Article

Sustainable Additive Manufacturing: Mechanical Response of High-Density Polyethylene over Multiple Recycling Processes

Nectarios Vidakis 1, Markos Petousis 1,* and Athena Maniadi 2

1 Mechanical Engineering Department, Hellenic Mediterranean University, 71410 Heraklion, Crete, Greece; vidakis@hmu.gr
2 Department of Materials Science and Technology, University of Crete, 70013 Heraklion, Crete, Greece; maniadi@materials.uoc.gr
* Correspondence: markospetousis@hmu.gr; Tel.: +30-2810-37-9227

Abstract: Polymer recycling is nowadays in high-demand due to an increase in polymers demand and production. Recycling of such materials is mostly a thermomechanical process that modifies their overall mechanical behavior. The present research work focuses on the recyclability of high-density polyethylene (HDPE), one of the most recycled materials globally, for use in additive manufacturing (AM). A thorough investigation was carried out to determine the effect of the continuous recycling on mechanical, structural, and thermal responses of HDPE polymer via a process that isolates the thermomechanical treatment from other parameters such as aging, contamination, etc. Fused filament fabrication (FFF) specimens were produced from virgin and recycled materials and were experimentally tested and evaluated in tension, flexion, impact, and micro-hardness. A thorough thermal and morphological analysis was also performed. The overall results of this study show that the mechanical properties of the recycled HDPE polymer were generally improved over the recycling repetitions for a certain number of recycling steps, making the HDPE recycling a viable option for circular use. Repetitions two to five had the optimum overall mechanical behavior, indicating a significant positive impact of the HDPE polymer recycling aside from the environmental one.

Keywords: additive manufacturing; 3D printing; recycling; high density polyethylene (HDPE); material characterization

1. Introduction

In 2019, approximately 359 million tons of plastic products were produced worldwide due to increasing demand of lightweight, durable parts. Due to poor waste management globally, nine billion tons of plastic ends up in the environment annually [1–4]. Polymer resins such as polystyrene (PS), low density polyethylene (LDPE), high density polyethylene (HDPE), and polyethylene terephthalate (PET) are used to produce the most plastic parts available globally. More specifically, literature reports that HDPE wastes accounted for 22% worldwide in 2019 accompanied by a production of 51.33 million tons of plastic [4]. Comparing HDPE production to PET, PET wastes amounted to 11% of the total waste generated (41.56 million tons in approximation) [4].

Polyolefins are produced mainly from oil and natural gas by a process of polymerization of ethylene and propylene, respectively. Their versatility has made them one of the most popular plastics in use today. LDPE, HDPE, and PP are the major types of thermoplastics used globally in applications such as containers, pipes, toys, bags (LDPE), gas pipes (HDPE) [5], industrial wrappings, housewares and film [6,7], electrical components, and automotive parts (PP) [8]. As one of the most versatile plastic materials around, HDPE plastic is also used in a wide variety of applications in industries to replace heavy parts with more lightweight ones that are able to withstand similar forces while offering rigid strength, corrosion resistance, and environmental friendliness. HDPE is also considered as
a material suitable for sustainable and affordable manufacturing, offering high recyclability and cost efficiency [5].

However, certain economic and technical factors must be taken into consideration to evaluate the feasibility and the benefits of recycling plastics such as HDPE. More specifically, polymer degradation and degradation temperature/time must be taken into account along with the possible introduction of contaminants during sorting, cleaning, re-pelletizing/shredding, and reprocessing stages of each recycling step. The above critical parameters can lead to manufacturing faults and finished products of inferior quality.

Current research in the field seeks to optimize and improve the properties of recycled polymers and especially those of polyolefin [6–12]. Regarding the 3D printing of thermoplastics and recycled thermoplastics, there are several studies available that focus on the mechanical properties of fused filament fabrication (FFF) specimens [9,13,14], while the studies on recycled additive manufacturing materials in general are very limited [9]. Regarding the recycling of HDPE, this study presents a global recycling circular economy model (Figure 1) with the four main parameters affecting the process of recycling. All these parameters are crucial for the economy of the recycling process and the material’s behavior after it is recycled.

