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RESEARCH ARTICLE

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Feasibility of incorporating recycled multilayer LDPE/EVOH films into flexible packaging

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Abstract

Multilayer films are widely used in the food industry because they allow the combination of properties of different materials to increase product shelf life without sacrificing product value. Despite the associated benefits, these films are difficult to recycle due to material immiscibility and difficult layer separation. Thus, the purpose of this research is to investigate the feasibility of incorporating recycled plastic material from the multi-material film industry into extruded films for food packaging. To simulate the thermomechanical recycling, two different post-industrial polymeric wastes of low-density polyethylene (LDPE) (with linear low-density polyethylene (LLDPE) or with ethylene vinyl alcohol (EVOH)), were reprocessed four times in a single-screw extruder. Moreover, these polymeric wastes were blended with virgin LDPE to improve the extrusion process control and to assess their effect on the blend properties. The extrusion experiments demonstrate that reprocessing of multilayer films and processing of their blends with virgin LDPE is feasible. Moreover, the mechanical and thermal properties of the corresponding films are not negatively affected when compared with those of virgin LDPE films. Nevertheless, the optical properties of recycled EVOH films, were lower than those of virgin LDPE or recycled LLDPE, with high haze and low gloss.

Highlights

- Two multilayer film scraps were reprocessed and blended with virgin LDPE.
- Thin film samples were produced by blown film extrusion.
- Films containing EVOH presented the worst optical properties.
- The presence of EVOH did not negatively affect the extrusion process.
- Mechanical and thermal properties were not significantly affected.

KEYWORDS

blown film, packaging, recycled multilayer film

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1 | INTRODUCTION

In recent years, the packaging sector has been dominant in the production of primary plastics and, consequently, also dominant in the production of plastic waste.[1,2] Despite the decrease of packaging deposited in landfills and the increase in recycling, more than 18% of this waste still ends in landfills.[2] These packages can be of different types and made in a variety of ways; however, this study focuses only on food packages made of flexible films.

Plastic film packages may combine different types of materials in coextruded or laminated films, improving its aesthetic, functional, mechanical, and/or barrier properties; however, it becomes difficult to recycle them.[3,4]

Polymers, such as ethylene vinyl alcohol (EVOH), polyamide (PA), and polyvinylidene chloride (PVDC), are often used in the inner layers of flexible food packaging, made up mostly of polyethylene (PE), to improve the mechanical and oxygen barrier properties of the package.[5-9] Although advantageous, the combination of PE with polymers normally used for oxygen barrier in multilayer films is extremely critical in terms of recycling cost.[10,11] The chemical structure of these polymers differs in polarity making them incompatible.[12] For this reason, the use of a compatibilizer between the PE and EVOH layers is always necessary, with these films requiring a minimum of five layers: PE/tie layer/EVOH/tie layer/PE.[13] When recycling thermomechanically, this incompatibility causes poor adhesion between phases, resulting in a recycled material with lower properties and less added value.[14,15] Despite the incompatibility of these materials, recent studies have shown that the tie layer used to bond PA and PE in the film serves as an effective compatibilizer throughout the recycling process.[16]

The use of recycled plastic packaging not only mitigates its adverse impact on the environment but also reduces the need for its eventual incineration. Several studies have been developed for this purpose, to evaluate the properties of plastic films with the incorporation of different percentages of recycled material, to ensure the maintenance of the packaging quality for the food industry.[17,18]

The study of Turriziani et al. (2023) evaluated the mechanical properties of two different recycled materials from multilayer films (A and B), with 5 and 7 wt.% of polyolefin compatibilizer with maleic anhydride (PCA-MA). The samples were produced by blown film extrusion with a thickness of 50 μm. Recycled films with PCA-MA exhibited greater elongation at break and tensile strength than films without the compatibilizer, but only in the transverse direction. This study demonstrates that while the compatibilizer offers advantages for the mechanical properties of the film, the recycled materials without it also exhibit good mechanical properties compared with virgin PE.[8]

Meran, Ozturk, and Yuksel (2008) investigated the change in mechanical properties of samples with varying percentages of postconsumer recycled material compared with the corresponding virgin materials. Tensile strength decreased by 35.5% in the sample with 100% recycled LDPE, compared with the sample without recycled material, but only 11.9% in the sample with 20% recycled.[19] Another significant outcome from this study was that these recycled materials did not cause any problems during processing. Studies carried out with samples of multilayer blown films showed different results. According to Chytiri et al. (2006) the introduction of different percentages of recycled material (0%, 50%, and 100%) in the inner layer of a five-layer LDPE and LLDPE film, did not affect the mechanical properties of the film.[20] These findings could be explained by the fact that the layer containing recycled material is inserted between two layers of virgin material.

Most of the studies found in the literature presented a mechanical recycling with a single material, without exploring the recycling of multilayer films with different materials. Additionally, the characterized samples are not frequently produced by industrial techniques of packaging production.[21–23]

The motivation for the development of the current research stems from the need to achieve scientific knowledge in the plastic film sector, particularly for the food market, aiming to promote the production of high-performance packaging with the incorporation of recycled material to reduce the deposition of plastic waste in landfills.

In this study, plastic films for the development of new packages were produced using tubular film extrusion technique. This processing technique enables the production of films with some biaxial orientation and low thickness, widely employed in the industry for manufacturing food packaging.[24] With the samples produced by this processing technique, it was possible to evaluate the properties of two different types of recycled multilayer films with different materials. Moreover, the effect of repeated thermomechanical reprocessing on these recycled materials was also assessed and compared with the film produced using a typical reference material for flexible film production, such as LDPE. Due to the increase in the standard deviation in the tensile tests results, and the increase in haze in the recycled EVOH films, mixtures were made with virgin and recycled material. The incorporation of a fixed percentage of virgin polymer in the recycled material can also improve film reproducibility in the industrial process, minimizing composition variations.

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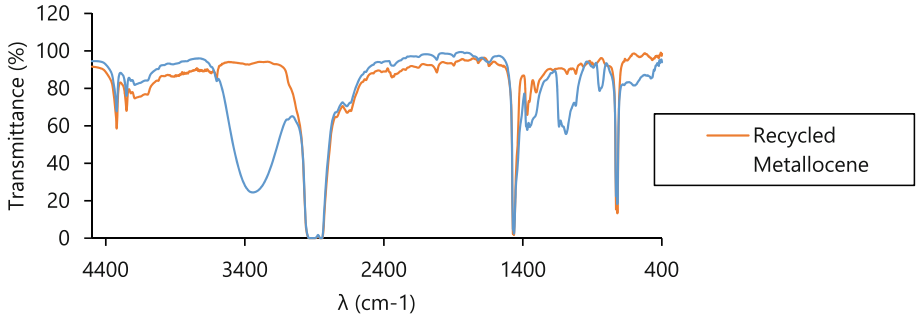

FIGURE 1 FTIR spectra of 'recycled Metallocene' and 'recycled EVOH'.

