Recycling of Mixed Plastic Fractions: Mechanical Properties of Multicomponent Extruded Polyolefin Blends Using Response Surface Methodology

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ABSTRACT: A software program for experimental design and response surface methodology (RSM) was used to set up and evaluate the effect of modifiers in mixed polyolefin fractions. Sheet extruded blends of low-density polyethylene (LDPE), linear low-density polyethylene (LLDPE), 2 different high-density polyethylenes (HDPE) and 2 different polypropylenes (PP) were prepared and characterized by tensile tests and tear tests. Ethylene–vinyl acetate copolymer (EVA), a very-low-density polyethylene (VLDPE) and a poly(1-butene) (PB) were used as modifiers at levels up to 20 wt % in order to improve the toughness of PE–PP blends. LLDPE, in which 1-butene is the comonomer, is most compatible with the PPs. The VLDPE generally gives the most substantial gain in tensile strength and tear propagation resistance, and it also has the ability to smooth out effects of variations in the composition of PE–PP mixtures. For HDPE–PP blends, the EVA and PB are also effective modifiers. PB acts as a plasticizer in the PP phase whereas VLDPE forms a separate rubbery phase. The RSM is a valuable tool in, for example, recycling operations, where mixed plastic fractions often appear. © 1998 John Wiley & Sons, Inc. J Appl Polym Sci 70: 2381–2390, 1998

 $Key\ words:$ recycling; polyolefin blends; modifiers; upgrading; response surface methodolgy (RSM)

INTRODUCTION

The polyolefins are a very versatile group of polymers. They all have the same basic structure: aliphatic hydrocarbons, which gives them some common characteristics, such as hydrophobicity, low density, and excellent electrical insulation properties. Mechanical properties shows a very wide range of values and can be varied further by blending.¹ Examples of commercial polyolefin blends are low-density polyethylene–linear lowdensity polyethylene (LDPE–LLDPE) in film blowing,² thermoplastic elastomers based on polypropylene (PP) mixed with ethylene-propylene copolymers,³ poly[1-butene] (PB) as modifier in PP for improvement of melt-flow properties and heat sealability or in polyethylenes (PE) for production of easy-to-open packaging.⁴ Polyolefin blends are an important group of materials from the viewpoint of recycling, that is, utilization of production scrap from coextrusion lines and postconsumer packaging recycling, where it is desirable to be able to tolerate some level of mixing.^{5,6} The morphology and mechanical properties of polyolefin blends is a complex issue that is governed by many factors, that is, repeating units, comonomers, tacticity, molecular weight distributions, and thermomechanical history. Structural dissimilarities of a pair of polyolefins doesn't have

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to be large for phase separation to occur in the molten state,^{7–9} but all polymers within this group seems to have at least some mutual solubility in the melt as well as in the amorphous phase after solidification. This statement is made based on the observations that phase-separated polyolefin pairs, also highly dissimilar pairs, such as PP–PE, still affect the crystallization behaviors of one another by nucleation,^{10–12} retardation,¹³ or changing the spherulite structure.¹⁴ The blends of olefin copolymers can cocrystallize if the difference in monomer composition is not too substantial (isomorphism),^{15,16} but cocrystallisation of olefin homopolymers with different repeating units is very rare.¹¹

The present works characterize the mechanical properties of multicomponent blends of various PEs, PPs, ethylene-vinyl acetate (EVA), and PB, with the PEs as the main components, using response surface methodology (RSM). Several 2-component and some 3-component blends of these polymers have been studied in detail over the past decades. The degree of miscibility in the PP-PB system has been a matter of some controversy.⁷ PB homopolymers and copolymers, with a few wt % ethylene, are used commercially as modifiers for PP in order to improve impact resistance, tear strength, and transparency and to lower the seal initiation temperature.^{4,17}

