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ORIGINAL PAPER

Effect of structure on light transmission in isotactic polypropylene and random propylene-1-butene copolymers

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Abstract The structure of semi-crystalline isotactic polypropylene and random isotactic copolymers of propylene and 1-butene at the nanometer and micrometer scales is controlled by the pathway of melt-crystallization, and the concentration of chain defects. Rapid cooling of the melt results in formation of mesomorphic nodules, being not organized in a higher order superstructure. Slow melt-crystallization, in contrast, allows formation of lamellae. These lamellae arrange within spherulites of a size which is decreased in copolymers. Analysis of the light transmission reveals a distinctly higher transparency in non-spherulitic preparations. Spherulitic samples exhibit a lower transparency, increasing with decreasing size of spherulites. Annealing of quenched preparations at elevated temperature leads to an increase of the crystallinity and of the dimensions of crystals, without affecting their habit, and their higher order organization. The transparency is only slightly decreased in these specimens. It can be demonstrated that samples of largely different transparency but identical crystallinity can be generated without the use of optical clarifiers/nucleation agents.

Keywords Propylene-1-butene random copolymer · Physical structure · Optical transparency · Atomic force microscopy · Polarizing optical microscopy · UV/VIS spectroscopy

Introduction

The physical structure and therefore the properties of isotactic polypropylene (iPP) can be adjusted in a wide range by the condition of crystallization. Fast cooling/

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quenching of the melt to ambient temperature results in formation of mesomorphic nodules [1–5]. These transform on subsequent heating into monoclinic crystals without changing their habit [6–8]. Slow cooling of the melt, in contrast, results in formation of crystalline lamellae, which on heating may reorganize/stabilize by increasing the ratio between volume and surface area [8–10]. The enthalpy-based crystallinity of both quenched and slowly cooled preparations is about 40–50%, and can be increased by annealing to about 70–75% [11]. Analyses of higher order structures in samples which were melt-crystallized at different rate of cooling revealed absence of spherulites in quenched preparations, and presence of spherulites in slowly cooled, or at low supercooling crystallized specimens [5, 12–14].

The crystallization behavior of iPP is furthermore controlled by the molecular architecture. Random insertion of small amount of 1-alkene comonomers into the iPP chain, in general, is connected with a reduction of the maximum achievable crystallinity, the formation of less stable, defective crystals, and a reduction of the crystallization rate [15–20]. These effects are well-adjustable by the comonomer concentration, and can therefore be used to tailor the mechanical, thermal, or optical properties of the iPP homopolymer. In a recent investigation, we confirmed that the effect of cooling rate on structure formation of random copolymers of propylene with a maximum concentration of about 10 mol-% of either ethylene or 1-butene is qualitatively similar as in case of the iPP homopolymer [20]. Quenching resulted in formation of isometric nodules of a size of 15–20 nm, being almost independent on the comonomer concentration. Slow cooling led to formation of lamellae with a thickness which was decreasing with increasing comonomer concentration. From the literature is known, that the crystalline lamellae are arranged within spherulites, which, however, get increasingly disordered and reduced in size with increasing concentration of co-units [21, 22]. Reports about higher order organization of nodules in quenched but semi-crystalline preparations are not available. Since in particular the structure at the micrometer scale has a major effect on optical properties of semi-crystalline polymers, we attempt with the present study to establish correlations between the pathway of crystallization, the physical structure at the length-scales of both crystals and spherulites, and the light transmission of both isotactic polypropylene and random propylene-1-butene copolymers.

Interaction of visible light with polymer films mainly includes scattering at the air/polymer interface due to surface roughness, and volume scattering at internal heterogeneities. Specular reflection/gloss at the surface, and absorption are negligible. Surface scattering and volume scattering cause haze and loss of transparency, being important characteristics of polymer films which are used, e.g., for packaging [23–25].

Surface roughness of polymer films may be a result of inhomogeneous elastic flow of the melt after the exit of the die in processing/extrusion, or a result of crystallization. Surface roughness/scattering, regardless whether it is caused by elastic-flow instabilities, or by crystallization, has been identified as major source for loss of clarity in case of oriented polymer films [26–30]. Volume scattering, in contrast, seems in this case of minor importance.

