

Physicochemical and Mechanical Properties of Experimental Coextruded Food-Packaging Films Containing a Buried Layer of Recycled Low-Density Polyethylene

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Migrational, permeation, and tensile properties of experimental five- and eight-layer coextruded and laminated films containing a middle buried layer of recycled low-density polyethylene (LDPE) comprising 40–50% bw of the multilayer structure were determined. Respective films containing 100% virgin LDPE as the buried layer were taken as controls. Results showed that the percentage of recycled LDPE in the multilayer structure did not affect overall migration values to distilled water, 3% acetic acid, and iso-octane. In all cases, overall migration values were lower than the upper acceptable limit (10 mg/dm²) set by the European Union. Transmission rate values to O₂, CO₂, and water vapor were also not affected by the percentage of recycled LDPE in the multilayer structure. On the basis of O₂ transmission rates, low-barrier, barrier, and high-barrier multilayer structures were produced. Likewise, tensile properties (tensile strength, percent elongation at break, and Young's modulus) were not affected by the percentage of recycled material in the multilayer structure. Finally, all experimental films produced no adverse effects in taste or odor of the food-contacting phase tested. The above findings are discussed in relation to the high quality of the primary LDPE scrap used throughout this work in combination with the functional barrier hypothesis. On the basis of the present results it is proposed that primary LDPE scrap may be used as a middle layer comprising 40–50% bw of multilayer food-packaging films without any compromise in migrational, barrier, mechanical, and organoleptic properties.

KEYWORDS: Multilayer films; recycled LDPE; migrational; permeation; mechanical properties

INTRODUCTION

Environmental protection in conjunction with waste reduction considerations have oriented industry, research, and government authorities toward recycling and/or reuse of packaging materials, especially plastics (1, 2). Technical processes such as blending or coextrusion of recycled plastics with virgin polymer resins are being developed for the most widely used polymers, namely, polyethylene, polypropylene, poly(ethylene terephthalate), and polystyrene (3–7).

The main question raised with plastics packaging materials containing recycled scrap is their performance in terms of their migration, permeability to fixed gases and water vapor, and mechanical properties when used in specific food-packaging applications where such properties may be critical in terms of the safety and quality of packaged foods (4, 8–12).

Currently in both the United States and European Union no regulations or directives exist controlling the use of recycled plastics materials for food-packaging applications. The U.S.

FDA has published guidelines for the use of recycled plastics for food-packaging applications on the basis of existing U.S. laws (1). In Europe regulations are planned in the form of EU directives. At present, Article 2 of Directive 89/109/EEC referring to food contact materials is applicable. This requires that food contact materials should not release substances in amounts that could endanger human health or lead to unacceptable changes in the food's composition or to deterioration in the food's organoleptic characteristics.

Coextruded multilayer structures based on LDPE, PA, and/or EVOH are increasingly being used in food-packaging applications today because of their unique properties combining the high barrier to fixed gases and flavor compounds of EVOH, the mechanical strength of PA, and the superior heat sealability and moisture barrier of polyethylene (13). Of these materials polyethylene in the form of LDPE, LLDPE, and HDPE is by far the most widely used food-packaging material (comprising ~70% bw of all plastics used) and is thus the first and most probable candidate for recycling.

Among the various approaches to the recycling of plastics packaging materials adopted by the Environmental Protection