The present study focuses on exploring the modification and preservation of the properties of recycled (postindustrial) materials, while evaluating their re-processability and blending with virgin LDPE, for potential integration into food packaging films.

2 | EXPERIMENTAL

2.1 | Materials

Dow LDPE 352 E, a low-density polyethylene blown film extrusion grade (density = 925 kg/m^3, melt flow index (MFI) = 2.0 g/10 min (190°C/2.16 kg) and Vicat softening temperature of 96.0°C), was used as the virgin polymer in the blend with recycled materials. The film producer company involved in this research, Vizelpas, supplied the recycled materials recovered from multimaterial multilayer films used in two food packages made with different materials, namely, 'recycled Metallocene' – consisting of LDPE, butene LLDPE and metallocene LLDPE; and 'recycled EVOH' – consisting of LDPE, butene LLDPE, metallocene LLDPE, and polyethylene grafted maleic anhydride (PE-g-MA) as adhesive and EVOH.

To characterize the composition of the materials recovered from industry, samples were analyzed by Fourier-transform infrared spectroscopy (FTIR) and differential scanning calorimetry (DSC). The EVOH content in the 'recycled EVOH' was determined by the dissolution of the soluble fraction (PE) in xylene. The experimental protocols used are described in Section 2.2.

The FTIR spectra of the 'recycled Metallocene' and 'recycled EVOH' are shown in Figure 1. The spectrum of the 'recycled Metallocene' reveals peaks at 1466 and 721 cm^{-1}, characteristic of scissoring and rocking vibrations of CH_2 groups, respectively, present in the chemical structure of PE.[25,26] The spectrum of recycled EVOH shows peak at 3338 cm^{-1}, characteristic of O-H bond present in the chemical structure of EVOH, as well as peaks at 1466 and 721 cm^{-1} (related to PE structure) as in the previous case.

TABLE 1 Sample codes of recycled reprocessed materials.

Sample code	Recycled reprocessed materials
1 M	1st 'recycled Metallocene'
2 M, 3 M, 4 M, 5 M	2nd, 3rd, 4th and 5th reprocessing of 'recycled Metallocene'
1E	1st' recycled EVOH'
2E, 3E, 4E, 5E	2nd, 3rd, 4th and 5th reprocessing of 'recycled EVOH'

DSC results indicate that recycled Metallocene has two melting peaks at 109.5 and 126.9°C, corresponding to the melting temperature of LDPE and LLDPE, respectively. For recycled EVOH two melting peaks at 111.7 and 183°C were observed, corresponding to the melting temperatures of LDPE, and EVOH, respectively. The first peak on the DSC of recycled EVOH shows a shoulder to the right of the LDPE melting peak indicating the presence of another polymer with similar melting temperature, such as LLDPE.

After dissolution of LDPE, LLDPE, and PE-g-MA in xylene, 9.0 wt.% EVOH content was obtained.

These recycled materials were used for the production of blends with virgin LDPE and for the production of pellets with four additional reprocessing cycles, obtained through filament extrusion and subsequent pelletizing.

2.2 | Reprocessing and films production

The supplied recycled material was reprocessed four additional times, resulting in a total of five reprocessing cycles, to evaluate the evolution of mechanical, optical, and rheological properties as the number of reprocessing cycles increased. The sample codes used for these samples are listed in Table 1.

The reprocessing was carried out in a PeriPlast single-screw extruder with a screw diameter (D) of 20 mm and L/D = 20, coupled to a filament extrusion die. The recycled Metallocene was reprocessed at 180°C for all

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heating zones, while the recycled EVOH, was reprocessed with a temperature profile of 200 – 230 – 220°C, given the EVOH melting temperature, both at a screw speed of 40 rpm. The filament of the reprocessed materials was cooled by forced air convection and subsequently pelletized. These pellets were dried at 60°C during 4 h before being processed on the blown film extrusion line.

The reprocessed materials were produced in a tubular film extrusion line equipped with a Periplast single-screw extruder with D = 25 mm and L/D = 25. The extrusion head attached to this equipment terminates with an annular die with a diameter of 50 mm and a 1.25 mm gap. In all the cases, a temperature profile of 180 – 200°C – 220°C in the extruder, 220°C in the extrusion head, a screw speed of 50 rpm, blow-up ratio (BUR) of 1.8 and a take-up ratio (TUR) of 11, were employed.

For the production of films with incorporation of recycled materials, 20, 40, and 50 wt. % of virgin LDPE granules were mixed with 80, 60, and 50 wt. %, respectively, of the recycled Metallocene and the recycled EVOH in a rotating drum mixer, as described in Table 2.

These blends were utilized for film production via extrusion, as described above. The quality of the films produced was assessed through tensile, haze, and brightness tests to evaluate the impact of incorporating virgin LDPE.

The thickness of the films was measured using a micrometer (Digimatic thickness gauge 547–301, Mitutoyo, Japan) with a measuring accuracy of 0.01 mm. The thickness of all the produced films was 50 ± 10 μm.

TABLE 2 Sample codes of recycled blends produced with incorporation of virgin LDPE.

Sample code	Recycled Metallocene (wt.%)	Recycled EVOH (wt.%)	LDPE (wt. %)
50% M + 50% LDPE	50		50
60% M + 40% LDPE	60		40
80% M + 20% LDPE	80		20
50% E + 50% LDPE		50	50
60% E + 40% LDPE		60	40
80% E + 20% LDPE		80	20

2.3 | Characterization techniques

2.3.1 | FTIR

The FTIR spectra of the recycled blends were obtained using the Jasco FT/IR-4100 equipment.

2.3.2 | Determination of EVOH content

To determine the EVOH content (insoluble fraction) two samples weighing 0.1007 g and 0.0948 g of recycled EVOH were placed on a 120-mesh wire pouch, previously weighed, that was fold to form a cage. The content in the wire cage was dissolved in 100 mL of refluxing xylene at 46°C during 12 h. After dissolution, the pouch was placed in a vacuum oven at 150°C and then weighed to calculate the percentage of EVOH. This method is an adaptation of the method used for the determination of gel content of ethylene plastics present in the ASTM D2765-16 standard.

