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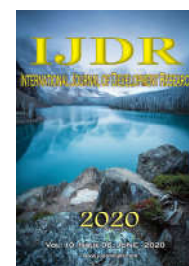
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RESEARCH ARTICLE

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RECYCLABILITY BY EXTRUSION MOLDING OF HIGH-DENSITY POLYETHYLENE FROM PLASTIC BAGS

¹Monteiro, J.R.G., ²Sireli, L., ¹Libano, E.V.D.G., ¹Bastos, D.C. and ^{*1}Pereira, P.S.C.

¹Escola de Engenharia, Fundação Centro Universitário Estadual da Zona Oeste, UEZO, 23070200, Rio de Janeiro – RJ, Brasil;

² Instituto de Macromoléculas Professora Eloisa Mano-IMA, Universidade Federal do Rio de Janeiro, 21941-598, Rio de Janeiro – RJ, Brasil

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*Corresponding author:

Pereira, P.S.C.

ABSTRACT

High-density polyethylene (HDPE) is one of the main materials used in the world, applied in the most varied human activities. Because of its extreme resistance, it takes years to degrade in nature, causing buildup of large volumes of material in landfills, when it does not end up in aquatic habitats due to the lack of awareness by society. Therefore, the recycling of this material is important. In this context, the present study describes the recycling of HDPE from plastic bags with four extrusion cycles in a twin-screw extruder. Changes in physical, chemical, thermal, and mechanical properties with reprocessing cycles were investigated by Fourier-transform infrared spectroscopy (FT-IR), X-ray diffraction (XRD), thermogravimetry/derivative thermogravimetry analysis (TG/DTG), differential scanning calorimetry (DSC) and tensile and flexion testing, as well as by determination of density and melt-flow index (MFI). The results indicated that post-consumer HDPE from plastic bags can be reused without affecting the mechanical properties for at least four processing cycles, enabling use in various products with satisfactory mechanical performance.

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INTRODUCTION

Mechanical recycling of plastics is attractive and environmentally friendly by minimizing waste and the consequent pollution, with lower energy consumption than that to produce virgin material (Nunes *et al.*, 2016). According to Hamad *et al.* (2013), in a review article of polymer recycling, mechanical recycling and chemical recycling are the most widely practiced methods, but, from an industrial point of view, mechanical recycling is most suitable because of its low cost and reliability. However, despite the advantages, recycling rates remain small (approximately 14%) in the plastic packaging field on a global scale (Sanchez *et al.*, 2020). Use of plastic products can only be reduced to certain extent but use of virgin material for manufacturing can be reduced by using recycling and management techniques. Post-consumer plastics (polyolefins) are especially useful materials to replace ceramics, wood, and metals because they are very functional, hygienic, light and economical. Examples of applications include construction of decks, marine pilings, pier and dock

surfaces, fences, and park benches (Singh *et al.*, 2016; Singh *et al.*, 2017). Recycling consists of four tasks, namely collection, separation, reprocessing and marketing. According to ASTM, there are four types of recycling: primary and secondary (mechanical recycling), tertiary (chemical, pyrolysis) and quaternary (incineration) (Singh *et al.*, 2016). Most plastic consumer goods are labeled with standardized resin codes marked inside a triangle (ASTM, 2013), identifying the chemical composition of the main polymer, which is used to sort and recycle compatible materials. These include polyethylene terephthalate (PET, #1); high-density polyethylene (HDPE, #2); polyvinyl chloride (PVC, #3); low-density polyethylene (LDPE, #4, which also currently includes linear LDPE [LLDPE]); polypropylene (PP, #5); polystyrene (PS, #6); and other polymers (#7). These codes are rarely present or legible in recovered plastic debris or small plastic fragments, so identification of the polymer must be accomplished using chemical testing.

From a technical perspective, the final quality is the main issue for mechanically recycled products (Junget *et al.*, 2018). Many studies have been published about recycling of plastics with or without adding organic and inorganic loads in the polymer

matrix (Martins *et al.*, 2019; Nunes *et al.*, 2016; Turku *et al.*, 2018). Nunes *et al.* (2016) studied a high-impact polystyrene (HIPS) and its nanocomposites with 3 and 5 wt% Cloisite 20A, processed in four extrusion cycles in a single-screw extruder. Changes in nanostructure, thermal, flow and mechanical properties and morphology of neat HIPS and its nanocomposites with reprocessing cycles were investigated. Mechanical properties showed a tendency to increase and decrease, or vice versa, during the extrusion cycles due to competition between crosslinking and chain scission reactions. Turku *et al.* (2018) tested the influence of accelerated weathering, xenon-arc light and freeze-thaw cycling on wood plastic composites extruded from recycled plastic. The results showed that, in general, weathering had a stronger impact on samples made from plastic waste compared to a sample made from virgin material. After weathering, the mechanical properties, tensile and flexural strength, were reduced. Incorporation of carbon black retained the properties during weathering, reducing the wettability of the sample, diminishing the change of mechanical properties, and improving color stability.