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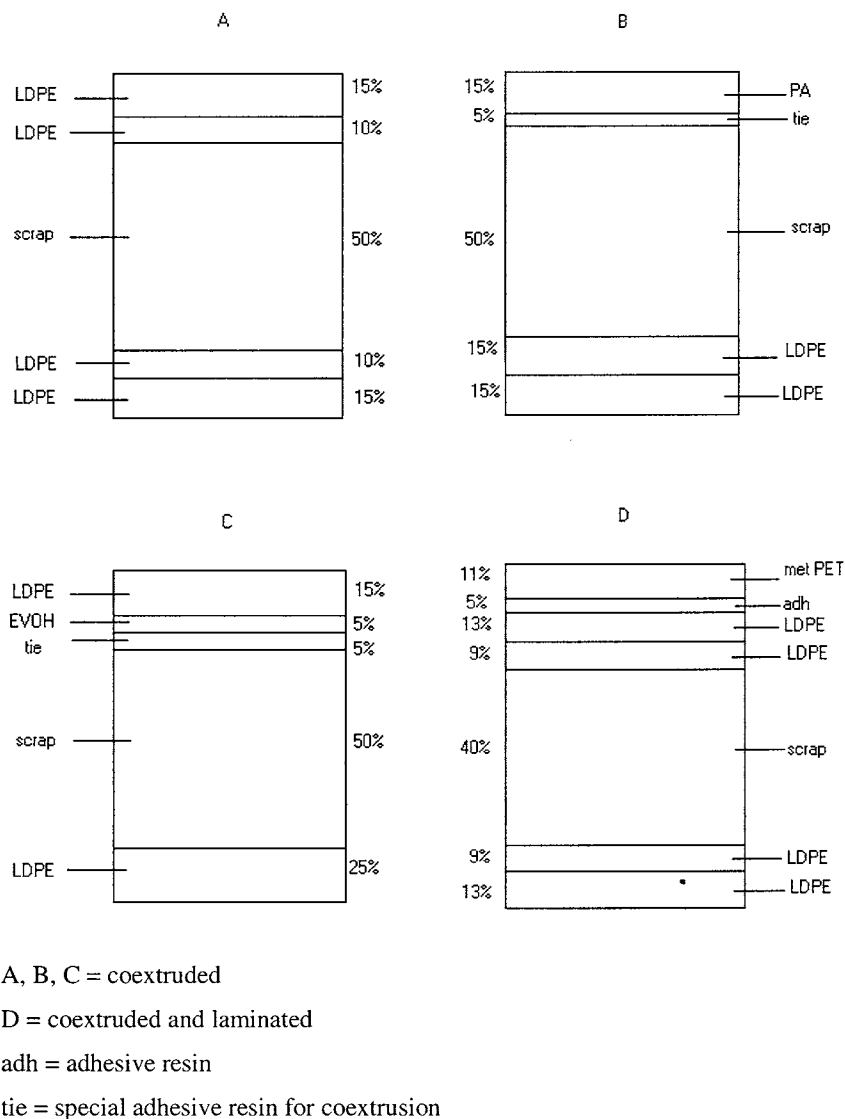


Figure 1. Structure of coextruded and coextruded/laminated experimental films (percent values indicate bw contribution of each layer to the multilayer structure).

Agency (EPA) in the United States, that is, primary, secondary, and tertiary recycling, the first involves the reuse of "in house" scrap produced during the manufacture of food-packaging containers (1). According to the EPA, such scrap material is not expected to pose a health hazard to the consumer when incorporated into a new food container provided good manufacturing practices are followed (1).

If consumer safety is of utmost importance in such applications, product quality retention in terms of the product's organoleptic properties is of equal importance. Studies have shown that organoleptic changes (mainly taste and odor) occur in foodstuffs packaged in containers made of recycled materials even when migration limits are not exceeded (3).

Even though polyethylene is rather permeable to low molecular weight substances and is not considered to be a good barrier to migration (1), the present work was undertaken with the objective to determine the effect of incorporating a buried recycled layer of LDPE, the product of primary recycling, in a five-layer coextruded and an eight-layer coextruded/laminated film structure on (a) overall migration, (b) permeability to O_2 , CO_2 , and H_2O , and (c) tensile properties of experimental low-barrier, barrier, and high-barrier food-packaging films. In all

experimental multilayer films the food-contacting layer was virgin LDPE.