In the present work we focus on the analysis of the effect of crystallization on the optical clarity. For this reason, non-oriented samples were prepared by quiescent melt-crystallization, and in absence of flow. Crystallization-induced scattering is therefore expected to be caused by presence of crystals and spherulites only. It has been demonstrated in numerous studies that crystallization-induced haze and loss in see-through clarity in semi-crystalline polymers mainly must be attributed to the presence of spherulites [31–35]. In contrast, only a few studies focused on the evaluation of the effects of the crystallinity and of the crystal size, independent of presence/absence of spherulites, on the light transmission [36, 37]. With the samples available to us, such an independent evaluation of the effect of variation of structure at the various length-scales on the light transmission is possible. In short, we prepared isotropic, non-spherulitic and spherulitic specimen of variable crystallinity and analyzed the light transmission by UV/VIS spectroscopy.

Experimental

Materials

We used an isotactic polypropylene homopolymer from Montell Polyolefins with a mass-average molar mass of 373.000 g mol^{-1}, and a polydispersity of 6.2. Detailed information about the crystallization and melting behaviors, and about the structure is provided elsewhere [5, 7, 8, 11, 20]. Random isotactic propylene-1-butene copolymers (iPP-But) with an average 1-butene content of 6 and 10.9 mol-% were purchased from Sigma Aldrich. The mass-average molar mass and the polydispersity of these copolymers are 225.000 g mol^{-1} and 3.1, respectively. In an initial study, these copolymers were investigated regarding the effect of cooling rate on the structure and morphology of crystals [20], which is now extended to analysis of the optical transmittance as a function of both the chemical structure and the pathway of crystallization, with the latter controlling the physical structure.

Sample preparation

Quenched films with a thickness of 100 μm were prepared by compression-molding in contact with cleaned microscope-cover glasses, using a Perkin-Elmer FTIR film press, and an ice–water mixture as quenching medium. Subsequently, the samples were aged at ambient temperature. Slowly cooled preparations of identical thickness of 100 μm were prepared in a temperature-programmable Collin press, using a rate of cooling of 10 K min^{-1}. The surfaces of the films were in this case in contact with polyester film during preparation. Annealing at elevated temperature was performed for a period of 60 min in a Heraeus vacuum oven. The annealing temperatures were 433, 413, and 403 K for iPP and the copolymers with 6 and 10.9 mol-% 1-butene, respectively. These temperatures were selected due to the close proximity to the melting temperature, as was measured by differential scanning calorimetry (DSC) using a heating rate of 20 K min^{-1}. During annealing, the films were in contact with microscope-cover glasses. Additional samples of lower thickness of only 20 μm were prepared for microscopic studies, following a procedure as described elsewhere [20].

Instrumentation

Wide-angle X-ray scattering (WAXS) measurements were performed to gain information about the structure of crystals in the various samples of different history of crystallization, and chemistry. WAXS data were collected in symmetric transmission mode on an URD 63 diffractometer from Seifert-FPM, using Ni-filtered CuK_α radiation, and a scintillation counter for registration.

Differential scanning calorimetry was applied for estimation of the crystallinity. Heat-flow rate data were recorded either with a DSC 7 from Perkin-Elmer, or a DSC 820 from Mettler-Toledo, with the instruments being calibrated according to standard procedures [38]. The estimation of the crystallinity was based on the measurement of the enthalpy of fusion, recorded on heating at 20 K min^{-1}.

Atomic force microscopy (AFM) was applied for evaluation of the crystal morphology at the nanometer scale. We used a Quesant Universal scanning probe microscope with a 5 μm × 5 μm scanner, and aluminum coated silicon tips NSC14 with a force constant of 5 N m^{-1} and a resonant frequency of 160 kHz (MicroMasch). The instrument software was used to estimate the long period in semi-crystalline preparations containing lamellae.

The spherulitic superstructure at the micrometer scale was analyzed by polarizing optical microscopy using a Leica DMRX microscope, operated in transmission mode. The images were directly obtained on the films of 20 and 100 μm thickness, with the latter used for analysis of the light transmission by UV/VIS spectroscopy.

Light-transmission data were collected as a function of wavelength between 200 and 800 nm on a Perkin-Elmer Lambda 900 UV/VIS/NIR spectrometer. The spectra were normalized to a film thickness of 100 μm using Lambert's law, to account for minor variations of the film thickness [27]. Repeated measurements allowed to estimate an experimental error of the light transmission of ±2%.

