Multi-material distributed recycling via Fused granular fabrication: rHDPE and rPET case of study

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5 Abstract

The high volume of plastic waste and the extremely low recycling rate has created a serious challenge worldwide. Local distributed recycling coupled to additive manufacturing (DRAM) offers a solution by economically incentivizing local recycling. A new DRAM technology capable of processing large quantities of plastic waste quickly is fused granular fabrication (FGF), where solid shredded plastic waste can be reused directly as 3D printing feedstock. This study presents an experimental assessment of multi-material recycling printability, using two of the most common thermoplastics in the beverage industry polyethylene terephthalate (PET) and high-density polyethylene (HDPE) and the feasibility of mixing PET and HDPE to be used as a feedstock material for large-scale 3-D printing. After the material collection, shredding, and cleaning its characterization, and optimization of parameters for 3D printing was performed. Results showed the feasibility of printing a large object from rPET/rHDPE flakes reducing the production cost up to 88%. .

6 Keywords: keyword1, keyword2

7 1. Introduction

- 8 The disposal of plastic waste is one of the most challenging current environmental concerns
- given its systemic complexity (Evode et al., 2021). The mass of micro- / meso- plastics in
- the oceans are expected to exceed the mass of the global stock of fish by 2050 (MacArthur,

11 2017). More critically, the global plastic annual production is expected to reach 1100 met12 ric tons by the same year (Geyer, 2020). The societal awareness on plastic recycling have
13 received substantial attention by scientific, policymaker and general public (Soares et al.,
14 2021). Unfortunately, the statistical analysis on the centralized recycling process proves that
15 it has been largely ineffective (godswillImpactsPlasticPollution2019a?) as only 9% of
16 the plastic that has been produced has been recycled from the total stock produced since
17 1950 (Geyer et al., 2017). Therefore, it remains an open challenge to identify alternatives to
18 valorize discarded plastic material.

Distributed recycling and additive manufacturing (DRAM), is an innovative technical ap-19 proach to recycle plastic wastes (Cruz Sanchez et al., 2020; Dertinger et al., 2020). DRAM was first practiced with recyclebots, which are waste plastic extruders that made filament 21 for conventional fused filament-based 3-D printers (Baechler et al., 2013; Woern et al., 2018; 22 Zhong and Pearce, 2018). Past research demonstrated that using distributed recycling fits 23 into the circular economy paradigm (Despeisse et al., 2017; Ford and Despeisse, 2016); where consumers directly recycle their own waste into consumer products from open source designs, from toys for children (Petersen et al., 2017) to adaptive aids for those with arthritis (Gallup et al., 2018). Distributed manufacturing is now in wide use (Pearce et al., 2022). In this 27 way DRAM-based recycling is done in a closed loop supply chain network (Santander et al., 2020). This type of recycling aims to reduce the environmental impact by the reduction of the transportation from the waste source to recycling facilities (M. A. Kreiger et al., 2014). In that sense, it aims to propose innovative closed-loop strategies using waste materials as 31 raw resources (Romani et al., 2021).

Fused filament fabrication (FFF, which is also known as Fused Deposition Modelling – FDM©-) is the most-widespread and established extrusion-based AM technology due to the open source proliferation from the self-replicating rapid prototyper (RepRap) project (Bowyer, 2014; Jones et al., 2011; Sells et al., 2009). This is due to its simplicity, versatility, low-cost, and ability in the construction of geometrically complex objects in the industrial and prosumer domains (romani2022?). Indeed, the open-source 3-D approach for 3-D printers