In literature, only a few studies are available on the mechanical properties of bulk and recycled HDPE [5–11] and HDPE composites [12–18], even less on the mechanical properties of HDPE 3D printed specimens [19], and no literature is available on the recycling of FFF HDPE specimens.

Around 89% of plastics used worldwide are hydrocarbon materials such as polypropylene (PP) and polyethylene (PE), and limited knowledge is available regarding the 3D printing of these semi-crystalline polyolefin thermoplastics. The main cause for this issue is the crystallization of PE and PP, as literature suggests. The crystallization of these polymers is the mechanism causing heavy thermal shrinkage and warpage upon cooling of the melt [20,21], thus making FFF considerably more difficult, as it affects the adhesion of the 3D printed object to the build plate. Additionally, this affects the bonding between

![Figure 1. Global recycling circular economy critical parameters and global recycled plastics market volume in 2020 (market volume data source: Technavio. Global Recycled Plastics Market 2018–2022 (IRTNTR21968). Global Recycled Plastics Market 2018–2022.](image-url)
adjacent filament strands, making the 3D printing of these materials almost impossible or only achievable with structural faults.

Thus, detailed literature is not yet available on the mechanical properties of HDPE via 3D printing. The current research work investigates the behavior of virgin and recycled HDPE throughout six recycling steps with specimens made via FFF to identify the recycling impact on the mechanical strength of the produced specimens and structural and morphological changes derived from the alteration of crystallinity and thermal properties.

The overall goal of this study was to prove that HDPE polymer can be successfully recycled multiple times through recycling of HDPE filament and pellets in order to used as a material for building parts with FFF 3D printing. To achieve that, the effect of the thermomechanical treatment during the recycling process and the effect of 3D printing on mechanical and thermal properties of recycled HDPE specimens were studied. It was found that recycling can introduce internal structuring changes in the polymer that can improve the mechanical properties of HDPE. Mechanical testing (tension, flexion, impact, and microhardness) was used to assess the effects of degradation processes in the mechanical properties of the specimens that can occur in the reprocessing of specimens. For the corresponding thermal properties, thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) were performed, while scanning electron microscopy (SEM) was conducted for the evaluation of their morphological characteristics in all recycling steps studied. It was found that, up to the fifth recycling step, the HDPE polymer properties overall improved, showing that the HDPE polymer was not affected by the recycling process. After the fifth recycling step, the HDPE polymer properties started to degrade.

2. Materials and Methods

2.1. Materials

For this research work, high-density polyethylene (HDPE) was used, procured from Plastika Kritis S.A. under the brand name Kritilen. The HDPE was in fine powder form. The same raw material was used throughout the entire experimental course. The virgin material was not blended with any other material or filler at any stage of the present research. Regarding the properties of Kritilen virgin HDPE, the supplier indicates that the density of the material is 0.960 g/cm3, the melt mass-flow rate (MFR) (190 °C/2.16 kg) is 7.5 g/10 min, while the Vicat softening temperature is 127 °C. Moreover, no plasticizers or additives were utilized.

2.2. Methods

2.2.1. Recycling Simulation and Experimental Course Parameters

For the current research project, all the process steps are depicted in Figure 2 below. Initially, HDPE powder was used to produce virgin HDPE filament via extrusion processing. The produced filament passed through extensive quality control inspection to ensure a stable diameter of 1.75 mm and a smooth surface finish. Part of the initial HDPE filament was employed to produce specimens via FFF. Specimens were comprehensively evaluated and analyzed regarding their dimensional stability and their physical properties (i.e., color, surface finish, structural faults). Tensile, flexion, impact, and micro-hardness experiments were conducted in accordance with international standards to evaluate the mechanical properties of the 3D printed virgin HDPE specimens. Additionally, parts of the specimens were used to study thermal properties and changes of the virgin and the recycled HDPE and to perform a morphological characterization analysis.

