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Polypropylene/cycloolefin copolymer blends: effects of fibrous phase structure on tensile mechanical properties

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Abstract

Tensile mechanical properties of polypropylene (PP)/cycloolefin copolymer (COC) blends were studied using an Instron tensile tester. As COC was expected to impart enhanced mechanical properties to the blends, their modulus, yield strength, tensile strength and tensile energy to break were measured as functions of blend composition. With regard to the reported sensitivity of the COC structure to thermal history, the influence of annealing at two different temperatures was also tested. The attention was primarily concentrated on blends with the volume fraction of COC in the interval $0 < v_2 < 0.40$, where COC formed (short) fibres almost uniaxially oriented in the direction of injection moulding. In the interval $0.40 < v_2 < 0.75$, the blends consisted of partially co-continuous components. Two different models were applied in the analysis of mechanical properties, namely (i) the rule of mixtures for fibre composites and (ii) the equivalent box model for isotropic blends (employing the data on the phase continuity of components obtained from modified equations of the percolation theory). Experimental data on the studied mechanical properties were better fitted by the models for fibre composites. Annealing of the samples (75 °C for 45 days; 120 °C for 3 h) did not markedly affect the tensile modulus, yield stress, and stress at break of the blends. On the other hand, the strain at break was markedly reduced by the annealing up to $v_2 = 0.2$; COC and the blend with 75% of COC ruptured in a brittle manner without yielding.

Keywords: Polymer blends; Polypropylene; Cycloolefin copolymer

1. Introduction

Preparation of polymer blends is one of the most cost-effective ways for the upgrading of existing polymers [1–6]. As potential applications of various polymeric materials are codetermined by their mechanical properties, it is desirable to know the relationships between the morphology (phase structure) and physical properties of intended blends. As generally known, polypropylene (PP) shows relatively low modulus, yield strength and resistance to creeping. Thus, search for 'reinforcing' polymeric components, which could be easily blended with PP, is still an interesting problem to be solved. Preparation of blends without compatibilisers may be difficult because the compatibility of PP with other polymers is limited [2,5]. Recently, amorphous ethylene–norbornene copolymers obtained with metallocene-based catalysts [7,8] have been marketed [9]. They rank among new polymer materials with remarkable properties, such as a high glass transition temperature (T_g), transparency, heat resistance, chemical resistance to common solvents, low moisture uptake, high water barrier, good mechanical properties, etc. Available products—usually denoted as cycloolefin copolymers (COC)—have recently attracted much attention in the field of basic and applied material science [9–16]. Studies of mechanical properties of ethylene–norbornene copolymers encompass dynamic mechanical thermal analysis (DMTA) [13,15], stress–strain measurements [9,13] flexural creep [9], micro-hardness [13], impact strength [10], etc. Rising percentage of norbornene accounts for increase in the yield or tensile strength and decrease in the strain at yielding and break [9, 12]; the phenomenon of yielding is preserved up to about 40% of norbornene in copolymers. Interestingly enough, increase in T_g caused by annealing was attributed [12] to the growth of rigid amorphous phase due to the short-range ordering of norbornene chain segments.

Because of its olefinic character, COC is expected [17, 18] to be compatible with polypropylene and other polyolefins; for this reason, we have attempted to prepare [16] the PP/COC blends without special compatibilisers. Of

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available COC products of Ticona [9], we have used Topas 8007, i.e. the copolymer with the lowest fraction of norbornene (about 30%), which displays yielding and relatively high strain at break (10%). Our intention was to prepare blends with co-continuous 'upgrading' component COC, because numerous studies [19–28] have shown that a co-continuous components affects physical properties of blends much more than a dispersed (discontinuous) component. In the case of mechanical properties, a co-continuous 'reinforcing' polymer is expected to account for a higher modulus, yield strength, resistance to creep, etc. The effects of a reinforcing component are even higher, if it is present in the form of fibres. Thus, a number of attempts have been made to prepare blends with fibre-like [29–32] or fibrillar [32–35] reinforcing component. However, for this end liquid crystalline polymers (LCP) were mostly employed [29–31]. Moreover, blend extrudates usually underwent extensive tensile deformation (drawing) as the last operation in the processing cycle.