MATERIALS AND METHODS

Materials. Five-layer coextruded and eight-layer coextruded/laminated films containing a middle buried layer of recycled LDPE, the product of primary recycling, were produced on a five extruder pilot scale coextrusion line (Alfa Marathon Manufacturing Co., Mississauga, Canada) and a Bieloni Castello lamination unit (Bieloni Castello, Milan, Italy). Both coextruded and laminated low-barrier, barrier, and high-barrier structures are given in **Figure 1** and **Table 1**. The recycled layer contained either 50% virgin plus 50% recycled LDPE or 100% recycled LDPE. Control samples contained 100% virgin LDPE in the respective layer. Film types A–C were 80 μm in thickness, whereas film type D was 95 μm in thickness.

LDPE was used for its high barrier to water vapor and its excellent sealability. PA and EVOH were used for their barrier and high-barrier to O_2 , respectively. Met PET was used for its high barrier to both O_2 and water vapor.

Migration Testing. Overall migration testing was carried out using distilled water, 3% acetic acid, and iso-octane as an alternative fatty food simulant (2, 14). Iso-octane in contrast to vegetable oils (olive

Table 1. Overall Migration Values from Experimental Coextruded/Laminated Films into Food Simulants

material ^a	film type	overall migration					
		distilled water		3% acetic acid		iso-octane	
		mg/L	mg/dm ²	mg/L	mg/dm ²	mg/L	mg/dm ²
LDPE/LDPE-LLDPE*/LDPE 100% virgin/LDPE-LLDPE*/LDPE	A	4.1 ± 0.6	0.21 ± 0.03	6.2 ± 0.9	0.31 ± 0.04	74.7 ± 5.2	3.7 ± 0.3
LDPE/LDPE-LLDPE*/LDPE 50–50% virgin–recycled/LDPE-LLDPE*/LDPE	A	3.8 ± 0.9	0.19 ± 0.04	7.0 ± 1.0	0.35 ± 0.05	82.5 ± 11.5	4.3 ± 0.6
LDPE/LDPE-LLDPE*/LDPE 100% recycled/LDPE-LLDPE*/LDPE	A	3.6 ± 0.5	0.17 ± 0.02	5.5 ± 0.7	0.28 ± 0.03	79.0 ± 4.5	3.9 ± 0.2
PA/tie/LDPE 100% virgin/LDPE/LDPE	B	10.5 ± 1.7	0.52 ± 0.08	15.1 ± 2.4	0.76 ± 0.12	81.0 ± 6.0	4.1 ± 0.3
PA/tie/LDPE 50–50% virgin–recycled/LDPE/LDPE	B	10.2 ± 0.8	0.52 ± 0.04	11.6 ± 2.2	0.58 ± 0.11	72.6 ± 2.5	3.6 ± 0.1
PA/tie/LDPE 100% recycled/LDPE/LDPE	B	9.6 ± 1.2	0.48 ± 0.06	13.0 ± 0.8	0.65 ± 0.04	70.2 ± 4.0	3.5 ± 0.2
PA/EVOH/tie/LDPE 100% virgin/LDPE	C	6.6 ± 0.3	0.33 ± 0.02	13.5 ± 1.9	0.68 ± 0.10	99.0 ± 9.2	4.9 ± 0.5
PA/EVOH/tie/LDPE 50–50% virgin–recycled/LDPE	C	7.1 ± 0.5	0.38 ± 0.02	14.5 ± 2.9	0.72 ± 0.14	102 ± 6	5.3 ± 0.3
PA/EVOH/tie/LDPE 100% recycled/LDPE	C	7.8 ± 1.5	0.40 ± 0.08	11.5 ± 1.6	0.58 ± 0.08	94.1 ± 3.0	4.8 ± 0.1
metPET//LDPE/LDPE-LLDPE*/LDPE 100% virgin/LDPE-LLDPE*/LDPE	D	8.1 ± 1.9	0.41 ± 0.10	124 ± 9	5.3 ± 0.4	102 ± 12	4.1 ± 0.6
metPET//LDPE/LDPE-LLDPE*/LDPE 50–50% virgin–recycled/LDPE-LLDPE*/LDPE	D	6.9 ± 0.8	0.34 ± 0.04	101 ± 12	5.0 ± 0.6	73.1 ± 7.5	3.6 ± 0.4
metPET//LDPE/LDPE-LLDPE*/LDPE 100% recycled/LDPE-LLDPE*/LDPE	D	4.9 ± 1.5	0.38 ± 0.02	81.5 ± 10.2	4.7 ± 0.5	81.0 ± 5.7	3.8 ± 0.3