The remaining quantity of the HDPE filament was then shredded into pellets and fed into the extrusion system, repeating the above process chain in order to determine the mechanical and the thermal properties of the 3D printed HDPE specimens of each recycling step.
More specifically, six recycling processes (recycling steps) were implemented. The implemented recycling simulation process is depicted in Figure 2 below with all the steps followed. This process aimed to isolate any effects of the thermomechanical treatment on the polymer’s mechanical properties during the recycling process from other parameters, such as aging, contamination, dirt, etc.

With this process, the filament suffered one additional thermomechanical treatment in each recycle course, the one of 3D printing, when passing through the printer’s nozzle. Therefore, in total, in each recycle course, the material was thermomechanically treated with the number of recycle courses plus one more for the 3D printing process.

HDPE filament in all recycling steps in this work was extruded through a Noztek Pro (Noztek, Shoreham-by-Sea, UK) single screw desktop extruder preheated at 230 °C to remove any humidity enclosed inside the extruder. The working temperature of the extruder was set to 230 °C throughout the current research work. This working temperature was derived experimentally while consulting the TGA analysis beforehand, with the extrusion system producing a constant 1.75 mm diameter filament with fine surface quality while maintaining a constant and uninterrupted extrusion flow. The filament diameter tolerances are defined by the extruder vendor, which states that the diameter tolerances are 1.75 ± 0.04 mm.

For the experimental procedures and testing, fused filament fabrication specimens were manufactured using the same 3D printing parameters (as shown in Figure 3) on a Wanhao Duplicator i3 3D printer (Wanhao Ltd., Zhejiang, China). All specimens were tested for the determination of their physical/mechanical properties in room temperature of 23 °C.
2.2.2. Tensile Specimens Fabrication and Testing

Specimens for tensile experiments were built according to the American Society for Testing and Materials (ASTM) D638-02a international standard. Overall, 42 FFF specimens were built consisting of seven specimens for each recycling step. The tensile experiments were performed using an Imada MX2 (Imada Inc., Northbrook, IL, USA) tension/flexion test apparatus in tension mode set up using standardized grips. According to the international standard, the tensile test machine chuck was set to a 10 mm/min speed for testing.

2.2.3. Flexion Specimens Fabrication and Testing

Regarding the specimens for the flexural tests, 3D printing specimens were built according to ASTM D790-10 standard (with dimensions of 64 mm length, 12.4 mm width, and 3.2 mm thickness). In total, 42 specimens were constructed consisting of seven specimens for each case. The flexural tests were performed using an Imada MX2 (Imada Inc., Northbrook, IL, USA) in three-point bending set up. The machine chuck was set at a 10 mm/min speed for testing according to the corresponding standard.

2.2.4. Impact Specimens Fabrication and Testing

As for impact specimens, they were constructed according to the ASTM D6110-04 standard, measuring 80 mm in length, 8 mm in width, and 10 mm in thickness. Overall, 42 specimens were built. The specimens were all built with the ASTM D6110-04 standard’s impact notch. In these series of experiments, a Terco MT 220 Charpy’s apparatus (Terco inc., Stockholm, Sweden) was employed. The apparatus’s hammer was released from the same height for each experiment case in order to evaluate each specimen’s impact energy. The initial hammer’s angle was set at 162° for all the implemented experiments.

2.2.5. Micro-Hardness Measurements

Regarding the micro-hardness measurements, the ASTM E384-17 standard was followed. Proper surface finish of the test specimens was ensured prior to the evaluation. The method applied in this case for measuring the micro hardness was the micro-Vickers one, with 0.1 kg selected force scale (0.981 N) and 10 s indentation time. The typical Vickers diamond pyramid was used as indenter (apex angle of 136°), which was forced onto a polished surface of the specimens. The area of the remaining indentation after the retraction of the diamond pyramid was calculated directly by the device from the remaining imprint’s mean average of the diagonals visible in the device’s microscope. Experiments were held with the aid of an Innova Test 400-Vickers (Innovatest Europe BV, Maastricht, The Netherlands) apparatus.
2.2.6. Thermal Analysis

Thermogravimetric analysis (TGA) was performed to obtain information about the critical degradation temperature of the HDPE virgin material selected for this research in order to identify the appropriate extrusion and 3D printing temperature. The measurements were taken via a Perkin Elmer Diamond TG/TDA (Waltham, MA, USA) with a heating cycle of room temperature (32 °C) to 600 °C with a heating step of 10 °C/min. Nitrogen was employed as purge gas.

