Phase Structural Analyses of Polyethylene Coatings on High-Density Papers. III. Determination of the Crystallite Thicknesses by T_1 Measurements

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ABSTRACT: Two different extrusion-coating qualities of polyethylene, namely LDPE and HDPE, were coated on high-density papers. Differences were observed with respect to their response to storage and low temperature heat treatment. HDPE does not respond to storage at ambient temperature and heat treatment in the same way as LDPE. The LDPE-coating exhibits an increase in the monoclinic crystalline fraction at the paper surface as a result of heat treatment. The nature of this response appears to be a result of adhesion to a paper surface, the properties of this surface, orientation of polymer chains, and chain mobility differences. The increase of the monoclinic fraction is shown to relate to an increase of the mean crystallite thickness and initiation of new crystallites at the paper surface. © 2003 Wiley Periodicals, Inc. J Appl Polym Sci 91: 235–241, 2004

Key words: extrusion; polyethylene (PE); morphology

INTRODUCTION

The polyethylene extrusion coating process of high-density paper initiates the formation of a monoclinic crystalline phase by adhesion to the paper surface and orientation of the polymer chains by shear forces and pressure.[1] It has previously been demonstrated from solid-state ^{13} C spectra of bulk high-density polyethylene samples, to which stress impacts were applied, that the fraction of monoclinic crystalline component may increase by several percent.[2]

The surface properties of the paper, with respect to differences in surface tension to the polymer, have been shown to be important in this process, as we have previously described. In the thin polyethylene coating (approx. 30 μm) the main fraction of the monoclinic crystalline phase was shown to be located in an uneven crystalline layer at the paper surface. Experiments showed that the monoclinic crystalline fraction increased in LDPE by a low-temperature heating (55°C) and cooling procedure.[3] This result is not in accordance with the continuous transformation of monoclinic crystallites to orthorhombic crystallites that are reported to begin at approximately 50°C and to be completed at 80°C.[4] In the present work we have measured the mean crystalline lamella thickness prior to and after heat treatment to investigate if this increase in crystalline fraction is mainly a result of thickening of the crystalline lamella or formation of new crystallites.

The measurements of the mean lamellae crystallite thickness of extrusion coatings of polyethylene were performed by high-resolution solid-state ^{13}C NMR spectroscopy by the method of ^{13}C spin-lattice relaxation time (T_1) measurements. From the measured T_1 values the crystallite thickness is estimated.[5–7]

The T_1 values can be related to chain diffusion[8–17] within the crystals. The chain diffusion itself has been assigned to 180° jumps of the chain stems in the crystallites. For instance, Klein et al.[16] investigated whether chain diffusion was the process responsible for the ^{13}C longitudinal relaxation, and the extent to which various structural and morphological features, namely molecular weight and lamellar thickness, affected the diffusion rate. They described the chain diffusion process as follows: “Random thermal fluctuations allow cilia in the amorphous phase, or a section of chain comprising a loose chain fold, to migrate into the lamella, and sections of chain can similarly escape from the lamella into the interfacial/amorphous region. The morphology is considered to be in dynamic equilibrium; the total crystallinity is constant in time

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(at temperatures below that required for annealing) but exchange between crystal and interface is continually occurring." They reported that the rate of chain diffusion seemed independent of molecular weight for samples in which it was varied by more than an order of magnitude. Diffusion was faster for pressure annealed (PA) samples than for samples that were slow cooled (SC). Crystallization by pressure annealing produces a significant increase in the lamellar thickness. The higher diffusion coefficients for the PA samples were attributed to morphological differences in the samples, as it has been shown that the process of pressure annealing reduces the amorphous phase entanglement density. Therefore, there is less constraint to the diffusion in these materials than in the conventional SC samples. This also explains the reduction in the rate of recovery for the SC samples because the presence of the entanglements would lead to a limit in the extent of migration of the crystalline stems.

Commercial extrusion coating grades of low-density polyethylene (LDPE) and high-density polyethylene (HDPE) without any additives have been used in this work. The high-density paper (HDP) is made of highly beaten spruce wood pulp fibers. The fibers are fibrillated and water swelled giving a paper of high density and a smooth surface. There are a variety of production parameters influencing the paper properties. The paper samples used in this work are from commercial production.

