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Development of drawn polypropylene-based blends and composites

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Abstract. We developed highly oriented polypropylene tapes with solid-state drawing, and studied the effect of additives—two grades of talc, low-density polyethylene (LDPE), and an amorphous poly- α -olefin (APAO). The PP compounds were prepared in a co-rotating twin-screw extruder, and sheet films were extruded. The specimens cut out from the sheet films were drawn in a tensile testing machine, and the drawing temperature of 130 °C was maintained by the heating chamber of the tensile testing machine. A draw ratio of $\lambda=10$ was set. We investigated the work of drawing and prepared DSC and tensile tests. Both types of talc acted as strong nucleating agents and increased the tensile strength and modulus of the tapes. APAO reduced the crystalline melting enthalpy only to a small extent. Both APAO and LDPE reduced the tensile strength and modulus and increased the strain at break of the tapes.

1. Introduction

Polypropylene (PP) is one of the most widely used polymers in the world, mainly due to its low price. However, given that it is a commodity plastic, PP has modest mechanical properties. These properties can be greatly enhanced by drawing, as during the drawing process, the PP molecules align to the drawing direction; thus, tensile strength and modulus can be increased by an order of magnitude in the drawing direction, at the cost of reducing strain at break [1].

Polymers can be drawn from the melt (or solution) or in the solid state. During the latter, polymers are first extruded to the desired shape (filament or sheet film), and after solidification, they are drawn between rolls, usually at an elevated temperature but always below the melting temperature of the given polymer [2]. Polypropylene is a semi-crystalline polymer, which contains amorphous and crystalline phases. The molecules in the crystalline phase, originally folded in lamellae, start to move upon drawing and form fibril-like structures while going through micro-necking, as Peterlin described [3]. The molecules in the amorphous phase start to align upon drawing in a more or less parallel manner. Perfect alignment, however, is prevented by the tie molecules, which go through both the amorphous and the crystalline phases. These molecules tighten upon drawing, and eventually, they become taut tie molecules, considerably reducing the capability of the relative movement of the phases, thus they reduce the achievable draw ratio [3].

Additives can be used for two main reasons in the production of drawn PP tapes: (i) to reduce the price of the product with the addition of a cheaper additive, or (ii) to specifically improve at least one property of the product. These properties can be mechanical properties, dyeability, maximal draw ratio, melt viscosity, etc. [4].



Although both polyethylene (PE) and polypropylene are polyolefins, they are insoluble in each other, and they crystallize in a different lattice, which is orthorhombic for PE, and usually monoclinic (α -PP) or hexagonal (β -PP) for polypropylene [5]. PP can, however, facilitate epitaxial crystallization of the polyethylene phase of drawn PP/PE blends [6]. Marcincin et al. investigated drawn blends of PP and low-density polyethylene (LDPE) [7]. They observed that low (below 20 wt%) LDPE content caused an increment in mechanical properties. They attributed the increment to the morphology of the blend, as the LDPE phase melted during the drawing process, and long, fibre-like polyethylene phases were formed upon cooling. The shape of these phases caused a geometrical constraint to crystal growth, thus forming an orientation and strengthening the drawn tape [7]. Similar results were reported for PP/HDPE (high-density polyethylene) blends [8].

Amorphous poly- α -olefins (APAOs) were developed to substitute atactic polypropylene (aPP), which originally was a by-product of the production of isotactic polypropylene. They mostly consist of propylene repeating units, residing in an atactic manner, similar to aPP, and they are used as melt adhesives and seals [9]. Although in PP/APAO blends APAOs are almost exclusively atactic and they only occur in the amorphous phase of the PP, the relatively small content of APAOs reportedly increased the crystalline ratio of PP. This was attributed to the fact that small APAO molecules can facilitate the molecular movements of the PP, thus making it easier for the PP molecules to be arranged and form crystalline phases [10, 11].

Talc is a naturally occurring magnesium silicate and a widely used additive in the polymer industry. The incorporation of talc particles reportedly improved tensile strength and modulus at the cost of reducing strain at break. Talc is also an efficient nucleating agent in PP [12, 13].