has enabled the technology to evolve in a radical manner for manufacturing and prototyping adding value to the recycled material (Cruz Sanchez et al., 2020). There are large efforts to 40 find sustainable feedstocks for 3-D printing (Pakkanen et al., 2017a). Several studies in the 41 literature have increase the spectrum of recycled filament materials such as PLA (Anderson, 42 2017; Cruz Sanchez et al., 2017a), ABS (Mohammed et al., 2017b, 2017a), PET (Vaucher et al., 2022; Zander et al., 2018), HDPE (Baechler et al., 2013; Chong et al., 2017; Mohammed et al., 2017b) PC (Gaikwad et al., 2018). In fact, using a comparative life cycle assessment 45 in a low density population case study of Michigan, USA, (M. a. Kreiger et al., 2014) argued 46 that about of 100 billion MJ of energy per year could be saved in a distributed approach, 47 for the 984 million pounds of HDPE that are recycled in the U.S. There is thus considerable evidence that DRAM can reduce the energy consumption and greenhouse gases of the manufacturing processes. 50

Most DRAM studies have been using mono-material for the fabrication of feedstock for 51 FFF. There are, however, several examples of mixed materials including wood waste and 52 recycled plastic (Löschke et al., 2019; Pringle et al., 2018) and textile fibers and recycled 53 plastic (Carrete et al., 2021). Recently, (Zander et al., 2019) reported the manufacturing of composite filament from recycled PET/PP and PS/PP blending through compatibilizer 55 copolymer such as SEBS. Their results revealed the technical printability of polypropylene blend composite filaments from a thermo-mechanical characterization perspective. Increasing the performance window of blending materials by compatibilization which could be a relevant path for recycling plastics in a local level and isolated areas contexts (e.g. during humanitarian 59 crises (Lipsky et al., 2019), supply chain disruptions (Attaran, 2020) and/or isolated off-grid 60 situations using solar-powered 3-D printers (mohammed2018?)). Likewise, (Vaucher et al., 2022) studied the evaluation of the microstructure, mechanical performance, and printing quality of filaments made from rPET and rHDPE varying the wt% of HDPE material from 0 to 10%. They confirmed the increase in the Young's modulus from 1.7 GPa of the pure PET to 2.1 GPa for all the HDPE concentrations. Additionally, the maximum stress of the bends were augmented with high HDPE concentrations. Values were lower than virgin PET

filament, yet similar to commercial recycle ones. The addition of rHDPE at higher levels, however, helped to meet the brittle-ductile transition in 15% despite the low interfacial tension of both polymers, allowing the printing of quality parts.

While former studies have proven been successful in FFF, a new approach to DRAM is fused 70 granular fabrication (FGF) or fused particle fabrication (FPF), where the material-extrusion AM systems print directly from pellets, granules, flakes, shred or grinder material (Fontana et al., 2022; Woern et al., 2018). In the context of recycling, this could reduce the number of melt/extrusion cycles that degrade the material needed in the filament fabrication process (Cruz Sanchez et al., 2017b). The FGF technique opens up the potential of use recycle materials as well as print large-scale objects either with a conventional cartesian 3-D printer (Woern et al., 2018), delta 3-D printer (Grassi et al., 2019) or hangprinter (Rattan et al., 2023). Research groups corroborate that plastic waste can be used as feedstock materials 78 for FGF/FPF. (Alexandre et al., 2020) assessed the technical and economical dimensions of virgin and shredded PLA printed in a self-modified FGF machine and compared with FFF. The investigation showed that the use of FGF reduced printing cost, time and its mechanical 81 performance was comparable with the obtained using the traditional FFF technique. Likewise, (Woern et al., 2018) found comparable properties between PLA, ABS, PP, and PET 83 recycled and virgin materials. Later publications demonstrated the technical and economic feasibility through the printing of complex objects validating the possibility of recycle plastic with FGF in both conventional and common FFF materials (Byard et al., 2019), but also recycle PC (Reich et al., 2019) and rPET (Little et al., 2020,). Few researchers, however, 87 have addressed the problem of the direct printing of recycled multi-materials, which might be a key step forward needed to facilitate the ease of sorting and recycling post-consumer plastic waste materials.

