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Araştırma Makalesi ● Research Article

Sustainability of Flexible Plastic Packaging for Instant Coffee: Evaluation of Environmental Impact and Recyclability

Hazır Kahve İçin Esnek Plastik Ambalajın Sürdürülebilirliği: Çevresel Etkilerin ve Geri Dönüştürülebilirliğin Değerlendirilmesi

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ANAHTAR KELİMELER

Hazır kahve
Geri dönüşüm
Yaşam Döngüsü Düşüncesi
Sürdürülebilir ambalaj
Çok katmanlı ambalaj

ÖZ

Brezilya'da hazır kahveler yaygın olarak PET(polietilen tereftalat)/Al(alüminyum)folyo/LDPE(düşük yoğunluklu polietilen) bazlı dik poşetlerde pazarlanmaktadır. Alüminyum folyo etkili bir nem bariyeri tabakasıdır. Ancak tüketici sonrası esnek ambalajlar genellikle malzeme ayırma zorluğu ve geri dönüşüm hatlarındaki uyumsuzluk nedeniyle çevre sorunlarına neden olmaktadır. Bu çalışmanın amacı, hazır kahve paketlemeye yönelik alternatif malzemelerin bariyer özelliklerini, su buharı ve oksijen iletim hızını (WVTR/OTR) ve geri dönüştürülebilirlik potansiyelini değerlendirmektir: (1) LDPE/HDPE(yüksek yoğunluklu polietilen)/LDPE, (2) BOPP(iki yönlü polipropilen)/BOPPmet(metalize)/PP, (3) PET/PETmet/LDPE ve (4) PET/BOPPmet/LDPE. Malzemelerin WVTR'si 0,37 g su m-2 gün-1'e (25°C/%75RH) kadardır ve OTR'si 3,95 mL (NTP) m-2 gün-1'e kadardır; bu durum pazar cirosu yüksek olan ürünün raf ömrünü garanti edebileceklerini göstermektedir. Geri dönüşüm testleri yalnızca (1)'in PE geri dönüşüm hatlarıyla uyumlu olduğunu göstermektedir. (3) ve (4)'teki geri dönüştürülmüş filmler bazı önemli mekanik özelliklerde artış gösterdi. (1) ve (4) değerlendirilen çevre boyutlarındaki en yüksek azalmayı gösterdi.

KEYWORDS

Instant coffee
Recycling
Life Cycle Thinking
Sustainable packaging
Multilayer packaging

ABSTRACT

In Brazil instant coffees are widely marketed in stand-up pouch based on PET (polyethylene terephthalate)/Al(aluminum) foil/LDPE (low-density polyethylene). Aluminum foil is an efficient moisture barrier layer. However, the post-consumer flexible packaging usually causes environmental problems due to the difficulty of materials separation and incompatibility in the recycling lines. The aim of this study was to evaluate the barrier properties, water vapor and oxygen transmission rate (WVTR/OTR) and recyclability potential of the alternative materials for packing instant coffees: (1)LDPE/HDPE(high-density polyethylene)/LDPE, (2)BOPP(bioriented polypropylene)/BOPPmet(metallized)/PP, (3)PET/PETmet/LDPE and (4)PET/BOPPmet/LDPE. WVTR of the materials was up to 0.37g of water m-2 day-1 (25°C/75%RH) and OTR was up to 3.95mL (NTP) m-2 day-1 indicating they can guarantee the shelf-life of the product with high market turnover. Recycling tests showed that only (1) is compatible with PE recycling lines. Recycled films from (3) and (4) showed increase in some important mechanical properties. (1) and (4) showed the highest reductions of environmental aspects evaluated.

1. Introduction

Circular economy combines economic development with the best use of natural resources and it is a trend in all sectors of society. In terms of packaging, it means relying less on

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virgin raw materials, prioritizing more durable, recyclable 'and renewable' inputs (Ellen MacArthur Foundation, 2021). The search for plastic packages that replace non-recyclable materials has motivated large companies to figure out viable and efficient solutions for packaging. Besides, the new options must maintain the quality of food. This demand paves the way for increasingly demanding markets and consumers in terms of consumption of products with low environmental impact (Food Packaging Forum, 2019).

From the point of view of purchase intention, Rokka and Uusitalo (2008) studied the attributes of packaging that guide consumers in the act of purchasing products, including sealing capacity, branding and recyclability. In a similar study, Wang et al. (2021) pointed out a significantly positive effect of the recyclable packaging on the choice behavior of consumers who participated in the survey, which indicates that ecologically correct packaging that includes being manufactured with potentially recyclable materials is an important factor that drives intention of purchasing. Factor for which consumers are willing to pay more (Klaiman et al., 2016).

The replacement of packaging materials, especially for options with reduced thickness, which represent a reduction in resources, or options made by single material is one of the main goals of manufacturers, but it does not always bring benefits. Thickness reduction, for example, may represent the use of multilayer films that sacrifice material recyclability (Barlow and Morgan, 2013). For some food products, this substitution is relatively easy; for others, more demanding in terms of protection, such as dry and powdered products, it is a challenge to overcome. Instant coffees fall into the last category.

Moisture gain plays a critical role in affecting the quality and shelf life of instant coffee, as due to its high hygroscopic behavior, it is susceptible to deterioration when exposed to moisture contents ranging from 7% to 8%, which cause agglomeration of the powder or of the granules and makes it pasty or paved (Robertson, 2013). To maintain the quality of the product, Brazilian legislation establishes a maximum humidity of 5% (on a wet basis) as a specific requirement for the class of soluble products (ANVISA, 2005), which is achieved using packaging materials with a functional barrier layer to water vapor.

In Brazil, instant coffee is sold in flexible stand-up pouch packages, used as refill for glass jars, and in small sticks for individual doses, both manufactured in multilayer film (PET/Al foil/LDPE) containing aluminum foil as a functional barrier layer for gas permeation, moisture and aroma loss. Flexible packaging has a lower production cost and greater mechanical resistance than rigid packaging, which reduces product losses in the filling, storage, retail outlets and final disposal. The laminated and stand-up pouch structure of the PET/Al foil/LDPE films ensures that the products are placed vertically on the retail outlets shelves, allowing the consumer to see the product easily (Andregheti, 2015).

Flexible packaging made up of single material is potentially recyclable and has efficient recycling systems. However, the flexible plastic packaging currently employed to pack instant coffee does not appeal to sustainability and represents a problem from an environmental point of view due to the difficulty of separating the materials of the structure and its incompatibility in the recycling lines for plastic materials (Tartakowski, 2010; Barlow and Morgan, 2013; Teixeira, 2013). The film structure is laminated with aluminum, which limits its mechanical recycling to thermo-press recycling due to the impossibility of prior separation of the structure layers.