Differential scanning calorimetry (DSC) was also performed to obtain information about the effect of recycling on the melting point (Tm) and the shift in the degree of crystallinity of the samples. The measurements were taken via a Perkin Elmer Diamond DSC (Waltham, MA, USA) with a temperature cycle of 50 °C to 300 °C with a heating step of 10 °C/min and then cooling back down to 50 °C. The heating was performed on air.

2.2.7. Morphological Characterization

For the morphological characterization of specimens’ internal/external structure and interlayer fusing, SEM characterization was conducted. The SEM analysis was performed using a JEOL JSM 6362LV (Jeol Ltd., Peabody, MA, USA) electron microscope in high-vacuum mode at 5 kV acceleration voltage on uncoated samples.

3. Results

3.1. Tension Results

In Figure 4, the tensile mechanical behavior results of virgin and recycled HDPE are presented.

![Figure 4](image)

**Figure 4.** Recycling step comparative graphs (a) tensile experimental setup; (b) tensile stress vs. strain graphs of a specific specimen from each recycling step (in all cases, specimen 2 was selected); (c) average value and deviation of the tensile strength results for all the recycling steps studied; (d) and average value and deviation of the tensile modulus of elasticity values for the recycling steps studied.

More specifically, Figure 4a depicts the tension experimental setup, while Figure 4b shows the tensile stress vs. strain curves of a specific specimen from each recycling step (in all cases, specimen 2 was selected). Figure 4c presents the average value and the deviation of the tensile strength results for all the recycling steps, while Figure 4d depicts the average value and the deviation of the tensile modulus of elasticity values for the recycling steps studied.
3.2. Flexion Results

In Figure 5, the flexural mechanical behavior results of virgin and recycled HDPE are presented.

More specifically, Figure 5a depicts the flexion experimental setup, while Figure 5b shows the flexure stress vs. strain curves of a specific specimen from each recycling step (in all cases, specimen 2 was selected). Figure 5c presents the average value and the deviation of the flexural strength results for all the recycling steps, while Figure 5d depicts the average value and the deviation of the flexural modulus of elasticity values for the recycling steps.

![Figure 5](image1.png)

Figure 5. Recycling step comparative graphs (a) flexion experimental setup; (b) flexural stress vs. strain graphs of a specific specimen from each recycling step (in all cases, specimen 2 was selected); (c) average value and deviation of the flexural strength results for all the recycling steps studied; (d) and average value and deviation of the flexural modulus of elasticity values for the recycling steps studied.

3.3. Impact Results

Regarding the impact testing, the results are summarized in Figure 6a, in which the average value and the deviation of the calculated impact strength for all the recycling step studied are presented. The Charpy’s experimental setup is depicted in Figure 6b.

![Figure 6](image2.png)

Figure 6. Recycling step comparative graphs (a) calculated average value and deviation of the impact strength for all the recycling step studied; (b) Charpy’s impact test experimental setup.
3.4. Micro-Hardness Results

Regarding the micro-hardness testing, the average value and deviation of the Vickers micro-hardness for all the recycling steps studied is shown in Figure 7a, while the Vickers micro-hardness experimental setup is depicted in Figure 7b.

![Figure 7](image_url)

**Figure 7.** Recycling step comparative graphs (a) Vickers micro-hardness average value and deviation for all the recycling steps studied; (b) micro-hardness Vickers experimental setup.

3.5. Thermal Analysis Results

Regarding the thermal analysis, the TGA results for the pure HDPE polymer are presented in Figure 8a, while the comparative results for DSC of first, third, and sixth recycling steps are depicted in Figure 8b.