This study explores the potential of direct 3-D printing two immiscible polymers commonly used in the beverage sector through a distributed recycling process for its easily implementation operation at the local level. To demonstrate the feasibility of the process, the bottled water plastic most used in France of roughly 90% of PET (body of the bottle) and 10% of

HDPE (cap) now called rPET90//rHDPE10, is used as a test material. The experimental process of collection, characterization, and printing of the recycled material is described and 96 the results are discussed in the context of widespread DRAM adoption at the community 97 level. 98

2. Materials and Methods

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In this study, water of bottles coming from the French brand Cristaline was used as a 100 feedstock. A framework of the process used is shown in Fig. 1.a. Post-consumer bottles were collected by receptacles placed in partnership schools in Lorraine, France. To convert the complete water bottles with its cap into 3DP feedstock material, the labels were removed before shredding in a cutting mill Retsch MS300 using a 3 mm grid. After shredding, the obtained flakes were sifted with a 3 mm sifter. Next, they were cleaned with hot water in an ultrasonic machine at 60°C for 1h to remove contaminants. Lastly, they were dried in a conventional oven overnight at 80°C (Taghavi et al., 2018; Van de Voorde et al., 2022) to avoid degradation of the material. Washing conditions were the same for all the samples, therefore, the effect of contaminants was not considered. The resultant material is shown in Fig. 1.b. 110



Figure 1: figure1

The material composition was calculated as a function of the mass of the bottles and caps separately. The percent (%) of bottle-cap was found to be ~90%rPET and ~10% rHDPE.

The complete bottle was shredded without separation of both materials thus this percentage is constant for all the samples.

2.0.1. Material particle size analysis -Granulometry-

In order to ensure the particle size suitable for printing, the characterization of the granulate particles were developed using the open-source ImageJ software (imagej2023?). The size characteristics of the particles were evaluated between four different samples; vPET (used as a reference) and the raw material sifted in three different sizes 1.5 mm, 3 mm and 5 mm.

2.0.2. Fourier-transform infrared spectroscopy -FTIR-

FTIR spectroscopy was carried out to determine the nature of the bottle and determine if there were impurities, plasticizers or additives that could be detected. The analysis were made on samples of rPET and rHDPE separately and then a printed sample of both materials to determine if there was possible to observe a chemical bonding. Every sample was measured in two different points, three times in each point then curves were normalized and analyzed with the Origin Pro 8 (https://www.originlab.com/origin?). The Fourier transform infrared spectra have been recorded in the range of \$4000 cm-1 to 375 cm-1 with resolution 4 cm-1-using Bruker IFS 66V spectrophotometer.

2.0.3. Differential scanning calorimetry

Differential scanning calorimetry analysis were performed with a DSC-1 Mettler Toledo with STARe software operating under nitrogen atmosphere at heating rate and cooling rate of 10~°C/min. rPET, rHDPE and rPET90//rHDPE10 samples were investigated using three cycles: first heating from 20°C to 270 °C, cooling to 20 °C and reheating to 270°C. The rHDPE sample was analyzed following similar cycles but with the maximum temperature set at 250°C and pBC with temperatures from -20 to 270°C. Glass transition temperature (Tg) of rPET was determined during the first heating cycle, while rPET90//rHDPE10 (Tg) during the second heating cycle along with the melting point of all materials. Crystallization

temperature (Tc) of the each of the materials was determined during the cooling cycle. The degree of crystallinity (Xc) was calculated from the second cycle for recycled materials and first cycle for the blend as expressed in equation (1) (aghavi2018a?; pan2020?):

$$X_c(\%) = \frac{\Delta H_m}{w \cdot \Delta H_m^{\circ}}$$

Where, ΔH_m is the latent heat of melt, w is weight percentage of polymer in the blend, and ΔH_m° is the reference heat of 100% crystalline PET (140 J/g) and HDPE (293 J/g), respectively, provided in the literature [(pan2020?); (kratofil2006?)).