The environmental impact of plastic packaging is indicated by energy consumption, which includes contributions from the energy consumed to manufacture the polymer (called embodied energy) and the energy required to manufacture the film (about 10% of embodied energy). Thus, the plastic packaging consumes a lot of energy for manufacturing and generate a substantial negative environmental impact (Barlow and Morgan, 2013).

Unfortunately, polymers are considered low commercial value waste and returning the material to the reuse cycle is considered logistically expensive. Films are often rejected from recycling waste streams due to their lightness and diversity of polymers; besides multilayer films cannot be separated in a single polymer type. However, if recycled, they can avoid unnecessary energy consumption and be beneficial to the environment by replacing, in some cases, the use of virgin resin, resulting in a reduction in the amount of waste generated (Barlow and Morgan, 2013; Kaiser, 2018).

Therefore, the objective of this work was to evaluate the recyclability and environmental impact of four options of potentially recyclable flexible plastic structures to pack instant coffee, which were selected based on the water vapor permeability rate required by this class of product.

2. Material And Methods

2.1. Flexible material structures

Four alternative plastic material structures without aluminum foil for instant coffee were evaluated in this study, as follows:

1 - LDPE 33 µm/HDPE 33 µm/LDPE 32 µm – 81.98 g m^{-2}

2 - BOPP 29 µm/BOPPmet 16 µm/PP 26 µm – 98.31 g m^{-2}

3 - PET 12 µm/PETmet 12 µm/LDPE 67 µm – 103.19 g m^{-2}

4 - PET 10 µm/BOPPmet 11 µm/LDPE 53 µm – 92.75 g m^{-2}

2.2. Water vapor transmission rate (WVTR)

Water vapor transmission rate of flexible plastic structures was evaluated employing Permatran-W 3/33 model - WVTR analyzer, from Mocon (Minneapolis, USA) according to ASTM F1249-20 (2020, US) performed at 38 ºC/80% RH – international ambient conditions and by gravimetric method according to ASTM E96/E96-M-22ae1 (2022) performed at 25 °C/75% RH – Brazilian ambient conditions.

2.3. Oxygen transmission rate (OTR)

Oxygen transmission rate of flexible plastic structures was evaluated employing Oxtran model 2/60, from Mocon (Minneapolis, USA) performed at 23 °C and 0.21 atm of partial pressure of oxygen according to ASTM F1307 (2014).

2.4. Recyclability

The recyclability of the studied materials was evaluated based on the recyclability assessment protocols available for polyethylene and polypropylene films (RecyClass, 2021a; RecyClass, 2021b; The Association of Plastic Recyclers, 2020) in order to determine whether the structures evaluated are compatible with post-consumer PE and PP film recycling chains.

Approximately 40 kg of each studied film was crushed and incorporated in two different proportions (12.5% and 25.0%) into the control resins employed to produce the recycled films. Control resins were selected considering the polyolefin with higher content in the structure, which are: structure 1: 100% HDPE, structure 2: 100% PP and structures 3 and 4: 80% LLDPE/20% LDPE. The methodology consisted of three steps: 1) Pre-treatment of the flakes, 2) Extrusion of pellets and 3) Conversion of pellets to obtain recycled films.

2.4.1. Pre-treatment of the flakes

Control and studied films were separately grinded in a Grinder Wortex GSG 300/800 model (Campinas, Brazil) to flakes of 10 to 20 mm. Printed structures were washed to evaluate the impact of inks in wet washing operations, as follow: 1 g:24 mL water ratio at room temperature and stir at 1000 rpm for 10 min, followed by rinsing at 500 rpm for 5 min.

Flotation test 1g:24 mL water ratio at room temperature and stir at 500 rpm for 10 min was adopted to determine if the flakes can be separated by density in float/sink tank used in the recycling operation.

Drying for moisture content determination by heating 2 kg of studied materials in an Eletrolab oven 122 FC model (São Paulo, Brazil) at 60 ºC and weighting after 6 h e 12 h.

2.4.2. Extrusion of pellets

Pellets composed by 100% control flake (A.0), 75% control flake and 25% studied material (A.25) and 50% control flake and 50% studied material (A.50) were extruded in an Extruder and Recycler Wortex model Challenger II Generation – 600 mm L/D 42 (Campinas, Brazil). The pellets were evaluated regarding to:

Relative density according to ASTM D792-20 (2020, US). 1g of pellet was pressed and had its specific mass determined in water by weighting in analytical balance Sartorius (Göttingen, Germany) with 10-5 g accuracy at ambient temperature (23°C).

Melt flow rate of pellets was obtained according to ASTM D1238-20 (2020, US) in a Melt flow tester from CEAST (Pianezza, Italy). A standard load of 2.16 kg was applied for all materials at a die temperature of 190 ºC (PE materials) and 230 ºC (PP materials) and flow of material was observed for time. The material weight that flowed through die cavity for time gave the MFI in g 10 min-1.

Thermal properties were evaluated by Differential Scanning Calorimetry (DSC) carried out using a DSC 250 model from TA Instruments (New Castle, USA) (ASTM D3418-15, US). Approximately 5 mg of the material were analyzed in a hermetic pan. Initially the thermal history of the sample was destroyed by heating from 25 ºC to 300 ºC and then holding at 300 ºC for 5 minutes, and then the sample was cooled to 25 ºC and held for 5 minutes. A second heating up to 300 ºC was performed using the same conditions. Both heating and cooling were done at a rate of 10 ºC per minute.

2.4.3. Production of recycled films

Blown films (90 µm thickness) were produced with the following compositions: B.0 - 50% A.0 pellet and 50% virgin pellet, B.25 - 50% A.25 pellet and 50% virgin pellet and B.50 - 50% A.50 pellet and 50% virgin pellet. The films were extruded in a 5-layers Coex Extruder Dr Collin Blown Film BL180/400 model (Germany) and evaluated by:

Thickness was obtained according to ISO 4593-93 (2020, Switzerland) standard using a digital indicator equipment Mitutoyo ID-H0530E model (Kawasaki, Japan). Five specimens of 10 cm² area were evaluated. Total of 25 determinations were done.

Mechanical properties of films obtained from the recycling process was evaluated by:

Tensile strength and elongation - Maximum tensile strength (TS) and elongation at break (E) in transverse direction (TD) and machine direction (MD) of studied materials were determined in an INSTRON instrument 5966–E2 model (Norwood, USA) with load cell 100 N, speed 500 mm min-1, claws distance 50 mm for structures 3 and 4 – B.25 and B.50 (TD and MD); load cell 1 kN, speed 12.5 mm min-1, claws distance 125 mm for structure 1 – B.0 (TD); load cell 1 kN, speed 500 mm.min-1, claws distance 50 mm for structures 1 and 2 – B.25 and B.50 (MD) and structures 2, 3 and 4 – B.0 (TD and MD); load cell 1 kN, speed 50 mm.min-1, claws distance 100 mm for structure 1 – B.0 (MD) and structures 1 and 2 – B.25 and B.50 (TD). Tensile test was performed following the ASTM D882-18 (2018, US) standard. Test specimens with 15 mm width and 100 mm length were used.