In our previous paper [16], phase morphology of PP/COC blends was studied by means of the scanning electron microscopy (SEM) and scanning transmission electron microscopy (STEM). Looking for the critical volume fraction of COC, at which this component assumes (partial) continuity, our interest was mainly focused on the composition interval up to 50% COC. Surprisingly enough, studied blends were found to have fibrous morphology. In the 90/10, 80/20 and 70/30 blends, the PP matrix contained fibres of COC, whose average diameter increased with the COC fraction in the range 0.25–0.80 μm. In the 60/40 blend, the COC component formed both fibres (average diameter 2.6 μm) and larger elongated entities in the PP matrix. The 50/50 blend consisted of co-continuous COC and PP components, while in the 25/75 blend, PP fibres were embedded in COC matrix. In all blends, the fibres were almost uniaxially oriented in the injection direction. Many COC fibres were partly pulled out from the PP matrix on the fracture surfaces perpendicular to the direction of injection. Some COC fibres were broken at the level of fractured surface, which evidences a noticeable interfacial adhesion between PP and COC. The microphotographs thus indicated that COC fibres were long enough to be broken instead of pulled out from the PP matrix at existing interface adhesion. According to the available literature, spontaneously formed and stable fibrous morphology of polymer blends is rather exceptional. COC fibres were not a product of additional drawing, but they were formed during mixing and/or injection moulding (the latter process brought about the uniaxial orientation of the COC fibres). An average fibre aspect ratio was estimated to be at least 20.

As COC forms short fibres in our PP/COC blends [16, 36], we consider measurement and analysis of the tensile mechanical properties of 'fibrous' blends a new and interesting topic. Such blends are expected to have properties different from those of the 'standard' isotropic heterogeneous blends. The objective of this paper is (i) to estimate the effects of the fibrous phase structure on tensile properties, (ii) to compare these properties, wherever possible, with the prediction of existing models, and (iii) to check the influence of annealing on the tensile mechanical properties of the prepared blends.

2. Models used

2.1. Equivalent box model for heterogeneous isotropic materials

Standard polymer blends are heterogeneous isotropic materials with the three-dimensional continuity of one or more components so that simple parallel or series models or the models for orthotropic or quasi-isotropic materials are not applicable. In our previous papers [19–21,27,28,37,38] we have proposed and verified a predictive format based on the combination of the equivalent box model (EBM) and the concept of phase continuity. The EBM in Fig. 1 operates with partly parallel (subscript p) and partly serial (subscript s) couplings of components. This EBM is a two-parameter model as of four volume fractions v_{ij} only two are independent; its volume fractions are interrelated as follows:

$$v_1 = v_{1p} + v_{1s}; \qquad v_2 = v_{2p} + v_{2s}; \qquad v_1 + v_2 = v_p + v_s = 1 \tag{1}$$

The blocks in the EBM are presumed to have physical properties of the neat components; the EBM is likely to fail if the mixing process produces a significant change in the structure and properties of a constituent. As the EBM is not

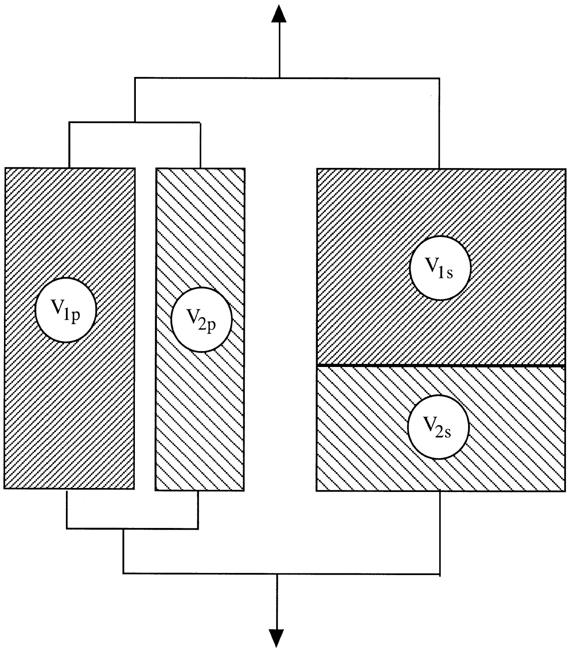

Fig. 1. Equivalent box model for a binary blend (schematically).

a self-consistent model, the predictive format requires two steps: (1) to derive the equations for the properties under consideration; (2) to calculate v_{ij} by using another appropriate model, e.g. modified equations [19–21,27,28,37,38] rendered by the percolation theory [39–41]. An essential feature of the proposed scheme is that all simultaneously predicted properties of a blend are related to a certain phase structure through an identical set of input parameters.