The main objective of this research is to investigate the effect of LDPE, APAO, and talc on the drawability of PP and the properties of drawn polypropylene tapes. These results can later be a basis of the investigation of a combination of additives, with which the tensile strength and modulus of the drawn PP tapes can be increased without a significant decrement in the strain at break.

2. Materials and methods

2.1. materials

We used Tipplen H681F (MOL Petrolkémia Zrt., Tiszaújváros, Hungary) polypropylene homopolymer as matrix material. This polypropylene grade has a Melt Flow Index (MFI) of 1.7 g/10 min (measured at 230 °C with 2.16 kg).

To improve the properties of the drawn polypropylene tapes, we used the additives listed below:

- HTP05 and HTP1 talc grades (IMI Fabi S.p.A., Postalesio, Italy). Both HTP05 and HTP1 are uncoated, talcs in powder form, and their average particle size is 1.4 and 1.8 μm , respectively, and 99% of their particles are smaller than 10 and 11 μm , respectively. The density of both talc grades is 2.8 g/cm³.
- Tipolen MF 243-51 (hereinafter referred to as LDPE) low-density polyethylene (MOL Petrolkémia Zrt., Tiszaújváros, Hungary). The MFI value of this LDPE grade is 20 g/10 min (measured at 190 °C, with 2.16 kg).
- Vestoplast 708 (hereinafter referred to as APAO) amorphous poly- α -olefin (Evonik Industries AG, Essen, Germany) was also used as an additive. This APAO grade has a mass average molecular weight (M_w) of 75000 g/mol, and a viscosity of 8 \pm 2 Pa·s.

2.2. Methods

The PP matrix and the additives were mixed in an LTE 26-44 co-rotating twin-screw extruder (Labtech Engineering Co., Samutprakarn, Thailand). The rotation speed of the screw of the extruder was 90 1/min, and the rotation speed of the screw of the volumetric dosing unit was 12 1/min. The zone temperatures of the extruder were 160, 170, 175, 180, 180, 180, 185, and 190 °C from hopper to die, and the temperature of the die was 190 °C. The extruded filaments were pelletized with an LZ-120/VS pelletizer (Labtech Engineering Co., Samutprakarn, Thailand). We prepared compounds containing 5,

10, and 15 wt% of APAO and LDPE, and compounds containing 0.5, 1.0, and 1.5 wt% of talc. The neat PP reference was also extruded to provide the same thermal history.

0.5 mm thick sheet films were prepared from the compounds with a 25-30C single-screw extruder and an LCR300 sheet film line (Labtech Engineering Co., Samutprakarn, Thailand). The rotation speed of the screw was 90 1/min, the zone temperatures of the extruder were 170, 180, 190, 195, and 200 °C from hopper to die, and the temperature of the die was 200 °C. Take-up speed was 0.6 m/min, and the first roll of the sheet film line was tempered to 85 °C. The rotation speed of the winder was set according to the diameter of the roll (between 6 and 11 1/min).

Dumbbell specimens were cut out from the sheet films (figure 1). The dimensions of the specimen were determined by the geometrical constraints of the heating chamber of the tensile testing machine; we were able to achieve a draw ratio of $\lambda=10$. We designed the specimens with the measuring length of 30 mm, and with the width of 10 mm.

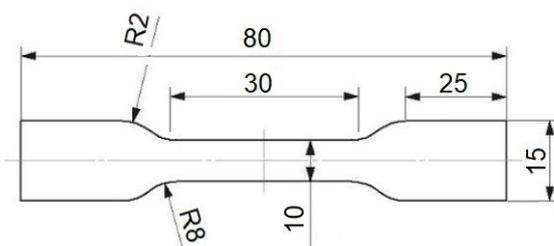


Figure 1. The specimens used in the drawing process.

The specimens were drawn in the heating chamber of a Z250 tensile testing machine (Zwick GmbH, Ulm, Germany). The machine was equipped with 100 kN wedge grips and a 20 kN load cell. The drawing temperature was 130 °C and drawing speed was 600 mm/min. The specimens were drawn to the draw ratio of $\lambda=10$. We stored the specimens in the heating chamber for at least 10 minutes prior to drawing to set their temperature, and left at least 2 minutes between closing the chamber door and the start of the drawing to maintain a constant temperature of 130 °C in the chamber. Force–strain curves during the drawing were recorded with the built-in software of the tensile testing machine.