The use of an inorganic solid waste from the manufacture of fluidized-bed catalytic cracking catalysts as flame retardants in polymer-matrix composites of recycled HDPE was carried out by Martins *et al.* (2019). The incorporation of inorganic residue in the polymer material increased mechanical properties and fire resistance. The aim of this work was to process high-density polyethylene from plastic bags in four extrusion cycles in a twin-screw extruder. Changes in physical, chemical, thermal, and mechanical properties with reprocessing cycles were investigated by Fourier-transform infrared spectroscopy (FT-IR), X-ray diffraction (XRD), thermogravimetry/derivative thermogravimetry analysis (TG/DTG), differential scanning calorimetry (DSC), tensile and flexion testing, as well as by determination of density and melt-flow index (MFI).

MATERIALS AND METHODS

Raw Materials: The post-consumer high-density polyethylene (HDPE_{pc}) was supplied by the recycling company Peterlu (Seropédica, Rio de Janeiro) in pellet form, and was used as received.

Materials Processing: The processing of HDPE was carried out in a twin-screw extruder (TeckTrill Mod DCT-20), equipped with 10 temperature zones, ranging from 70 to 190°C from the feed to die, and rotating at 302 rpm. Figure 1 shows the scheme of the four processes performed (extruder 1, 2, 3 and 4). The samples were called: HDPE_{pc}, HDPE_{pc1}, HDPE_{pc2}, HDPE_{pc3} and HDPE_{pc4}, from the post-consumer to the fourth extrusion cycle, respectively.

Characterization

Density and melt-flow index (MFI): The materials were characterized according to density (ASTM D792-2013) and melt flow index, MFI (ASTM D1238-2013). For density analysis, the pellets were pressed for 300 s at 190 °C and 15000 MPa and cooled in a cold press for 60 s. The MFI analysis was performed with the polymer in pellet form.

Thermogravimetry and derivative thermogravimetry (TG/DTG): Thermal stability in HDPE samples was evaluated

using thermogravimetry/derivative thermogravimetry (TG/DTG), in a TA Instruments Q500 analyzer, performed under a nitrogen atmosphere, between 30-700 °C, at 10 °C/min. The initial (T_{onset}), maximum (T_{max}) and final (T_{final}) degradation temperatures were determined, as well as the presence of burnt residue. The TG/DTG test was also performed with the polymer in pellet form.

Differential scanning calorimetry (DSC): Differential scanning calorimetry (DSC) was performed with a TA Q1000 calorimeter. The samples were analyzed according to the following cycles. In the first cycle, the sample was heated from 0 °C to 200 °C, at a heating rate of 10 °C/min, leaving the material at 200 °C for 2 minutes under nitrogen atmosphere. In the second cycle, cooling was performed at a rate of 10 °C/min, to 0 °C, followed by a second heating cycle under the same conditions as the initial one (except for the waiting time of 2 minutes at 200 °C); finally, a second cooling cycle was carried out under the same conditions as the first one. The X_c value was determined based on the ratio between ΔH of the recycled HDPE and ΔH of the 100% crystalline HDPE (290 J/g). The DSC test was performed with the polymer in pellet form.

X-ray diffractometry (XRD): The X-ray analysis was performed in a Rigaku Miniflex diffractometer, using a nickel filter, $CuK\alpha$ radiation of $\lambda = 1.5418 \text{ \AA}$, at 20 kV, current of 20 mA, with 2θ between 2-30°, and data collected at 0.05 °/min. The specimens for XRD testing were obtained by pressing the pellets for 300 s, at 190 °C and 15000 MPa followed by cooling in a cold press for 60 s.

Fourier-transform infrared spectrometry (FT-IR): Fourier-transform infrared spectra were acquired using a Nicolet 6700 FT-IR spectrometer (Thermo Scientific). The samples were mounted on an attenuated total reflectance (ATR) accessory equipped with a ZnSe crystal prior to scanning. The spectra were obtained with accumulation of 120 scans and resolution of 4.182 cm^{-1} . The specimens for the FT-IR test were obtained by pressing the pellets for 300 s at 190 °C and 15000 MPa followed by cooling in a cold press for 60 s.