Tensile moduli of the parallel and series branches of the EBM [19–21,28] are the following: $E_p = (E_1 v_{1p} + E_2 v_{2p})/v_p$; $E_s = v_s/[(v_{1s}/E_1) + (v_{2s}/E_2)]$. The resulting tensile modulus of two-component systems is then given as the sum $(E_p v_p + E_s v_s)$:

$$E_b = E_1 v_{1p} + E_2 v_{2p} + v_s^2/[(v_{1s}/E_1) + (v_{2s}/E_2)] \tag{2}$$

A linear stress–strain relationship indispensable for the modulus measurements can be granted for glassy and/or crystalline polymers only at very low strains, typically below 1%, where virtually all blends show interfacial adhesion sufficient for the transmission of acting stress. At higher strains (usually 3–6%), the applied tensile stress exceeds the linearity limit and attains the value of yield strength, thus inducing yielding and plastic deformation of constituents. In our previous papers [19–21,28], we have derived the following equation for S_{yb} of the EBM visualised in Fig. 1:

$$S_{yb} = S_{y1} v_{1p} + S_{y2} v_{2p} + A S_{y1} v_s \tag{3}$$

where $S_{y1} < S_{y2}$ characterise the parent polymers and A the extent of interfacial debonding. Two limiting values of S_{yb}, identified with the lower or upper bound, can be distinguished by means of Eq. (3): (i) interfacial adhesion is so weak that a complete debonding occurs between the fractions of constituents coupled in series before yielding is initiated ($A = 0$ at the yield stress); (ii) interfacial adhesion is strong enough to transmit the acting stress between constituents so that no debonding appears in the course of yielding ($A = 1$). However, if two components differing in the yield strength are coupled in series, the series branch (Fig. 1) yields at S_{y1} or S_{y2}, whichever is lower. Experimental experience [28,42,43] shows that formally analogous equations can be used for evaluation of the yield as well as tensile strength of particulate systems; thus, we will tentatively apply Eq. (3) for S_{ub} by replacing S_{y1} and S_{y2} by the tensile strengths S_{u1} and S_{u2}, respectively.

Employing a universal formula provided by the percolation theory [39] for the elastic modulus of binary systems, we can calculate [19–21,28] v_{ij} by using the following equations:

$$v_{1p} = [(v_1 - v_{1cr})/(1 - v_{1cr})]^q \tag{4a}$$

$$v_{2p} = [(v_2 - v_{2cr})/(1 - v_{2cr})]^q \tag{4b}$$

where v_{1cr} or v_{2cr} is the critical volume fraction (the percolation threshold) at which the component 1 or 2 becomes partially continuous and q is the critical exponent; the remaining v_{1s} and v_{2s} are evaluated by using Eq. (1). In the marginal zone $0 < v_1 < v_{1cr}$ (or $0 < v_2 < v_{2cr}$), where only component 2 (or 1) is continuous, simplified relations can be used for the minority component, i.e. $v_{1p} = 0$, $v_{1s} = v_1$ (or $v_{2p} = 0$, $v_{2s} = v_2$), to obtain an approximate prediction of mechanical properties. Most ascertained values of q are located in an interval of 1.6–2.0 so that $q = 1.8$ can be used also as an average value [39–41]. For three-dimensional cubic lattice, the percolation threshold $v_{cr} = 0.156$ was calculated [40,41]. In general, the patterns predicted by using 'universal' values $v_{1cr} = v_{2cr} = 0.156$ and $q_1 = q_2 = 1.8$ should be viewed as a first approximation that may not be in a good accord with experimental data because real v_{1cr} and v_{2cr} of polymer blends frequently differ from 0.156 and from each other.