Morphology and mechanical properties for blends of PP and HDPE, ^{6,10,12,16,18,19} LDPE, ^{5,20–22} or $LLDPE^{5,22-24}$ have been studied extensively. PP blends with HDPE as the major component are generally considered to exhibit rather poor compatibility.^{5,6,10} Several workers have reported that impact resistance at room temperature is reduced with a factor around 3 (the percentage of reduction of the notched Charpy or Izod impact strength per percentage of PP) up to PP levels around 20%.^{5,6,18} The results, of course, depend on which grades were used and how the samples were prepared. This is clearly illustrated by Sherman²⁵ who prepared strong composites, where PP acts as a reinforcement in HDPE, by cold drawing, followed by heating up to a temperature above the melting point of HDPE but below the melting point of PP. Poor mechanical compatibility has also been reported for blends of PP and LDPE or LLDPE, 20,26,27 but modification of melt mixing conditions can turn brittle blends into highly ductile ones.²¹ Teh et al.²⁸ have studied tubular film extrusion of octene-based LLDPE and found moderate changes in the mechanical properties up to 20% addition of PP. Tensile strength, dart impact, and tear resistance was reduced by PP, but these changes could be successfully counteracted by addition of 5% EVA (containing 28 wt % vinyl acetate). Other materials that have been evaluated as compatibilizers or modifiers for various PE–PP blends are ethylene–propylene random copolymers,^{5,16,18,20,23} ethylene–propylene block copolymers,^{5,23} styrene–ethylene–butadiene block copolymers,²⁷ and very-low-density polyethylene (VLDPE).⁵ These polymers serve to reduce the overall crystallinity of the material, thereby shifting the properties from stiff–brittle towards soft– tough^{16,18,20}; and they can also, at least in the chase of ethylene–propylene copolymers, provide a finer dispersion of the PE and PP phases.^{5,15}

The object of this work is to study mechanical properties by RSM of multicomponent blends of PEs, PPs, EVA, and PB, with the polyethylenes as main components. Mechanical compatibility of extruded blends of various PE and PP grades that are common in packaging applications is examined in this article, as well as modification of these materials with EVA, VLDPE, and PB.

EXPERIMENTAL

Materials

All materials used in this study are commercial resins. Table I presents general information about the grades, as provided by the suppliers, and the abbreviations that will be used in this text. Both PPs are nonnucleated homopolymers, but the PB is a copolymer containing 2 wt % ethylene.

Experimental Design

The experimental planning and evaluation was made using Modde 3.0 software, supplied by Umetri AB (Umeå, Sweden). This is a Windows software for response surface methodology, that is, statistical experimental design and multivariate analysis. It is an aid for investigation and optimization of complex products and processes in which many factors affect the results. The factor that, where varied, was the percentage of each of the 9 polymers in the blends. An experimental set-up was designed considering the following constraints.

1. The sum of the polypropylenes was not allowed to exceed 30%.

Material	Abbreviations	Supplier	Grade	Density (kg/m ²)	MFR (dg/min)	
HDPE (blow molding)	HDb	Borealis	HE8331	955	$0.2^{\rm b}/24^{\rm d}$	
LLDPE (1-butene-based)	LLD	Borealis	LE6520	919	1.2^{b}	
LDPE	LD	Borealis	LE1804	922	$2.1^{ m b}$	
HDPE (injection molding)	HDi	Borealis	HE7012	962	$12^{\rm b}$	
PP (extrusion)	PPe	Neste	VB3247C	908	$3.2^{\rm c}$	
PP (injection molding)	PPi	Borealis	HF135M	908	$18^{\rm c}$	
Poly(ethylene-co-vinyl acetate) ^a	EVA	DuPont	Elvax 3165	940	$0.7^{ m b}$	
Very-low-density polyethylene [poly(ethylene- <i>co</i> -1-octene)]	VLD	DuPont–Dow Elastomers	Engage 8150	868	0.5^{b}	
Poly(1-butene)	PB	Shell	DP8220	897	2.0^{b}	

Table I General Information About the Materials Used in the Experiments

 $^{\rm a}$ 18 wt % vinyl acetate.

^b 190°C; 2.16 kg.

° 230°C; 2.16 kg.

^d 190°C; 21.6 kg.

- 2. The sum of the injection-molding grades was not allowed to exceed 30%.
- 3. The sum of the modifiers, EVA, PB, and VLDPE, was not allowed to exceed 20%.