![Figure 8](image_url)

**Figure 8.** (a) Thermogravimetric analysis (TGA) data for pure high-density polyethylene (HDPE); (b) differential scanning calorimetry (DSC) comparative data for first, third, and sixth recycling step.

DSC analysis was used for defining the degree of crystallinity and the shift in the melting temperature (Tm) of the studied materials. The degree of crystallization was calculated by the following equation [22]:

\[
X_c(\%\text{crystallinity}) = \frac{\Delta H_m}{\Delta H_o} \times 100\%
\]  

where \(\Delta H_m\) is the melting enthalpy (the area under the melting curve), and \(\Delta H_o\) is a theoretical value of the melting enthalpy of 100% crystalline HDPE. The value \(\Delta H_0 = 293\) J/g was used in a degree of crystallinity calculation.

3.6. Morphological Characterization Results

In the following Figure 9, the SEM images of the tensile specimens’ internal area at the specimens’ neck for each recycling step studied are presented, while Figure 10 presents the fracture areas of the impact specimens for the first (Figure 10a) and the sixth recycling step (Figure 10b).
Figure 9. SEM images of the tensile specimens’ internal area at the neck of the (a) first recycling step; (b) second recycling step; (c) third recycling step; (d) fourth recycling step; (e) fifth recycling step; and (f) sixth recycling step.
4. Discussion

The mechanical properties of bulk, injection molded, and virgin HDPE polymer have been thoroughly studied and discussed in literature [5–10]. Regarding the mechanical properties of 3D printed HDPE specimens, there is no complete research or literature available yet due to the low printability of the HDPE material. The overall results of the current study for the virgin HDPE mechanical properties are in good agreement with the literature [5,18,19].

The overall results on the mechanical properties of virgin and recycled HDPE showed an increase in the mechanical properties until the fifth recycling step, proving there is a valid cause to recycle HDPE, even through FFF, as an end manufacturing process option.

Figure 11 below summarizes the overall results of this study. The complete results of this research showed that each recycled course increased the mechanical strength of the virgin HDPE material. More specifically, regarding the tensile strength results, it was shown that there was an 23.13% increase in the tensile strength when comparing recycling steps first to fifth. The same increase was present on the tensile modulus of elasticity with a value of 27.26% between the first and the fifth recycling step. Literature reports a tensile strength value of 30.2 MPa on virgin HDPE injection molded specimens, while the recycled specimens had no difference in the tensile strength [5].
Figure 11. Overall results on the mechanical properties of virgin and recycled HDPE in the six recycle courses studied.

Tensile modulus of elasticity followed the same pattern with a reported value of 1150 MPa [5]. Baligid et al. studied the mechanical properties of HDPE sintering parts and found a base tensile strength value of 6.37 MPa and a tensile modulus of 171.31 MPa on specimens with 20 µm layer thickness [18]. Schirmeister et al. focused on the printability and the mechanical properties of virgin HDPE and reported tensile strength of 25.2 MPa for FFF made specimens. Moreover, they reported a value of 970 MPa tensile modulus of elasticity [19]. Elongation of 600% found in tensile experiments is also in agreement with literature [22], confirming the viscoelastic behavior of the HDPE in FFF specimens. This behavior is also visible in Figure 4 where the yielding zone, the strain softening zone, and the strain hardening zones were clear. No literature is yet available on the mechanical properties of recycled HDPE FFF specimens.

Regarding the flexural strength, the maximum value was calculated on the second recycling step. There was an increase of 20.02% when comparing the virgin HDPE to the second recycling step. An increase of 21.6% was also determined regarding the flexural modulus of elasticity when comparing the first and the third recycling step. No literature is available yet on the flexure strength of HDPE FFF specimens or on recycled ones.

Impact strength experiments, on the other hand, showed a significant increase of 60.93% when comparing first to second recycling steps. No literature is available on the impact strength of HDPE FFF specimens or on recycled ones. Micro-hardness Vickers results showed an increase of 29.88% when comparing first recycling step to third. Changes of hardness may be directly attributed to the changes in the degree of crystallinity, as previously presented in Table 1.