2.2. Models for fibrous structures

As we have reported in our previous paper [16], prepared PP/COC blends with weight fractions of COC up to 0.4 have a structure resembling short fibre composites. Thus in this interval the models for (short) fibre composites are to be applied. Longitudinal tensile modulus of aligned short fibres is routinely calculated by means of the Halpin–Tsai equation [42,44–46] that is usually expressed in the following form:

$$E_b = E_1(1 + ABv_2)/(1 - Bv_2) \tag{5a}$$

where $A = 2L/d$ is given by the ratio of the fibre length L and the fibre diameter d, while $B = [(E_2/E_1) - 1]/[(E_2/E_1) + A]$. For very high aspect ratios, Eq. (5a) is reduced [45] to the rule of mixtures

$$E_b = E_1 v_1 + E_2 v_2 \tag{5b}$$

Similarly enough, the tensile strength S_{ub} of a composite with uniaxially oriented short fibres reads [45]

$$S_{ub} = S_1' v_1 + S_{u2} v_2 [1 - (L_{cr}/2L)] \tag{6a}$$

where S_{u2} is the tensile strength of fibres, S_1' is the stress attained in the matrix when the strain-at-break of fibres is reached, and L_{cr} is the critical length of short fibres. Obviously, if $L \gg L_{cr}$, Eq. (6a) can be simplified as follows (rule of mixture):

$$S_{ub} = S_1' v_1 + S_{u2} v_2 \tag{6b}$$

The minimum volume fraction that ensures fibre-controlled composite failure is usually indicated as $v_{2\ \min}$ [44,45]:

$$v_{2\ \min} = (S_{1u} - S_1')/[(1 - L_{cr}/L)S_{2u} + S_{1u} - S_1'] \tag{7a}$$

where S_{1u} is the tensile strength of the matrix. For this volume concentration of short fibres, S_{ub} passes through a minimum.

For $L \gg L_{cr}$, Eq. (7a) is reduced to the form valid for the composites with continuous fibres:

$$v_{2\ \min} = (S_{1u} - S_1')/(S_{2u} + S_{1u} - S_1') \tag{7b}$$

As long as $v_2 < v_{2\,\mathrm{min}}$, the composite will not fracture when all fibres break because the remaining matrix cross-section can still support the load. As a consequence the composite strength will be lower than the matrix strength and given by:

$$S_{\mathrm{ub}} = S_{1\mathrm{u}}(1 - v_2) = S_{1\mathrm{u}}v_1 \qquad (7c)$$

For $v_2 > v_{2\,min}$, the composite strength can be predicted by using Eq. (6b).

3. Experimental

3.1. Materials

Polypropylene Moplen C30G was the product of Basell, Ferrara, Italy: melt flow index (230 °C, 2.16 kg) = 5.7 g/10 min; density: 0.903 g/cm^3; crystallinity: 52%; $T_g = -10$ °C. Amorphous cycloolefin copolymer produced under the trade name Topas 8007 was the product of Ticona-Celanese, Frankfurt, Germany, consisting of 30% of norbornene and 70% of ethylene: MFI (190 °C, 2.16 kg) = 1.7 g/10 min; density: 1.020 g/cm^3; $T_g = 75$ °C.

3.2. Blend preparation

A series of the PP/COC blends was prepared with 5, 10, 15, 20, 25, 30, 40, 50, 75 weight% of COC. Polymers were mixed in a Banbury mixer (chamber 4.3 l; 164 rpm) at 190 °C for 3.5 min. Produced pellets were used for feeding a Negri–Bossi injection moulding machine to produce test specimens for the measurements of mechanical properties. Two types of the test pieces were prepared: (1) ASTM D638 (length: 210 mm; thickness: 3.3 mm; gauge length: 80 mm; gauge width: 12.8 mm; barrel temperature: 242 °C; injection pressure: 20 MPa); (2) ISO 527 (170; 4; 80; 10 mm; 215 °C; 30 MPa). Specimens used for the testing of mechanical properties were stored for more than 6 months at room temperature to avoid any interfering effect of the physical ageing during measurements.

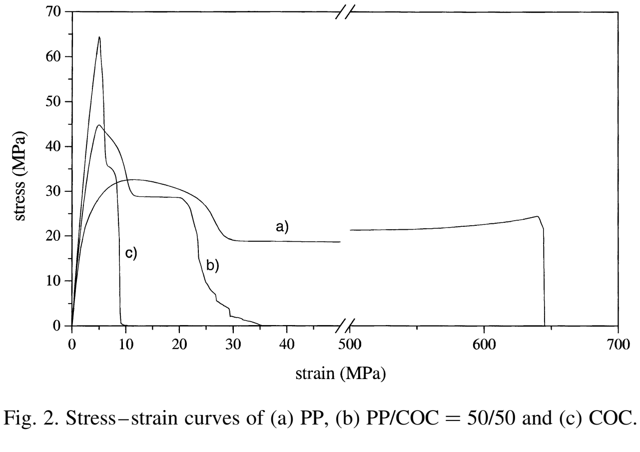

Fig. 2. Stress–strain curves of (a) PP, (b) PP/COC = 50/50 and (c) COC.

