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Properties of Simulated Plastic Waste Mixtures in Upcycling Processes: An Experimental Evaluation

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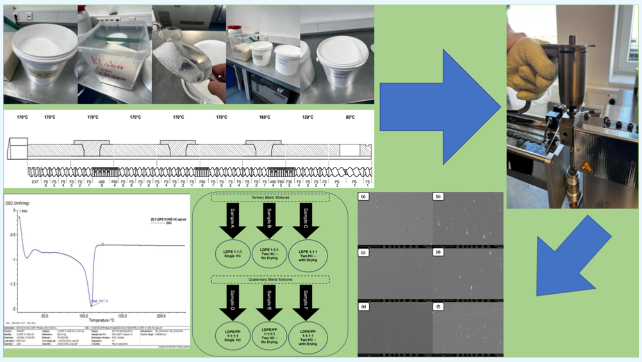
ABSTRACT: Current production of virgin plastics stands at approximately 0.4 billion tonnes annually, with significant applications in packaging, construction, and the automotive industry. Despite the utility of plastics, their waste management remains a challenge. This study focuses on upcycling plastic waste (PW) through mechanical recycling, which adds value to discarded plastics and aligns with sustainable practices. We have investigated the mechanical and thermal properties of ternary and quaternary blends of the most common polyolefin polymers, low-density polyethylene and polypropylene (PP), simulating typical PW mixtures. The blends were prepared using twin screw extrusion for melting, mixing, and strand extrusion. The strands from the extruder were guided through a water bath, optionally dried, and pelletized, to determine the processing conditions variations. Aliquots of the blends were directly taken from the extruder to form specimens from the melts. Additionally, the produced pellets were remolten to form specimens, resulting in a second heating cycle (HC). Thermal stability, calorimetric properties, surface morphology, and mechanical characteristics of the blends were analyzed. Results indicate that ternary blends exhibit higher thermal stability but lower mechanical strength compared with quaternary blends. The addition of PP slightly reduced the onset temperature in quaternary blends. The melting temperatures of the blends show minimal change with additional HCs or drying, suggesting the maintenance of thermal properties. The ternary blends exhibit consistent mechanical properties regardless of the processing conditions, but the breakup force and tensile modulus of the quaternary blends were lower with additional HCs. The surface morphological studies revealed increased cavitation with two HCs and improved surface smoothness with drying. These findings support the potential of mechanical recycling to produce marketable plastic grades from mixed PW.



1. INTRODUCTION

Increase in demand on the variety of polymeric grades is tied directly to their production capacity, population growth, and the dependency on durable plastic products with high-end and acceptable market standards.[1] The inexpensive and durable plastic articles produced and used on a daily basis make it even harder to replace in our modern day and age. This supports the fact that the properties of such products are still upheld with the highest regard in the production cycle for consumer satisfaction. The global market is still dependent on plastics for a plethora of applications, as judged by the rapid increase in its global production capacity. The current production rate of all virgin plastics is some 0.4 billion tonnes annually,[2] which covers the main applications sectors, such as packaging (44%), construction (18%), and the automotive industry (8%).[3] To date, and due to the recent Covid-19 pandemic whereby plastics played a major role, 1.6 million tonnes of plastics are produced on a daily basis with a recycling rate >10%.[2] The global medical sector, which covers the high demand of personal protective equipment, reached a production capacity of 69 billion units on a monthly basis.[4] Some 50% of all plastic waste (PW) is still managed through landfilling, which is the least favorable option for the management of solid waste.[5] The majority of the remainder (waste) is either incinerated (with or without an energy recovery configuration) or ends up in various environmental sinks.[6]

The mismanagement of PW is quite irksome for urban environmental development and sustainable practices. About 150 million tonnes of plastics exist in the marine environment,[1] and an estimated rate of 8 million tonnes of macro-PW that potentially turns into microplastics (MPs) enter the oceans on an annual basis.[7] PW is typically managed through a hierarchy of methods, which starts with primary (i.e., in-house or within facility) recycling solutions for plastic scrap, secondary

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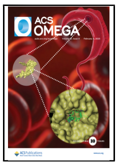

Table 1. Summary of Main Mechanical Recycling of Plastics Studies Utilizing Blends of Common Grades of Plastics and Reporting the Impact on Properties Examined