^a / = coextruded; // = laminated; tie = special adhesive resin for coextrusion; * LDPE-LLDPE = blend consisting of 70% LDPE and 30% LLDPE

oil and sunflower oil) can be easily evaporated, and thus overall migration can be determined directly by weighing a residue after solvent evaporation.

Several researchers (15, 16) have reported iso-octane to be a suitable alternative fatty food stimulant for overall migration testing under various time/temperature conditions.

Rectangular strips of each film sample (surface area = 1 dm²) were placed in two-side contact (total contact surface area = 2 dm²) with 100 mL of food simulant (distilled water, 3% aqueous acetic acid, or iso-octane) in glass beakers. Beakers were covered with Parafilm to avoid evaporation of the simulant during the contact period and kept in a thermostatically controlled chamber (Binder model KBF) at 40 ± 0.5 °C for 10 days. For iso-octane the temperature/time of plastic/simulant contact was 20 ± 0.5 °C for 2 days (14). The film samples were then removed, and the simulant was placed in a 250-mL preweighed round-bottom flask and evaporated on a rotary evaporator with distilled water in the heating bath. The round-bottom flask containing the residue of evaporation was kept in a thermostatically controlled chamber at 105 ± 1.0 °C for 1 h followed by 1 h in a desiccator and then weighed. An analytical balance Sartorius model BP221S capable of weighing to 0.1 mg was used. The overall migration was calculated in milligrams per square decimeter of film surface area taking into account the exposed surface area of the test sample and in milligrams per liter of simulant. Blank samples were run simultaneously, and corrected migration values were calculated for each simulant. For each plastics sample three determinations were performed in duplicate, and reported migration value was the mean of six determinations.

Permeability Measurements. Oxygen transmission rates for all films were measured using the Oxtran 2/20 oxygen permeability tester (Mocon Controls) at relative humidity (RH) = 60%, *T* = 23 °C and were expressed as mL/m²·day·atm. Water vapor transmission rates were measured using the Permatran W 3/31 water vapor permeability tester at RH = 100% and *T* = 23 °C and were expressed as g/m²·day. CO₂ permeation rates were measured using the Permatran C-200 CO₂ permeability tester at RH = 0%, *T* = 23 °C, and expressed as mL/m²·day·atm. For each plastics sample three determinations were performed in duplicate, and reported permeability value was the mean of six determinations.

Measurement of Mechanical Properties. Tensile strength, percent elongation at break, and Young's modulus were measured on an Instron model 4411 universal testing machine according to the D-882 ASTM official method (17) under the following conditions: Film sample dimensions were 200 mm × 15 mm, cross head speed was 500 mm/min, and initial grip distance was 50 mm. For each plastics sample 10

determinations were performed, and reported mechanical property value was the mean of 10 determinations.

Organoleptic Evaluation. Extracts of films were made and given to a test panel to detect changes in odor and taste when compared to the control solution. Film extracts were prepared according to the NBN S29-001 standard (18). A specimen of the film (1 dm²) was submerged in an NaHCO₃ solution (0.042%) for 24 h and stored in the dark at room temperature. The films were then placed in fresh solution and stored again under the same conditions; this was repeated once more. The third solution comprises the test solution. Dilutions of 1:1, 1:2, and 1:3 were prepared. Normally the first contact solution is rejected, but in the present case all solutions including the first were considered for organoleptic evaluation. The test panel received two numbered samples—a control and a test solution—and additionally a flask with the control. All solutions were evaluated and recorded as follows: A perceived difference was assigned as 1, whereas no difference was assigned as 0.