2.3.3 | DSC

For the DSC test one pellet (4–5 mg) from each sample was placed in aluminium crucibles with pierced lid under a nitrogen flow rate of 50 mL/min using a DSC Netzsch equipment. The procedure was repeated three times for each material. The test was performed from 20 to 230°C at heating rate of 10°C/min and cooled down at rate of 20°C/min. Each sample was subjected to two heating–cooling cycles, as recommended in ISO 11357-1. The melting and crystallization temperatures of the different materials were determined from the 2nd heating endothermic peak, and first controlled cooling exothermic peak, respectively.

2.3.4 | XRD

X-Ray diffraction measurements were carried out on film samples at 23 ± 2°C using a diffractometer, Bruker AXS D8 Discover, equipped with a CuKα generator (λ = 1.5404 Å) operating at 40 kV and 40 mA. Measurements were conducted in a 2θ range from 5° to 60° with a 0.02° step and a counting time of 1 s per step. The crystallinity of the samples was calculated from their scans by dividing the area of the crystalline peaks by the total area under the diffraction curve, using the Bruker AXS DIFFRAC.EVA V4.2.2 software.

2.3.5 | SEM

Scanning Electron Microscopy analysis was carried out on film samples coated with gold, fractured in liquid nitrogen, using a Nano SEM – FEI Nova 200 equipment.

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2.3.6 | MFI

The MFI of reprocessed granules from 'recycled Metallocene' and 'recycled EVOH' were carried out in a MFI Daventest equipment at temperatures of 190, 210 and 230°C with a 2.16 kg weight, according to ISO 1133-1.

2.3.7 | Tensile tests

The tensile tests of the films produced with reprocessed recycled material and the films produced with recycled material incorporating virgin LDPE were performed using a ZWICK universal mechanical testing equipment with a load cell of 5kN, at a speed of 50 mm/min, at 23 ± 2°C. From the extruded film bubble, five specimens 25-mm wide and 160-mm long, were collected in accordance with ASTM D882-10 in the machine direction (MD) and in the transverse direction (TD).

2.3.8 | Optical properties

The haze of the films was determined using a XL-211 Hazegard System transmittance meter, according to ASTM D1003-00. Five measurements were performed for each sample.

Gloss measurements of the films were analyzed according to ISO 2813 standard using a BYK Micro-TRI-gloss S gloss meter, with 20° and 60° measurement geometries.

2.4 | Statistical analyses

The results of tensile tests and haze measurements are presented as mean ± standard deviation. Statistical analysis was performed using IBM SPSS Statistics software, employing one-way analysis of variance (ANOVA) with comparison procedures assuming equal variances with Tukey method for differences of means, with a confidence level of 95%.

3 | RESULTS AND DISCUSSION

3.1 | Reprocessing study

As can be seen in Figure 2, and corresponding Table S1, the melting and crystallization temperatures of the first and fifth reprocessing cycles of recycled Metallocene and EVOH are similar. Additionally, a shoulder attenuation in LDPE melting peak from sample 1E to 5E, can be observed in the DSC curves, which may indicate improved homogenization of this recycled material after reprocessing.

From the analysis of the MFI results depicted in Figures 3 and 4 and in Table S2, it can be noticed that material's fluidity is almost unaffected by the successive reprocessing cycles (from 1 to 5) for the recycled EVOH, only the results of the tests at 230°C show slightly differences among the recycled cycles, increase until the third and then decreases. This can be due to thermal degradation during the MFI test, since the test has a preheating

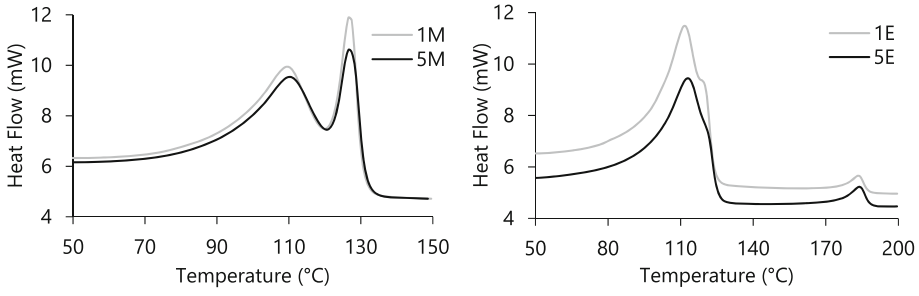

FIGURE 2 DSC second heating curves of the first and fifth reprocessed recycles.

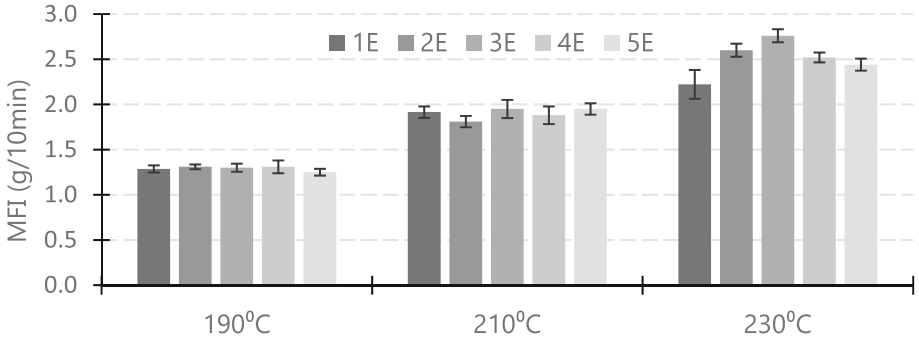

FIGURE 3 MFI of reprocessed recycled EVOH (E).

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of 5 min, at this temperature, thermal degradation might occur. Due to tertiary carbons present in EVOH, degradation occurs mainly through chain scission rather than crosslinking, which explains the increase of MFI as the recycled cycle increases. For recycled Metallocene, at all tested temperature, there is a slight decreasing trend in the MFI as the number of reprocessing cycles increases, which can be associated with a slight crosslinking of LDPE and LLDPE, well-known effect for reprocessing recycled PE 27. Nevertheless, it is not possible to associate the cross-link reaction with LDPE or LLDPE since this is a recycled material and the composition is not known. The consistent rheological performance is of great importance as it allows to keep the processing conditions throughout the reprocessing cycles.