reference	plastic types studied	mechanical recycling method/preparation	main findings
Satya and Sreekanth[27]	recycled polypropylene (rPP) and recycled high-density polyethylene (rHDPE)	extrusion/palletizing of feedstock and injection molding for specimen preparation (two heating cycles−HC− are introduced)	○ mechanical properties (i.e., tensile strength, Young's modulus, and hardness) of both rPP and rHDPE were lower than their virgin grades○ rPP showed better properties, in comparison to rHDPE. Addition of fillers was recommended to enhance properties
Jubinville et al.[28]	virgin HDPE was processed six times to simulate mechanical recycling	melt mixing and injection molding of HDPE and then reactive extrusion to prepare maleated MA-g-HDPE resin	the recycling of HDPE increased its viscosity which led to an increase in its tensile strength (due to the thermo-mechanical degradation mechanism)
Eriksen et al.[25]	polyethylene (PE), polyethylene terephthalate (PET), and PP reprocessed/reclaimed PW	extrusion and palletizing for the 25 different sorted samples	○ PET was determined to be highly suited for closed-loop recycling, meeting bottle standards for the Danish case considered○ PE tensile strength was higher compared to virgin PE○ PP mechanical properties were also higher compared to PP virgin grades
Jin et al.[29]	virgin low-density polyethylene (LDPE) was extensively recycled 100 times	extrusion/palletizing of feedstock	○ simulated extensive mechanical recycling did not show much effect on melting and crystallization temperatures of LDPE○ thermal degradation had a high impact on rheological properties of recycled LDPE. This was attributed to cross-linking and chain scission
Van Velzen et al.[30]	six reclaimed/recycled and sorted types of HDPE	extrusion/palletizing of feedstock	the highest impact resistance was determined from the single grade PE bottles
Stanic et al.[31]	blends of seven PP types, two long-chain branched PP and five linear polypropylene grades	reactive extrusion with two peroxides	○ mechanical properties were not changed to a large degree in comparison to the original material○ potentially, elongation at break could be increased○ upcycling could be achieved with blending
Bahlouli et al.[26]	blends of two virgin PP types processed up to 12 times	extrusion/palletizing of feedstock	○ reported past findings that PP requires up to five HCs to start showing effect on measured properties○ morphology analysis showed embrittlement of the amorphous matrix by the chain scission and cavitation○ higher stability was shown for blends with talc
Bertin and Robin[32]	blends of virgin and reclaimed LDPE and PP	extrusion/injection molding	compatibilizing agents were used to improve the following properties: elongation at break and impact strength
Santos and Pezzin[33]	PP/recycled PET composites	extrusion/injection molding	mechanical properties improved with the addition of PET

(mechanical recycling utilizing physical methods), and finally, chemical and energy recovery technologies.[8−10] Although recovery of fuels and energy has received quite an increase in attention lately due to the fact that light fuel-cuts, diesel, and heat are commonly produced from such techniques,[8,11−13] the fact remains that (i) such technologies require capital costs that make them (in general) a luxury in our modern energy intensive times, (ii) geopolitical issues have made auxiliary (background) energy to support these techniques somewhat out of reach, whereby various western European countries have started considering coal once again as a major component to their energy basket, and finally, (iii) mechanical recycling is more established and can produce with ease marketable grades of polymeric materials that are required on a daily basis.[14−21]

The management of waste, and in particular PW, in small terrain countries via upcycling methods is essential. This is due to the fact that land scarcity presents an issue, in addition to, having an economical and sustainable practice to boost local economies too.[22] Prime examples of such countries include Kingdoms of Bahrain and Brunei, States of Qatar and Kuwait, and South Korea. Upcycling is defined as the process of adding value to discarded PW.[1] It is also typically associated with the aesthetic aspects of the final product nowadays. Mechanical recycling fits this definition perfectly, as it involves reforming and reshaping the original form of the PW. However, physical blending is essential to add value to the process.[8] It also provides numerous advantages over other technologies, such as incineration whereby greenhouse gases (GHGs), dust, and toxins are not emitted by its application.[23] Mechanical recycling provides various other advantages too, such as its ability to have PW act as an additive for reinforcement and provide market-grade products ready for commercial use, especially using melt blending via heat extrusion.[24] Control over processing conditions will also enable the recycler to overcome common problems faced, such as the deterioration of the polymeric matrix, loss of mechanical integrity, and the desirable appearance of the final product.[24−26] Table 1 reviews studies reporting the properties of common plastic grades used globally on the market.

A number of major technical findings could be deduced from past related literature reports on the subject matter. The lifecycle of plastics, reaching its end point (i.e., discarded items/grave or even energy utilization as a recovery option), is tied directly to GHGs emission, namely, the carbon footprint of the process (i.e., 56 GT by 2050).[34] It is imperative to start utilizing upcycling processes, including mechanical and thermo-mechanical options, with an ultimate design of end products to reduce environmental burdens associated with such. This is related to the energy consumption and losses in small- to medium-scale recycling options available on the market for main plastic groups, which were reviewed extensively by Uzosike et al.[35] It was recommended to start reducing losses of energy in extrusion processes and utilize renewables more effectively in powering up recycling facilities. Furthermore, the advantages of mechanical recycling plastics outweigh their disadvantages and could easily be applied as a viable option to boost circularity in local economies and reduce environmental burdens.[25,35] In addition, mechanically recycled blends show a somewhat loss of properties when compared to the original virgin-grade polymers (see summary in Table 1). Talc, PET fibers, and reinforcement with glass powder have shown popularity as a filler to mechanical recycling blends, despite cost addition to the compounding process.[26,33,35] Jin et al.[29] have shown loss of rheological properties of LDPE due to loss of thermal integrity with mechanical processing. This also indicates the degressive nature of the process, which has to be considered carefully (and recommended to be nature- and application-specific) for each blend.