EXPERIMENTAL

Materials

High-density papers

The extrusion coating was performed on two samples of high-density papers supplied by Nordic Paper. Both paper samples were produced in a full-scale production plant of 100% highly refined sulphite pulp of Standard brand. The two samples are produced under the same conditions, except that one of them is treated with a steam box prior to calandering to reduce surface roughness. In the production, all the samples were coated with carboxy methylcellulose (CMC) (12–14g/liter water) and sized with alkyl keten dimer water suspension (AKD).

High-density polyethylene

Grade CG8410 from Borealis (M_w = 85,000, M_n = 15,000, MWD = 5.9) was used. CG8410 is here termed as HDPE but has small amounts of butene added as comonomer so the polymer is in fact an ethylene-1-butene copolymer. This is a polyethylene grade especially developed for extrusion coating of paper and board. It has a density of 941 kg/m^3 (that indicates that this polymer is of medium density grade, MDPE, but here this polyethylene grade is denoted as Borealis does) and a melt flow rate at 7.5 g/10 min at 190°C/2.16 kg according to vendor specifications.

Low-density polyethylene

The LDPE used (grade CA7230, Borealis, M_w = 155,000, M_n = 20,000, MWD = 8.0) was developed for extrusion coating. It has a density of 923 kg/m^3 and a melt flow rate of 4.5 g/10 min at 190°C/2.16 kg.

Extrusion coating

The samples were extrusion coated at Borealis' pilot plant in Finland.

Extrusion conditions

HDPE

Coating weight : 30 g/m^2, line speed: 100 m/min, temperature setting: 315°C, chill-roll temp. 18°C, press-roll: 250 and 400 kPa, corona treating: 8.5 kW. LDPE: coating weight: 30 g/m^2, line speed: 100 m/min, temperature setting: 320°C, chill-roll temp. 18°C, press-roll: 250 and 400 kPa, corona treating: 8.5 kW.

NMR measurements

^{13}C spin-lattice relaxation times (T_1) were measured at 50.3 MHz by the crosspolarization magic angle spinning method (CPMAS) using a Bruker Avance DMX 200 NMR instrument at ambient temperature. The samples were prepared by putting a strip (approx. 1.5 × 6 cm) of coated paper, tightly rolled up, into a 2.5-mm zirconia rotor. Spectral parameters used: contact time for the crosspolarization (CP) process: 1 ms, magic angle spinning (MAS) rate: 7 kHz, recycle delay: 6 s, pulsewidth: 3 μs (^{13}C, ^{1}H), proton decoupling: ~20 G, 1024–2048 scans per spectrum, 30 kHz sweep width and 1024 data points and quadrature detection. The inversion recovery experiment for the T_1 determinations were performed with 64 scans per spectrum and 32 delay times, from 10 μs to 500 s.

Line-fitting procedure

The calculations of the mass fractions were done by line fittings. They were performed with NUTS software (2D version, Acorn NMR Inc.) that performs a Simplex fit. The FIDs were four times zero-filled prior to Fourier transform. The ^{13}C NMR signals were decomposed to three or four peaks with 100% Lorentzian line shape. During the line fitting performance of the HDPE-spectra, the chemical shifts for the amorphous and monoclinic crystalline phases were fixed to 31.0

and 34.4 ppm, respectively, relative to the orthorhombic crystalline peak that was set to 33.0 ppm.[1,2,18] In the deconvolution of spectra of ethylene–1-butene copolymer two lines, a narrow and a broad, both of orthorhombic crystallinity, can be fitted in refs.1 and 19. In the present work one orthorhombic line was fitted for improved quantification of the monoclinic crystalline fraction.[2] The program fit/optimized the chemical shift of the interfacial phase. LDPE-spectra analysis was performed similarly as for HDPE, but the fitting of the interfacial PE was neglected for some of the spectra.[2] Only the chemical shift of the monoclinic crystalline lines was fixed.

Heat-treatment of the laminates

The extrusion-coated samples (from the process quench cooled) were heat treated at temperatures under the melting point of the polymer. This was simply done by putting the zirconia rotor with the respective sample into a heat cabinet at 55 or 110°C for 8 min to ensure uniform temperature throughout the samples. The samples were cooled at ambient temperature for about 20 min before NMR measurements.

RESULTS AND DISCUSSION

The samples investigated by T_1 measuremets in the present work have been chosen from a set of extrusion-coated paper samples that have been previously analyzed.[2] Table I gives the results of the actual samples with respect to sizes of morphological phases of the extrusion coating.

