as melting of the β-form, this finding implies that the molten hexagonal form recrystallised into the monoclinic phase.

In establishing the role of mould temperature, relaxation should play an important role during crystallization. Huo et al. [22] has reported that at low temperatures is impossible to eliminate the shear-induced orientation and fully stretch iPP chains before the beginning of iPP crystallization. As follows from our results, the contribution of β-phase was greater when lower mould temperatures were applied. A possible explanation is that at higher temperatures, the relaxation is faster and crystallization is slower. Our results show that the rate of crystallization of the PP matrix modulated by mould temperature plays an important role in the $\beta \rightarrow \alpha$ phase transition.

Moreover, it is very interesting that chemical modification of wood with anhydrides and sodium hydroxide, resulted in a significant decrease in the shear force in the interface region, manifested as lower content of β-form than in the system with unmodified wood. In addition, the adhesion improved following the chemical treatment of wood with acid anhydrides, increases the content of hexagonal form relative to that in the system with mercerized wood. Xie et al. [53] studied PP crystallisation in PP composites with sisal prepared by injection moulding. The addition of a compatibilizing agent, SEBS, reduced the contribution of hexagonal phase in PP. One of the possible explanations of this fact can be a probable change in the surface topography of wood fillers. Numerous literature data confirm that surface roughness of different fillers is one of the possible causes of development of transcrystalline layers, as has been observed for flax [3], carbon and Kevlar fibers [54].

The surface morphology of pine wood was examined by SEM. Wood topography study was used to explain the different abilities of the wood samples to induce the appearance of polymorphic form of iPP. Figures 6a–g illustrate the surface topography of pine wood revealed by the SEM method.

Small ridges, sharp edges and grooves are usually present on the unmodified wood surface (Fig. 6a). It is suggested that the presence of grooves and roughness of the surfaces enhances the nucleating ability. A very different picture of wood surface emerges after the mercerization process (Fig. 6b) and chemical modification with the anhydrides (Fig. 6c–g), as a result of which the wood surface is smooth with only small irregularities. The mercerised wood has smooth surface because of the removal of wax and fats. Such smooth surfaces reduce the nucleation ability. However, after chemical modification of wood with anhydrides the roughness of the wood surface increased, which led to its greater nucleation ability in comparison to that of mercerized wood. Analysis of wood by SEM indicated that differences in the nucleation activities of wood fillers are associated with differences in topography of their surface.

The differences in the surface topography (grooves and roughness) between the unmodified and chemically modified wood can explain the mechanism of α-row nuclei formation on the surface fillers in the composite materials. Varga and Kocsis [28] studied interfacial morphologies and the structure in carbon fiber–iPP composites. They speculated that during shear-induced crystallization of iPP, the α-row nuclei formed in situ could induce the growth of β-form. Also, Somani et al. [23, 55] found that formation of oriented α-crystals occurred immediately after the application of the shear field and subsequently, the growth of primarily unoriented