Upcycling is also a lucrative route in reducing costs associated with PW management, whereby in many cases, it is cheaper to produce virgin polymer grades than to recycle them.[1] This is why it is quite important to understand the effect of common plastic thermal mixtures produced in mechanical recycling. It is also paramount to increase the efficiency of upcycling by understanding the effect of processing conditions typically used in recycling facilities, such as air drying and the introduction of various heating cycles (HCs) to thermoplastics, which are common practices conducted in recycling facilities. Polyolefin (PO) polymers make up the major share of the municipal component of PW. This is due to the fact that the majority of “single use” plastics are used for packaging. These include HDPE, LDPE, LLDPE, and PP, in addition to PET and PS.[36] The main component of typical PW is comprised of PE which is reported in some cases to make up some 98% of the collected PW,[37−39] and kerbside collection of waste has previously shown that PO polymers constitute some 40% of the total waste component.[24]

Therefore, this study was initiated to assess and analyze the mixtures of the most common plastic grades typically found in PW mixtures from various sources, determining the change of properties with various blends and processing conditions to understand the future potential of upcycling such blends using mechanical treatment (recycling) methods. This involves the following specific objectives of the work at hand: (1) acquirement and thermal blending of the two most common PO polymers found in PW mixtures, i.e., PE and PP, (2) preparation and compounding of these blends to simulate PW mixtures, (3) determination of the properties of three PE-based blends (ternary mixture) and three PE and one PP blend (quaternary mixture) to simulate common PW, (4) comparison of the blend’s morphological and thermal properties against single virgin grades, and finally, (5) assessment of the circularity potential of plastics upcycling and regulations associated with such mixtures.

2. EXPERIMENTAL SECTION

2.1. Materials. The materials used in this study were acquired as virgin commercial grade polymers for both LDPE and PP, to simulate ternary and quaternary blends of typical recycled PW. Three LDPE grades were used in this work. The first grade was an LDPE Klüber Cleanplus LDN (IMCD Deutschland GmbH, Germany) with a melting point (T_m) reported as 114 °C and a processing (melt) temperature range between 160 and 240 °C. The second LDPE was a Lupolen 1800H (LyondellBasell, Wesseling, Germany) grade with a T_m of 108 °C and melt temperature between 180 and 230 °C (ρ = 0.919 g·cc^{-1}). The third LDPE used was a Lupolen 1800S (LyondellBasell, Wesseling, Germany) grade with a T_m of 106 °C and melt temperature of 180−230 °C (ρ = 0.917 g·cc^{-1}). Finally, the PP grade used was a Moplen HP501H (Homo PP, LyondellBasell, ALIBIS, Germany), with a T_m of 160 °C (ρ = 0.900 g·cc^{-1}). All materials used were in pellet form (≈2−3 mm) used as delivered as per the pictorial depiction in Figure S1.

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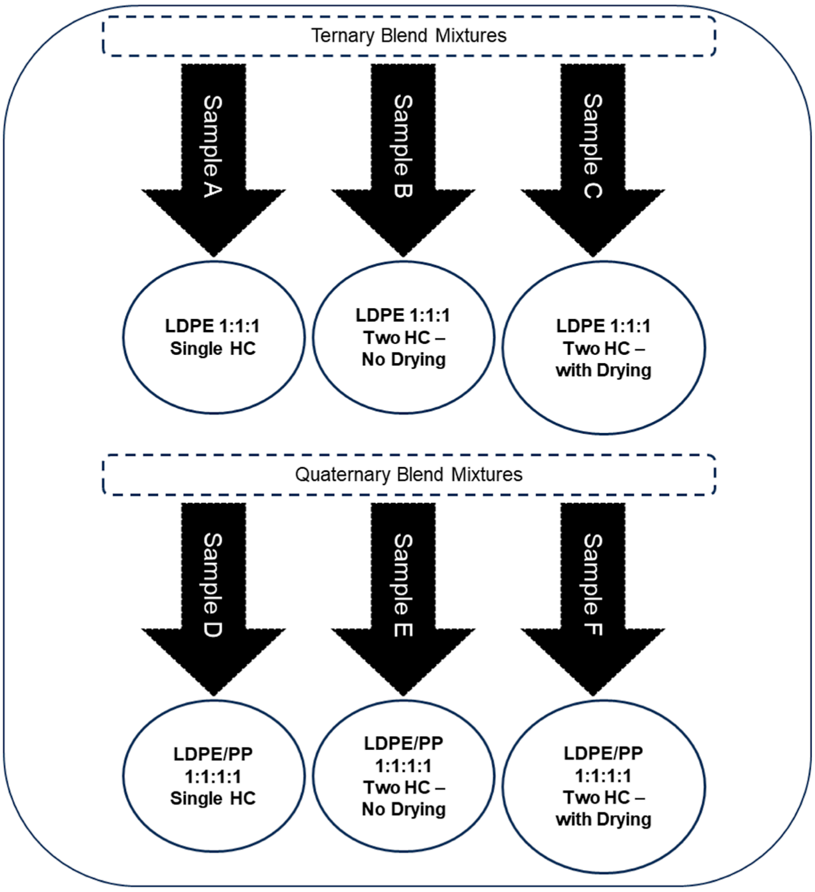

Figure 1. Schematic representation of blend mixtures conducted for this work.