Table II presents the composition of the blends included in this study. This is identical with the experimental design suggested by the software, except for blends 48 and 49, which were added in order to increase the applicability of the model to blends of HDb, PPi and modifiers. Based on the experimental results, models for each response (tensile strength, modulus, etc.) were generated by "Modde". The models are polynomial functions of the factors (HDb, LD, LLD, etc.) with a constant, first-degree coefficients, second-degree coefficients, and coefficients for interaction parameters (e.g., HDb \times LLD).

Processing

The materials were compounded and repelletized using a counter-rotating, intermeshing, twin-screw extrusion compounder, a Brabender DSK 35/9D *D*, 35 mm; *L*, 9D; barrel temp, 200–210°C; screw speed, 60 rpm). The compounded materials were then extruded into sheets of approximately 0.5 mm thickness using an Axon single-screw extruder (*D*, 18 mm; *L* 20D; barrel temp profile, 170–190–200–200°C; screw speed, 120 rpm) equipped with a slot die (width, 52 mm; gap, 0.55 mm; temp, 180°C). The haul off speed was 2.3 m/min, and the throughput was approximately 2.7 kg/h.

Analyses

Tensile testing was performed at 23°C according to ISO 1184 on Dumbbell specimens with 33-mm-long and 6-mm-wide narrow sections that were punched out of the extruded sheets with the long axis coinciding with the machine direction. The rate of grip separation was 200 mm/min. The yield strength was calculated at an offset of 5%. Five specimens of each material composition were tested. Blends 19, 37, 45, and 49 had a tendency not to break in the narrow section of the specimens but instead in the necking front at high extensions when the necking had propagated beyond the narrow section. This problem was overcome by measuring the elongation at break and tensile strength for these blends on specimens with an only 3-mmwide narrow section. The length of the narrow section, initial grip distance, and speed was the same for these tests as for those with the standard specimens.

Tear propagation resistance (TPR) was measured in accordance with ASTM D1938. The longitudal slits of the test specimens, from which the tears propagate, were cut parallel to the machine direction. Five specimens of each material composition were tested. For measurements of both TPR and tensile properties, an Instron 5566 tensile tester was employed.

Dynamic mechanical properties in the tensile mode was scanned from -50 to 100° C using a

Blend	HDb (%)	LLD (%)	LD (%)	HDi (%)	PPe (%)	PPi (%)	EVA (%)	VLD (%)	PB (%)
01	100								
02		100							
03			100						
04	70			30					
05		70		30					
06	70					30			
07		70				30			
08			70			30			
09	70				30				
10		70			30				
11			70		30				
12			40	30	30				
13	80						20		
14		80					20		
15			80				20		
16	80							20	
17		80						20	
18			80					20	
19	80								20
20		80							20
21			80						20
22	50			30			20		
23			50	30			20		
24		50		30				20	
25		50		30					20
26			50	30					20
27		50				30	20		
28			50			30	20		
29	50					30		20	
30			50			30		20	
31			50			30			20
32			50		30		20		
33		50			30			20	
34			50		30				20
35		20		30	30		20		
36	20			30	30			20	
37	20			30	30				20
38		33.3	33.3		10	10	6.7	6.7	
39		35	35	15			5	5	5
40		27.5	27.5	15		15	5	5	5
41	35		35			15	5	5	5
42	28.3		28.3	15	15		6.7	6.7	
43	35	35			15		5	5	5
44	33.3	33.3		10		10	6.7	6.7	
45	30	30		12	12	6			10
46	20	20		15	15	15	5	5	5
47	28.3	28.3	28.3				5	5	5
48	50					30			20
49	50					30	20		
50	20	20	20	15	15				10
51	16.7	16.7	16.7	15	15			10	10
52	16.7	16.7	16.7	15	15		10		10
53	23.3	23.3	23.3	12	12	6			
54	18.3	18.3	18.3	12	12	6	5	5	5

 Table II
 Composition of the Characterized Blends



Figure 1 Scatter plot of yield strength versus the E modulus for all polyolefin blends studied.