Dart impact - Dart impact resistance of studied materials was determined according to ASTM D1709-22 (2022, US), in

Davenport equipment serial FD nº 50/150 (London, England). The specimen (125 mm diameter) was fixed by means of vacuum and metal rings. The dart with a head diameter of 38 mm and with the necessary weight increments was positioned at 660 mm from the specimen. The impact of the dart occurred in the center of the specimen. The mass needed to cause failure in 50% of the analyzed specimens was determined (P50).

Tear strength - Tear propagation resistance was determined according to ASTM D1922-15 (2020, US) standard. Elmendorf equipment ED 1600 model Regmed (Osasco, Brazil) was used, which consists of a calibrated pendulum, whose movement causes the propagation of the tear in the specimen. The work required to perform this tear is measured by the potential energy loss of the pendulum. This loss is indicated by a pointer on a graduated scale from 0% to 100% of the capacity, in this case, 1600 gf for the equipment used.

Surface appearance and Amount of gels and specks larger than 200 µm in 500 cm² were evaluated by visual observation.

Total light transmission was determined based on ASTM D1003-21 (2021, US). Spectra were obtained on an Analytik Jena UV-Visible Specord 210 spectrophotometer (Jena, Germany) in the range 200 nm to 800 nm at a speed of 120 nm min-1 using an integration sphere as accessory. Three specimens with approximately 10 mm x 30 mm from different parts of the films were evaluated.

Stereomicroscopy to obtain images 10x magnification of films surfaces was performed in Leica equipment M165C model (Heerbrugg, Switzerland) with LAS EZ Leica Application Suite Software Version 3.0.0.

Scanning electron microscopy (SEM) was performed in ZEISS equipment DSM 940a model (Oberkochen, Germany) with 200x magnification. Pieces of cross section of films were cut in a rotating microtome Leica 2245 model (Buffalo Grove, US), with a cut thickness of 100 microns, and later fixed on aluminum stubs with carbon adhesive and covered with two layers of gold (40 mA / 40 s), in metallizer Balzers SCD 050 model (Fürstentum, Liechtenstein). The microphotographs taken in SEM system were complemented by the secondary electron detector (relief contrast, image topography, SE) and the backscattered electron detector (BSE), which composes images providing an indication of the material composition by the similarity of hue, that is, regions with the same hue have a similar chemical composition. The images of BSE regions with elements of lower atomic weight have a darker shade compared to the other regions.

2.5. Environmental profile

To estimate the environmental performance of the four studied materials and the material employed nowadays in the market to pack instant coffee a Life Cycle Thinking was applied. The estimation of the potential environmental impacts was based on the guidance of ISO 14040 and 14044 standards (ISO, 2006).

2.5.1 Goal and scope

The goal of the study was to evaluate possible environmental gains due to the substitution of the current packaging of instant coffee by some of the four studied materials.

The scope was to evaluate the production of the materials of each film (extraction of raw materials and production of the plastics or aluminum) and the end-of-life stage of the packaging (recycling or landfill). The other life cycle stages were considered equal to all materials and then were not accounted for.

2.5.2 Functional unit

The functional unit adopted was one m² of film.

2.5.3 System boundary

The system under study included the production and disposal of the primary packaging taking into account the attributional approach. Since this study has a comparative basis among the materials, the other life cycle stages were considered unchanged and then they were excluded from this evaluation. Secondary data obtained from recognized databases available in GaBi 6 Product Sustainability software were used for materials (PE International A.G.) and packaging production (ELCD database 2.0) as described in Table 1.

Table 1. Secondary data employed in the Life Cycle Thinking study.

Material/Stage	Dataset	Database
PET	Polyethylene Terephthalate Granulate (PET) via DMT, production mix, at plant - DE	PE International
Aluminum foil	Primary production, production mix, at plant, 5 to 200 µm - EU-27	PE International
LDPE	Polyethylene film (LDPE) technology mix, production mix, at producer - RER	Plastics Europe
HDPE	Polyethylene High Density Granulate (HDPE) Mix, technology mix, production mix, at plant - DE	PE International
PP film	Polypropylene film (PP) technology mix, production mix, at producer - RER	Plastics Europe
Landfill	Landfill of plastic waste - EU-27	PE International

Datasets corresponding to a European average were preferentially selected, which are considered representative for Brazilian production since the technologies employed in Brazil are quite similar to the European countries once many Brazilian industries import equipment from Europe. The disposal scenario refers to the Brazilian situation in the year

2021 with a recycling rate of 23.4% for post-consumer plastics (ABIPLAST, 2022). This recycling rate is obtained from door-to-door selective collection in Brazilian municipalities that have a selective collection system, in which consumers dispose of solid waste in two groups (recyclable - all types of packaging material and organic - food waste).

2.5.4 Environmental impact categories

The environmental impact categories adopted in this study are climate change (global warming potential for a 100-year perspective - GWP_{100}, excluding biogenic carbon), fine particulate matter formation - PMFP (BR specific), freshwater eutrophication - FEP (BR specific), terrestrial acidification - TAP (BR specific), water depletion - WDP, fossil depletion - FFP and freshwater ecotoxicity - FETP, and abiotic depletion (ADP fossil), which were estimated according to the ReCiPe 2016 v 1.1 Midpoint (Hierarchist perspective), since this method has characterization factors that are globally or Brazilian oriented. Furthermore, the impact category primary energy demand (PED) from renewable and non-renewable resources (net calorific value), which considers direct and indirect fuel consumption was calculated using the GaBi 6 Product Sustainability software program. Data modeling was performed by means of the GaBi 6 Product Sustainability software program (PE…, 2015).

2.6. Statistical Analysis

The results were statistically evaluated by means of analysis of variance (ANOVA) and the Tukey test to compare the averages ($p < 0.05$).

3. Results and Discussion

3.1 Water vapor and oxygen transmission rate (WVTR and OTR)

Table 2 shows water vapor and oxygen transmission rate of alternative materials evaluated.

Table 2. WVTR and OTR of alternative materials evaluated in this study.*

Material	WVTR (g water.m^{-2}.day^{-1})		OTR (mL (NTP) m^{-2} day^{-1})
	at 38°C/90% RH International ambient conditions	at 25°C/75% RH Brazilian ambient conditions	at 23ºC and 1 atm of partial pressure gradient of oxygen
1	0.51 ± 0.04^c	0.11 ± 0.00^c	0.20 ± 0.02^d
2	0.22 ± 0.05^c	0.04 ± 0.00^c	0.37 ± 0.01^c
3	1.97 ± 0.45^b	0.19 ± 0.03^b	1.07 ± 0.00^b
4	3.41 ± 0.18^a	0.37 ± 0.07^a	3.95 ± 0.12^a

* mean ± standard deviation, 1 = LDPE/HDPE/LDPE; 2 = BOPP/BOPPmet/PP; 3 = PET/PETmet/LDPE; 4 = PET/BOPPmet/LDPE; ^{a,b,c,d} means followed by the same letter, in the column, do not differ at 95% confidence level ($p < 0.05$).