2.2. Specimens Preparation.

Melt blending and extrusion were used to prepare the ternary and quaternary mixtures with different processing conditions in order to investigate their effect on the properties of the final products. The ternary blend was produced by incorporating the three LDPE grades in a 1:1:1 ratio using a 100 g sample of each material, mixed manually at laboratory conditions (Figure S2). The melt blending was conducted with a parallel corotating twin screw extruder (Thermo Scientific Process 11, Germany, $D = 11$ mm, $L/D = 40$, Thermo Fisher Scientific) using a three mixing stages standard screw configuration (Figure S3). The extrusion profile is shown in Figure S4, maintaining actual measured temperatures between 80 and 170 °C for the eight zones of the extruder unit (200 rpm, 300 $g \cdot h^{-1}$ throughput), and a die head temperature (DHT) of 170 °C was maintained throughout the extrusion process. The residence time equaled 1 min measured using a Kollicoat IR Red pigment anticipating first appearance (Figure S5). To investigate the effect of processing conditions, investigating simulated recycled blends of PW, three batches were produced from the ternary blend. The first is a single HC whereby the extruded material was not cooled down using the water bath nor palletized but injected directly into the injection molding machine (HAAKE MiniJet Pro Injection molding machine, Thermo Fisher Scientific) to produce the desired specimen for further characterization (Figure S6). The second batch of the prepared mixture was produced by cooling down the extruded blend to room temperature (RT) using a water bath and pelletized in a knife mill (Thermo Scientific VariCut 11 Strand pelletizer, Thermo Fisher Scientific) to approximately 2 × 2 mm pellets (Figure S7). The extrudate was then fed into the MiniJet injection molding machine to produce desired specimens, hence introducing two HCs for the prepared recyclate which is a common practice in PW processing subjecting it to more than one HC. The third batch of the mixture was prepared with the aim of investigating the effect of drying using 0.5 bar air pressure at the exit of the water bath. The other conditions from the second mixture preparation stages were maintained. The quaternary blend was achieved by incorporating the PP with a 1:1 ratio similar to that of the previous LDPE ternary procedure (Figure S8). Three batches were produced as well for the quaternary mixture using an extrusion temperature profile maintained between 100 and 220 °C (DHT = 220 °C) and an RPM of 200 at 300 $g \cdot h^{-1}$ throughput (Figure S9). The three batches, following the previous procedure depicted for the ternary mixture, were a single HC injected directly into the specimen mold from the extruder's die, palletized two HC mixture with air drying, and palletized two HC mixture without air drying (Figure S10). The residence time measured for the quaternary mixture was 1.2 min, determined using the same procedure. Figure 1 summarizes the blend mixtures and ratios/compositions produced and used in this study. Finally, specimens were introduced into the injection molding machine to produce specimens for further analytical testing. The cylinder temperature for the ternary mixture was set to 170 °C and 70 °C for the mold at 300 bar pressure held for 5 s. The quaternary mixture was produced using a set temperature for the cylinder equal to 200 °C and 90 °C temperature for the mold at 150 bar pressure held for 5 s (Figure S11). The experimental data compiled for the preparation are shown in

Table S1. The mold chosen was a dog-bone ISO 572−2−1BA with a thickness of 2 mm, midlength of 30 mm (total length 75 mm), and a width of 10 mm (see Figure S11). Figures S12 and S13 show a depiction of the screw configuration and heating zones used in the preparation stages of both the ternary and quaternary blends.

2.3. Thermogravimetric Analysis. Thermogravimetric analysis (TGA) was conducted to investigate the thermal stability of the materials studied by using a Shimadzu (Model TMA DTA 60 Hz) thermogravimetric setup for each conducted test. A sample weight of approximately (∼10 mg) was used with a start set point at laboratory (room) temperature (∼25 °C) until 800 °C in an inert environment with a N_2 flow rate of 50 mL·min^{-1} utilizing a 10 K·min^{-1} temperature ramp (β). A calibrated platinum pan was used for each experimental.

2.3.1. Differential Scanning Calorimetry. The thermal and calorimetric results were studied using differential scanning calorimetry (DSC) (NETZSCH 240F1 Phoenix), which was calibrated and used following standard practice for plastics measurements.[38] The sample weight used in each experimental run was about 6−7 mg with a single cycle of a temperature ramp (β) equal to 10 K·min^{-1} from room temperature (RT) to 500 °C under a constant N_2 flow rate of 50 mL·min^{-1}. T_m for each specimen was monitored on the exothermic (upward) curves obtained.

2.3.2. Scanning Electron Microscopy. Morphological studies were conducted on the samples prepared, using the manually cut sections from the middle part of each sample's mold. 5 kV voltage was used on different magnifications for the analysis using a scanning electron microscope (Phenom XL Benchtop SEM, Thermo Fisher Scientific) postcoating each individual sample prior with gold sputtering (5 nm) using a sputter coater (LUXOR gold coater, Luxor-Tech) (Figure S14). Samples were analyzed using a G301 0.5″ aluminum stub on carbon tape in the SEM setup.