Results and discussion

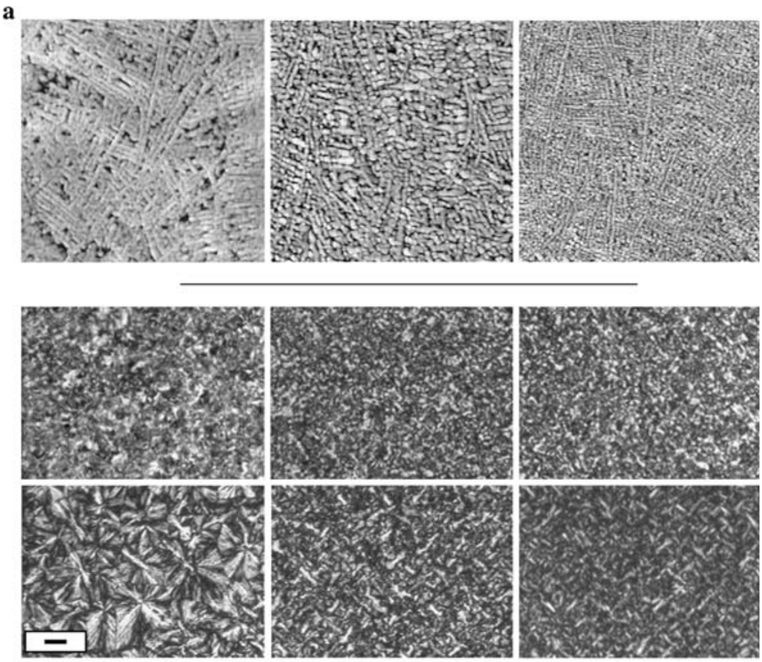
Effect of constitutional chain defects on structure and light transmission in slowly cooled and quenched films

The effect of constitutional chain defects on the crystallization behavior and the light transmission was evaluated on example of random isotactic copolymers of propylene and 1-butene. It has been shown in a preceding study that the addition of 1-butene at concentrations less than about 10 mol-% leads to an only minor reduction of the crystallinity on non-isothermal crystallization at low cooling rate [20]. Monoclinic lamellae are partially replaced by orthorhombic crystals, and the unit cell is expanded due to inclusion of chain defects into the crystals. Fast cooling resulted in formation of mesomorphic nodular domains. These results are confirmed with the present investigation, and are now completed by observation of optical micrographs. Figure 1a shows in the top row AFM phase-mode images, and in the center and bottom rows polarizing optical micrographs of iPP-But copolymers with 0 (left), 6 (center), and 10.9 mol-% (right) 1-butene, being melt-crystallized at slow cooling. The polarizing optical micrographs in center row of Fig. 1a were obtained on films which also were used for the measurement of UV/VIS spectra. The thickness of these films was 100 μm, which, however, did not allow an unambiguous detection of an effect of addition of 1-butene in iPP on the spherulitic superstructure. We assume that differences of the superstructure at the micrometer scale cannot clearly be detected due to superimposed spherulites in the path of light in the microscope. Therefore, additional films with a thickness of only 20 μm, that is, with a thickness less than the size of spherulites were prepared. The structures of these thin films are shown in the bottom row.

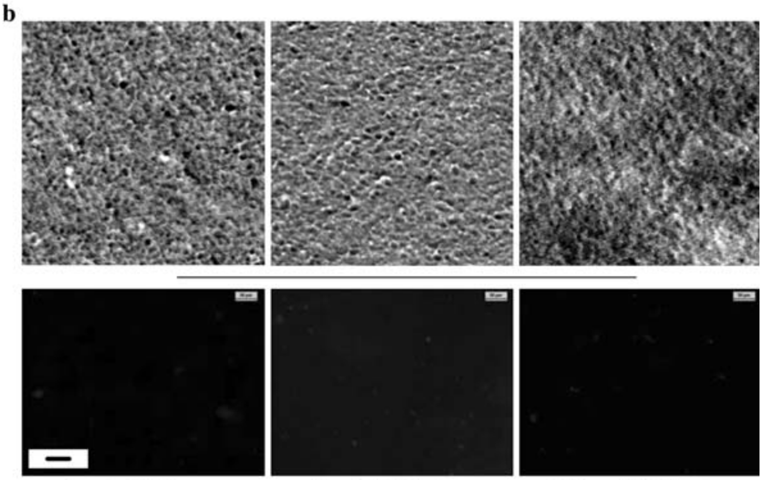
Figure 1b, correspondingly, shows the structure after quenching and subsequent aging at ambient temperature. The AFM micrographs represent an area of $1 \times 1\ \mu m^2$, and the scaling bar, related to the optical micrographs, indicates a length of 50 μm. Since the optical micrographs are featureless, there was no need to investigate films of lower thickness of only 20 μm, as in the case of slowly cooled preparations.