The work of drawing was determined by calculating the area under the force–strain curves recorded during the drawing process. We expressed the work of drawing in terms of the area of the cross-section of the specimens (the work of drawing was divided with the area of the specimens) to make the results comparable.

Differential scanning calorimetry was performed on both drawn and undrawn samples. We conducted the tests using a Q2000 DSC device (TA Instruments, New Castle, United States) with one heating ramp in the temperature range between 0 and 200 °C, with a heating rate of 10 °C/min in a 50 ml/min nitrogen atmosphere.

Tensile tests were conducted with a Z005 tensile testing machine (Zwick GmbH., Ulm, Germany) on both drawn and undrawn samples. In the case of the drawn tapes, we cut out a 140 mm long sample from the middle of the tape, and the tensile tests were performed on these specimens with the measuring length of 60 mm. For undrawn samples, we used the specimens specified in Figure 1. The test was performed with the drawing speed of 20 mm/min at room temperature in both cases. As the drawn width of the drawn samples could not be measured with conventional methods—their width decreased greatly during the drawing process—we measured their width using a VHX-5000 light microscope (Keyence Ltd., Osaka, Japan). At least five specimens were tested in each case.

3. Results and discussion

3.1. Work of drawing

The specific drawing works of the different compounds are listed in table 1. In the case of the talc grades, an unambiguous tendency cannot be drawn due to the relatively large standard deviation. It can be

concluded, however, that talc did not have a huge effect on the drawing work of the PP—especially compared to that of the APAO and LDPE.

Both LDPE and APAO acted as a plasticizer, even though they probably caused this effect differently. APAOs—similar to atactic polypropylene—are soluble in the amorphous phase of the PP matrix [10], but LDPE is not, consequently, it forms separate phases when mixed with PP when the melt is cooled [7]. At the temperature of drawing (130 °C), the polyethylene phase was in the melt state, and was consequently easily deformed upon drawing. This greatly reduced the specific drawing work. The APAO used consisted of relatively small molecules—compared to that of the PP matrix—and these small APAO molecules facilitated the movement of the PP molecules during the drawing process, thus reducing the work of drawing.

Table 1. Specific drawing work of the tapes.

Additive	Additive content (wt%)	Specific work of drawing (J/mm ²)
-	-	2.17±0.09
HTP05 talc	0.5	2.13±0.17
HTP05 talc	1.0	2.11±0.12
HTP05 talc	1.5	2.22±0.04
HTP1 talc	0.5	1.99±0.16
HTP1 talc	1.0	2.17±0.15
HTP1 talc	1.5	2.11±0.13
APAO	5	2.04±0.16
APAO	10	1.76±0.13
APAO	15	1.46±0.05
LDPE	5	1.75±0.11
LDPE	10	1.64±0.20
LDPE	15	1.48±0.04