Polymer Laboratories Mk II Dynamic Mechanical Thermal Analyzer. The frequency was 1 Hz, and the heating rate was 3°C/min. Test specimens were cut in the machine direction of the extruded blends 01, 06, 16, 19, 29, and 48.

RESULTS AND DISCUSSION

The tensile tests revealed that polyolefin blends who have a high E modulus also have high yield strength (σ_y) . Figure 1 shows a scatter plot of σ_y versus E for all sample compositions included in this study. There is a general relation of the type $\sigma_y = \alpha E^{\beta}$, where α equals 0.2625 and β equals 0.6389, that fits well to all samples.

Figure 2 shows how the flexibility of HDb and a 70/30 HDb–PPi blend is increased by addition of the modifiers, expressed as reduction of E modulus. In both cases, VLD reduces the E modulus



Figure 2 *E* modulus as a function of the modifier addition level for HDb and 70/30 HDb–PP blends.



Figure 3 Tensile strength as a function of the modifier addition level for HDb and 70/30 HDb–PP blends.

the most. For HDb, a 20% addition of VLD results in approximately 45% reduction of the *E* modulus, so it is clear that VLD has a large inhibitory effect on the ability for HDb to crystallize.

Figure 3 shows how the modifiers affect the tensile strength when added to HDb, a 70/30 HDb–PPi blend or a 70/30 HDb–PPe blend. The 2 PP grades have very different effects when added to HDb. PPe increases the tensile strength, whereas PPi decreases it. Also, the elongation at break is much lower for the HDb–PPi blend (see 06 in Table III) than for the HDb–PPe blend (09 in Table III). All 3 modifiers increase the tensile strength substantially.

Figure 4 illustrates the effect of PPs and modifiers on the tensile strength of LLDPE, which is the toughest of the pure PEs. Both PPs increase the tensile strength, which is contradictory to the results presented by Teh et al.²⁸ An important difference is that the comonomer of the LLDPE grade used in this study is 1-butene, which means that the polymer has pendent ethyl groups, whereas Teh et al.²⁸ used a 1-octene-based LLDPE with pendent *n*-hexyl groups. The mechanical compatibility between PP and LLDPE seems to be favored when the side groups are short.

Figure 5 presents the tensile strength for LDPE and its blends with PPs and modifiers. The mechanical compatibility between LDPE and the PPs is poor, especially for PPi. In PEs produced by free radical mechanism, *n*-butyl branches, formed by backbiting through a 6-membered ring-shaped transition state, are dominating,²⁹ that is, with longer side groups than in the butene-based LLDPE. So, based on this study, it cannot be clearly determined if it is the length of the shortchain branches or the presence of long-chain

Polyolefin Composition (%)				Unmodified			10% VLD			20% VLD				
HDb	LLD	LD	HDi	PPe	PPi	E (MPa)	$\begin{matrix} \sigma_b \\ (\mathrm{MPa}) \end{matrix}$	TPR (N/mm)	E (MPa)	$\begin{matrix} \sigma_b \\ (\mathrm{MPa}) \end{matrix}$	TPR (N/mm)	E (MPa)	$\begin{matrix} \sigma_b \\ (\mathrm{MPa}) \end{matrix}$	TPR (N/mm)
30	10	20	10	10	20	656	22.9	68.0	537	25.3	75.3	399	28.2	82.3
40	10	15	5	10	20	725	21.7	67.0	585	25.1	74.9	430	28.9	82.6
40	0	15	10	15	20	736	22.0	69.2	587	25.1	78.0	427	28.8	86.6
20	10	40	0	10	20	614	21.8	58.3	507	24.5	69.7	380	27.3	77.1
40	5	10	10	0	35	934	17.5	36.1	747	23.8	54.7	554	29.7	70.2
60	5	15	10	0	10	810	20.3	81.9	640	24.7	85.8	457	29.3	93.2
30	10	10	30	10	10	767	23.2	89.6	620	24.9	89.7	458	27.4	94.4
30	25	15	0	30	0	825	24.4	53.5	660	26.9	67.1	482	29.7	79.0
Mean	value					758	21.7	65.5	610	25.0	74.4	449	28.6	83.2
Standa	ard devi	ation				101	2.1	16.6	75	0.9	10.9	54	0.9	8.1

Table III Mechanical Properties for Examples of Polyolefin Blends Unmodified and Modified with 10 or 20 Wt % VLD

branches in LDPE that accounts for the difference in mechanical compatibility with PPs. The tensile strength of LDPE–PP blends can be drastically increased by modification with VLD.