Table 1. Basic thermal characteristics of the high-density polyethylene samples as obtained and calculated from DSC.

| Sample               | Tc (°C) Crystallization Temperature | Tm (°C) Melting Temperature | ΔHm (J/g) Melting Enthalpy | Xc (%) Crystallinity Degree |
|----------------------|------------------------------------|-----------------------------|---------------------------|----------------------------|
| HDPE first cycle     | 116.00                             | 142.60                      | 98.87                     | 33.67                      |
| HDPE third cycle     | 110.10                             | 145.50                      | 91.44                     | 31.14                      |
| HDPE sixth cycle     | 100.50                             | 146.30                      | 77.38                     | 26.40                      |

Research suggests that decrease in crystallinity leads to reduction of the effect that material molecules can pack and form crystal regions. The changes in crystallinity that are evident from the recycling procedure suggest that there might be a simultaneous occurrence of chain branching or polymer cross-linking along with chain scission when using extrusion systems in the recycling process [22,23]. Furthermore, changes in the degree
of crystallinity may result in changes of mechanical properties, e.g., creep, modulus, and hardness, presented in continuation.

Regarding the morphological analysis of the virgin and the recycled materials, it was shown from Figure 9 that, throughout the recycling steps, the interlaying fusing changed. More specifically, it was evident that, when comparing the first recycling step to the sixth, the layering fusion became more pronounced to the extent that the layers on the sixth recycling step specimens were not visible. This is in agreement with the increase noticed in mechanical toughness, because better interlayer fusion leads to higher mechanical stability due to uniform stress transfer. Morphological evaluation of unstressed 3D printed specimens seems to have been manufactured with good adhesion between the layers. However, it is worth mentioning that the 3D printed samples of fifth and sixth recycling steps showed some inhomogeneity in the printed layer thickness as well as some displacement of the additively printed sample’s layers due to possibly some degradation that HDPE underwent, e.g., shortening of the polymeric chains that may affect the rheology of HDPE in the melt state.

When observing the fracture areas of the impact specimens of the first recycling step (Figure 10a,c) and the sixth recycling step (Figure 10b,d), it was observed that the virgin FFF specimens behaved more ductile than those of the sixth recycling step, which broke in a more brittle manner. This was derived from the “peaks”, the “craters”, and the microfibers that were visible on the first recycling step specimen in Figure 10c and that were significantly less visible on the specimen of the sixth recycling step (Figure 10d).

Regarding the degradation of the HDPE polymer and the increase in the mechanical properties, researchers suggest that there are two competing mechanisms of degradation affecting the mechanical properties and the overall material behavior during the reprocessing of HDPE. Those are chain scission and side-chain branching [22–24]. In addition to this, some also suggest the presence of cross-linking [24,25]. Results from other studies [22] suggest that chain scission and chain branching may occur simultaneously from the start of the reprocessing due to chain scission caused by the shear forces introduced in the extrusion system.

5. Conclusions

This research focused on the effect of the thermomechanical treatment during the recycling process on mechanical and thermal properties of virgin HDPE polymer over a specific number of recycling steps. In total, six recycling steps were conducted, and tensile, flexural, and impact properties were determined on FFF specimens of each recycling step.

1. The above findings prove that the overall mechanical behavior of the recycled HDPE polymer is generally improved over the recycling steps for a certain number of repetitions, making HDPE a suitable polymer to be used in circular use. An optimum overall mechanical behavior was found between the second and the fifth recycling step, indicating a significant positive impact of the HDPE polymer recycling and circular use besides the environmental and the economy sectors, as mentioned in the introductory part of this work.

2. It became evident that the recycling steps altered the mechanical properties of HDPE polymer, resulting in an average 22% increase in all mechanical properties studied herein between the second and fifth recycle courses, while the polymer seemed to be slowly degrading after the fifth recycling step.

3. The crystallinity of the HDPE polymer decreased with increased extrusions cycles, and cross linking and branching predominated over chain scission during multiple extrusions, thus the increase in mechanical properties.