Mechanical property testing: The tensile and flexural properties of the samples were measured at 23 °C according to the respective standards ASTM D 638-2003 and ASTM D 790-2003. The specimens for the tensile and flexure test were produced by injection molding in an Arburg Allrounder 270s. The injection temperature was adjusted to 250 °C, with an injection pressure of 10 MPa. The tensile and flexural tests were performed at speeds of 50 mm/min and 14 mm/min, respectively, with an Instron universal testing machine (model 1101). All the properties are the average values of 7 specimens.

RESULTS AND DISCUSSION

Density and melt-flow index (MFI): Figure 2 (a) shows the density behavior of the HDPE_{pc} after four extrusion cycles. The results indicate a slight increase in density over the reprocessing cycles, suggesting an increment in the ordered arrangement of HDPE (Nunes *et al.*, 2016). Figure 2 (b) shows the MFI values after multiple extrusion paths. The MFI of the first and second extrusion cycles are quite similar. The MFI values exhibit, as a tendency, a slight decrease from the third extrusion cycle. According to the literature, the decrease in

MFI can be attributed to scission followed by crosslinking reactions in the HDPE chains (Johnston and Harrison, 1996; Nunes *et al.*, 2016).

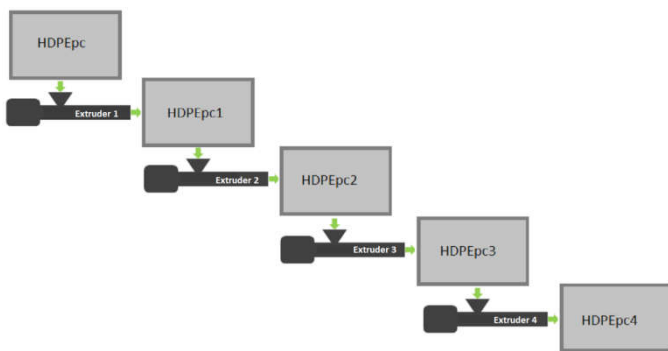


Figure 1. Processing Scheme

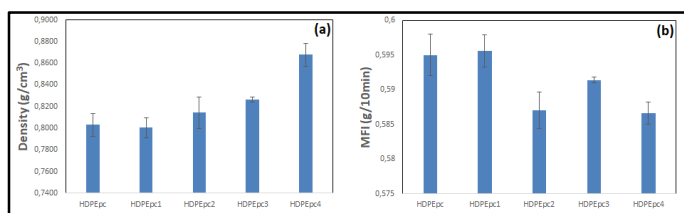


Figure 2. Density (a) and MFI (b) of HDPEpc for multiple extrusion paths

Thermogravimetry and derivative thermogravimetry (TG / DTG): The thermograms of the materials are shown in Figures 3 and 4, and Table 1 reports the values of the initial degradation temperature (T_{onset}), the temperature of maximum degradation (T_{max}) and the residual content of the materials after the different cycles.

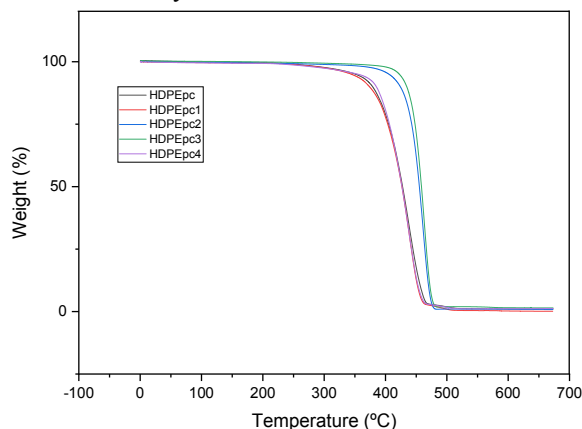


Figure 3. TGA curves of HDPEpc after the different cycles

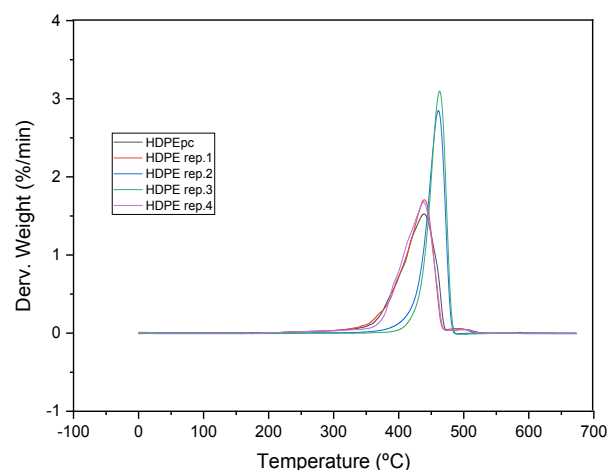


Figure 4. DTGA curves of HDPEpc after different cycles

Table 1. T_{onset} and T_{max} values of HDPEpc after the different cycles

Sample	T_{onset} (C°)	T_{max} (C°)	Residues (%)
HDPEpc	418	459 e 519	0,6
HDPEpc1	424	462 e 512	0,2
HDPEpc2	456	478	0,6
HDPEpc3	462	482	1,1
HDPEpc4	423	461 e 528	1,3