2.1. Melt Flow Index -MFI-

The melt-flow index (MFI) of rPET90//rHDPE10 flakes was determined using a Instron CEAST MF20. The analysis was performed using three samples of ~5 g at 255 °C using a 2.16 Kg weight according to ASTM D1238. The process was repeated three times. The average value of the three results was then reported with gr/10 min unit.

149 2.2. Density

In order to calculate the material's density, first; the volume was found measuring the dimensions of a solid $50x50x50 \ mm$ cubic geometry fabricated injecting rPET90//rHDPE10 flakes into a square mould with a known volume using open-source desktop injection (Holipress, Holimaker, France) machine (https://holimaker.fr/holipress?/). Then the model was weighed, and the mass was obtained. Finally, density was calculated as expressed in equation (2). To ensure the accuracy the test was performed twice and the average value was reported in g/cm3.

$$$\$ = V/m$$

\$\$ Where, ρ is the density, V is the volume, and m the mass.

59 2.3. Case of study

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2.3.1. Establishing optimal parameters

Establishing optimum combinations of parameters is essential for better quality and 161 mechanical properties of the printed parts (Jaisingh Sheoran and Kumar, 2020). Ac-162 cording to (Oberloier et al., 2022a), particle swarm optimization (PSO) is an effective 163 and time-effective method for this purpose. The optimization of the 3-D printing 164 parameters for the rPET90//rHDPE10 material in the GigabotX was performed us-165 ing the open-source PSO Experimenter platform (available in Linux), following the methodology developed by (Oberloier et al., 2022a). Three process benchmark ar-167 tifacts were printed; line, plane, and cube. They were modeled in Onshape v1.150 168 (https://www.onshape.com/en?/) and sliced CAD using Prusaslicer v2.52.0169 (https://github.com/prusa3d/PrusaSlicer/releases/tag/version 2.5.0?). The 170 geometry models and dimensions are shown in Fig. 2.

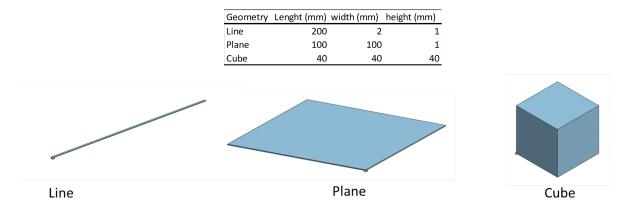


Figure 2: Dimensions and CAD models of the geometries used for parameters optimization.

Four parameters were assessed: the temperature of the bottom (nozzle) and bed, the printing speed and extrusion multiplier (Oberloier et al., 2022b). The initial parameters for the line are presented in Table 1.a while other parameters were obtained in preliminary experimental work shown in Table 1.b. Finally, the PSO tuning parameters were found in the previous PSO work (Oberloier et al., 2022a) Table 1.c.

c) Variable Value Description Ky 0.5 The emphasis given to the velocity component Kp 1

Table 1: Overall header

(a)

(b) Line optimization initial parameters

Parameters	Value	Units
Layer height	0.5	mm
Width	2	mm
Т2	230	$^{\circ}\mathrm{C}$
Т3	220	$^{\circ}\mathrm{C}$
Cooling	0	%
Infill density	2	%

(c)

(d) Fixed parameters to perform printing parameters optimization based on PSO

Variable	Min	Max	Guess	True/False	Description
Т1	255	270	260	TRUE	Temperature Zone 1 on GigabotX
Tb	80	90	85	TRUE	Bed temperature
Ps	10	25	15	TRUE	Printing Speed
E	0.5	2	1	FALSE	Extrusion Multiplier

The emphasis given to a particle's personal best position Kg 2 The emphasis given to the swarm's group best position

FGF 3D printing

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To print the raw material obtained, a 3-heat-zone modified open-source printer (Gigabot XL re:3D, Houston, TX, USA) was used. The machine is a single screw extrusion-based 3-D printer capable of direct printing pellets, flakes or granules, with a nozzle size of 1.75 mm. For this study, a chair was printed to evaluate the ability for the material to 3-D print and the machine capacity to print large-objects such as a piece of furniture. The ideal parameters found for the cube geometry were used to print the final part. Results and discussion

187 Material characterization

Both polymeric components of the bottle as well as the blend were characterized and analyzed to determine their properties using different methods as explained in the previous section.