RESULTS AND DISCUSSION

Overall Migration Testing. Overall migration values for all 12 experimental films are given in **Table 1**. On the basis of these values the following observations can be made: (a) the percentage of recycled LDPE in the multilayer structure does not affect (*p* > 0.05) overall migration values in all three food simulants; (b) overall migration values for a given packaging material increase in the order water < 3% acetic acid < iso-octane; (c) overall migration values drastically increase for the coextruded/laminated samples (three last samples) in the presence of 3% acetic acid; (d) migration values in all cases are lower than the upper acceptable limit (10 mg/dm²) set by the EU (19).

Observation a can be justified in terms of the composition of the recycled LDPE layer in conjunction with the functional barrier hypothesis. This type of scrap (preconsumer scrap) is produced “in house” by the food-packaging converter as a “waste” of the blown film extrusion process and is thus a material that the manufacturer has complete control over. Having thus eliminated the possibility of scrap contamination and by introducing in parallel a layer of virgin material between the packaged food and the recycled layer, it is not surprising that

Table 2. Transmission Rates of O₂, CO₂, and Water Vapor through Various Experimental Films

material ^a	film type	transmission rate		
		PO ₂ ^b (cm ³ /m ² ·day·atm)	PCO ₂ ^c (cm ³ /m ² ·day·atm)	PH ₂ O ^d (g/m ² ·day)
LDPE/LDPE–LLDPE*/LDPE 100% virgin/LDPE–LLDPE*/LDPE	A	2822 ± 181 ^e	11288 ± 424	0.54 ± 0.02
LDPE/LDPE–LLDPE*/LDPE 50–50% virgin–recycled/LDPE–LLDPE*/LDPE	A	2555 ± 128	9891 ± 482	0.51 ± 0.02
LDPE/LDPE–LLDPE*/LDPE 100% recycled/LDPE–LLDPE*/LDPE	A	2677 ± 103	11779 ± 515	0.56 ± 0.03
PA/tie/LDPE 100% virgin/LDPE/LDPE	B	26.9 ± 1.4	108 ± 4	1.48 ± 0.06
PA/tie/LDPE 50–50% virgin–recycled/LDPE/LDPE	B	28.9 ± 1.8	117 ± 7	1.36 ± 0.07
PA/tie/LDPE 100% recycled/LDPE/LDPE	B	25.3 ± 2.0	107 ± 6	1.27 ± 0.08
PA/EVOH/tie/LDPE 100% virgin/LDPE	C	1.33 ± 0.07	5.6 ± 0.3	1.10 ± 0.05
PA/EVOH/tie/LDPE 50–50% virgin–recycled/LDPE	C	1.26 ± 0.08	5.3 ± 0.3	1.09 ± 0.04
PA/EVOH/tie/LDPE 100% recycled/LDPE	C	1.19 ± 0.04	5.1 ± 0.2	1.10 ± 0.04
metPET//LDPE–LLDPE*/LDPE 100% virgin/LDPE–LLDPE*/LDPE	D	1.12 ± 0.04	4.5 ± 0.2	0.24 ± 0.02
metPET//LDPE/LDPE–LLDPE*/LDPE 50–50% virgin–recycled/LDPE–LLDPE*/LDPE	D	1.18 ± 0.06	4.9 ± 0.3	0.12 ± 0.01
metPET//LDPE/LDPE–LLDPE*/LDPE 100% recycled/LDPE–LLDPE*/LDPE	D	0.97 ± 0.04	4.3 ± 0.3	0.15 ± 0.02

^a / = coextruded; // = laminated; tie = special adhesive resin for coextrusion; * LDPE–LLDPE = blend consisting of 70% LDPE and 30% LLDPE. ^b RH = 60%, T = 23 °C. ^c RH = dry, T = 23 °C. ^d RH = 100%, T = 23 °C. ^e Mean ± SD.

overall migration values from structures containing the scrap LDPE did not differ from those containing the virgin LDPE.