Souza (2022) showed that headspace gas composition of the Brazilian packaging systems does not have modified atmosphere, i.e., Brazilian instant coffee products are packaged in air. This study demonstrates that O_2 inside packaging is not a critical parameter for instant coffee products, which means that despite traditional stand-up pouch packaging with aluminum foil has an efficient gas barrier it can be replaced by another structure. Structures **1** and **2** present good oxygen barriers ranging from 0.20 to 0.37 mL (CNTP) m^{-2} day^{-1}, as shown in Table 2.

WVTR of the materials evaluated in this study indicates structures **1** and **2** do not differ significantly from each other ($p < 0.05$) in the two ambient conditions. Statistical analysis also demonstrates the same difference among the WVTR of the four materials at 38°C/90% RH and at 25°C/75% RH.

Flexible packaging composed by PET/ Al foil (11 μm to 15 μm)/LDPE currently used to pack instant coffee has a WVTR lower than 0.001 g water m^{-2} day^{-1} at 38°C/80% RH. According to Robertson (2013), flexible packaging for instant coffee containing aluminum foil (12 μm thickness) provide shelf life of up to 12 months. When instant coffee is packaged in other plastic materials that do not have such efficient O_2 and moisture barrier this time is very short.

Metalized layer (approx. 30 nm) applied to polymeric films, often PP or PET also increases the barrier of the material (Barlow and Morgan, 2013). Alves and Bordin (1998) studied the shelf life of individual packages (25 g and 50 g) for instant coffee at 30 °C/80% RH in three plastic structures: LDPE, BOPP/pearled BOPP and metallized PET/LDPE. These films presented mean WVTR of 6.1 g water m^{-2} day^{-1}, 1.2 g water m^{-2} day^{-1} and 0.9 g water m^{-2} day^{-1}, respectively.

Thickening and mixing of materials, such as polyolefins, can also increase the barrier properties of film structures. Flexible food packaging has thicknesses ranging from 10 μm to 250 μm depending on the combination of strength, durability and functional barrier demand for its application. The use of PE or PP with 70% of the total film thickness brings a gain in the mechanical properties of the film, in particular the hardness that gives resistance to piercing and splitting (Barlow and Morgan, 2013).

According to Souza (2022), the minimum WVTR necessary to pack instant coffees is 0.042 g water m^{-2} day^{-1} for spray dried product and 0.057 g water m^{-2} day^{-1} for freeze-dried product. These results were predicted at 25°C/75% RH for critical moisture of 5% (ANVISA, 2005), packaging area of

0.0256 m^2, product weight of 50 g and storage time of 365 days. This study also indicated that increasing the packaging area to 0.0585 m^2 (product weight of 200 g) WVTR values change to 0.073 g water m^{-2} day^{-1} and 0.100 g water m^{-2} day^{-1} for spray dried product and freeze-dried product, respectively. Therefore, flexible material **2** could be employed to pack instant coffee of 50 g or higher, while the other structures could be used for 200 g packs with lower shelf life.

3.2. Recyclability

3.2.1. Flakes

Washing test: Printed flakes obtained from materials **3** and **4** do not shed color, suspended particles and fibers in the washing water, which indicates zero impact in wet washing operations.

Flotation test: Material **1** showed mostly flakes in suspension, but no apparent phase separation. Materials **2**, **3** and **4** showed flakes suspended, without phase separation and dispersed in liquid medium. PET is a high-density polymer, then, structures **3** and **4** were expected not float. However, BOPP is a low-density polymer and produces light films (material **2**). The expectation was that BOPP-based films would float easily. One possibility for this result is the higher amount of BOPP film per gram of water than other films due to the smaller thickness, which implies in higher area of film g^{-1} of water and then, higher volume of cutted film g^{-1} of water. Higher amount of material turns harder the phase separation in material/water ratio indicated by the flotation procedures. Therefore, the results indicated only flakes from structure **1** can be easily separated by density in float/sink tank used in the recycling operation.

Drying:

Table 3 shows moisture content results of flakes of alternative materials

Table 3. Flakes moisture content.*

Flakes from alternative plastic flexible material	Moisture content (%)
1 = LDPE/HDPE/LDPE	0.12 ± 0.03^{c}
2 = BOPP/BOPPmet/PP	0.14 ± 0.00^{bc}
3 = PET/PETmet/LDPE	0.20 ± 0.03^{a}
4 = PET/BOPPmet/LDPE	0.17 ± 0.02^{ab}

*mean ± standard deviation; ^{a,b,c} the means, followed by the same letter, in the column, do not differ at the 95% confidence level ($p < 0.05$).

Flakes from alternative materials showed moisture content from 0.12% to 0.20%. Films **3** and **4** retained greater amount of water due chemical nature of PET present in these structures. PET molecules are polar and form hydrogen bonds with H_2O molecules. Excess moisture in the flakes can lead to problems in the final product from recycling process, e.g., spots and blisters. In extruded films, it causes the formation of "fisheye", gels, rupture of the extrusion bubble, among others. According to Shen and Worrall (2014) flakes with less than 0.1% moisture by weight are ready to be reprocessed. It indicates that flakes evaluated, mainly **3** and **4**, should be dried before processing.

3.2.2. Pellets

Table 4 shows some properties of the pellets evaluated.

As **A.0** pellets were processed from 100% flake control, they are adopted as standards in comparison to mixed recycled materials (**A.25** and **A.50**). RecyClass (2021a, b) and The Association of Plastic Recyclers (2020) protocols established some recommendations for the evaluated parameters to check if pellets containing recycled materials can be used in PE and PP recycling lines.

For relative density all pellets attended recycling protocols recommendations, although the materials obtained from structures **3** and **4** showed a significant difference ($p < 0.05$). Pellets from structures **1** and **2** showed only one value for thermal transition temperatures (T_m and T_c) because LLDPE-HDPE and BOPP-PP have thermal behavior very similar showing a single band in DSC curves. Pellets from structures **3** and **4** presented three values for T_m and T_c because they composed mostly by LDPE ($T_{m,c,1}$) - LLDPE ($T_{m,c,2}$) blend and PET ($T_{m,c,3}$). Resins from structures **3** and **4** did not attend the specification since PET has melting temperature > 150°C. Thermal analysis of pellets from alternative flexible plastic structure **2** did not indicate presence of polyethylene what approves the material. However, its melt flow rate did not attend the specification since **A.25** and **A.50** showed values with higher than 15% deviation in respect to **A.0**. Maybe a lower percentage of alternative material in the blend, e.g., A.10, could show melt flow rate compatible with PP lines recycling.