2.4. Mechanical Properties. Mechanical profiling by obtaining stress vs strain curves was conducted for the studied specimens at RT (22−25 °C), to determine the effect of blending at different conditions following the guidelines of ASTM D638−14. Experiments were conducted using an Autograph Model Trapezium X, Shimadzu Universal Testing Machine (UTM) using a speed of 1 MPa·s^{-1} with a 20 kN load cell. The measured properties include force at break (MPa), tensile modulus (MPa), maximum stress (MPa), and maximum strain (%).

3. RESULTS AND DISCUSSION

3.1. Thermal Stability. The thermal degradation properties for all tested materials are depicted in Table 2, with experimental thermograms including the first derivative curve (DTG) appended in Figures S15−24. TG data sets are commonly used to study the thermal stability of materials, kinetics of degradation, weight loss behavior, governing mechanism of deterioration, and unit design.[38] Figures S15−18 show the virgin grades thermograms whereby the onset temperature (T_{os}) for the three used virgin LDPE grades was between 310 and 350 °C (see Table 2). This is slightly lower than past reports on LDPE T_{os} but in general agreement with PO polymers degradation behavior.[38−41] Past work on virgin PP was in large agreement with the results obtained in this work,[25,41] which showed a T_{os} of 390 °C (Table 2).

Table 2. Thermal Properties of All Samples Obtained from the Thermal Analysis[a]

specimen code	$T_{os}{}^1$ (°C)	$T_{if}{}^2$ (°C)	$T_{es}{}^3$ (°C)
virgin materials/polymers			
LDPE LDN	310	471	740
LDPE 1800H	360	471	735
LDPE 1800S	350	468	750
Homo PP HP501H	390	452	620
ternary blends			
A: LDPE one HC	360	468	775
B: LDPE two HC/no drying	325	464	775
C: LDPE two HC with drying	350	469	790
quaternary blends			
D: LDPE/PP one HC	340	445	720
E: LDPE/PP two HC/no drying	350	451	790
F: LDPE/PP two HC with drying	350	441	680

[a]Note to reader: (1) Estimated as onset temperature after 5% of degradation,[39−41] (2) estimated as the inflection (point) temperature on the DTG curve, and (3) estimated as the end set (maximum) temperature with no monitored change in weight %.

Minor differences were observed for the ternary blends obtained and studied in this work. The T_{os} for the samples exposed to one HC had the highest T_{os} (360 °C) when compared to the samples exposed to two HCs without drying (325 °C) and with drying (350 °C). The addition of PP in the quaternary blends reduced T_{os} significantly for the samples exposed to a single HC (340 °C), especially when compared to those of the ternary blends and the other quaternary blend samples (Figures S19−S24). There were no significant changes in the inflection point (T_{if}) between the blends studied, and the residue amount determined experimentally at the end of each run was about 1%. Past work showed that addition of waste to virgin PO polymer blends had minimal effect on the T_{if} and T_{os}.[38]

Eriksen et al.[25] studied PE samples' thermograms noting that 5 to 10% degradation occurred when lids and labels were not present, as to PE samples without contaminants. This is a direct indicator that a monopolymeric (homogeneous) matrix degradation occurs when contaminants are not present and when HCs are reduced in number due to further cross-linking and heat exposure.[28] This was also apparent when compared to the quaternary blends, as the PP slightly reduced T_{os} (Table 2). On the other hand, the increase in HCs number has increased the quaternary blends T_{os}. PP is susceptible to chain degradation, loss of stabilizers, and loss of amorphous regions when degraded.[41] This has shown a clear effect on the release of free radicals and cross-linking after termination to increase the bulk nature of the blend and increase the amount of energy required to deteriorate it. The results obtained herein can also point toward the applicability of processing quaternary blends in thermal or thermo-chemical units, more advantageously over ternary ones, due to reduced T_{os} obtained which can further reduce their processing costs due to lower energy requirements. Furthermore, ternary blends have shown more thermal stability than quaternary blends, which is attributed to the fact that single (mono) PE blends are highly stable compared to heterogeneous mixtures and blends.

3.2. Calorimetric Thermograms Analysis. The melting temperatures (T_m) of the studied specimens were studied on the DSC calorimetric thermograms obtained. The virgin grades used were studied at the first stage, and their calorimetric data are shown in Figures S25−S28. The T_m observed conformed with the manufacturer's reported data. Plastics of PO nature are semicrystalline materials with a structure that facilitates high degree of degradation due to their susceptible nature to oxygen (O_2) accessibility in their matrix.[42] Al-Salem et al.[38] showed in their study that PE waste and its blends had a notable degree of rearrangement of the crystalline phase due to the high mobility of the amorphous region in the polymer's matrix, loss of chain ends of the polymeric chains, and high ability of surface contraction on the materials surface. These reasons combined can affect the crystalline zone studied using the DSC analysis and the melting behavior of the samples.