Slow cooling leads to formation of monoclinic/orthorhombic lamellae, and a spherulitic superstructure in the iPP homopolymer and in the copolymers with 6 and 10.9 mol-% 1-butene. Addition of co-units results in a slight decrease of the long period from 20 nm in the iPP homopolymer, to 17 and 15 nm in the copolymers with 6 and 10.9 mol-% 1-butene, respectively. The thickness of lamellae, seemingly, does not change significantly on addition of 1-butene co-units. The size of spherulites, in contrast, decreases from about 50–100 μm in the iPP homopolymer, to perhaps 20–30 μm in the iPP-But copolymers. Additionally, we observed a slight decrease of the crystallinity from 57% in the iPP homopolymer to 46 and 40% in iPP-But copolymers with 6 and 10.9 mol-% 1-butene, respectively. Quenching, in contrast, inhibits formation of lamellae, and of spherulites, regardless of the concentration on 1-butene. Instead, we observed formation of mesomorphic isometric domains/nodules, which are not spatially organized, as is concluded from absence of a characteristic extinction pattern in polarizing optical microscopy. The content of mesomorphic nodules was estimated by calorimetry, and is 50, 34, and 31% for iPP, and iPP-But copolymers with 6 and 10.9 mol-% 1-butene. The calculation of the mesophase content of quenched preparations at room temperature by calorimetry is based on the determination of the enthalpy of melting of monoclinic crystals which were formed on heating the mesophase. Erroneous inclusion of enthalpy of fusion of mesophase on integration the DSC curve, or enthalpy of transition from the mesophase to crystals is negligible since the specific enthalpy of fusion of crystals and mesophase differs by less than 20% [39, 40].

Figure 2 shows the light transmission as a function of the wavelength of iPP and iPP-But random copolymers, prepared by non-isothermal melt-crystallization at largely different rate of cooling. Slowly cooled films exhibit a light transmission between 50 and 70%, with the higher values obtained in the copolymers. Apparently, the increase of the transparency in iPP-But copolymers can only be due to a reduced crystallinity, in combination with a reduced size of spherulites. The



0 mol-% 1-butene 6 mol-% 1-butene 10.9 mol-% 1-butene



0 mol-% 1-butene 6 mol-% 1-butene 10.9 mol-% 1-butene

Fig. 1 **a** AFM phase-mode images (*top row*), and polarizing optical micrographs (*center* and *bottom rows*) of iPP and iPP-But random copolymers. Samples were melt-crystallized at a rate of cooling of <10 K min^{-1}. The concentration of 1-butene is 0 (*left*), 6 (*center*), and 10.9 mol-% (*right*). The AFM images represent an area of 1 × 1 μm^2, and the scaling bar, related to the optical micrographs, indicates a length of 50 μm. The optical micrographs of the *center* and *bottom rows* were obtained on films with a thickness of 100 and 20 μm, respectively. **b** AFM phase-mode images (*top row*), and polarizing optical micrographs (*bottom row*) of iPP and iPP-But random copolymers. Samples were quenched in ice-water, and subsequently aged at ambient temperature. The concentration of 1-butene is 0 (*left*), 6 (*center*), and 10.9 mol-% (*right*). The AFM images represent an area of 1 × 1 μm^2, and the *scaling bar*, related to the optical micrographs, indicates a length of 50 μm

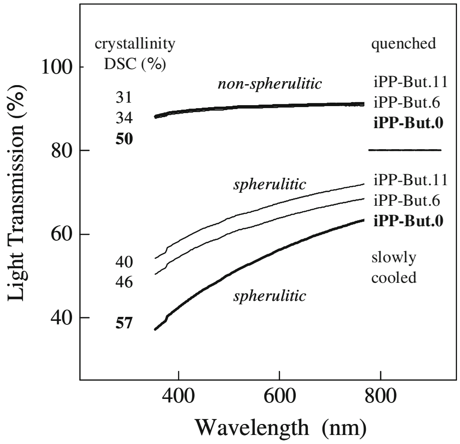


Fig. 2 Light transmission of iPP and iPP-But random copolymers as a function of the wavelength. Samples were non-isothermally melt-crystallized at low rate of cooling (*bottom curves*), or quenched and subsequently aged at ambient temperature (*top curves*). The structure of all preparations is shown in Fig. 1a, b. The figure additionally includes information about the enthalpy-based crystallinity of the different samples