Plastic with melt flow rate from zero to 1 g.10 min^{-1} are suitable for extrusion of film or plastic bags, as well as thermoformed into low trays. Melt flow rate from 0.3 to 5 g 10 min^{-1} are suitable for blow molding to produce bottles (Eriksen et al., 2019). Therefore, structure **2** could be suitable to produce blow molding plastic products while the other structures could be employed for extrusion of films or bags. Statistical analysis of melt flow rate results indicated significant differences ($p < 0.05$) between the evaluated pairs. However, the reduction or increase of the values obtained with the increase of the percentage of recycled material incorporated is not a trend, it depends on the structure/composition of the alternative film.

Table 5. Films properties. *

Identification		Thickness (µm)	Tensile strength (MPa)		Elongation at break (%)		Dart impact (g)	Tear strength (gf)**		Surface appearance	Amount of gels and specks
			TD	MD	TD	MD		TD	MD		
1 - B.0		**92.3 ± 10^{ab}**	**28.7 ± 1.5^{a}**	**30.0 ± 0.7^{a}**	**17.4 ± 3.8^{b}**	**115.4 ± 120^{b}**	**58**	**90 ± 3.3^{c}**	**69 ± 3.1^{c}**	Smooth surface, gels and stains, translucent	33
1	**B.25**	89.5 ± 3.9^{b}	29.1 ± 2,2^{a}	28.7 ± 2.5^{a}	25.6 ± 14^{b}	231.3 ± 120^{b}	**101**	113 ± 4.5^{b}	88 ± 3.8^{b}		34
	B.50	96.2 ± 9.0^{a}	28.6 ± 0.6^{a}	27.5 ± 0.9^{a}	159.5 ± 64^{a}	**505.8 ± 215^{a}**	**125**	**163 ± 5.6^{a}**	**121 ± 11^{a}**		27
2 - B.0		**95.9 ± 6.9^{c}**	**28.9 ± 1.1^{a}**	**29.4 ± 1.0^{a}**	**649.5 ± 139^{b}**	**617.0 ± 28^{a}**	**90**	**117 ± 6.7^{a}**	**86 ± 6.3^{a}**	Smooth surface, gels and stains, translucent	11
2	**B.25**	135.0 ± 22^{b}	*22.3 ± 1.2^{b}*	*22.2 ± 1.9^{b}*	575.0 ± 112^{c}	*20.5 ± 5.8^{b}*	*65*	*74 ± 2.9^{b}*	*48 ± 1.8^{b}*	Rough surface, incrustations, gels and stains, gray coloring, translucent	24
	B.50	174.6 ± 39^{a}	*19.0 ± 0.5^{c}*	*20.2 ± 1.6^{c}*	**1314.8 ± 99^{a}**	553.0 ± 117^{a}	*37*	*66 ± 1.0^{c}*	*40 ± 1.1^{c}*		**133**
3 and 4 - B.0		**95.3 ± 4.2^{c}**	**31.3 ± 2.4^{a}**	**31.2 ± 3.2^{a}**	**889.6 ± 28^{a}**	**828.8 ± 44^{a}**	**759**	**> 1600**		Smooth surface, gels and stains, translucent	19
3	**B.25**	115.2 ± 17^{b}	*18.7 ± 2.0^{b}*	*19.4 ± 1.4^{b}*	786.8 ± 29^{b}	731.5 ± 32^{b}	592			Rough surface, incrustations, gels and stains, green coloring, translucent	13
	B.50	154.0 ± 32^{a}	*12.2 ± 0.9^{c}*	*13.3 ± 3.2^{c}*	712.8 ± 19^{c}	699.8 ± 30^{b}	*402*				**44**
4	**B.25**	116.0 ± 17^{b}	*16.8 ± 1.9^{b}*	*17.6 ± 3.5^{b}*	767.3 ± 24^{b}	727.3 ± 38^{b}	*400*			Rough surface, incrustations, gels and stains, brown coloring, translucent	**103**
	B.50	173.2 ± 39^{a}	*10.4 ± 1.7^{c}*	*10.9 ± 2.8^{c}*	686.6 ± 35^{c}	639.1 ± 70^{b}	*402*				**225**
PE film recommendation (RecyClass, 2021a)		Process adjusted to 100 µm for all films***	No more than 25% delta to B.0							-	Based on application
PE film recommendation (The Association of Plastic Recyclers, 2020)			No more than 25% delta drop to B.0							No more than 25% count increase to B.0 For B.0: Gels - up to 30 Specks - up to 5 Holes - zero tolerance	
PP film recommendation (RecyClass, 2021a)			No more than 25% delta to B.0							-	Based on application

* mean ± standard deviation, TD = transverse direction and MD = machine direction, 1 = LDPE/HDPE/LDPE; 2 = BOPP/BOPPmet/PP; 3 = PET/PETmet/LDPE; 4 = PET/BOPPmet/LDPE, B.0 = 50% control pellet and 50% virgin pellet; B.25 = 50% A.25 pellet and 50% virgin pellet; B.50 = 50% A.50 pellet and 50% virgin pellet, **1600 gf is maximum potential energy, *** value chosen based on the current flexible package for instant coffee (stand up pouch); ^{a,b,c} the means, followed by the same letter, in the column compared to the respective A.0, do not differ at the 95% confidence level ($p < 0.05$).