When both HDPE coated paper (sample 3) and LDPE coated paper (sample 12A) are heated up to 110°C and cooled, we observed that the respective mass fractions of the orthorhombic crystalline phase were increased while the monoclinic crystalline phase reduced in size. The heat treatments of this HDPE-coated paper at 55°C and (see Table II) did not make any significant changes in the sizes of the different morphological mass fractions. On the other hand, although the heat treatment of a laminate of LDPE (sample 12B) at 55°C did not cause any significant changes of the orthorhombic crystalline fraction, the monoclinic fraction is increased significantly. Here, we should have expected a decline of the monoclinic fraction rather than an increase due to transformation of the monoclinic crystallites to orthorhombic.[20] Here, we observe (Table I) that there are still monoclinic fractions left after heating to 110°C for both LDPE and HDPE (3 and 6%, respectively). If there are residual fractions of monoclinic crystallinity that have not been transformed, this may be due to reduced polymer chain mobility as a result of bonding/interactions (adhesion) with the paper surface.

TABLE I
Results of Solid-State CPMAS ^{13}C NMR Analyses before and after Reheating

				Mass fractions, morphological phases (half-height width in Hz)				Mass fractions, morphological phases after Reheating[a,c] (half-height width in Hz)			
Sample	Paper grade	PE grade	P on press Roll. kPa	Amorph. (31 ppm)	Interph. (31.6 or 31.7 ppm)	Orthorh. cryst. (33.0 ppm)	Monocl. cryst. (34.4 ppm)	Amorph. (31.0 ppm)	Interph. (31.6–31.7 ppm)	Orthorh. cryst. (33.0 ppm)	Monocl. cryst. (34.4 ppm)
3	Standard	HDPE: CG8410	250	0.26 (94)	0.21 (111)	0.47 (47)	0.07 (129)	0.26 (131)	0.13 (108)	0.59 (47)	0.03 (72)
12A	Standard[b]	LDPE: CA7230	250	0.54 (114)	—	0.36 (76)	0.10 (187)	0.42 (140)	—	0.51 (48)	0.06 (118)
12B	Standard[b]	LDPE: CA7230	250	0.53 (114)	—	0.36 (75)	0.11 (171)	0.45 (114)	—	0.36 (71)	0.17 (268)
15	Standard/steamed	LDPE: CA7230	400	0.44 (102)	—	0.36 (74)	0.21 (273)	—	—	—	—

The mass fraction and the half-widths (in parentheses) of the deconvoluted lines are given of the respective morphological phases of polyethylene (LBPE and HDPE) of the coated paper samples. The coating thickness is approximately 30 μm (corresponding to 30 g/m^2 paper).

[a] Cooled at ambient temperature.

[b] The fitting of the interfacial lines were not included, see text.

[c] Sample 3 and 12A: heated to 110°C, sample 12B:heated to 55°C.

TABLE II
Results of Solid-State CPM As ^{13}CNMR Analysis of Polyethylene Coating after Storage at Ambient Temperature Prior to Heat Treatment at 55°C

				Mass fractions, morphological phases (half-height width in Hz) after storing at ambient temperature				Mass fractions, morphological phases after reheating, 55°C (half-height width in Hz)[a]			
Sample	Paper grade	PE grade	P on press roll. kPa	Amorph. (31 ppm)	Interph. (31.3 to 31.7 ppm)	Orthorh. cryst. (33.0 ppm)	Monocl. cryst. (34.4 ppm)	Amorph. (31.0 ppm)	Interph. (31.6–31.7 ppm)	Orthorh. cryst. (33.0 ppm)	Monocl. cryst. (34.4 ppm)
3	Standard	HDPE: CG8410	250	0.28 (108)	0.11 (86)	0.49 (45)	0.11 (162)	0.29 (105)	0.11 (85)	0.50 (44)	0.10 (150)
12A	Standard	LDPE: CA7230	250	0.27 (182)	0.07 (72)	0.41 (69)	0.24 (308)	0.27 (129)	0.06 (73)	0.42 (59)	0.24 (230)
15	Stand./steamed	LDPE: CA7230	400	0.29 (161)	0.11 (71)	0.39 (78)	0.21 (255)	0.22 (100)	0.10 (93)	0.38 (70)	0.30 (266)
15[b]	Stand./steamed	LDPE: CA7230	400	0.42 (125)	—	0.37 (74)	0.21 (260)	0.33 (109)	—	0.38 (71)	0.29 (262)

The mass fractions and the line widths at half-height (in parentheses) are given of the respective morphological phases. The coating thickness is approximately 30 μm (corresponding to 30 g/m^2 paper).