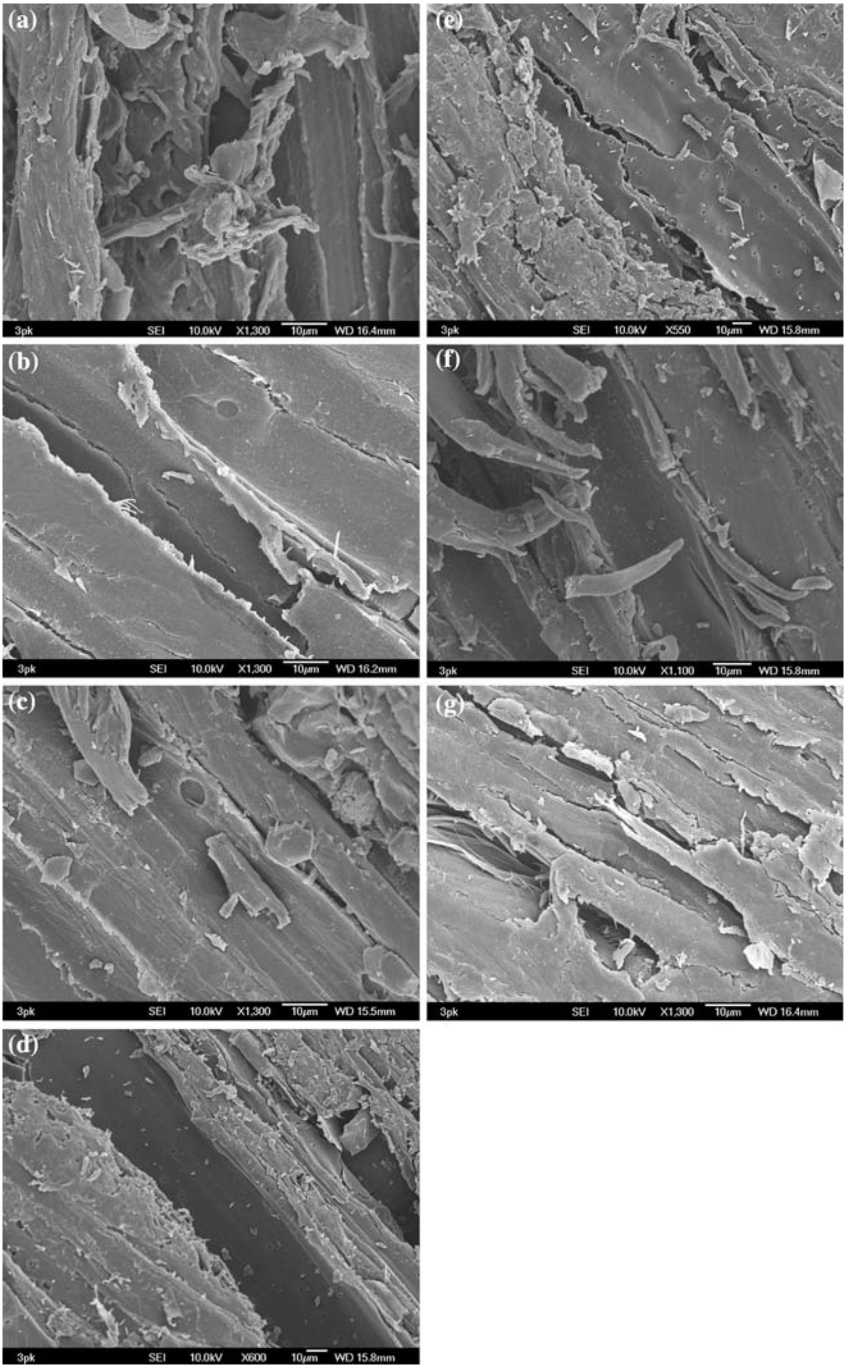

Fig. 6 The SEM photographs of (**a**) unmodified pine wood, (**b**) wood after mercerization, wood after chemical modification with the anhydrides: **c** succinic, **d** propionic, **e** maleic, **f** crotonic, **g** phthalic

β-crystals was observed. They showed that β-crystals grew only after the formation of oriented α-crystals in the sheared iPP melt.

On the basis of literature data and our observations in this work, we conclude that is very probable that molten PP located in the filler (called "the inside polymer") can be shifted together with the filler. In this situation, the shear forces between the filler and "the inside polymer" will not be present (the same velocity of components) and in consequence creation of the α-phase only (termed α-row nuclei by Varga and Kocsis) at the surface will be observed. The efficiency of the α-row structure formation depends on the roughness of the fillers controlled by the chemical treatment. The increase in the surface roughness is attributed to the crystallization of α-form crystal. In our composites, the surface roughness of wood is much diversified (by chemical treatment), which can lead to changes in the nucleation activity of α-row nuclei and consequently influence the formation of hexagonal phase. Our experiments confirmed that the highest content of the hexagonal form of iPP was found in the composites with unmodified wood (highest surface roughness). In another scenario, if wood filler is after chemical treatment, the nucleation of oriented α-row crystallites is decreased by the smooth surface. Such topography will certainly affect the nucleation activity of the wood surface. The occurrence of transcrystallisation in wood–PP composites is strongly dependent on the type of chemical modification of lignocellulosic materials and was earlier described in [56]. It has been found that PP transcrystalinity forms most easily on the unmodified wood and is more difficult on chemically modified wood.

Also Pompe et al. [57] reported three regions of different thickness of interface fibre/polymer. The region near the surface of the fibre includes the interface between the fibre and the transcrystalline part of PP matrix. The thickness of this region was determined by the roughness of the fibre.

On the grounds of our observations and literature data, we put forward a hypothesis that one of the reasons for the β-phase formation is the difference in mobility of both components during solidification. In other words, the observed polymorphism can be considered as an effect of shearing forces in the neighbourhood of wood surfaces. The differences in amounts of hexagonal form for various modifications of wood suggest that the formation of β-form is connected with the different ability of surface fibre nucleation and with the crystal growth rate.

As a continuation of this study, we plan to investigate the impact of chemical composition of wood on its nucleation activity.

Conclusions

The shear-induced supermolecular structure of isotactic PP developed in a compression moulding (performed with a compression mould constructed according to the new design proposed) and by injection technique was characterised by WAXS and SEM methods. The results permit drawing the following conclusions.

1. As follows from the WAXS studies, the polymorphism of PP matrix developed due to melt-shearing, strongly depends on the pulling temperature as well as the

type of chemical modification of wood. The modification process of wood caused significant decrease in the ability of the iPP hexagonal phase formation. The composites containing wood modified by acid anhydrides were characterized by higher contents of β-phase relative to those with wood subjected to mercerization process.

2. The formation of hexagonal phase is particularly intense at low pulling temperatures. The contribution of β-iPP polymorph depends on the pulling temperatures of wood. With increasing pulling temperature, the amount of β-phase decreases rapidly and reaches a minimum at a characteristic temperature, (termed: the critical temperature).

   The critical temperature of pulling, above which the β-iPP phase is not formed has been established as 180 °C for the composites with unmodified wood and 160 °C for the composites with modified wood.
3. In the samples obtained by the injection technique, the amount of β-form strongly depends on the type of chemical modification of wood. The composites containing wood modified by acid anhydrides or by alkali were characterized by lower contents of β-form (about 15–20%) relative to those in the systems without any modifications (25–30%). Moreover, the PP matrix was characterized by distinctly the lowest amount of hexagonal phase (below 10%).
4. Results of WAXS investigation provide important information on the influence of mould temperature in the injection method on the structure of composite materials. According to our results, with increasing rate of cooling (at low mould temperature), the content of β-form in the composites increases.
5. As follows from the SEM studies, the wood surface texture affects its activity. In the unmodified wood, there is a lot of pores and sharp edges that can all act as active places for the nucleation of the semicrystalline matrix crystallization, in contrast to the texture of the surface of chemically modified wood. The efficiency of α-row structure formation depends the degree of roughness of fillers controlled by chemical treatment. In our composites, the surface roughness of wood is much diversified as a result of chemical treatment and these differences can lead to changes in the nucleation activity of α-row nuclei and consequently influence the formation of hexagonal phase.

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