Figures S29−S31 show the ternary blend DSC analysis (i.e., samples A, B, and C). The increase in HCs and the effect of drying showed a minimal effect on the T_m observed which was determined between 110 and 111 °C. This indicates that the blending of ternary LDPE blends has no major impact on the melting behavior and can produce products with somewhat similar thermal properties and market standards. This is also in line with the previous TGA thermograms obtained and discussed in the previous section, which showed no major effect on the T_{os} for the ternary blends. A similar behavior and results were noted for the quaternary blends which are compiled in Figures S32−34. This indicates that the addition of PP has no significant impact on the melting behavior of the blends. The morphology changes are not necessarily tied to the large change of T_m especially with PP blends, which were reported to fluctuate slightly (between one and two degrees) in past findings of different blending cycles.[26] This fluctuation is attributed to lowering the average molar mass of the blend reprocessed and exposed to a number of HCs. Jin et al.[29] also reported no major differences on T_m for reprocessed PE, but rheological properties were affected nonetheless due to the matrix exposure to HCs. However, extensive extrusion in mechanical recycling of the blends in our case can reduce crystallinity.[29] This also points toward the fact that cross-linking is prevalent over chain scission in extensive mechanical recycling. The mechanism of thermo-mechanical degradation in mechanical recycling of PE was previously described in detail by Jubinville et al.,[28] whereby cross-linking is described as the recombination of groups which increase chain length of the polymeric matrix. Chain scission promotes separation of the C−C bonds. In the case of PP, tertiary carbon groups must be accounted for in such mechanisms. The use of fillers is also recommended for the design of plastic products with prolonged longevity and durability for the blends studied at hand to manipulate the crystalline zone of PE and PP.[28]

3.3. Morphological Studies.

The morphology of the studied specimens was analyzed and studied by subjecting the ternary and quaternary blends to SEM analysis. Figure 2 shows SEM micrographs of the ternary blends studied in this work. It was clearly noted that the addition of heating loops or cycles (HCs) creates more cavitations and disassociation in the polymeric matrix coupled with a blistering effect typically created from the shape molding or extrusion die.[37−39] Comparing the drying effect between samples B and C (Figure 2c−f), smoother and better surfaces were created by drying. Bulk crystalline zones were also noticed in the micrographs which increased with the HCs number. This could be attributed to the creation of a molten state and reshaping the matrix a second time.

Figure 3 shows the quaternary blend samples micrographs. At first glance, the samples were clearly immiscible with prominent crystalline zones clearly visible. This indicates that LDPE and PP are very hard to be physically characterized as homogeneous and their blends are well noticeable. In addition,

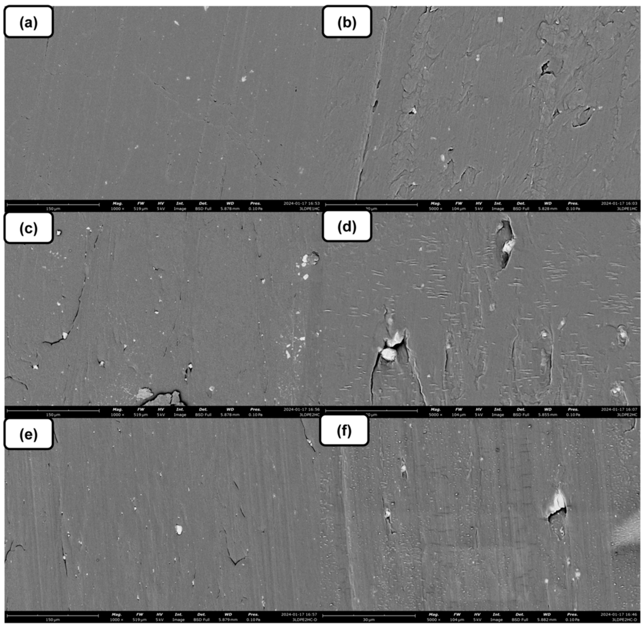

Figure 2. SEM micrograph of the studied specimens showing the ternary blends, in particular sample A (LDPE) blended with one HC at (a) 1000× magnification and (b) 5000× magnification, sample B (LDPE blended with two HCs with no drying) at (c) 1000× magnification and (d) 5000× magnification, and sample C (LDPE blended with two HCs exposed to drying) at (e) 1000× magnification and (f) 5000× magnification.