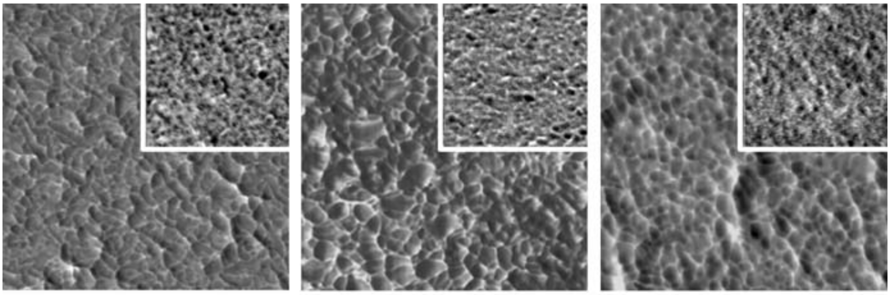
remaining parameters of the structure, that is, the structure, shape and size of crystals, are almost independent on the concentration on 1-butene.

Quenched films show a transparency of 90%, being independent on the concentration on 1-butene. The increase of the transparency, compared to slowly cooled preparations, probably, cannot exclusively be explained by variation of the crystallinity. Slowly cooled films exhibit crystallinities between 40 and almost 60%, decreasing with increasing content on 1-butene co-units. Quenching, in combination with subsequent aging at ambient temperature still leads to crystallinities between 30 and 50%. The increase of the transparency in quenched samples can then only be explained by absence of spherulites and/or changes of the structure and morphology of crystals from non-isometric monoclinic lamellae to isometric mesomorphic nodules. Such a structure, optically, is less heterogeneous than a spherulitic structure. Consequently there is less light scattering and increased percentage of transmitted light in forward direction.

Effect of annealing on structure reorganization and light transmission in initially quenched films

Figure 3 shows AFM images of initially quenched and at ambient temperature aged films of iPP and iPP-But random copolymers, after additional annealing at a temperature close to final melting. The insets show the AFM structure before annealing at a magnification which allows a direct, quantitative comparison with the structure of the annealed films. Annealing of the quenched films leads to an increase of the AFM-size of nodules from initially 15–20 to 30–40 nm after annealing. The habit of the domains/crystals does not change qualitatively. WAXS analyses revealed furthermore that on heating/annealing the internal mesomorphous structure of the domains transferred into a monoclinic/orthorhombic structure.

Annealing is not connected with a change of the higher order organization of crystals. The optical micrographs of the initially quenched films, as were shown with the bottom images in Fig. 1b, are not changed as a result of annealing, that is, formation of spherulites is not observed.



0 mol-% 1-butene 6 mol-% 1-butene 10.9 mol-% 1-butene

Fig. 3 AFM phase-mode images of quenched and subsequently at elevated temperature annealed films of iPP and iPP-But random copolymers, representing an area of 1 × 1 μm^2. The insets show the AFM structure before annealing, and represent an area of 0.5 × 0.5 μm^2

Figure 4 shows the light transmission as a function of the wavelength of quenched films of iPP and random iPP-But copolymers, before (top three curves) and after annealing at elevated temperature (lower three curves). Annealing does not result in a major loss of the optical transparency. The reduction of the light transmission due to annealing is less than 3–5% at wavelengths larger about 500 nm. A larger effect, that is, a decrease of the transparency by about 5–10% is only observed near the UV-region. The bold drawn curves obtained on the quenched and subsequently annealed samples are not labeled regarding the 1-butene concentration since there is not observed a systematic effect, that is, we consider the observed differences being caused by experimental errors. Figure 4 includes