Observation b can be interpreted in terms of the chemical affinity of the nonpolar iso-octane to the also nonpolar polyethylene comprising >50% of the material in each structure.

Observation c can be probably related to the inefficient lamination of the metallized PET to the coextruded structure permitting aqueous acetic acid to wet the metallized (buried) layer of the PET films, causing delamination and partial destruction of the aluminum layer. This was apparently the case in the present study after 3% acetic acid/film contact for 10 days at 40 °C, resulting in a drastic increase in overall migration values. Even in this exceptional case overall migration values were lower than the upper acceptable limit for migration (10 mg/dm²) set by the EU.

In similar migration work, Begley and Hollifield (4) reported that a two-layer LDPE package construction, that is, an outer layer of recycled material and an inner layer virgin material, appears to offer significant advantages from a migration standpoint for the use of recycled polymers in food packaging over a single layer of packaging material in the case of specific contamination of such a material. On the basis of data presented by these authors the initial concentration of a contaminant in a single layer of LDPE could not exceed 2.5 μg/cm³; otherwise, the food would become contaminated above the dietary threshold value (3 ppb), whereas the respective value for a two-layer LDPE (recycled/virgin) material would be 300 μg/cm³. In other words, the virgin food-contacting LDPE layer in a two-layer construction adds a safety factor with regard to acceptable migration by ~100–120 times. This is the case with film type C in the present work. In film types A, B, and D two virgin LDPE layers separate the scrap layer from the food-contact medium, providing an additional safety factor.

Franz et al. (5) studied overall migration from three-layer coextruded polypropylene cups containing a middle layer of recycled PP into 3% acetic acid and 35 and 80% ethanol and found no statistically significant differences between control and test specimens after contact for 10 days at 40 °C. Further testing involving redissolution of overall migration residues (into 80% ethanol) and subsequent GC analysis showed practically the same peak profile both for the “virgin” material cup and for the cup containing the recycled material.

Incarnato et al. (12) studied overall migration from recycled

PP containers in 15% ethanol and 3% acetic acid and reported comparable overall migration values (2–3 mg/dm²) between containers made of virgin and recycled HMW or LMW polypropylene. The same authors, however, reported significantly higher migration values from containers made of recycled LMW PP into iso-octane, as compared to those from virgin LMW PP containers.

Lox et al. (3) studied overall migration from experimental PP films containing 20–60% recycled PP into water, 3% acetic acid, 15% ethanol, and olive oil and found no effect of the percentage of scrap on the migrational behavior of the PP films.

Finally, Devlieghere et al. (20) studied migration of first-generation recycled HDPE into distilled water, 15% ethanol, 3% acetic acid, 95% ethanol, and iso-octane and reported that migration values (between 0.01 and 9.3 mg/dm²) of the recycled and virgin materials were comparable. Even in fatty food simulants such as 95% ethanol, overall migration of the recycled HDPE seemed to be equal and in fact a little lower than the migration of the virgin polymer.

Permeability Measurements. Transmission rate values to O₂, CO₂, and water vapor for all experimental films are given in Table 2. Experimental film structures were chosen to produce low-barrier (all purpose), barrier, and high-barrier packaging materials. On the basis of the present permeation values, the following observations can be made: (a) the percentage of recycled LDPE in the multilayer structure does not affect (*p* > 0.05) the permeability of experimental films to O₂, CO₂, and water vapor; (b) as expected, the use of PA in film type B significantly increased the barrier properties of the multilayer structure, whereas the use of EVOH and metallized PET in film types C and D further enhanced the barrier properties of the multilayer structure; (c) oxygen as well as water vapor permeation values of laminated structures show significant discrepancies, which can be directly related to insufficient lamination of the metallized PET film to the multilayer coextruded structure; (d) barrier properties to O₂, CO₂, and water vapor comparable to those of EVOH were obtained when using met PET.