The WAXD spectra for the first and fifth reprocessed recycled materials and virgin LDPE film samples are depicted in Figure 5. All samples exhibit three main crystalline diffraction peaks characteristics of PE at 2θ of 21.5°, 23.7°, and 36.2°, corresponding to the 110, 200 and 020 lattice planes of the orthorhombic crystal lattice, respectively.[27-29] The characteristic EVOH diffraction peaks mentioned in other research works do not appear in any of the EVOH recycles. Since the characteristic diffraction peak of EVOH is at 21.24°, this peak is overlaid by the high intensity peak of LDPE, which hinders a clear distinction of the individual peaks.[30,31]

Table 3 shows that the amount of EVOH present in the 'recycled EVOH' (9 wt.%) did not change the crystalline form of PE, a fact that was also observed by Du et al. (2004).[29] On the contrary, the recycled Metallocene has slightly decrease in the degree of crystallinity, which might be associated with the cross-linking reactions.

SEM micrographs of recycled EVOH samples in Figure 6 reveal circle and elongated particles of EVOH detached from the PE matrix, which demonstrate poor adhesion between PE and EVOH. Films with recycled Metallocene exhibit a homogeneous morphology.

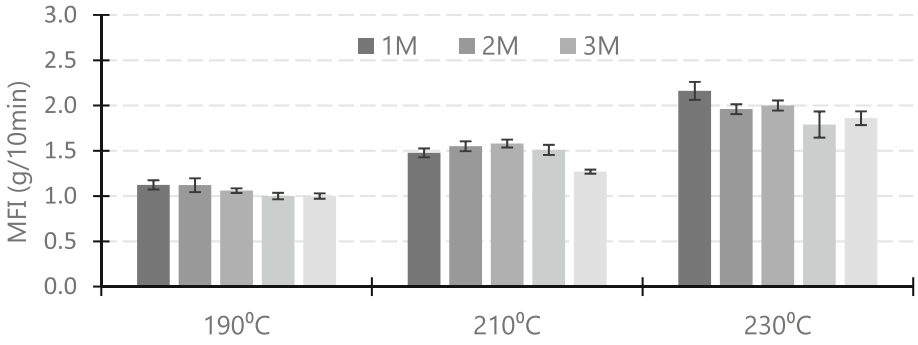

FIGURE 4 MFI of reprocessed recycled Metallocene (M).

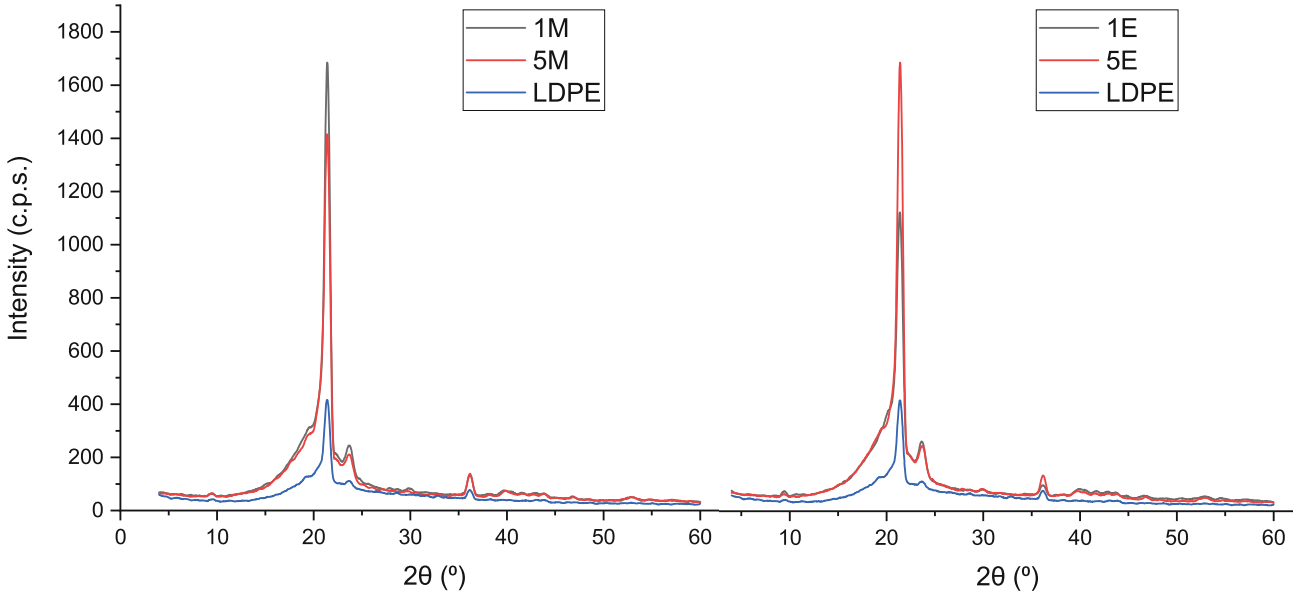

FIGURE 5 On the left, WAXD spectra for the 1st and 5th reprocessed recycled Metallocene and LDPE and on the right WAXD spectra for the 1st and 5th reprocessed recycled EVOH and LDPE.

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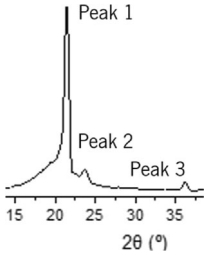
The mechanical properties (Figures 7 and 8, and Tables S3 and S4) of the films produced with reprocessed material generally demonstrate that all the reprocessed systems have better mechanical properties than virgin LDPE, including higher Young's modulus, maximum stress, and strain at break. This improvement is attributed to the presence of materials with higher mechanical performance in the recycled films. The properties of the recycled Metallocene improve with increasing cycles, which might be related to a progressive better homogenization of the materials present in the recycle, as already discussed in the DSC results, and possibly to a

TABLE 3 Diffraction peaks and crystallinity of the 1st and 5th reprocessed cycles and virgin LDPE.