Table 4. Physical and thermal properties of the pellets.*

Identification		Relative density ($g.cm^{-3}$)	Melt flow rate ($g.10\ min^{-1}$)	Thermal property**	
				T_m (°C)	T_c (°C)
A.0		0.952 ± 0.002^{a}	1.004 ± 0.008^{a}	135.1 ± 0.7	118.0 ± 0.5
1	**A.25**	0.947 ± 0.005^{ab}	0.989 ± 0.004^{b}	133.1 ± 0.6	117.6 ± 0.7
	A.50	0.942 ± 0.002^{b}	0.925 ± 0.003^{c}	133.0 ± 0.6	117.6 ± 0.7
A.0		0.895 ± 0.002^{a}	1.475 ± 0.010^{c}	147.3 ± 0.7	114.2 ± 0.4
2	**A.25**	0.894 ± 0.005^{a}	**2.201 ± 0.004**^{b}	153.2 ± 0.9	118.6 ± 0.2
	A.50	0.894 ± 0.002^{a}	**2.881 ± 0.013**^{a}	157.5 ± 0.7	121.2 ± 0.2
A.0		0.919 ± 0.012^{a}	0.301 ± 0.006^{b}	$T_{m,1}$ 108.5 ± 1.2 $T_{m,2}$ 121.1 ± 0.1	$T_{c,1}$ 94.8 ± 0.6 $T_{c,2}$ 107.0 ± 0.1
3	**A.25**	0.859 ± 0.018^{b}	0.472 ± 0.002 ^{a}	$T_{m,1}$ 109.2 ± 0.7 $T_{m,2}$ 122.1 ± 0.5 **$T_{m,3}$ 230.4 ± 6.0**	$T_{c,1}$ 95.4 ± 0.4 $T_{c,2}$ 107.3 ± 0.3 $T_{c,3}$ 175.2 ± 9.6
	A.50	0.809 ± 0.018^{c}	0.239 ± 0.006^{c}	$T_{m,1}$ 109.5 ± 0.7 $T_{m,2}$ 122.2 ± 0.8 **$T_{m,3}$ 228.5 ± 5.0**	$T_{c,1}$ 94.4 ± 0.4 $T_{c,2}$ 107.8 ± 0.3 $T_{c,3}$ 175.1 ± 8.6
4	**A.25**	0.910 ± 0.016^{a}	0.482 ± 0.006^{a}	$T_{m,1}$ 109.8 ± 0.7 $T_{m,2}$ 121.8 ± 0.1 **$T_{m,3}$ 224.1 ± 2.8**	$T_{c,1}$ 96.2 ± 0.4 $T_{c,2}$ 106.6 ± 0.1 $T_{c,3}$ 167.8 ± 7.8
	A.50	0.902 ± 0.007^{a}	0.203 ± 0.007^{c}	$T_{m,1}$ 107.3 ± 5.1 $T_{m,2}$ 121.2 ± 2.6 **$T_{m,3}$ 223.6 ± 3.4**	$T_{c,1}$ 95.9 ± 0.4 $T_{c,2}$ 106.0 ± 0.2 $T_{c,3}$ 159.8 ± 11.9
PE pellet recommendation (RecyClass, 2021a)		-	< 0.5 $g.10\ min^{-1}$ delta to A.0	T_m < 150 °C No more than 2.5% of PP for A.50	
PE pellet recommendation (The Association of Plastic Recyclers, 2020)		< 0.996 $g.cm^{-3}$	< 0.75 $g.10\ min^{-1}$ 25% delta to A.0	T_m < 150 °C	
PP pellet recommendation (RecyClass, 2021b)		< 0.920 $g.cm^{-3}$	< 15% deviation in respect to A.0	No more than 2.5% of PE for A.50	

* mean ± standard deviation, 1 = LDPE/HDPE/LDPE; 2 = BOPP/BOPPmet/PP; 3 = PET/PETmet/LDPE; 4 = PET/BOPPmet/LDPE; A.0 = 100% control; A.25 = 75% control flake with 25% alternative material; A.50 = 50% control flake with 50% alternative material, ** T_m = melting temperature; T_c = crystallization temperature; ^{a,b,c,} the means, followed by the same letter, in the column compared to the respective A.0, do not differ at the 95% confidence level ($p < 0.05$).

3.2.1. Films

Mechanical properties and appearance

Table 5 shows the thickness, mechanical properties and appearance of the films in Appendices. The significant difference ($p < 0.05$) observed in the thickness values for recycled films **B.25** and **B.50** of the processing of alternative materials **2**, **3** and **4** is related to the number of incrustations on the surface of the materials, being more prominent in films with a higher percentage of recycled material (B.50). These films also have a rough surface appearance and large amount of gels and spots with dimension larger than 200 µm.

Gels and specks can weaken the film quality. RecyClass (2021a, b) protocols do not establish recommendation for amount of gels and specks but only mention they must be in accordance with the intended application. However, The Association of Plastic Recyclers (2020) protocol for PE films establish maximum number of defects no more than 25% count increase to **B.0**. Based on these criteria and using maximum number of defects equal to 35 (gels plus specks), **B.50** films from structures **2**, **3** and **4** indicate they are non-recyclable. **B.25** film from structure **3** also presented large amount of visual impurities.

Regarding mechanical properties, recycled films obtained from alternative structure **2** presented the greater change in performance according to evaluated parameters with reduction of values in tensile strength (DM and DT), elongation at break (DT), dart impact and tear strength (DM and DT). For tensile strength, only recycled films from structure **1**, made of just polyethylene, showed values according to protocols recommendations in both directions. If we consider the criteria of 25% deviation in relation to **B.0** only in the test of performance drop (The Association of Plastic Recyclers, 2020) all films from structure **1** are accepted in recycling PE lines which was expected because they are made of monomaterial.

Compatibilizers and inorganic fillers have the function of improving the homogeneity of materials (Horodytska et al., 2018, Tartakowski, 2010, Wyser et al., 2000), that is why they are usually employed for recycling multilayer films. Therefore, recycling multilayer films in polyolefins recycling lines without the use of compatibilizers is a huge challenge.

Despite the significant decline ($p < 0.05$) some mechanical properties of the recycled films as a whole, the films with a higher percentage of recycled material from alternative materials **3** and **4** showed good performance in terms of elongation at break and better performance for tear strength. These films have PET in their structures, which is responsible for their mechanical strength. These properties can be interesting for some applications.

According to ABNT NBR 14937-23 standard (2023, Brazil) which establishes the minimum quality parameters for plastic bags made of virgin HDPE resin, the requirements for mechanical properties are less stringent than the results for recycled films obtained in this study. For example, for plastic bags with capacity of up to 16 kg dart resistance is evaluated using 70 g weight, recycled films from structures **1**, **3** and **4** comply with this requirement.

Agricultural films are another interesting application for recycled films. Mulching films need to show high values of mechanical resistance in terms of tensile strength (> 20 MPa) and elongation at break (approx. 300%) to attend the requests imposed on their use. Recycled films **1** and **2 – B50** comply these parameters. Using adequate percentage of recycled material is possible to achieve these parameters in generating solutions for reusing of plastic waste (Briassoulis et al., 2004).

3.2.2. Total light transmission

Figure 1 shows total light transmission spectra from recycled films evaluated in this study.

Optical properties, in particular transparency, are important requirement for plastic materials. In the case of recycled films, it can be harmed by the presence of paints, metallization residues and incrustations resulting from the mixture of incompatible materials. The results in Figure 1 show that recycled **B.0** films have excellent light transmission for wavelength higher than 380 nm (visible range), reaching >85% transparency. Recycled films **B.25** and **B.50** obtained from alternative materials **1** and **2** show the best light transmission performance (> 75%) results in terms of the highest percentage of recycled material. Alternative material **1** is colorless while structure **2** is metallized, getting recycled films tinted gray. Structures **3** and **4** are printed and the color the recycled films were green and brown, respectively, reducing the transparency of the materials in the visible region (from 380 nm to 800 nm). Light barrier from 200 nm to 300 nm is characteristic to PET present in recycled films from structures **3** and **4**.