[a] Cooled at ambient temperature.

[b] Sample 15 with a three-component line fit.

By low-temperature treatment (55°C) of a sample coated with LDPE we observe an increase of the monoclinic fraction by approximately 6%. This surprising result was checked by additional experiments. The samples had, in the meantime, been stored at ambient temperature and the appearance of new CPMAS ^{13}C NMR spectra gave immediate evidence of a morphological change of the samples. Due to the spectral changes, deconvolution of the spectra actually gave more reliable results with respect to the fitting of the line representing the interfacial polyethylene phase compared to data of Table I. The line fitting procedure of four lines, if possible, is preferred (Figure 1). Even so, we see in Table II that the three and four component line fittings give approximately the same mass fractions for the crystalline phases.

Table II gives the mass fractions for the different morphological phases after storing at ambient temperature and then after low temperature heating treatment. Compared with Table I, storage of the HDPE coating seems to have resulted in a small increase of the orthorhombic and monoclinic crystalline mass fractions of approximately 2 and 4 %, respectively. A low-temperature heat treatment (55°C) of the sample has not resulted in significant changes of the mass fractions. For the LDPE coating of sample 12 we have a more significant increase of the crystalline fractions as a result of storage at ambient temperature. This extrusion coating exhibits increases in its two crystalline fractions, orthorhombic and monoclinic, of 5 and 14% (total increase of 19%), respectively (compare Table I and Table II). The subsequent low-temperature heat treatment for this LDPE coating (Table II) showed no significant effect on mass fractions, but the crystallites went through a perfection process as we see their line widths reduced in the NMR spectra. Low-temperature heat treatment of the LDPE-coated standard paper exhibited small effects after the sample had been stored at ambient temperature. For sample 15, that was coated and stored exactly in the same way as sample 12, we observe another effect. This sample has a paper that is steam box treated prior to calandering and has a smoother surface and a lower surface tension[2] (more comparable to the surface tension of a polyethylene film). This sample also experienced a higher press roll pressure. We observe in Table I that this has given a relatively high monoclinic fraction (21%) compared to the sample with paper without steam treatment and with lower press roll pressure (11%). Conversely, the orthorhombic fraction has the same size, demonstrating the importance of the paper surface characteristics. It seems that the storage effect is absent for this sample. On the other hand, the subsequent low temperature heat treatment had an effect on the size of the monoclinic crystalline fraction, increasing 9% to about 30%. In Table II, the three-component line fit is added for the comparison with data in Table I to follow up previous discussions.

T_1 measurements

These results gave the opportunity to perform the T_1 measurements to estimate how the crystallite lamella thickness changes for samples stored and heat treated. In general, more order and rigid structures, that is, more perfect crystallites, give longer T_1 values. In less perfect crystallites at room temperature, polymer chains can execute 180° flips with a frequency in the kilohertz domain. In more perfect structures such flips do not occur or occur in much slower time scale.[8,17] T_1 calculated from the slope of the decay curves[5] of their respective signals by setting the slope equal to $-1/T_1$. Thus, short T_1 values give steep decay curves of the respective morphological phases while long values give relative flat decay curves. Mean crystal thickness is then derived from the relationship between the T_1 values and previously derived correlation with crystal thickness.[5] Thicker lamellas give longer T_1 values (due to an approximately linear relationship between these variables). The decay curve representing the crystalline component is often sectioned (if possible) into two or three segments where each segment represents different groups of crystallites with different mean thickness values. From the appearance of the decay curves in the present work, it has been convenient to section the "crystalline segment" of the curve to represent two groups of crystallites. In the range up to about 150 s, the T_1–lamellae thickness correlation curve is very steep and not very well defined. Therefore, the assumed values for lamellae thickness will be approximate in this range, as will be the case for at least one of the components here. Trends however are more clearly defined.

The results of the T_1 measurements are given in Table III and examples of T_1 decay curves are given in Figures 2 and 3. For HDPE (Fig. 2) there are relatively small differences between the decay curves of the crystalline components prior to and after heat treatment (110°C). These differences imply that the mean lamella thickness of the two crystalline components is increased from approximately 80–100 to 120 Å and from 210 to 220 Å for the two components, respectively. The LDPE coating has a larger response on heat treatment (110°C) than HDPE. The crystallites increase significantly in mean thickness. The decay curve from the sample exposed to heat is flatter than the curve of the untreated sample (Fig. 3). The mean thickness increases from about 80–90 to 160 Å for one component and from 90 to 310 Å for the other (110°C) (see Table II).