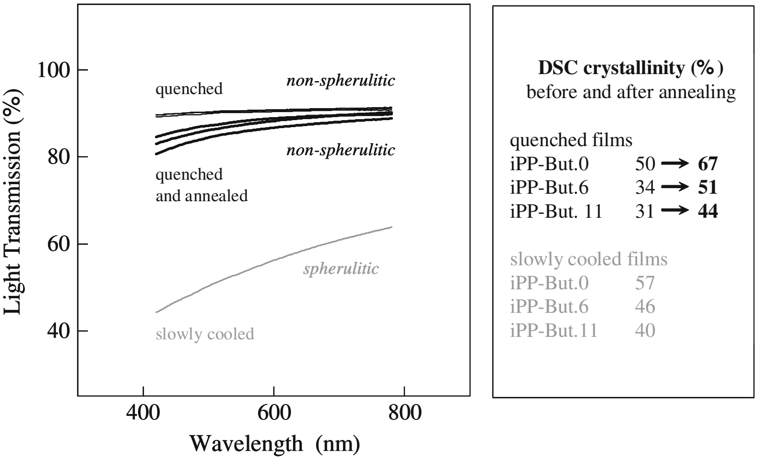


Fig. 4 Light transmission of iPP and iPP-But random copolymers as a function of the wavelength. Samples were quenched and subsequently aged at ambient temperature (*top curves*, *thin lines*), and additionally annealed at elevated temperature (*thick lines*). The structure of all preparations is shown in Figs. 1b (quenched samples) and 3 (quenched and annealed samples). The figure additionally includes light-transmission data of initially slowly cooled iPP for comparison (*gray curve*), and information about the crystallinity of the different samples

additionally crystallinity-data of quenched samples before and after annealing. The crystallinity increases distinctly as a result of annealing, to reach, or even exceed the level of the initially slowly cooled films. In detail, the crystallinity of slowly cooled iPP is 57%. Quenching of the same material led to a crystallinity of 50%, which increased by annealing to 67%. The increase of the crystallinity by annealing, however, is not connected with a major decrease of the transparency. It remains at high level of 80–90%, and is considerably larger than the transparency of slowly cooled, spherulitic iPP of similar crystallinity. The data of this experiment show clearly, that in iPP and random iPP-But copolymers, films of largely different transparency but adjustable crystallinity can be prepared.

We assume that the slight decrease of the transparency of initially quenched samples after annealing is due to an increase of the crystal size, and not primarily due to an increased crystallinity. This is concluded from the comparison of the crystallinity of non-annealed quenched iPP, which is 50%, with that of quenched and annealed iPP-But with 10.9 mol-% 1-butene, which is 44%. If the crystallinity would be the only controlling parameter, then quenched iPP needed to show a lower transparency than quenched and annealed iPP-But with 10.9 mol-% 1-butene, which is not true.

Conclusions

In the present study we investigated the effect of the physical structure of an iPP homopolymer and two specific random copolymers of propylene and 1-butene, at both the length-scales of crystals and of spherulites, on the light transmission. Variation of the cooling rate on melt-crystallization, and random insertion of 1-butene co-units into the iPP chain allowed to prepare semi-crystalline films of largely different superstructure. The light transmission, or see-through clarity,

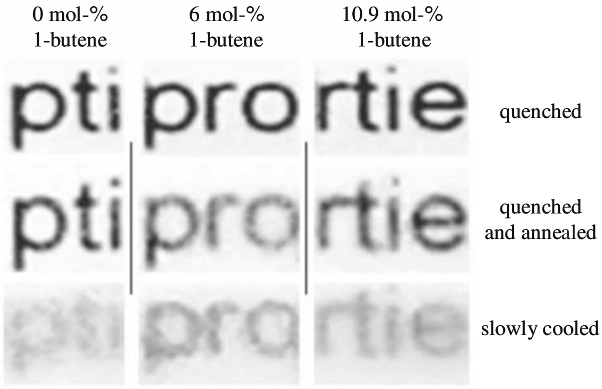


Fig. 5 See-through clarity of films of 100 μm thickness of iPP (*left*) and iPP-But random copolymers with 6 mol-% (*center*) and 10.9 mol-% 1-butene (*right*). The *top row* shows quenched films, the *center row* quenched and subsequently at elevated temperature annealed films, and the *bottom row* slowly cooled films

respectively, can be adjusted in a wide range between about 50 and 90% for films of 100 μm thickness, which is visualized in Fig. 5. The lower limit of transparency of about 50% was obtained in slowly cooled iPP, and increases as a function of 1-butene to about 60–65% (bottom row, from left to right). This effect is attributed to a decrease of the size spherulites in the copolymers. The maximum transparency of about 90% is obtained in non-spherulitic, quenched films, being independent on the concentration of 1-butene (top row). Annealing of the quenched films caused a slight decrease of the transparency which is due to an increase of the size of crystals (center row).

The most striking result of the present study is the observation that by variation of the pathway of crystallization, and by variation of the concentration of constitutional defects any combination of optical transparency and crystallinity can be adjusted. Since the crystallinity is expected to have major impact on mechanical properties, further investigations for evaluation of advantageous combinations of materials properties are needed.

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