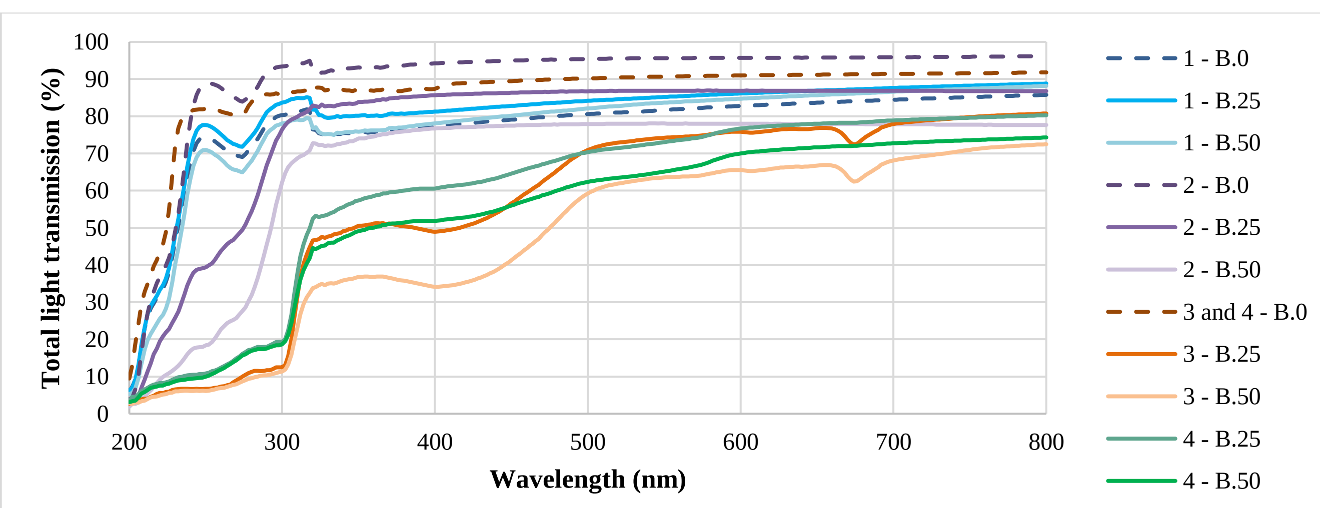

Figure 1. Total light transmission of recycled films: 1 = LDPE/HDPE/LDPE; 2 = BOPP/BOPPmet/PP; 3 = PET/PETmet/LDPE; 4 = PET/BOPPmet/LDPE, B.0 = 50% control pellet and 50% virgin pellet; B.25 = 50% A.25 pellet and 50% virgin pellet; B.50 = 50% A.50 pellet and 50% virgin pellet

3.2.3. Stereomicroscopy and Scanning electron microscopy (SEM)

Figure 2 shows stereomicroscopy and scanning electron micrograph for recycled films.

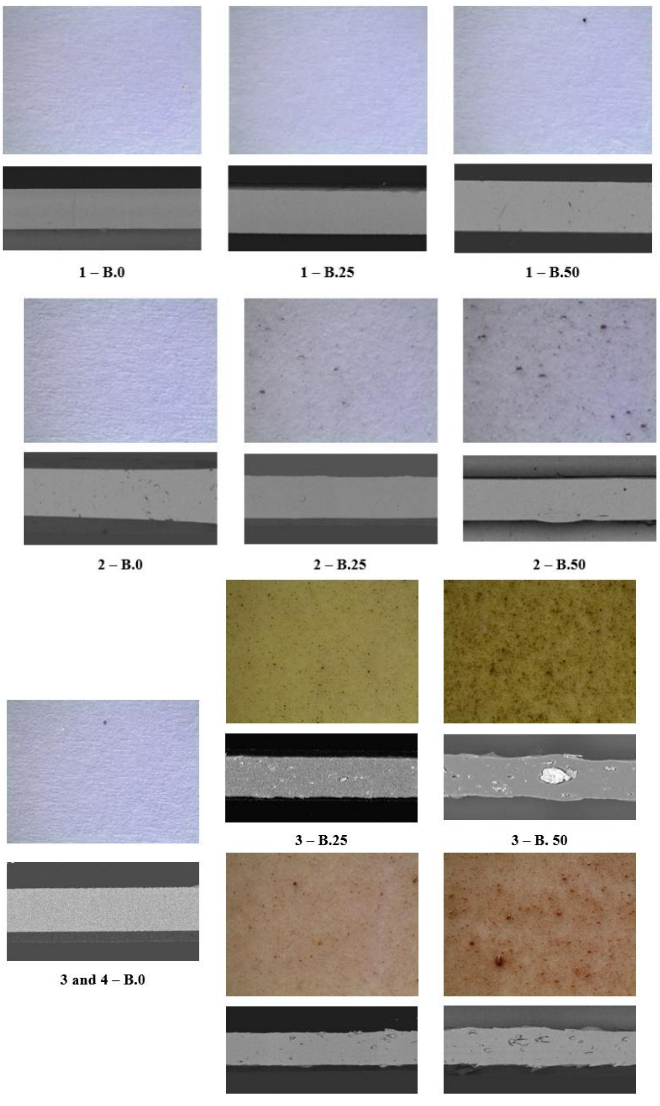

Figure 2. Stereomicroscopy and scanning electron microscopy (SEM) images of the recycled films (5 kV, 25 mm, 200x, scale: 100 μm): 1 = LDPE/HDPE/LDPE; 2 = BOPP/BOPPmet/PP; 3 = PET/PETmet/LDPE; 4 = PET/BOPPmet/LDPE, B.0 = 50% control pellet and 50% virgin pellet; B.25 = 50% A.25 pellet and 50% virgin pellet; B.50 = 50% A.50 pellet and 50% virgin pellet.

Results obtained in visual evaluation of the films appearance can be confirmed in the stereomicroscopy and electron microscopy images. **B.0** films have a flat surface while the recycled films, especially from structures **3** and **4**, have a rough surface. Roughness increases with the increase of the percentage of recycled material. Structure **1** is composed only by polyethylene and structure **2**, despite being multilayer and metallized, also is made up of a unique type of polymer (polypropylene) and after the recycling process, it becomes a homogeneous mixture. Alternative materials **3** and **4** are metallized and composed by a mixture of materials (PET, BOPP and LDPE), then reaching homogeneity is very difficult without adding modifiers. These results show that metallized BOPP is less critical to get homogeneity of the recycled films than metallized PET since films from structure **3** are more heterogeneous than structure **4**. Probably this heterogeneity is responsible for the lowest values of tensile strength of the recycled films **3 – B.50** and **4 – B.50**, as shown in Table 5.