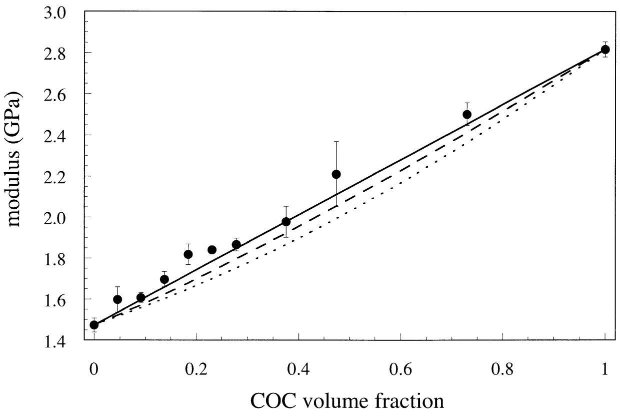

Fig. 3. Tensile modulus as a function of the COC fraction in blends. Full line: the rule of mixtures (5b); dashed line: the EBM (2) for $v_{1\mathrm{cr}} = 0.16$, $v_{2\mathrm{cr}} = 0.103$ and $q = 1.2$; dotted line: $q = 1.8$.

3.3. Stress–strain measurements

Instron tensile tester, model 4502, was used to measure tensile mechanical properties of studied blends. Tensile modulus was determined by using a strain gauge extensometer (Instron, model 2620; gauge length: 25 mm) on ASTM dog-bone shape specimens tested up to 1% strain at the cross-head speed of 1 mm/min (three specimens were tested for each blend). Tensile yield strength and strain, stress and strain at break and tensile energy to break were also ascertained on the ASTM test pieces tested up to the fracture at the cross-head speed of 40 mm/min, i.e. at the strain rate of 50%/min (six specimens were measured for each blend). All test were carried out at about 25 °C.

3.4. Annealing of PP/COC blends

Two different types of thermal treatment were used: (1) storage at 75 °C (i.e. at about T_g of COC) for 45 days followed by cooling at a rate of 2 °C/h; (2) storage at 120 °C (i.e. about 45 °C above T_g of COC) for 3 h followed by a fast cooling to room temperature (at a cooling rate of about

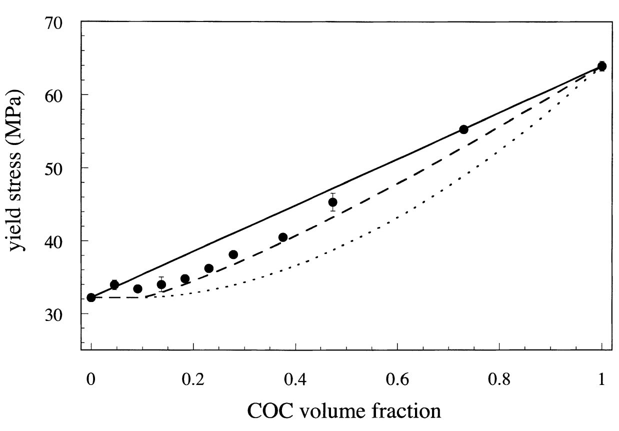

Fig. 4. Yield stress as a function of the COC fraction in blends. Full line: the rule of mixtures; dashed line: the EBM (3) for $A = 1$, $v_{1\mathrm{cr}} = 0.16$, $v_{2\mathrm{cr}} = 0.103$ and $q = 1.2$; dotted line: $q = 1.8$.