3.2. Differential scanning calorimetry

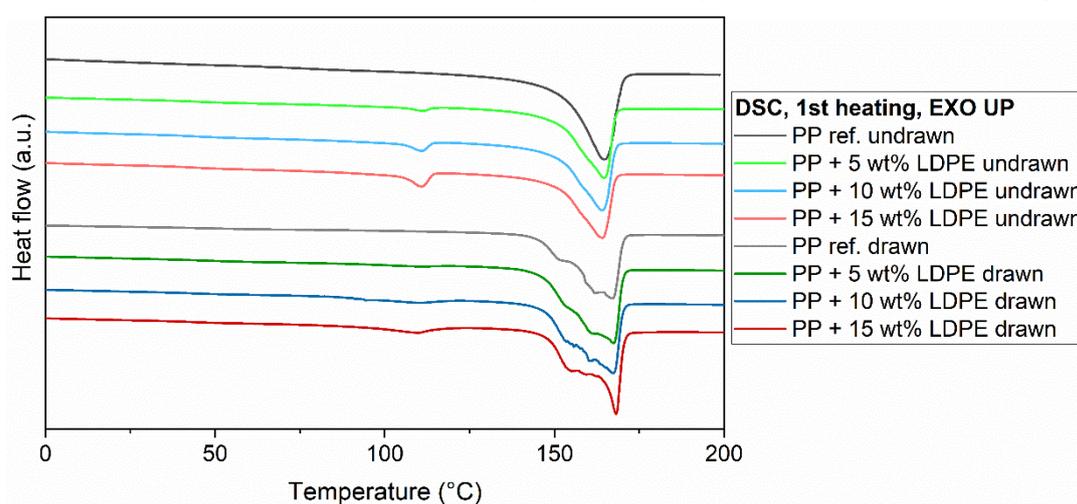
The drawing process increased both the melting temperature and melting enthalpy of the pure PP, as expected (table 2). APAO, however, reduced both values. The APAO applied is fully atactic, and thus it cannot form crystalline phases. It can reside exclusively in the amorphous phase of the PP matrix. However, the decrement of melting enthalpy is not proportional to the APAO content of the blend. It was reported [10] that small APAO molecules can facilitate the movement of large PP molecules, as discussed above. This effect of the APAO molecules helped the PP molecules to form a larger ratio of the crystalline phase, thus the overall melting enthalpy did not decrease as much as expected due to the addition of the fully amorphous APAO.

The DSC curves of PP/LDPE blends had two distinct melting peaks (figure 2), from which the one at the lower temperature belongs to the polyethylene phase and the other one to the PP phase. The crystalline melting temperature of the polyethylene phase decreased upon drawing. In the literature, if drawing took place above the melting temperature of PE, but below the melting temperature of PP, an increase was reported in the melting temperature of the polyethylene phase of drawn PP/PE blends both for LDPE [7] and HDPE [8]. This difference could have been caused by the high MFI value of the PE applied, which eased the deformation of the PE phase upon drawing. During the solidification of the PE phase, the geometrical constraints of these elongated PE phases reduced the ability of the PE molecules to form crystalline phases, which could have caused imperfect lamellae growth, and thus, lower melting temperature.

Table 2. Results of the DSC tests of drawn and undrawn samples.

Additive	Additive content (wt%)	Draw/undrawn (-)	Melting temperature ^a (°C)	Melting enthalpy ^a (J/g)
-	-	undrawn	164.6	86.4
-	-	drawn	167.6	115.9
HTP05 talc	0.5	undrawn	163.6	86.2
HTP05 talc	1.0	undrawn	163.8	88.0
HTP05 talc	1.5	undrawn	163.1	94.9
HTP05 talc	0.5	drawn	167.9	117.5
HTP05 talc	1.0	drawn	167.1	115.7
HTP05 talc	1.5	drawn	168.5	119.9
HTP1 talc	0.5	undrawn	164.0	91.2
HTP1 talc	1.0	undrawn	164.4	92.4
HTP1 talc	1.5	undrawn	164.3	91.1
HTP1 talc	0.5	drawn	167.0	119.3
HTP1 talc	1.0	drawn	168.3	119.8
HTP1 talc	1.5	drawn	167.8	113.7
APAO	5	undrawn	164.8	86.5
APAO	10	undrawn	164.3	86.7
APAO	15	undrawn	164.4	86.1
APAO	5	drawn	166.9	108.4
APAO	10	drawn	166.4	104.9
APAO	15	drawn	166.4	106.3
LDPE	5	undrawn	111.3; 164.5	1.6; 82.6
LDPE	10	undrawn	111.1; 164.0	4.7; 80.3
LDPE	15	undrawn	110.8; 164.2	9.5; 74.6
LDPE	5	drawn	106.2; 167.4	1.4; 111.8
LDPE	10	drawn	110.2; 167.4	5.4; 103.5
LDPE	15	drawn	109.5; 168.0	8.9; 96.7

^a In the case of the LDPE additive, the first value belongs to the LDPE phase, and the second to the PP phase

**Figure 2.** DSC curves of the drawn and undrawn PP/LDPE blends.

3.3. Tensile test

Typical tensile curves are presented in figure 3, and the results of the tensile tests are listed in table 3. HTP1 talc increased both tensile strength and modulus compared to that of the neat PP reference, except when talc content was 1 wt% (note that the melting enthalpy of this compound is also smaller than that of the others, so it is possible that this compound was not mixed homogeneously enough). HTP1 talc also

increased tensile strength and modulus, but to a smaller extent than HTP05, as it consists of larger particles, which presumably caused a lower level of reinforcement.