Miltz et al. (10) studied gas barrier properties of three-layer PET structures containing a buried layer of recycled PET and reported retention of gas permeability properties of multilayer structures when proper processing conditions were used.

Mechanical Properties. Results on tensile strength, percent

Table 3. Tensile Properties of Experimental Films

material ^a	film type	tensile strength (MPa)		% elongation at break		Young's modulus (MPa)	
		md ^b	cd ^c	md	cd	md	cd
LDPE/LDPE-LLDPE*/LDPE 100% r/LDPE-LLDPE*/LDPE	A	20.2 ^d ± 1.0	17.4 ± 1.1	268 ± 31	708 ± 51	184 ± 23	195 ± 5
LDPE/LDPE-LLDPE*/LDPE 50-50% virgin-recycled/LDPE-LLDPE*/LDPE	A	20.3 ± 1.4	15.8 ± 0.8	302 ± 37	632 ± 36	187 ± 20	227 ± 25
LDPE/LDPE-LLDPE*/LDPE 100% recycled/LDPE-LLDPE*/LDPE	A	21.0 ± 1.5	15.0 ± 1.1	329 ± 41	618 ± 44	195 ± 20	220 ± 24
PA/tie/LDPE 100% virgin/LDPE/LDPE	B	23.6 ± 0.9	20.7 ± 2.3	303 ± 12	352 ± 46	256 ± 36	313 ± 33
P/tie/LDPE 50-50% virgin/LDPE/LDPE	B	25.6 ± 0.9	19.9 ± 1.2	358 ± 20	333 ± 26	271 ± 27	306 ± 19
PA/tie/LDPE 100% recycled/LDPE/LDPE	B	23.7 ± 2.6	19.5 ± 0.9	332 ± 14	331 ± 10	283 ± 15	297 ± 13
PA/EVOH/tie/LDPE/100% virgin/LDPE	C	25.7 ± 1.8	22.4 ± 1.1	387 ± 31	380 ± 15	254 ± 14	241 ± 12
PA/EVOH/tie/LDPE 50-50% virgin-recycled/LDPE	C	27.5 ± 1.5	20.2 ± 0.6	379 ± 24	341 ± 13	281 ± 11	285 ± 29
PA/EVOH/tie/LDPE 100% recycled/LDPE	C	26.9 ± 1.4	19.0 ± 0.8	378 ± 23	328 ± 16	303 ± 34	281 ± 29
metPET//DPE/LDPE-LLDPE*/LDPE 100% virgin/LDPE-LLDPE*/LDPE	D	30.2 ± 2.9	31.9 ± 3.9	61.7 ± 4.2	52.3 ± 5.6	888 ± 30	924 ± 62
metPET//LDPE-LLDPE*/LDPE 50-50% virgin recycled/LDPE-LLDPE*/LDPE	D	30.6 ± 2.7	32.5 ± 5.3	60.3 ± 3.6	62.1 ± 9.2	903 ± 31	937 ± 127
metPET//LDPE-LLDPE*/LDPE 100% recycled/LDPE-LLDPE*/LDPE	D	31.6 ± 3.1	30.0 ± 3.2	63.6 ± 5.4	45.3 ± 4.3	861 ± 23	944 ± 42

^a / = coextruded; // = laminated; tie = special adhesive resin for coextrusion; * LDPE-LLDPE = blend consisting of 70% LDPE and 30% LLDPE. ^b Machine direction. ^c Cross direction. ^d All values for tensile strength and percent elongation at break are the mean of 10 readings ± SD. Cross head speed = 500 mm/min. Initial grip distance = 50 mm.

elongation at break, and Young's modulus values for all experimental films are given in **Table 3**. On the basis of these values the following observations can be made: (a) no statistically significant differences were recorded between structures containing virgin and recycled LDPE, which may be attributed to the quality of "in house" scrap used in this study; (b) as expected, incorporation of PA in the film structure increases all three parameter values. This effect regarding tensile strength and Young's modulus is especially pronounced in laminated to PET structures, given the excellent mechanical properties of PET. At the same time, percent elongation at break drastically decreases in the presence of PET (three last films).