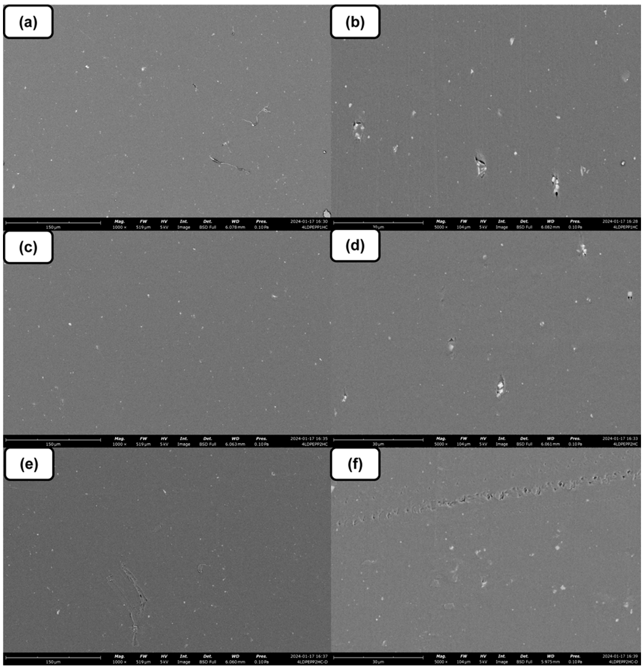

Figure 3. SEM micrograph of the studied specimens showing the quaternary blends, in particular sample D (LDPE/PP) blended with one HC at (a) 1,000× magnification and (b) 5,000× magnification, sample E (LDPE/PP blended with two HCs with no drying) at (c) 1000× magnification and (d) 5000× magnification, and sample F (LDPE/PP blended with two HCs exposed to drying) at (e) 1000× magnification and (f) 5000× magnification.

drying has also created (relatively) smoother surfaces (Figure 3c−f) between the samples exposed to single or dual HCs, relatively proportional to the injection molding temperature too. The mosaic patterns found on the LDPE/PP blends surfaces are also an indicator of incompatibility of both polymers on a physical blending level noticeable with the clear crystalline zones detected visible as white spots.[38]

The creation of voids was previously noted by Bahlouli et al.[26] on PP surfaces when exposed to a number of HCs with and without fillers. This was also supported by our findings depicted in Figure 3. LDPE/PP blends are generally known to be incompatible on a morphological level especially when samples are rich in LDPE (as it is in the studied case at hand).[32] Such blends rich in LDPE create a heterogeneous dispersion effect and have a two-phase mixture due to poor interfacial bond strengths between both polymers. Such an effect is responsible for weaker mechanical properties reported in previous well-documented studies.[43−45] It is therefore recommended to add compatibilizers if increase in mechanical properties is desired.[32]

3.4. Mechanical Characterization. Monitoring the stress−strain curves can be of immense importance to study the plasticity and behavioral changes. Figures S35−S40 show the ternary and quaternary blend stress−strain curves obtained from the mechanical profiling experiments conducted. A somewhat classical mechanical behavior was observed under tensile loading, while a linear elastic response was noted for a majority of the samples. A viscoelastic response before the yield point was not clear in the obtained data set pointing toward hardened samples in their nature.[37] The elastic region and the yield onset are governed by the glass transition temperature (T_g).[46] After which, the deformity of polymers is considered to be plastic.[26] The drop knee (break) observed after the yield point in the samples studied shows voidage or space creation permitting the disentanglement of macroscopic amorphous chains of the polymeric matrix.

The ternary blends show a clear and pronounced knee just after the yield point, whereby it could be observed that the ternary blend with one HC shows a lesser extension of the stress−strain curve in comparison to others (Figure S35). Quaternary blends were clearly more susceptible to breakage (Figures S38−S40), as a clear drop (e.g., failure) is detected in the curve after the yield point almost immediately. No necking was observed in all tested samples, which is attributed to the nature of the material and the shape of the standard mold and size of each test specimen.

Table 3 shows the force measured at break (MPa) for the studied samples. Pertaining to the ternary blends, it was observed that drying of the samples maintained the original value of the measured force (119 MPa) whereby the wet samples exhibited a higher force value of 132 MPa. Due to the physical immiscibility of the quaternary blend, previously discussed, the increase in HC helped reduce the force magnitude (Table 3). This could be attributed to the fact that increasing heating loops can help homogenize the studied samples, resulting in a more uniform polymer matrix, henceforth, requiring higher force to break. Drying of the samples also helped maintain the tensile modulus value for the ternary blend (0.119 MPa). Then non-dried (wet) samples were higher in the tensile modulus value (0.132 MPa). Typically, the increase in HCs which reflects recycling efforts reduces both stress and strain measured.[26] This was generally noted and clear, especially with the quaternary blends. This is directly attributed to the cross-linking created and uniformity of the polymer matrix with exposure to heat.[37]

Table 3. Mechanical Properties of the Studied Samples

sample code	force (MPa)	tensile modulus (MPa)	stress (Max.) (MPa)	strain (Max.) (%)
A: LDPE one HC				
	119.22	0.11922	13.6927	67.1862
std	8.54761	0.00855	0.76117	9.25224
B: LDPE two HC/no drying				
	132.66	0.13266	14.0741	65.4988
std	12.2558	0.01226	0.63571	8.28854
C: LDPE two HC with drying				
	119.75	0.11975	14.2481	63.3797
std	19.1655	0.01916	0.94713	8.42211
D: LDPE/PP one HC				
	429.883	0.42988	16.9712	167.12
std	83.7717	0.08377	0.96566	94.2989
E: LDPE/PP two HC/no drying				
	262.732	0.26273	15.9697	112.732
std	61.5967	0.0616	0.78008	93.1573
F: LDPE/PP two HC with drying				
	294.59	0.29459	16.3979	157.505
std	77.8614	0.07786	0.53392	73.9973