Figure 6 shows TPR for HDb and its blends with PPs and modifiers. Both PPs reduce TPR, but the effect is much stronger for the injectionmolding grade. Also in this aspect, VLD is the most effective modifier. PB reduces TPR drastically when added to HDb but has the opposite effect when added to a 70/30 HDb–PPi blend. This synergistic effect can be assigned to the good compatibility between PB and PP.

Figure 7 illustrates the effect of PPs and modifiers on the TPR of LLDPE. The 70/30 LLD–PPe blend has significantly lower TPR than pure LLD but not the 70/30 LLD–PPi blend. All modifiers, especially EVA, lower the TPR for the LLD–PPi



Figure 4 Tensile strength as a function of the modifier addition level for LLD and 70/30 LLD–PP blends.

blend; but for the LLD–PPe blend, the modifiers have only moderate effects.

Figure 8 shows TPR for blends of LD, PPs, and modifiers. Both PP grades reduce this property severely. A synergistic effect between PB and PPs, similar to that observed for blends with HDb, is apparent. VLD also increases TPR of LD–PP blends extensively but not to levels above the values for LD–VLD blends as in the case of tensile strength.

Figure 9(a) and (b) shows results from dynamic mechanical analysis (DMA) on samples containing HDb, PPi, and VLD or PB. The β -transition peak that is associated with the glass transition normally occurs at approximately 5°C for PP but is almost completely overshadowed by the large α -transition of HDb (the broad peak around 40°C). It can be apprehended as a shoulder on the



Figure 5 Tensile strength as a function of the modifier addition level for LD and 70/30 LD–PP blends.





Figure 6 Tear propagation resistance as a function of the modifier addition level for HDb and 70/30 HDb–PP blends.

E'' curve for the 70/30 HDb-PPi blend in the 0-10°C range. Samples containing VLD exhibit a large peak on the E'' curve at -40° C. Neither this transition, nor the β -transition of iPP is shifted in the blend containing both VLD and PPi, which indicates that these polymers are well separated. Blends of PP and PB, on the other hand, has been said to be compatible or miscible in the amorphous phase by Piloz et al.³⁰ and Siegmann³¹ based on DMA, which shows a single β -transition peak between the corresponding peaks of the pure components. Berticat et al.³² studied PP-PB blends using dilatometry and thermomechanical analysis (TMA). They came to the conclusion that the data fitted better to a 2-phase model and called the polymers pseudo-compatible. Gohil et al.³³ found the blends to be miscible only if the



Figure 7 Tear propagation resistance as a function of the modifier addition level for LLD and 70/30 LLD–PP blends.



Figure 8 Tear propagation resistance as a function of the modifier addition level for LD and 70/30 LD–PP blends.

content of 1 component was over 80%. Hsu and Geil¹¹ observed separate glass transitions in the DMA spectra along with a depression of the peak temperature for PP in the blends, which indicates partial miscibility. Lee and Chen³⁴ observed that injection-molded PP-PB blends with 25 or 50% PB exhibited a higher tensile strength and ultimate elongation than the pure components, an effect that vanished when the same materials were compression-molded with slow cooling. This behavior seems very reasonable in view of observations, reported by Marand et al.,⁷ of phase separation of a 70/30 PP-PB blend in the molten state by optical microscopy. The 70/30 blend exhibited 2 distinct peaks in the melting region of PP, after isothermal crystallization at 145°C, where the first peak was attributed to less perfect crystals formed by PP molecules dissolved in the PB-rich phase of the melt. Marand et al. also observed a single glass transition by DMA, indicating a higher degree of miscibility in the PP-PB system than Geil et al. The differences might be assigned to experimental procedures and also to the fact that Geil et al. used a PB with a much higher M_w (730 kg/mol) than Marand et al. (285 kg/mol). In Figures 9(a) and (b), we can see that PPi and PB has a single β -transition also in the ternary blend with HDb, but it is considerably broadened compared to the narrow peak at -18°C for PB the blend without PPi. However, the broadening of the β -transition peak can, to a large extent, be assigned to the fact that when it is shifted to higher temperatures by PPi, it is superimposed with the onset of the α -transition of HDb, so it can still be stated that PPi and PB has