APAO reduced tensile strength and modulus, but increased strain at break. The decrement in strength and modulus is less substantial than the increment in strain at break in the case of the drawn tapes. The reason for this can be that APAO did not considerably reduce the overall melting enthalpy—and thus, the crystalline ratio—of the blend.

It is difficult, however, to conclude the overall tendencies of the tensile properties due to the large standard deviations. These could be caused by the micro-cracks originally formed while the specimens were cut out before drawing. This can be overcome by cutting out the specimens more gently and with perhaps the use of a different method.

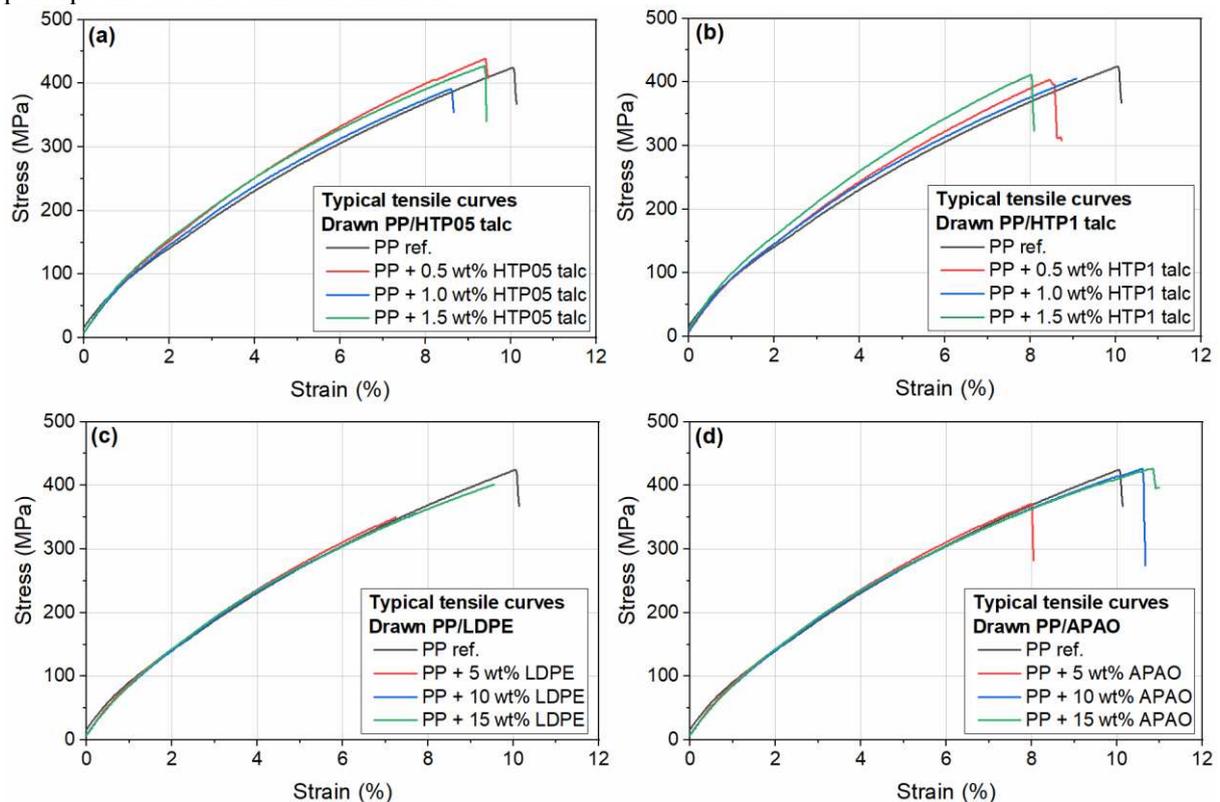


Figure 3. Typical tensile curves of drawn PP/HTP05 talc (a), PP/HTP1 talc (b), PP/LDPE (c), and PP/APAO (d).