	Peak 1		Peak 2		Peak 3		Crystallinity
Samples	**2θ (°)**	**Intensity (c.p.s)**	**2θ (°)**	**Intensity (c.p.s)**	**2θ (°)**	**Intensity (c.p.s)**	**(%)**
1 M	21.46	2010	23.82	287	36.12	158	54.1
5 M	21.46	1656	23.66	231	36.06	161	51.7
1E	21.36	1322	23.74	283	36.20	137	54.0
5E	21.40	1008	23.58	270	36.16	98	53.8
LDPE	21.42	481	23.64	131	36.18	102	39.5



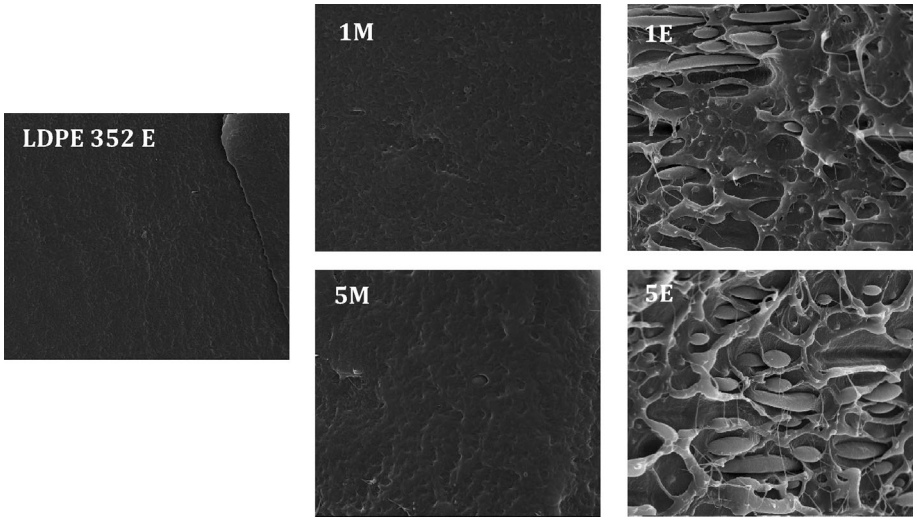


FIGURE 6 SEM micrographs, with 15,000x magnification, of virgin LDPE and recycled films.

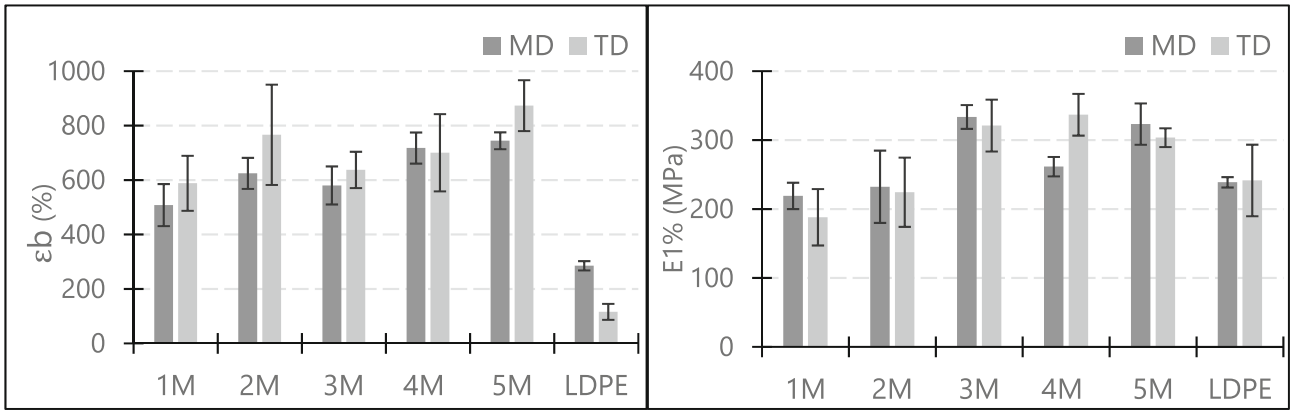


FIGURE 7 Strain at break (ε_b) and Young's Modulus ($E_{1\%}$) of films with recycled Metallocene.

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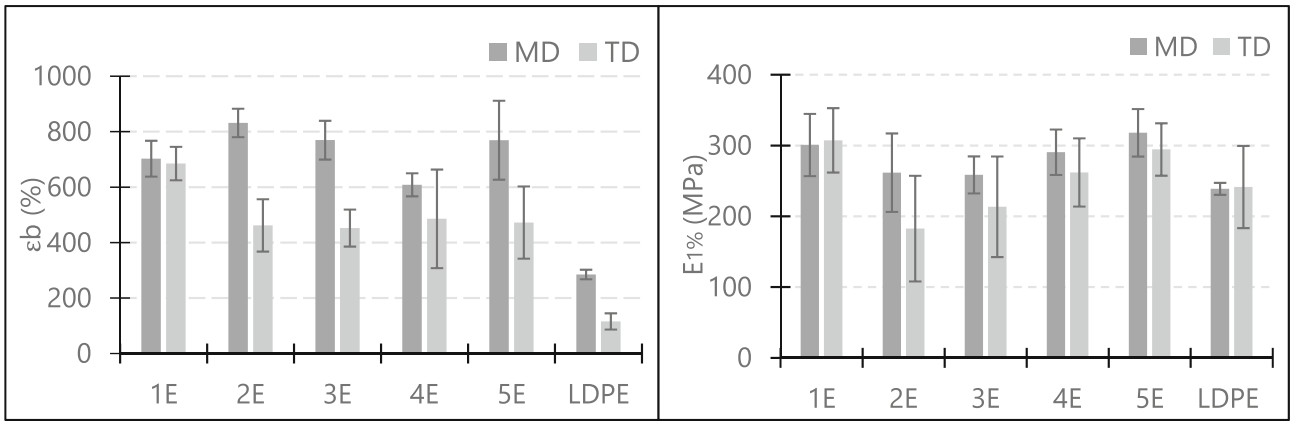
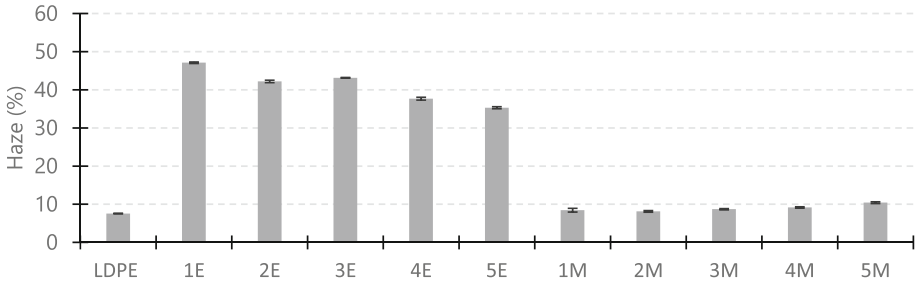


FIGURE 8 Strain at break (ε_b) and Young's Modulus ($E_{1\%}$) of films with recycled EVOH.