3.5. Circularity and Harmonization of Regulatory Aspects. The quality of the recycled plastic governs the ability of the recycling system to close the material loop. Any hindering factors that could potentially affect the recycling loop could be deemed dangerous in achieving *circular economy* for plastics in any country or region.[47] It is therefore quite essential to improve the quality of recycled plastics to a level that allows closed-loop recycling (i.e., recycling into similar products and properties).[25] Plastics contain chemical additives that orient the application of the polymer for a specific application.[48,49] The presence of said additives hinders the potential of closed-loop recycling and the establishment of a circular economy. The presence of such chemicals in PW can also present a technical challenge to recyclers, as waste is typically collected in bulk (i.e., commingled); the origin and type of these additives remains unknown.[25] It is therefore essential to produce recycled products and introduce them in the market with certain specifications and chemical compositions that can govern the collection stages and maintain a certain quality of the final product. PE and PP, as established prior, are considered to be the main components in PW. The change in the quality of their properties, especially mechanical ones, governs the standards required on the market and consumer demands.[39] This is also true in the case of food packaging, whereby various countries around the world have started to have certain specification on compositions of plastics introduced in the market, such as following the EU Guidelines for food packaging.[50] Countries in the developing world, such as the State of Kuwait, are still working on introducing various regulations to govern plastics on the market. These include biodegradable standards, launch of a waste management state plan, technical studies on applicable recycled blends for the local market, and boosting the local economy with recycled goods.[39,50] The results obtained in this study showed that LDPE can be a good candidate for upcycling through mechanical recycling means, as it was more stable with drying and better suited when compared to the morphology of the LDPE/PP blend. This points toward better stability with time against degressive media. Furthermore, the LDPE/PP blend can also be a good candidate for thermal treatment based on thermal and calorimetric data obtained and discussed prior. These aspects of the work deserve to have a regulatory body that looks at them with a further economic assessment study in the near future to boost local economies in developing countries with the aim of circularity. Furthermore, when dealing with real-life PW, contamination of the discarded and commingled materials should be under consideration. Plastics collected from municipal sectors would normally contain various contaminants that could hinder the recycling process. Based on a detailed assessment conducted on the washing water by Santos et al.,[51] collected PW will contain total suspended solids ($mg \cdot L^{-1}$) 5,338 against the amount 943 in prewashed PET. The amount is higher for HDPE/PP plastics. Suspended solids reach some 907 $mg \cdot L^{-1}$ against 498 $mg \cdot L^{-1}$ for prewashed HDPE/PP plastics. Heavy metals are also present as major containments, which include lead (Pb) and iron (Fe). Camacho and Karlsson[52] determined the following chemicals as major contaminants in recycled HDPE/PP: *o*-, *m*-, and *p*-xylene, as well as ethylbenzene. These were five times higher in the recycled specimens when compared with regular levels in commercial virgin batches. This raises two major issues when dealing with real-life PW, the environmental pollution caused by the washing process itself and the management of the recycling process, which can deal with contaminated plastics. It is therefore essential to regulate and monitor contaminants present in PW when dealing with the recycling processes of common PW.

4. CONCLUSIONS

This study demonstrates the viability and advantages of upcycling through mechanical recycling of PW. By analyzing ternary and quaternary blends of common polyolefin plastics, we showed that thermal and morphological properties could be maintained or even enhanced through controlled processing conditions. The incorporation of PP into LDPE blends slightly reduced thermal stability but did not significantly affect melting behavior. The onset temperature for the samples exposed to one HC had the highest onset temperature (360 °C) when compared to the samples exposed to two HCs without drying (325 °C) and with drying (350 °C). The addition of PP in the quaternary blends reduced the onset temperature significantly for the samples exposed to a single HC (340 °C), especially when compared with the ternary blends and the other quaternary blend samples. The increase in HCs and the effect of drying showed a minimal effect on the T_m observed which was determined between 110 and 111 °C. This indicates that the blending of ternary LDPE blends has no major impact on the melting behavior and can produce products with somewhat similar thermal properties and market standards. Also, mechanical properties of the LDPE blends were independent of the processing conditions in this study, as can be concluded from the stress vs strain measurement. However, the force needed to break the quaternary LDPE/PP blends, as well as the tensile modulus, was significantly reduced with the additional HC. This research highlights the importance of understanding processing conditions, such as the number of HCs and drying, to optimize the recycling process. Mechanical recycling offers a sustainable solution that minimizes GHG emissions and environmental impact compared with other waste management methods. The findings suggest that with appropriate processing, upcycled plastics can meet market standards, contributing to sustainable waste management practices. It is also essential to have a conceptualized workplan for the contaminants present in the discarded plastics to have an effective recycling process to meet the required standards.

■ ASSOCIATED CONTENT

Data Availability Statement

The data used in this analysis is made available in the Supporting Information file.

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.4c10458.

> Additional data, plates, and figures to support this work; polymeric grades used and weights and extrusion configuration; specimens preparation and mechanical recycling procedure; thermal and mechanical data sets; and compiled data summary for the samples prepared (PDF)

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Notes

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