3.3. Environmental profile

Changing the current structure of PET/Al foil/LDPE to any of the other four structures evaluated gets a reduction of the four environmental aspects evaluated as shown in Figure 3. In general, the alternative film options for the standup pouch have a lower environmental impact than the current film, which is mainly due to the elimination of aluminum foil from the structure of the films. The highest environmental impact values were obtained for the climate change (GWP), water depletion (WDP) and fossil depletion (FFP). Films **1** and **2** are recyclable since they are monomaterials (LDPE/HDPE/LDPE and BOPP/BOPP/PP, respectively). Therefore, a recycling rate of 23.4% was applied to the plastic waste generated by these films (ABIPLAST, 2022), which explains the lower environmental impact of these films compared to the current, **3** and **4** films, which are not recyclable.The higher environmental impacts reduction was obtained with the film of LDPE/HDPE/LDPE (structure **1**) that showed 0.19 kg $CO_{2\text{-eq}}$ m^{-2} of GWP, 8.64×10^{-5} PM2.5 m^{-2} of PMFP, 1.36×10^{-6} kg $P_{\text{-eq.}}$ m^{-2} of FEP, 7.09×10^{-4} kg $SO_{2\text{-eq.}}$ of TAP, 0.14 kg oil-eq. of FFP and 1.15×10^{-4} kg 1,4-$DB_{\text{-eq}}$ m^{-2} of FETP, which represents a reduction of 50%, 57%, 35%, 55%, 24% and 36% in relation to the structure of PET/Al foil/LDPE (current film), respectively.

The structure PET/BOPPmet/LDPE (structure **2**) showed the second-best reductions of the environmental impacts, with 0.28 kg $CO_{2\text{-eq}}$ m^{-2} of GWP, $1{,}23 \times 10^{-4}$ PM2.5 m^{-2} of PMFP, and 1.02×10^{-3} kg $SO_{2\text{-eq.}}$ of TAP, representing a reduction of 29%, 39% and 36% in relation to the structure of PET/Al/LDPE (current film), respectively.

Figure 4 shows the contribution of the several materials to the impact category GWP. As can be seen, LDPE is the major contributor to GWP. With exception of structure **2** that is all PP-based, LDPE is the material with the higher content in the structure of the films explaining its major contribution to this impact category.

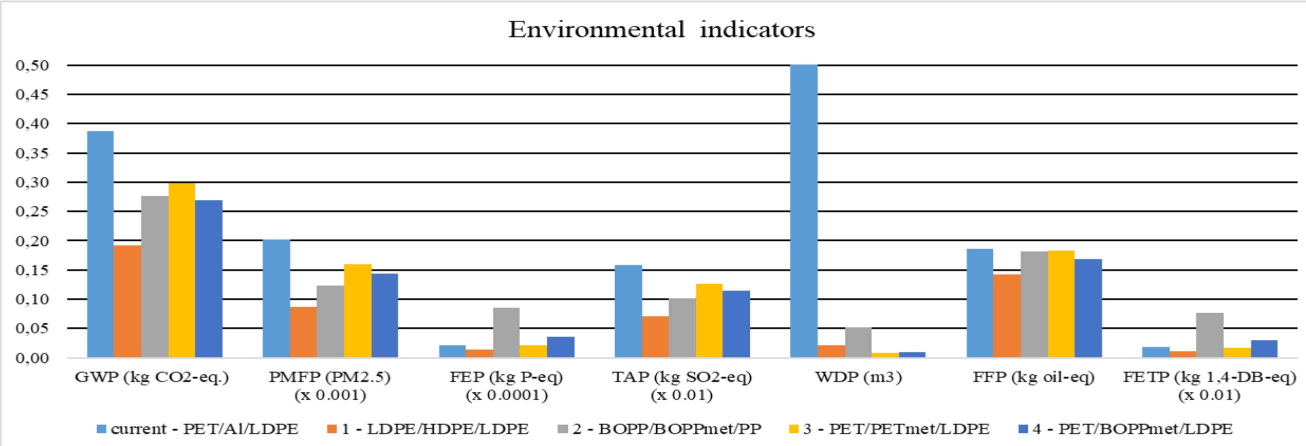

Figure 3. Environmental indicators of flexible plastic packaging for instant coffee. (Functional unit = 1 m^{-2}

As can be seen in Figure 4, aluminum has a large contribution (39%) to the impact category GWP. This is due to the production process of aluminum that demands a great amount of energy.

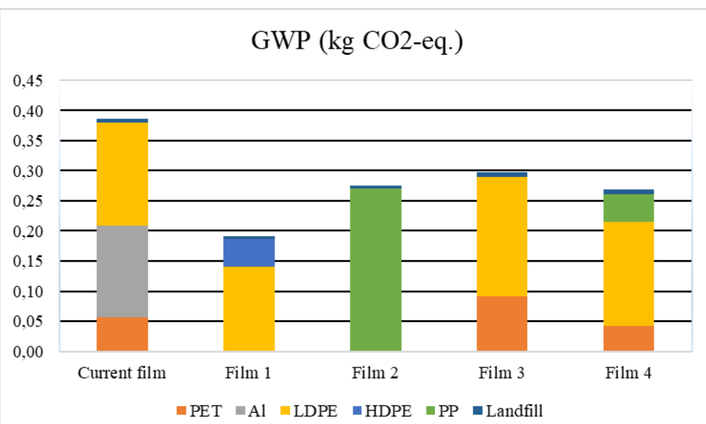

Figure 4. Contribution of the different materials to the impact category GWP.

4. Conclusions

Only alternative flexible plastic structure made exclusively by polyethylene (**1** = LDPE 33 µm/HDPE 33 µm/LDPE 32 µm) showed all pellet and film properties totally compatible with current established PE recycling lines according to the recycling protocols available. The good homogeneity of these recycled PE films was proven by appearance and transparence results. Although films from alternative materials **3** - PET 12 µm/PETmet 12 µm/LDPE 67 µm and **4** - PET 10 µm/BOPPmet 11 µm/LDPE 53 µm demonstrated incompatibility with PE recycling lines, they showed increase in important mechanical properties for using in some applications, such as high tensile and tear strength. Recycled films obtained from alternative materials **1**, **3** and **4** could be used for manufacture of plastic bags, and film **2** - BOPP/BOPPmet/PP – B.25 could be used as agricultural films. It is known that polyolefin recycling lines are well consolidated and functional processes. However, it is suggested to study the possibility of carrying out some changes in these processing lines so that multilayer flexible plastic materials can be incorporated considering the environmental gain that will be achieved, besides alignment with the circular economy. Regarding the environmental performance, the higher reduction of the environmental aspects evaluated in this study was also achieve with the adoption of structure **1** for packing instant coffee (50% less GWP, 57% less PMFP, 35% less FEP, 55% less TAP, 99% less WDP, 24% less FFP and 36% less FETP) in relation to the structure of PET/Al foil/LDPE (current film), followed by structure **4**. Therefore, based on the aspects evaluated in this study it is suggested the adoption of structure **1** for instant coffee in flexible packaging as a recyclable and lower environmental impact alternative in accordance with the goals of the circular economy.

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