FIGURE 9 Haze values of virgin LDPE, 'recycled EVOH' (E), and 'recycled Metallocene' (M).



minor crosslinking of polymer chains due to repeating the extrusion processes. The crosslinking effect resulting from simulated mechanical recycling through repeated or extensive extrusion has been observed in the studies by Gonzalez Gutierrez et al. (2013) and Yin et al. (2015).[32,33]

For the recycled EVOH, the mechanical properties do not exhibit the same trend with the number of cycles. Due to the presence of an incompatible material (EVOH), it results in a poorly distribution in the PE matrix, as observed in SEM micrographs. This leads to an increase in the standard deviation and irregular mean values as reprocessing cycles increase. Therefore, it is not possible to observe a consistent increase or decrease in the mechanical properties. Thus, the incorporation of virgin material in this recycled one, as described in point 4 of this study, is important to evaluate the stability of the film properties with recycled EVOH.

As indicated in Figure 9 and Table S5, haze decreases with the reprocessing cycles of 'recycled EVOH', potentially resulting from a better dispersion of this material in the PE matrix. However, the haze level in this recycled material remains significantly higher than that of LDPE (4.5–6.4 times higher), hindering clear visibility of the food through the film. In recycled Metallocene, the haze slightly increases with the increase in reprocessing cycles, potentially linked to the presence of crosslinks, as previously mentioned. This observation is corroborated by the study conducted by Esmailzadeh et al. (2022), which demonstrates that crosslinks lead to an increase in haze and a decrease in gloss of LDPE and LLDPE films.[34]

The increased reprocessing of recycled EVOH led to a slight increase of the surface gloss of the films, as it can be observed in Figure 10 and Table S6.

Since the gloss values obtained for recycled Metallocene exceeded 70 GU (measurement angle of 60°), characteristic of high-gloss surfaces, it was necessary to use the 20° measurement angle for a finer analysis of the results. The recycled Metallocene films, with low haze similar to virgin LDPE film, exhibited a high-gloss film surface. Up to the second reprocessing cycle, the recycled Metallocene showed lower gloss than 1 M and virgin LDPE film samples.

The gloss of these recycles correlates with haze and SEM results. As gloss increases, haze decreases, and vice versa. The recycled EVOH films exhibited a semi-gloss surface with high haze due to the clear immiscibility between LDPE and EVOH observed in SEM micrographs, while the films with recycled Metallocene achieved a high-gloss surface with low haze due to a compatible PE-based constitution. This effect has been documented in several studies on extruded monolayer films.[35-37]

The reprocessing of the recycles did not affect the mechanical properties of the two different recycles but did impact the optical properties of the recycled EVOH. As the reprocessing cycles increase, the film with recycled EVOH becomes brighter and less hazy, although not enough to compete with recycled Metallocene or

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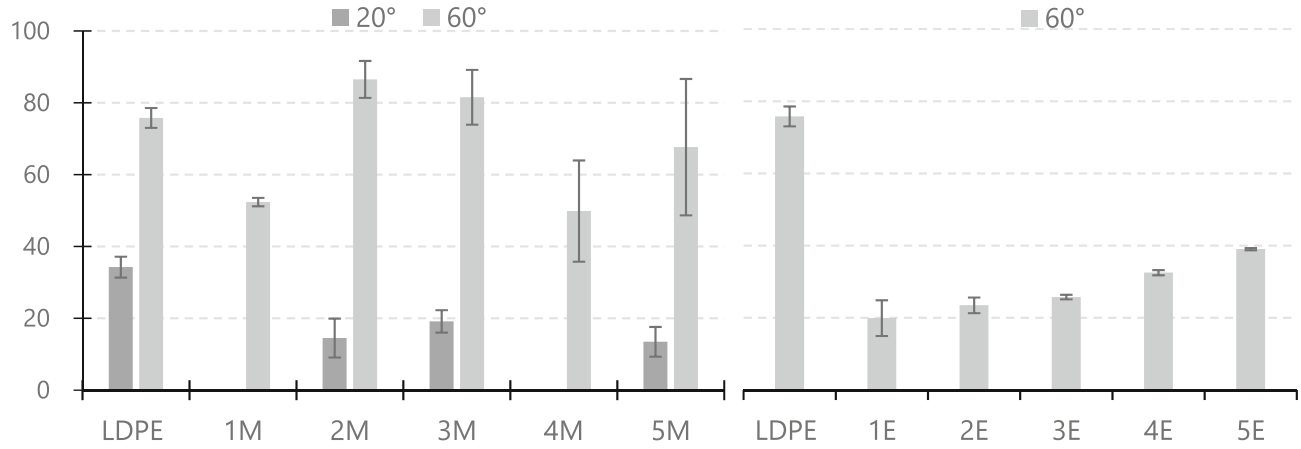

FIGURE 10 Gloss of 'recycled EVOH', 'recycled Metallocene', and virgin LDPE films.

Samples	Haze (%)	Samples	Haze (%)
1 M	8.4 ± 0.2^{A}	1E	47.1 ± 0.4^{B}
50% M + 50% LDPE	7.7 ± 0.2^{B}	50% E + 50% LDPE	34.2 ± 0.2^{D}
60% M + 40% LDPE	7.6 ± 0.2^{B}	60% E + 40% LDPE	38.0 ± 0.3^{C}
80% M + 20% LDPE	8.7 ± 0.2^{A}	80% E + 20% LDPE	48.4 ± 0.1^{A}
LDPE	7.6 ± 0.1^{B}	LDPE	7.6 ± 0.1^{E}

TABLE 4 Haze values of the films with the recycled EVOH (E) and recycled Metallocene (M), films with blends and virgin LDPE.

Note: Means that do not share a letter are significantly different.

TABLE 5 Gloss units of recycled/LDPE blends at 20° and 60° measurement geometries.

	Gloss units			Gloss units
Samples	20°	60°	Samples	60°
1 M	28.77 ± 1.76^{AB}	52.37 ± 1.17^{C}	1E	19.91 ± 0.72^{D}
50% M + 50% LDPE	27.19 ± 15.24^{B}	84.55 ± 27.65^{BC}	50% E + 50% LDPE	38.29 ± 4.95^{B}
60% M + 40% LDPE	48.04 ± 5.41^{A}	124.25 ± 21.19^{A}	60% E + 40% LDPE	30.72 ± 2.16^{C}
80% M + 20% LDPE	25.61 ± 3.13^{AB}	118.00 ± 3.61^{AB}	80% E + 20% LDPE	24.20 ± 0.64^{CD}
LDPE	34.26 ± 2.91^{AB}	75.79 ± 2.76^{BC}	LDPE	75.79 ± 2.76^{A}

Note: Means that do not share a letter are significantly different.

virgin LDPE. Incorporating a fixed percentage of virgin polymer with the recycled material in the industrial process can help to control and improve the optical properties of the film. Also, incorporating virgin material helps to reduce the influence of eventual variations on the composition on the recycled material, resulting in a higher film reproducibility.