The profile of the curves was similar to that of HDPEpc. All materials degraded in just one step, leaving a small amount of residue, which can be attributed to the probable presence of small amounts of additives in HDPEpc, such as lubricants or inorganic pigments. T_{onset} and T_{max} tended to shift to higher values with increase number of processing cycles, suggesting an improvement in the thermal stability of the materials from the second cycle onward. This result was not expected according to the literature (Camacho and Karlsson, 2002; Martín-Alfonso and Franco, 2015; Nunes *et al.*, 2016). Some materials showed two T_{max} peaks, probably due to the variability of the sample from recycling.

Differential scanning calorimetry (DSC): All materials showed similar thermal behavior. The main results are shown in Table 2. The values of T_m and T_c remained constant. Considering the second heating, there was no variation in the crystalline melting temperature of HDPE (123°C) after the different cycles.

Table 2. Thermal properties of HDPE after the different cycles

Sample	T_m (C°)	T_c (C°)	X_c (%)
HDPEpc	105 e 123	113 e 115	32
HDPEpc1	104 e 123	113 e 115	36
HDPEpc2	105 e 123	113 e 115	42
HDPEpc3	104 e 122	113 e 115	41
HDPEpc4	104 e 123	113 e 115	45

A second melting temperature ($\cong 104$) was observed in all curves. According to Camacho and Karlsson (2002), this bimodal behavior suggests an indicator of degradability. The T_c (113 and 115°) presented the same values, indicating there were no changes in crystallization in all samples. An increase in the degree of crystallinity (X_c) was observed after the different processing cycles as a possible consequence of thermo-mechanical degradation, where degradation by chain scission followed by crosslinking predominated (Strömberg and Karlsson, 2009; Peres *et al.*, 2016).

X-ray diffractometry (XRD): Figure 5 shows the XRD diffractograms of the HDPEpc samples after the different cycles. It can be seen that the HDPEpc1 and HDPEpc3 samples presented the three crystalline peaks at $2\theta = 20.6^\circ$, 22.9° and 38.55° , characteristic of the planes (110), (200) and (020), respectively, in the crystalline polyethylene region (Abdalsalam *et al.*, 2020; Lazrak *et al.*, 2019; McDaniel *et al.*, 2015). The peaks at 14.35° , 17.15° , 25.7° and 44.7° do not reflect the polymer structure and may be related to the presence of additives or some contaminant in the sample, as it is a recycled material (Mahmoud *et al.*, 2017).

Fourier-transform infrared spectroscopy (FT-IR): FT-IR analysis was used to confirm that the chemical composition of HDPEpc from plastic bags corresponded to virgin HDPE. Figure 6 shows the FT-IR results. The HDPEpc peaks are

similar to the spectra of virgin HDPE, formed by the repetition of the $-(CH_2)$ group: peaks at 2950 and 2850 cm^{-1} , corresponding to C-H stretching vibrations; peaks in the 1450-1350 cm^{-1} region, corresponding to CH_2 bending vibrations; and peaks in the region of 730 to 717 cm^{-1} , referring to CH_2 rocking vibrations. HDPE also exhibits C-C bending, observed with absorption bands in the 1000-1250 cm^{-1} region (Lazrak et al., 2019). The spectra remained constant in all extrusion cycles, indicating no change of the crosslink reaction.