The mobility of the low-density polyethylene chains seems to be high compared to high-density polyeth-

TABLE III
Results of the T_1 Measurements of the Polyethylene Coatings on High-Density Papers and Their Mean Lamellae Crystalline Thickness Prior to and after Heat Treatment

			T_1 values of morphological phases (seconds)			Cryst. lamellae thickn., L_R (Å)[b]	Reheat. temp. (°C)	T_1 values of morphological phases (seconds) after reheating			Mean cryst. lamellae thickn., L_R (Å)
Sample	Paper grade	PE grade	Amorf.	Interph.	Cryst.[a]			Amorf.	Interph.	Cryst.[a]	
3	Standard	HDPE: CG8410	0.47	2.07	81, 781	80–100, 210	55	0.37	5.10	96, 486	100, 170
3	Standard	HDPE: CG8410					110	0.61	7.89	173, 895	120, 220
12A	Standard	LDPE: CA7230	0.25	2.10	89, 78	90, 80–90	55[c]	0.22	1.74	108, 1478	110, 300
12B	Standard	LDPE: CA7230					110	0.74	10.70	362, 1596	160, 310
15	Standard/steamed[c]	LDPE: CA7230	0.25	3.99	150, 884	90–100, 220	55	0.18	2.23	83, 232	50–100, 130

[a] Two T_1 values measured for crystalline phases for samples 3, 12, and 15.
[b] Low T_1 values for the crystalline phases give uncertain calculations of lamellae thickness.
[c] T_1 measured after storage at ambient temperature.

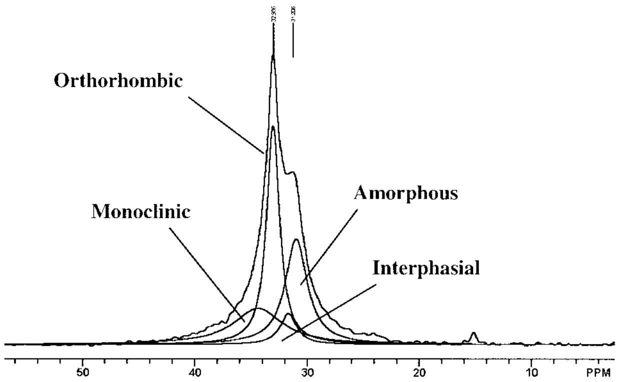

Figure 1 CP MAS ^{13}C NMR spectrum of LDPE extrusion coating on HD paper. The deconvolution lineshapes of respective morphological phases are shown.

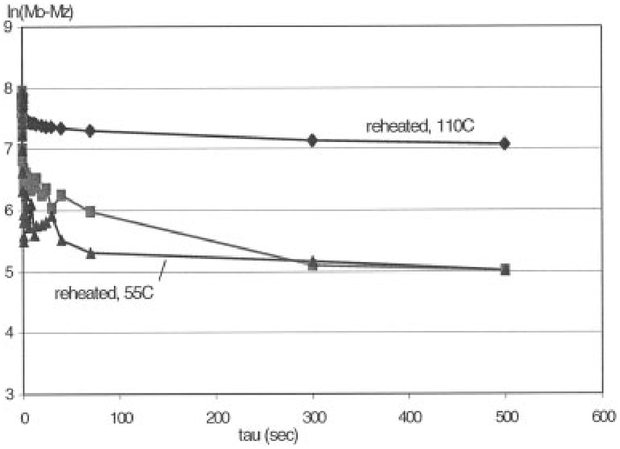

Figure 3 The figure shows the T_1 decay plot of the morphological omponents of LDPE extrusion coating (sample 12) prior to and after heat treatment at 55 and 110°C.

ylene. From Table I (sample 12) we see that even at a relatively low temperature of 55°C the crystalline fraction increases, but this rise in crystallinity is given by the monoclinic fraction. The orthorhombic fraction has about the same size but these crystallites have a somewhat higher degree of perfection as reflected by a smaller spectral half-height width. The T_1 measurement (see Table III) on this sample shows that the mean crystallite thickness of the two groups has increased. The first one is slightly increased; the second shows a mean thickness increase (after storage and heat treatment) from about 90 to 300 Å. Note that the orthorhombic and monoclinic fractions are increased from 36 to 41% and from 10 to 24%, respectively.