Figure 9 (a) Dynamic loss modulus and (b) dynamic tan δ at 1 Hz for samples containing HDb, PP, and VLD or PB.

a high level of mutual miscibility also in the blend with HDb.

Figure 10 gives an example of how experimental design and response surface methodology can be used to predict properties of mixed polyolefins with applications in recycling. Based on the measured data, a tensile strength of 22.8 MPa and a TPR of 69 N/mm has been predicted for a hypothetical polyolefin fraction comprised of 30% HDb, 20% LD, 10% LLD, 10% HDi, 10% PPe, and 20% PPi. Figure 10 shows the predicted values of tensile strength when 20% modifier is added. The highest value is in the lower left corner, which represents modification with only VLD; that is, VLD is the most effective modifier. No maximum is seen inside the triangle or on the sides; that is,



Figure 10 Tensile strength (MPa) for a blend of 30% HDb, 20% LD, 10% LLD, 10% HDi, 10% PPe, and 20% PPi, after modification with 20% EVA, VLD, PB, or combinations thereof.

there is no synergistic effect that would suggest that a combination of 2 or 3 modifiers would be preferable to modification with a single modifier. Figure 11 shows predicted TPR values for the same polyolefin fraction with 20% modifier. Again, VLD gives the toughest polyolefin blend.

Figure 12 shows values for tensile strength of a hypothetical "mixed rigid polyolefin container" fraction comprised of 60% HDb, 30% PPi, and



Figure 11 Tear propagation resistance (N/mm) for a blend of 30% HDb, 20% LD, 10% LLD, 10% HDi, 10% PPe, and 20% PPi, after modification with 20% EVA, VLD, PB, or combinations thereof.



Figure 12 Tensile strength (MPa) for a blend of 60% HDb, 10% PPe, and 30% PPi, after modification with 20% EVA, VLD, PB, or combinations thereof.

10% PPe, after addition of 20% modifier. The predicted value for the same blend without modifiers is 15.6 MPa, so the potential gain of using modifiers is much higher in this case. All 3 modifiers increase the tensile strength substantially.

Table III presents mechanical properties for 8 examples of mixed polyolefin fractions that might be encountered in a recycling plant, unmodified and modified with 10 or 20 wt % VLD. The *E* modulus is reduced, while the tensile strength and TPR is increased by VLD in all cases, as expected. Another important finding is that the standard deviations decrease drastically when VLD is added, which means that VLD also has the ability to smooth out the effect of variations in the composition of the unmodified polyolefin mixture.

CONCLUSIONS

Experimental design and response surface methodology can be used to predict properties of mixed polyolefins with applications in recycling. It was demonstrated that blends of a butene-based LLDPE and PP are much more compatible than LDPE–PP blends. Tensile strength and tear propagation resistance (TPR) of LDPE–PP blends can be greatly improved by addition of VLDPE. The injection-molding-grade PP causes much more severe embrittlement of HDPE than the extrusion grade. The VLDPE is an effective modifier for HDPE– PP blends, regardless of whether the PP is the injection-molding or extrusion grade. If it is desired to have less reduction of the stiffness, PB or EVA can be interesting alternatives to VLDPE as modifiers for HDPE–PP blends.

PB lowers the T_g of the PP phase when added to a HDPE–PP blend, which gives rise to synergistic effects between PB and PP. When added to HDPE, PB increases the tensile strength, while it reduces TPR. When added to a HDPE–PPi blend, PB increases both tensile strength and TPR.

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