Devlieghere et al. (20) studied the mechanical properties of recycled HDPE milk bottles that were used to blow-mold new bottles and reported insignificant differences in tensile strength and percent elongation at break between bottles made from recycled and virgin material. A slight decrease in Young's modulus values was observed in the virgin material as compared to the recycled material.

Tzankova Dintcheva et al. (21) studied the mechanical and rheological properties of a recycled blend of LDPE-LLDPE reprocessed into low-pressure pipe and reported comparable values for Young's modulus and percent elongation at break between pipe samples made from recycled and virgin material, respectively. This was achieved by the suitable addition of antioxidants, inert fillers, and modifier agents. Tensile strength values, however, of the recycled pipe remained below the respective value of the pipe made from virgin material.

La Mantio and Scaffaro (11) studied the mechanical and rheological properties of monolayer blends of virgin and recycled PET and reported that the above properties remain very close to those of the virgin material provided that careful drying is carried out before any melt operation. Incarnato et al. (12) studied the mechanical properties of recycled PP containers made of low molecular and high molecular weight PP with and without the addition of a stabilizer. These authors reported no significant differences in tensile strength and Young's modulus in injection molded PP containers after first and second reprocessing cycles; however, a decrease in percent elongation

at break in recycled containers was found, which was attributed to a decrease in molecular weight occurring during recycling. They also reported higher tensile strength, percent elongation at break, and Young's modulus values for recycled containers made from HMW PP as compared to those made of LMW PP.

Pattanakul et al. (22) similarly reported that percent elongation at break is the mechanical property most affected by degradation occurring during HDPE recycling.

Organoleptic Evaluation. Organoleptic evaluation was carried out on the basis of the rationale that potential volatile compounds migrating out of the plastic material would be impossible to determine in the overall migration experiment. Moreover, the threshold value of these substances is so low that only small amounts need to be present in order to be detected by sensorial testing.

Organoleptic evaluation (results not shown) on odor and taste gave no statistically significant differences ($p > 0.05$) between control and film samples containing 50 and 100% recycled LDPE in the middle layer. This finding supports the functional barrier hypothesis and can be attributed to the high quality of the scrap LDPE resin, over which the manufacturer had complete control in terms of its composition and its history.

Devlieghere et al. (20) investigated the possibility of recycling milk-contaminated HDPE bottles. New bottles were blown from untreated, caustic-washed, and caustic wash/steam-stripped/air-dried recycled material. Sensorial results showed that untreated and caustic-washed recycled material could not be used as a food-packaging material because a large number of compounds migrated into various food simulants tested, causing a distinct off-flavor. However, the steam-stripped/air-dried recycled material showed excellent sensorial properties and could not be distinguished from the respective virgin material.

Lox et al. (3) worked with blends of virgin and first-generation recycled PP and reported that PP scrap in amounts from 5 to 20-30% did not affect the organoleptic properties of a food-contacting phase. The difference between this finding and that reported in the present work can be interpreted by the differences in structure of the test films; that is, monolayer

versus multilayer, and of course the application of the functional barrier hypothesis.

Conclusion. From the present results it is clear that primary LDPE scrap may be used in multilayer structures as a buried layer comprising 40–50% bw of the composite film sample without any compromise in migrational, barrier, mechanical, and organoleptic properties.

ACKNOWLEDGMENT

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ABBREVIATIONS USED

LDPE, low-density polyethylene; PA, polyamide; EVOH, ethylene vinyl alcohol; PP, polypropylene; PET, poly(ethylene terephthalate); PS, polystyrene; HDPE, high-density polyethylene; HMW, high molecular weight; LMW, low molecular weight; tie, tie layer (special adhesive layer for coextrusion); adh, adhesive layer.

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