3.2 | Incorporation of virgin LDPE in recycled material

As it can be seen in Table 4, haze increased with the rise in the percentage of recycled material in the film (Table 5). These findings align with WAXD results presented in Table 6, where the degree of crystallinity decreases with the incorporation of LDPE. This decrease in crystallinity reduces the structural rearrangement within the polymeric matrix, resulting in increased transparency.

In recycled/LDPE blends, there is a decrease in brightness with the reduction in the percentage of LDPE, as can be seen in Table 3. The gloss results are in line with the turbidity results, as observed for the reprocessed materials.

The WAXD spectra for recycled EVOH, recycled Metallocene, their blends with 50 wt. % of LDPE, and virgin LDPE film samples are illustrated in Figure 11. As mentioned earlier regarding the reprocessed recycles, all samples exhibit three main crystalline diffraction peaks characteristics of PE.

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TABLE 6 Degree of crystallinity of recycled Metallocene and recycled EVOH (1 M, 1E), recycled /LDPE blend and virgin LDPE films.

	Peak 1		Peak 2		Peak 3		
Samples	**2θ (°)**	**Intensity (c.p.s)**	**2θ (°)**	**Intensity (c.p.s)**	**2θ (°)**	**Intensity (c.p.s)**	**Crystallinity (%)**
1 M	21.46	2010	23.82	287	36.12	158	54.1
50% M + 50% LDPE	21.44	849	23.72	165	36.20	104	48.1
1E	21.36	1322	23.74	283	36.20	137	54.0
50% E + 50% LDPE	21.40	976	23.62	211	36.24	137	45.6
LDPE	21.42	481	23.64	131	36.18	102	39.5

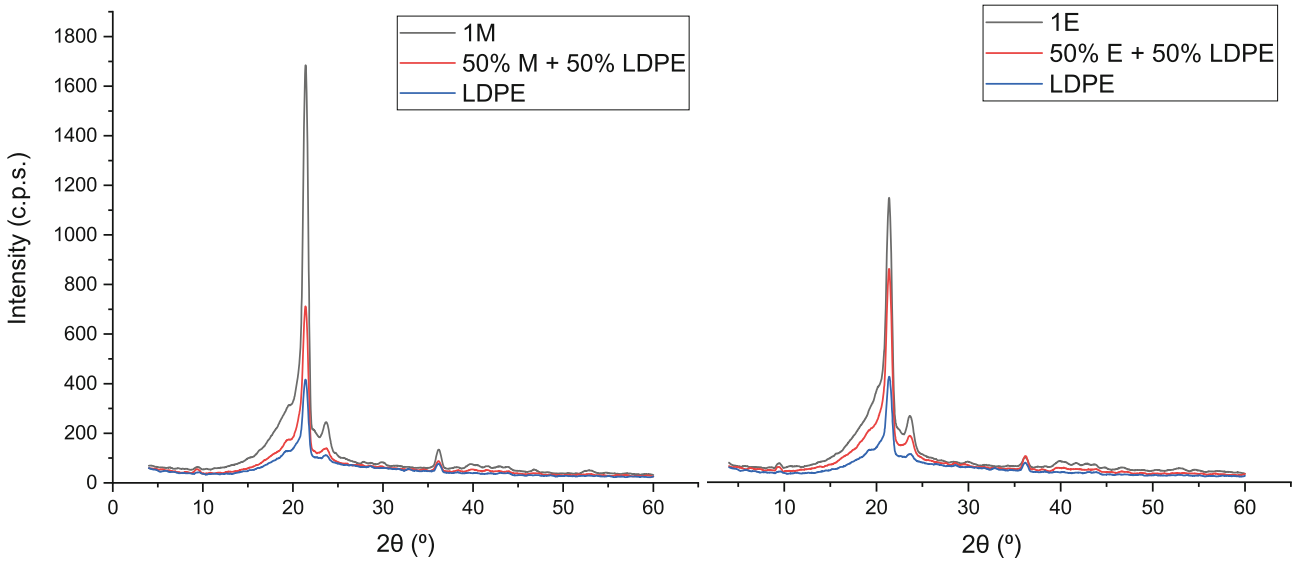

FIGURE 11 On the left, WAXD spectra of recycled Metallocene, 50%M + 50%LDPE, and virgin LDPE and on the right recycled EVOH, 50%E+50%LDPE, and virgin LDPE.

FIGURE 12 SEM micrographs, 15,000× magnification, of the films with 1 M, 1E and their blends with 50 wt.% LDPE.

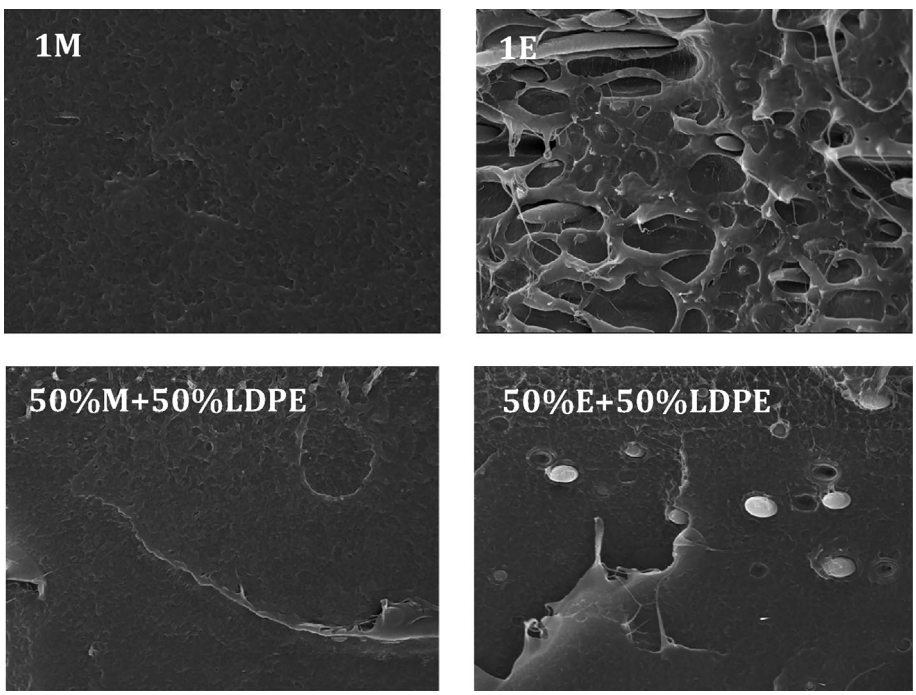

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TABLE 7 Tensile test results of films with LDPE and recycled Metallocene with LDPE.