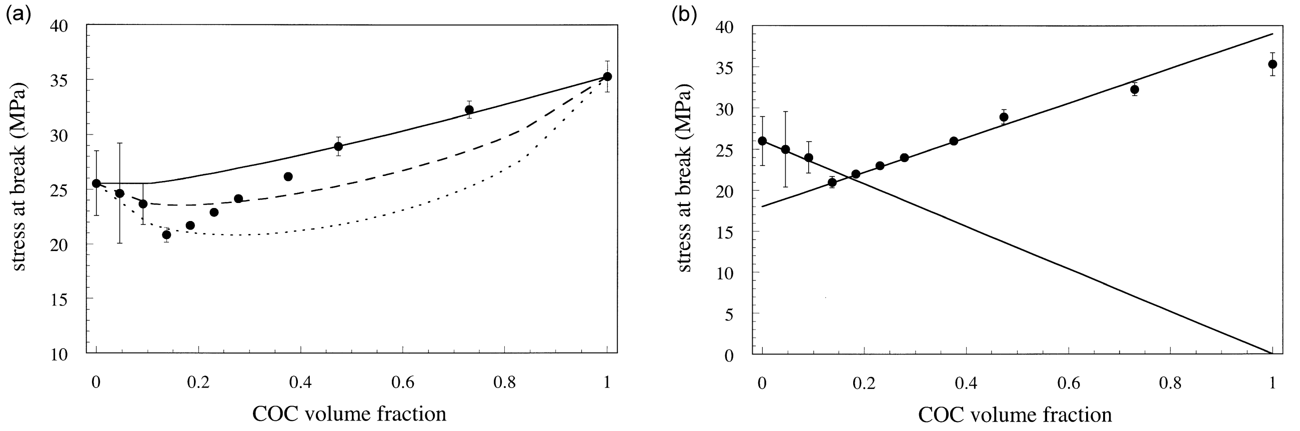

Fig. 5. Stress at break as a function of the COC fraction in blends. (a) The EBM (3) for $v_{1cr} = 0.16$, $v_{2cr} = 0.103$ and $q = 1.2$: full line: $A = 1$; dashed line: $A = 0.5$; dotted line: $A = 0$. (b) Full lines: the rule of mixtures (7)

2 °C/s). The treatment (1) was intended to imitate accelerated physical ageing in order to obtain preliminary information on the material properties after a long storage of blends at ambient temperatures. The second type of treatment was conducted to verify the resistance of blends to short-term action of elevated temperatures (temperature of annealing 120 °C was still much lower than the melting temperature of PP). It is important to note that the shape and dimensions of the test specimens were not changed by any of these treatments.

4. Results and discussion

Typical stress–strain curves found for PP, COC and their blend 50/50 are given in Fig. 2. PP has modulus $E_1 = 1.47 \pm 0.03$ GPa, yield stress $S_{y1} = 32.2 \pm 0.3$ MPa, yield strain $e_{y1} = 11.5 \pm 0.2\%$, stress-at-break $S_{u1} = 25.5 \pm 2.9$ MPa and strain-at-break $e_{u1} = 684 \pm 87\%$. On the other hand, COC as a 'reinforcing' component shows $E_2 = 2.82 \pm 0.04$ GPa, $S_{y2} = 63.9 \pm 0.7$ MPa $e_{y2} = 4.8 \pm 0.3\%$, $S_{u2} = 35.3 \pm 1.4$ MPa and $e_{u2} = 10 \pm 2\%$. The 50/50 blend is characterised by $E_b = 2.21 \pm 0.16$ GPa, $S_{yb} = 45.3 \pm 1.2$ MPa, $e_{yb} = 4.8 \pm 0.2\%$, $S_{ub} = 28.9 \pm 0.9$ MPa and $e_{ub} = 27 \pm 6\%$, which clearly evidences the contribution of the COC component to the tensile properties of the resulting blend.

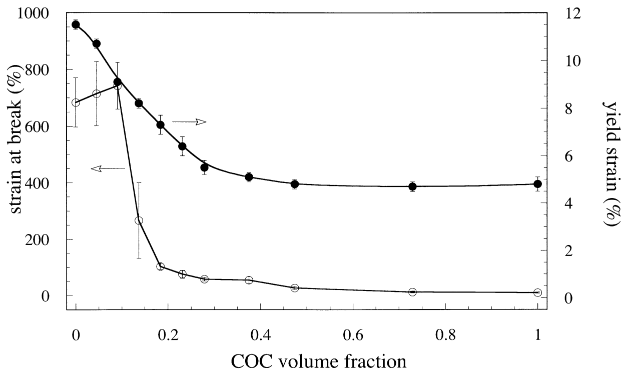

Fig. 6. Yield strain (full points) and strain at break (empty points) as functions of the COC fraction in blends.