Table 3. The results of the tensile tests.

Additive	Additive content (wt%)	Drawn/undrawn	Tensile modulus (GPa)	Tensile strength (MPa)	Strain at break (%)
-	-	undrawn	1.21±0.02	40.1±1.5	333.2±134.1
-	-	drawn	9.44±0.77	398.5±22.2	8.5±0.8
HTP05 talc	0.5	undrawn	1.25±0.11	39.2±0.9	341.1±187.7
HTP05 talc	1.0	undrawn	1.19±0.03	40.8±0.5	179.1±94.5
HTP05 talc	1.5	undrawn	1.22±0.08	39.1±0.9	421.5±94.4
HTP05 talc	0.5	drawn	10.15±0.14	414.2±31.2	9.4±1.8
HTP05 talc	1.0	drawn	9.82±0.18	389.5±60.4	8.1±1.9
HTP05 talc	1.5	drawn	10.21±0.10	421.2±25.1	9.3±1.5
HTP1 talc	0.5	undrawn	1.18±0.06	38.4±0.8	307.4±85.4
HTP1 talc	1.0	undrawn	1.23±0.10	38.2±0.8	406.8±86.3
HTP1 talc	1.5	undrawn	1.31±0.07	38.3±0.6	270.3±142.8
HTP1 talc	0.5	drawn	9.69±0.26	415.8±44.1	10.4±1.4
HTP1 talc	1.0	drawn	9.61±0.25	412.9±28.1	10.2±2.2
HTP1 talc	1.5	drawn	9.73±0.40	362.5±39.3	8.3±1.8
APAO	5	undrawn	1.01±0.08	35.7±0.2	134.3±62.1
APAO	10	undrawn	0.91±0.03	32.5±0.7	430.1±187.1
APAO	15	undrawn	0.87±0.06	31.0±0.4	404.5±129.7
APAO	5	drawn	9.38±0.21	374.2±25.5	10.4±1.7
APAO	10	drawn	8.75±0.20	384.1±41.2	10.3±1.9
APAO	15	drawn	8.60±0.34	386.5±49.1	11.9±1.8
LDPE	5	undrawn	1.16±0.10	40.9±3.0	626.4±102.6
LDPE	10	undrawn	1.00±0.05	35.4±2.1	632.9±234.7
LDPE	15	undrawn	0.94±0.04	33.1±1.2	511.5±127.5
LDPE	5	drawn	9.32±0.59	338.2±28.5	6.6±0.9
LDPE	10	drawn	10.50±0.72	401.2±59.4	7.3±2.2
LDPE	15	drawn	9.72±0.53	420.1±63.3	9.3±3.6

4. Conclusions

Polypropylene-based compounds were prepared with a twin-screw extruder with LDPE, APAO, and two different grades of talc as additives. The specimens cut out from the sheet film made from these compounds were drawn in the heating chamber of a tensile testing machine to the draw ratio of $\lambda=10$ at 130 °C. DSC and tensile tests were performed, and we also investigated the work of drawing. The following conclusions can be drawn from the results of the tests:

- Both LDPE and APAO reduced the specific work of drawing, as LDPE was in the melt state at the drawing temperature, and small APAO molecules facilitated the movement of the PP molecules. The talc grades showed only a slight effect on the work of drawing.
- Both talc grades acted as nucleating agents, but both APAO and LDPE reduced the crystalline melting enthalpy. The fully amorphous APAO, however, reduced the melting enthalpy less than it could be expected from its weight ratio in the blend. This could also be linked to the small APAO molecules facilitating the movement of PP molecules, thus making it easier for the PP molecules to form crystalline phases.
- Talc grades increased tensile strength and modulus and reduced strain at break—this was more significant in the case of HTP05 talc, but both LDPE and APAO had an adverse effect on tensile properties. APAO, however, increased strain at break more substantially than it reduced strength and modulus, as it reduced the crystalline ratio of the PP phase less than LDPE did.
- Due to the above-mentioned properties of the drawn PP/APAO blends, APAOs can improve the properties of highly drawn PP tapes. When combined with other additives, it can improve the strain at break of the drawn tapes without reducing their tensile strength and modulus.

5. Acknowledgments

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