The steam box-treated paper sample (sample 15) has, prior to low-temperature heat treatment (and storage), a mean crystallite thickness of 90–100 and 220 Å respectively, for the two defined crystalline components. The orthorhombic and monoclinic mass fractions are about 39 and 21%, respectively. The effect of low-temperature heat treatment is surprising. The mean crystallite thickness decreases to 50–100 Å and 130 Å. The orthorhombic fraction remains about the same size, but the monoclinic fraction is increased 9 to about 30%.

These effects may be explained in the following way. In sample 12 the increase of the monoclinic fraction is associated with building, and increased perfection, of the existing crystallites. The fraction is low after extrusion, but storing at ambient temperature increases the fraction. Sample 15 has a relatively high monoclinic fraction already after extrusion coating with higher pressure at the press roll. Increase of press roll pressure increases the monoclinic fraction if the surface tension of the paper is close to polyethylene.[2] The storage time has no effect, but by low-temperature heat treatment, the sample shows an even higher potential for increase of the monoclinic fraction. The paper surface is more optimal with respect to surface tension, smoothness, and more uniform contact with the polymer phase, and the increase of monoclinic crystallinity is a result of formation of new crystallites. The mean crystallite thickness is thus decreased. This phenomenon is likely to be a result of adhesion to a paper surface, orientation of polymer chains during extrusion coating, and the inherent molecular/chain mobility. The system is more optimal after extrusion coating and storage has, consequently, less effect. But when more energy is given to the system as heat, the molecular mobility[8] allows formation of crystallites of already oriented molecules at the paper surface as a transcrystalline layer.[22] The nucleation might be enhanced by the porous and "hairy" ultrastructure of the highly refined sulphite cellulose fibers, supposed to have a relative high content of hemicelluloses, of the papers coated.[23–25]

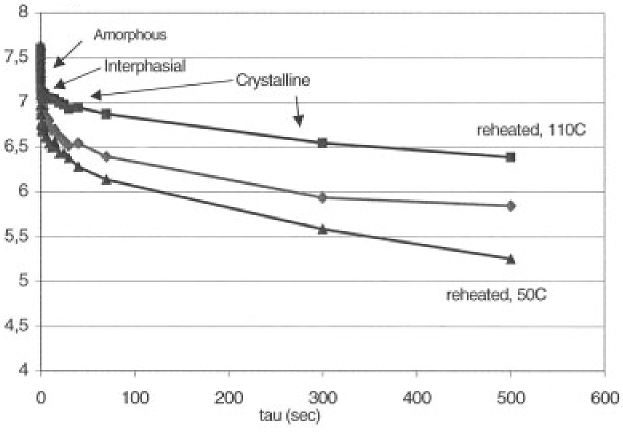

Figure 2 The figure shows the T_1 decay plot of the morphological components of HDPE extrusion coating (sample 3) prior to and after reheating to 50 and 110°C.

CONCLUSIONS

Two different extrusion-coating grades of polyethylene, namely LDPE and HDPE, were coated on high-density papers. The two polyethylene grades extrusion coated on high-density papers differ with respect to how they respond on storage and low-temperature heat treatment. The HDPE does not respond on storing at ambient temperature and heat treatment at low temperature in the same way that LDPE does. The LDPE-coating exhibits a higher degree of chain mobility at these low temperatures that seems to correlate with an increase in the monoclinic crystalline fractions at the paper surface. The nature of this response appears to be a result of the nature of the adhesion to the paper surface and the properties of this surface, orientation of polymer chains, and chain mobility. With a more rough paper surface the effect of storage gave the same effect as heat treatment at 55°C and the increase in the monoclinic fraction seems to be mainly the result of growth of the mean crystalline lamella thickness. A smother paper surface with a surface tension closer to that of polyethylene and a higher pressure makes the conditions more optimal for the formation of the monoclinic layer at the extrusion coating. In this case, storage at ambient temperature had no effect on the size of the monoclinic fraction. A subsequent low temperature heat treatment decreased the mean crystallite thickness, but increased the monoclinic crystalline fraction, likely as a result of initiation and formation of new crystallites at the paper surface (transcrystallisation) likely enhanced by a "hairy" and porous cellulose fiber surface structure.

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