Tensile modulus E_b as a function of blend composition (Fig. 3) can plausibly be fitted by the rule of mixtures (5b) valid for the composites with uniaxially oriented continuous fibres, while the prediction (2) of the EBM is lower. Thus the COC fibres spontaneously created in blends (for $v_2 < 0.5$) seem to be long enough to impart to the blends enhanced stiffness similar to that of long-fibre composites. The co-continuous structures constituted at $0.5 \leq v_2 \leq 0.75$ have the moduli also very close to those predicted by the rule of mixtures, which can be explained by the observation [16] that, depending on the composition, the co-continuous PP component contains COC fibres, while the co-continuous COC component contains PP fibres. It is to be noted that a relatively small difference between the tensile moduli of components accounts for the fact that the modulus E_b predicted by the EBM is only slightly lower than that predicted by the rule of mixtures.

Yield stress S_{yb} predicted by Eq. (3) for the EBM is expected to start increasing at $v_2 \geq v_{2cr}$, when the second components with $S_{y2} > S_{y1}$ becomes co-continuous. The experimental data in Fig. 4 are fitted by Eq. (3) quite

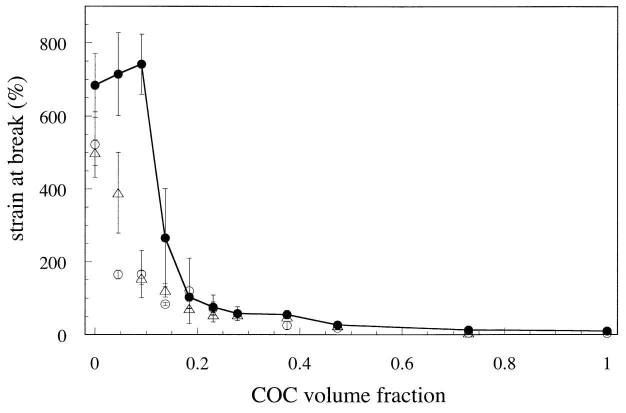

Fig. 7. Strain at break as a function of the COC fraction in aged blends. Full points: as-received specimens; empty points: annealing at 75 °C for 45 days, empty triangles: annealing at 120 °C for 3 h.

well if $v_{2cr} = 0.1$ and $q = 1.2$ are chosen. A relatively low value of q is related to the uniaxial orientation of fibres in blends which are no longer isotropic, but rather orthotropic. Monotonic increase in S_{yb} with v_2 is possible only when $A = 1$; for $A < 1$, a minimum on the S_{yb} vs. v_2 dependence is to occur [19]. Thus Fig. 4 evidences sufficient interfacial adhesion between PP and COC at yielding. The theory of (short) fibre composites does not consider yielding of reinforcing fibres so that no adequate equation is available for such a case. As can be seen in Fig. 4, the rule of mixtures overestimates the values of S_{yb}.

Tensile strength S_{ub} of the PP/COC blends (Fig. 5a) passes through a minimum at $v_2 = 0.136$ ($w_2 = 0.15$), which cannot be fitted by the EBM (Eq. (3)). However, if only the blends with the fibres of COC are considered (Fig. 5b), i.e. $v_2 < 0.5$, the experimental data obey quite well Eqs. (6b) and (7c) for fibre composites. Unfortunately, a quantitative analysis of the data is difficult because the values of S_{2u} and e_{2u} for the COC fibres spontaneously formed in the course of blend preparation are not known. An extrapolation of S_{2u} in Fig. 5b leads to a value of about 39 MPa, which is higher than the value of 35.3 MPa found for COC in bulk. This result is reasonable considering the fibrous structure of COC in the blends up to $v_2 = 0.5$.

Yield strain of the blends markedly decreases with the fraction of COC (Fig. 6) in the region $v_2 < 0.3$ and remains virtually constant for $v_2 > 0.4$, where it is equal to the value for neat COC. On the other hand, strain at break remains very high (almost equal to that of PP) for $v_2 = 0.1$; afterward, it drops rapidly to the value characterising COC. Thus, both yield strain and strain at break concurrently reflect the changes in the phase structure of the blends.

4.1. Effects of annealing on tensile properties of PP/COC blends

Table 1 summarises the values of the tensile modulus, yield stress and stress at break obtained from the tensile tests conducted on the samples annealed at 75 °C for 45 days and at 120 °C for 3 h. The selected annealing conditions do not have a significant systematic effect on these mechanical properties (for comparison see Figs. 3–6). Moreover it is interesting to observe that the trend of the stress at break as a function of the COC content still passes through a minimum, which suggests that the annealing treatments do not modify the fibrous structure of COC in the PP matrix. On the other hand, the strain at break was noticeably reduced for the blends in the region $v_2 < 0.2$, as evidenced by Fig. 7. Embrittlement of the annealed blends can be viewed as a result of the reduction of molecular mobility, which is in conformity with previous results [12,47]. It is worth noting that the annealing at 65 °C for 30 days (imitating an ageing treatment) performed with some selected compositions of the PP/COC blends has not induced any appreciable variation of the mechanical properties, including the strain at break.