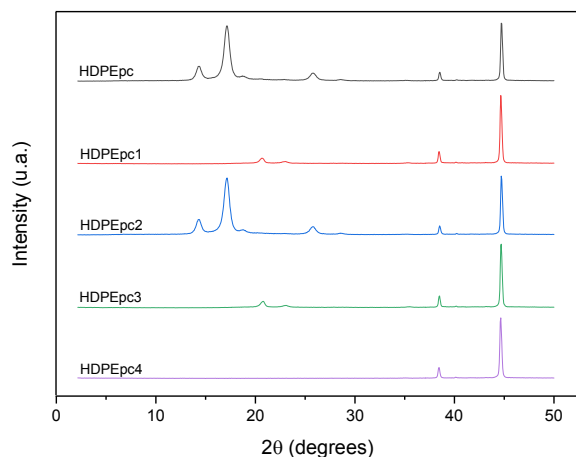


Figure 5. XRD patterns of HDPEpc after the different cycles

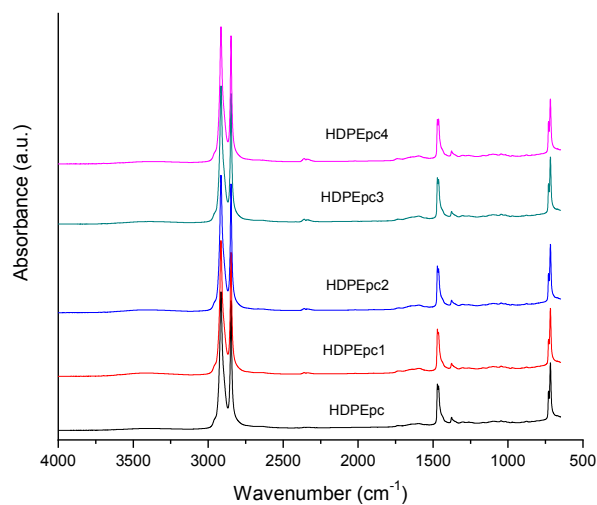


Figure 6. FT-IR spectra for multiple extrusion paths

Mechanical property testing: Table 3 shows the mean values of tensile strength, elongation at break and elastic modulus of samples after multiple extrusion cycles.

Table 3. Tensile properties of HDPE for multiple extrusion paths

Sample	Tensile strength (MPa)	Elongation at break (%)	Elastic modulus (MPa)
HDPEpc	13.2±0.16	225.0±9.76	227.4±15.34
HDPEpc1	13.2±0.11	230.5±4.09	246.2±6.64
HDPEpc2	13.1±0.21	232.7±8.81	244.7±9.28
HDPEpc3	12.5±0.32	192.7±22.58	241.9±2.51
HDPEpc4	12.2±0.06	181.9±0.18	241.3±7.98

A trend can be observed of a slight decrease in the value of tensile strength and elongation in the third and fourth extrusion cycles. This decrease can be attributed to a slight increase in rigidity (Young's modulus) of HDPE, possibly due to

crosslinking reactions (Boldizar et al., 2000; Nunes et al., 2016). The results, however, show that the investigated HDPEpc from plastic bags did not degrade to any significant extent during four reprocessing cycles. Table 4 shows the results of flexion testing. No significant changes were observed along the reprocessing cycles. The flexion test results corroborated the tensile tests. Therefore, the mechanical results indicated that HDPEpc can be reused without affecting its mechanical properties for at least four processing cycles, aiming to make products with satisfactory mechanical performance.

Table 4. Flexion properties of HDPE for multiple extrusion paths

Sample	Force (N)	Tensile strength (MPa)	Elastic modulus (MPa)
HDPEpc	16.0±0.4	10.3±0.3	285.3±23.5
HDPEpc1	15.7±0.3	9.9±0.3	309.3±7.0
HDPEpc2	15.8±0.0	10.0±0.1	315.5±16.8
HDPEpc3	16.4±0.9	10.3±0.6	311.17±14.5
HDPEpc4	16.2±0.3	10.2±0.3	323.1±13.3

Conclusion

In this study, the properties of post-consumer high-density polyethylene from plastic bags were analyzed with four extrusion cycles in a twin-screw extruder. Changes in physical, chemical, thermal, and mechanical properties with reprocessing cycles were investigated. The slight increase in density could suggest an increment in the ordered arrangement of HDPE. The MFI exhibited a tendency for slight decrease from the third extrusion cycle, which could be attributed to scission followed by crosslinking reactions in HDPE chains. There was an improvement in the thermal stability of the samples from the second processing cycle onward and an increase in the degree of crystallinity after the different processing cycles, a possible consequence of chain scission followed by crosslinking. X-ray analysis showed HDPE peaks and others that did not reflect the polymer structure and might have been related to the presence of additives or some contaminant in the samples, since they were made from recycled material. FT-IR results showed that the post-consumer material's peaks were similar to the spectra of virgin HDPE, formed by the repetition of the $-(CH_2)$ group. The spectra remained constant in all extrusion cycles, without indication of any change in the crosslinking reaction. The mechanical results indicated that HDPEpc from plastic bags can be reused without affecting their mechanical properties for at least four reprocessing cycles, to make products with satisfactory mechanical performance aiming at sustainability.

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