Samples		$E_{1\%}$ (Mpa)	σ_{max} (MPa)	ε_b (%)
LDPE	MD	238.76 ± 8.42^{A}	16.99 ± 0.42^{A}	284.96 ± 19.22^{B}
	TD	241.36 ± 58.02^{A}	9.76 ± 0.24^{C}	115.76 ± 32.8^{B}
50% M + 50% LDPE	MD	246.02 ± 32.15^{A}	15.85 ± 1.55^{AB}	590.12 ± 70.04^{A}
	TD	252.71 ± 30.31^{A}	12.55 ± 0.51^{AB}	765.81 ± 141.40^{A}
60% M + 40% LDPE	MD	225.45 ± 8.57^{A}	14.19 ± 1.15^{AB}	491.07 ± 110.28^{A}
	TD	246.13 ± 22.78^{A}	12.45 ± 0.57^{AB}	702.10 ± 106.88^{A}
80% M + 20% LDPE	MD	213.46 ± 13.89^{A}	13.59 ± 0.93^{B}	574.70 ± 46.11^{A}
	TD	242.21 ± 16.35^{A}	12.81 ± 0.78^{A}	774.70 ± 80.04^{A}
1 M	MD	219.01 ± 21.32^{A}	13.80 ± 3.00^{B}	507.90 ± 86.42^{A}
	TD	188.06 ± 45.73^{A}	11.65 ± 0.78^{B}	587.98 ± 113.02^{A}

Note: Means that do not share a letter are significantly different. Statistical analysis was performed separately for the samples collected in MD and TD.

TABLE 8 Tensile test results of films with LDPE and recycled EVOH with LDPE.

Samples		$E_{1\%}$ (Mpa)	σ_{max} (MPa)	ε_b (%)
LDPE	MD	238.76 ± 8.42^{AB}	16.99 ± 0.42^{AB}	284.96 ± 19.22^{D}
	TD	241.36 ± 58.02^{A}	9.76 ± 0.24^{C}	115.76 ± 32.8^{B}
50%E + 50%LDPE	MD	236.80 ± 15.68^{B}	16.52 ± 1.95^{B}	525.65 ± 107.99^{BC}
	TD	294.66 ± 23.24^{A}	13.31 ± 0.84^{AB}	754.35 ± 283.75^{A}
60%E + 40%LDPE	MD	280.58 ± 52.62^{AB}	15.73 ± 1.23^{B}	478.25 ± 125.12^{C}
	TD	249.80 ± 76.08^{A}	11.93 ± 0.91^{BC}	615.24 ± 213.45^{A}
80%E + 20%LDPE	MD	256.80 ± 21.72^{AB}	18.02 ± 1.18^{AB}	691.54 ± 75.75^{AB}
	TD	270.79 ± 23.42^{A}	14.99 ± 1.06^{A}	845.41 ± 87.46^{A}
1E	MD	300.72 ± 43.93^{A}	19.86 ± 2.23^{A}	702.57 ± 72.28^{A}
	TD	307.28 ± 45.53^{A}	13.24 ± 0.88^{B}	684.93 ± 67.46^{A}

Note: Means that do not share a letter are significantly different. Statistical analysis was performed separately for the samples collected in MD and TD.

As indicated in Table S3, the addition of 50 wt. % virgin LDPE reduced the crystallinity of both recycles. These findings are consistent with those obtained in Li et al. (2019) study, where the crystallinity of LLDPE is found to be slightly higher than that of LDPE.[38]

Comparing recycled EVOH (1E) with its blend containing 50 wt.% LDPE, as shown in Figure 12, the reduction of EVOH is clearly visible with the incorporation of virgin LDPE, while its dispersion in the LDPE matrix remains the consistent. Conversely, films with recycled Metallocene show no signs of incompatibility upon the addition of LDPE.

The tensile test results of the films produced with blends of recycled materials and virgin LDPE are presented in Tables 7 and 8. The tensile tests conducted on the recycled mixtures containing 20%, 40%, and 50% LDPE demonstrate positive results, even with a low incorporation of virgin LDPE.

The strain at break of both recycled blends increased considerably compared with LDPE film. However, the incorporation of virgin material did not lead to the expected control of mechanical properties or a decrease in the standard deviation of the tensile results of the samples. Therefore, in the case of applying this study in an industrial process, it may not be necessary to incorporate virgin material into the recycled one, as the results obtained for the two systems are very similar, particularly in terms of mechanical properties.

4 | CONCLUSION

The present study concludes that it is feasible to incorporate materials recovered from multilayer films, consisting mainly of LDPE, into the structural layers of conventional food packaging without compromising their mechanical and thermal properties, while maintaining the extrusion conditions.

Reprocessing the recycled material did not significantly alter the crystallization and melting temperatures,

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degree of crystallinity, MFI, or the stability of the tubular film production process. While Young's modulus of the recycled Metallocene increased with reprocessing, it remained constant for films with reprocessed recycled EVOH. Overall, all reprocessed recycles exhibited superior mechanical properties compared with virgin LDPE films. Additionally, LDPE/recycled blends demonstrated significantly higher strain at break values than LDPE film.

Blending 50 wt.% LDPE with recycled materials can effectively control half of the raw material content in the final film packaging, given that the recycled material may vary in composition across different batches. Regarding optical properties, the haze of films containing recycled EVOH was much higher than that of LDPE and Metallocene recycled materials, which exhibited similar haze and gloss values. While haze decreased with the reprocessing of recycled EVOH material, it slightly increased for recycled Metallocene. In LDPE blends with recycled EVOH, the haze increased with the proportion of recycled material in the film. It was also observed that as turbidity increased, gloss decreased, and vice versa across all samples.

Despite the favorable mechanical results of recycled EVOH, films with recycled Metallocene exhibited the best overall properties, as all polymers in this recycled material are fully compatible.

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CONFLICT OF INTEREST STATEMENT

The author(s) declare(s) that there are no conflicts of interest.

DATA AVAILABILITY STATEMENT

The data that supports the findings of this study are available in the supplementary material of this article.

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