5. Conclusions

PP and COC were found to be compatible polymers forming—under fortuitously selected conditions of mixing—blends with fibrous structures. Although no compatibiliser was used, the adhesion between components was fairly good. Tensile mechanical properties of the PP/COC blends were found to be markedly affected by the blend composition: increasing fraction of ‘reinforcing’ COC component in the blends accounted for an increase in the modulus, yield strength and tensile strength, while the yield strain, strain at break and tensile energy to break showed a rather intensive drop.

As in the composition interval $0 < w_2 \le 0.40$ COC formed fibres (almost uniaxially oriented in the direction of injection moulding), while in the interval $0.40 < w_2 \le 0.75$ the blends consisted of partially co-continuous components, two different models were applied in the analysis of mechanical properties: (i) the rule of mixtures for fibre composites; (ii) the equivalent box model (EBM) for isotropic blends where the data on the phase continuity of components were obtained from modified equations of the percolation theory. Tensile modulus as a function of blend composition was plausibly fitted by the rule of mixtures. Monotonic rise of yield strength with the COC fraction was in conformity with the prediction of the EBM under the assumption of good interfacial adhesion. On the other hand, tensile strength passing through a minimum was satisfactorily described by the model for fibre composites.

With regard to the reported sensitivity of COC to thermal history, the influence of annealing at two different temperatures was also tested. Annealing of samples (75 °C for 45 days; 120 °C for 3 h) did not profoundly affect the modulus, yield stress and stress at break of blends. On the other hand, the strain at break was markedly reduced; moreover, COC and the blend with 75% of COC ruptured in a brittle manner. Thus the annealing accounted for some embrittlement of the blends, probably owing to the reduction of the free volume and molecular mobility.

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Table 1
Tensile mechanical properties of various PP/COC blends annealed under different conditions

PP/COC (weight ratio)	Annealing conditions					
	Modulus (MPa)		Yield stress (MPa)		Stress at break (MPa)	
	75 °C/45 days	120 °C/3 h	75 °C/45 days	120 °C/3 h	75 °C/45 days	120 °C/3 h
100/0	1.57 ± 0.05	1.61 ± 0.03	32.2 ± 0.3	33.4 ± 0.1	22.1 ± 0.2	23.1 ± 0.1
95/5	1.62 ± 0.05	1.60 ± 0.09	34.0 ± 0.6	34.4 ± 0.1	20.6 ± 0.4	22.8 ± 2.0
90/10	1.64 ± 0.05	1.73 ± 0.07	33.4 ± 0.6	34.4 ± 0.4	19.2 ± 0.9	19.8 ± 0.1
85/15	1.70 ± 0.05	1.70 ± 0.04	34.0 ± 1.0	35.2 ± 0.1	20.8 ± 0.3	20.8 ± 0.1
80/20	1.77 ± 0.04	1.67 ± 0.01	34.8 ± 0.3	36.3 ± 0.2	22.3 ± 0.2	22.8 ± 0.1
75/25	1.83 ± 0.09	1.77 ± 0.08	36.2 ± 0.1	37.5 ± 0.4	23.4 ± 1.5	24.5 ± 0.1
70/30	1.86 ± 0.03	1.89 ± 0.19	38.1 ± 0.3	38.8 ± 0.1	25.1 ± 0.1	24.5 ± 0.4
60/40	1.94 ± 0.03	1.84 ± 0.02	40.5 ± 0.1	41.4 ± 0.5	26.8 ± 0.7	26.3 ± 0.2
50/50	2.06 ± 0.05	1.96 ± 0.02	45.4 ± 0.1	45.9 ± 0.4	28.9 ± 0.4	28.0 ± 0.2
75/25	2.53 ± 0.01	2.35 ± 0.09	55.3 ± 0.2	n.m.	57.1 ± 0.5	55.0 ± 1.0
0/100	2.60 ± 0.07	n.a.	64.0 ± 0.7	n.a.	61.7 ± 1.0	n.a.

n.m. = not measurable; n.a. = not available (the annealing temperature higher than the glass transition temperature of COC accounted for warping of test specimen.

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