4. It was also proven that, with the experimental 3D printing parameters used in this work, HDPE filament and recycled filament could be successfully utilized in 3D printing applications introducing improved mechanical stability and toughness to 3D printed parts without heavy warping and other printing issues.
References

1. Barnes, J. Out of Sight, Out of Mind: Plastic Waste Exports, Psychological Distance and Consumer Plastic Purchasing. * Glob. Environ. Chang.* 2019, 58, 101943. [CrossRef]

2. Bijaisoradat, O.; Luttapreecha, M.; Manuspiya, H. Eco Composites with Synergistic Combinations of Natural Rubber Latex and Wood Flour Toughened Recycled HDPE. *Mater. Today Commun.* 2020, 25, 101483. [CrossRef]

3. Luedtke, J.; Gaugler, M.; Grigsby, J.; Krause, A. Understanding the Development of Interfacial Bonding within PLA/Wood-Based Thermoplastic Sandwich Composites. *Ind. Crops Prod.* 2019, 127, 129–134. [CrossRef]

4. Plastics—The Facts 2019. An analysis of European Plastics Production, Demand and Waste Data. 2020. Available online: https://www.plasticseurope.org/application/files/1115/7236/4388/FINAL_web_version_Plastics_the_facts2019_14102019.pdf (accessed on 23 April 2019).

5. Enderle, H.F. Polyethylene: High-density. In *Encyclopedia of Materials: Science and Technology*; Buschow, K.H.J., Ed.; Pergamon: Oxford, UK, 2001; pp. 7172–7180.

6. Coulier, L.; Orbons, H.G.M.; Rijk, R. Analytical Protocol to Study the Food Safety of (Multiple-)Recycled High-Density Polyethylene (HDPE) and Polypropylene (PP) Crates: Influence of Recycling on the Migration and Formation of Degradation Products. *Polym. Degrad. Stab.* 2007, 92, 2016–2025. [CrossRef]

7. Simões, C.L.; Costa Pinto, L.M.; Bernardo, C.A. Environmental and Economic Assessment of a Road Safety Product Made with Virgin and Recycled HDPE: A Comparative Study. *J. Environ. Manag.* 2013, 114, 209–215. [CrossRef]

8. Achilias, D.S.; Roupakias, C.; Megalokonomos, P.; Lappas, A.A.; Antonakou, V. Chemical Recycling of Plastic Wastes Made from Polyethylene (LDPE and HDPE) and Polypropylene (PP). *J. Hazard. Mater.* 2007, 149, 536–542. [CrossRef]

9. Vidakis, N.; Petousis, M.; Maniadi, A.; Koudoumas, E.; Vairis, A.; Kechagias, J. Sustainable Additive Manufacturing: Mechanical Response of Acrylonitrile-butadiene-styrene over Multiple Recycling Processes. *Sustainability* 2020, 12, 3568. [CrossRef]

10. Alzerreca, M.; Paris, M.; Boyron, O.; Orditz, D.; Louarn, G.; Correc, O. Mechanical Properties and Molecular Structures of Virgin and Recycled HDPE Polymers used in Gravity Sewer Systems. *Polym. Test.* 2015, 46, 1–8. [CrossRef]

11. Strangl, M.; Ortner, E.; Buettnner, A. Evaluation of the Efficiency of Odor Removal from Recycled HDPE Using a Modified Recycling Process. *Resour. Conserv. Recycl.* 2019, 146, 89–97. [CrossRef]

12. Ávila, A.F.; Duarte, M.V. A Mechanical Analysis on Recycled PET/HDPE Composites. *Polym. Degrad. Stab.* 2003, 80, 373–382. [CrossRef]

13. Vidakis, N.; Petousis, M.; Maniadi, A.; Koudoumas, E.; Liebscher, M.; Tzounis, L. Mechanical Properties of 3D-Printed Acrylonitrile-butadiene-styrene TiO2 and ATO Nanocomposites. *Polymers* 2020, 12, 1589. [CrossRef] [PubMed]

14. Vidakis, N.; Petousis, M.; Maniadi, A.; Koudoumas, E.; Kenanakis, G.; Romanitan, C.; Tutunaru, O.; Suchea, M.; Kechagias, J. The Mechanical and Physical Properties of 3D-Printed Materials Composed of ABS-ZnO Nanocomposites and ABS-ZnO Microcomposites. *Micromachines* 2020, 11, 615. [CrossRef] [PubMed]

15. Adhikary, K.B.; Pang, S.; Staiger, M.P. Dimensional Stability and Mechanical Behaviour of Wood-Plastic Composites Based on Recycled and Virgin High-Density Polyethylene (HDPE). *Compos. Part B Eng.* 2008, 39, 807–815. [CrossRef]

16. Charoenvai, S. Durian Peels Fiber and Recycled HDPE Composites Obtained by Extrusion. *Energy Procedia* 2014, 56, 539–546. [CrossRef]

17. Deepak, D.; Goyal, N.; Rana, P.; Gupta, V.K. Effect of Varying Reinforcement Content on the Mechanical Properties of Hemp-Recycled HDPE Composites. *Mater. Today Proc.* 2019, 18, 5286–5291. [CrossRef]

18. Baligidad, S.M.; Chandrasekhar, U.; Elangovan, K.; Shankar, S.; Anjankumar, N.; Supriya, J. A Study on Influence of Process Parameters on the SIS Processability and Mechanical Properties of a HDPE. *Mater. Today Proc.* 2018, 5, 25494–25500. [CrossRef]

19. Schirmeister, C.G.; Hees, T.; Licht, E.H.; Mühlaupt, R. 3D Printing of High Density Polyethylene by Fused Filament Fabrication. *Addit. Manuf.* 2019, 28, 152–159. [CrossRef]

20. Fischer, J. *Handbook of Molded Part Shrinkage and Warpage*; William Andrew: Oxford, UK, 2012.

Author Contributions: Conceptualization, A.M.; methodology, A.M.; software, A.M.; validation, A.M., N.V. and M.P.; formal analysis, A.M.; investigation, M.P., N.V.; resources, N.V., M.P.; data curation, A.M.; writing—original draft preparation, A.M.; writing—review and editing, A.M., N.V., M.P.; visualization, N.V., A.M.; supervision, N.V.; project administration, N.V., M.P.; funding acquisition, N.V. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: The data presented in this study are available in the article.

Acknowledgments: Authors would like to thank Ioan Valentin Tudose from the Hellenic Mediterranean University for the SEM images.

Conflicts of Interest: The authors declare no conflict of interest.
21. Stürzel, M.; Mihan, S.; Mülhaupt, R. From Multisite Polymerization Catalysis to Sustainable Materials and All-Polyolefin Composites. *Chem. Rev.* 2015, 116, 1398–1433. [CrossRef]

22. Oblak, P.; Gonzalez-Gutierrez, J.; Zupančič, B.; Aulova, A.; Emri, I. Processability and Mechanical Properties of Extensively Recycled High Density Polyethylene. *Polym. Degrad. Stab.* 2015, 114, 133–145. [CrossRef]

23. Javierre, C.; Clavería, I.; Ponz, L.; Aisa, J.; Fernández, A. Influence of the Recycled Material Percentage on the Rheological Behaviour of HDPE for Injection Moulding Process. *Waste Manag.* 2007, 27, 656–663. [CrossRef]

24. Kealy, T. Rheological Analysis of the Degradation of HDPE during Consecutive Processing Steps and for Different Processing Conditions. *J. Appl. Polym. Sci.* 2009, 112, 639–648. [CrossRef]

25. Kartalis, C.N.; Papaspyrides, C.D.; Pfaendner, R.; Hoffmann, K.; Herbst, H. Mechanical Recycling of Post-used HDPE Crates Using the Restabilization Technique. II: Influence of Artificial Weathering. *J. Appl. Polym. Sci.* 2000, 77, 1118–1127. [CrossRef]