191 Material particle size analysis (granulometry)

The material after shredding has an irregular shape, which led to machine clogging and generated under-extrusion issues, lowering the quality of the prints. Therefore, granulometry

evaluation is essential to control these issues. Previous studies demonstrated that particles with areas smaller than 22 mm² were optimal for print without jamming or under-extrusion 195 problems (Woern et al., 2018). From the experiments conducted, however, particles with 196 areas above 10 mm² clogged in the feeding system and auger screw of the machine. There-197 fore, granulometry analysis was performed using three different mesh sizes. Figure 3 shows 198 the results obtained, where particles sifted at 5 mm had an average area similar to the reference. There are, however, particles with areas over 9 mm² which blocked in the feeding 200 and extrusion section. Particles sifted to 1.5 mm showed a distribution with areas from 0 201 to proximately 3 mm², this area was considered too low for printing. Small particles can 202 completely melt in the first heat zone obstructing the consistent flow of other particles and 203 not allowing the pressure needed to extrude the melted particles lower in the screw. Although 204 flakes of 3 mm show a more dispersed distribution and slightly lower area compared with the 205 reference, those particles were found to be optimal for printing. The final objects, however, 206 still displayed under-extrusion issues. For this reason, a crammer was implemented (Little et 207 al., 2020); which physically pushes particles to the auger to convey them from feeding tube 208 into the extruder. After the crammer implementation under-extrusion issues were significa-209 tively decreased. It was concluded that flakes with areas between 1.5 mm2 to 10 mm2 were 210 optimum for print using a crammer able to aid the feeding system. 211

Fig. 3. Granulometry analysis

213 Chemical analysis from FTIR

Chemical structure information of the materials was obtained using FTIR spectroscopy, which provided analysis of the characteristic spectral bands of the polymers. First, for the rPET (bottle) four characteristic bands can be observed Fig 4, one in 1713 cm-1 representing the C=O double bond, the C-O single bond ester at 1240 cm-1, 1093 cm-1 band corresponding to the methylene group and vibrations of the ester bond and finally, a band 722 cm-1 the CH2 rocking bending vibration. Similar results were obtained in the literature for PET coming from recycled water bottles, soda bottles and food containers (Zanders2018?). For

rHDPE (caps), four characteristic peaks were observed, the bond of C-H functional group in peaks 2915cm-1 and 2847 cm-1, main bending mode of the -CH2 in 1465 cm-1 and CH2 222 rocking bending vibration at 729 cm-1. These results confirmed the chemical structures of 223 starting materials. Additionally, other indicative resonances besides those associated with 224 the polymer structures were not observed, concluding that additives or plasticizers in sig-225 nificative quantities were not present in either sample. The spectrum of the printed blend (rPET90//rHDPE10) had the same characteristic peaks as the bottle, confirming the PET 227 dominant content. There are, however, observable differences between 1000 cm-1 and 720 228 cm-1 and in the bond of C-H (2915cm-1 and 2847 cm-1 peaks), confirming the presence of 229 HDPE (cap). The shifting observed can be attributed to the hydrogen interactions between 230 both materials. 231

Fig 4. FTIR spectra of rPET, rHDPE and their blend

Thermal analysis DSC

The thermal properties of both recycled materials and their blend were characterized via 234 DSC to have a starting point for the 3-D printing process parameter optimization. From the 235 representative heating and cooling thermograms shown in Fig. 5, two distinct endothermic 236 peaks are observed in the printed blend sample that are associated with fusion of the crys-237 talline fractions of rHDPE and rPET, which confirms the immiscibility of both materials. 238 Moreover, a significant reduction in the enthalpy of fusion and crystallization of the rHDPE in the blend is attributed to the low percentage of HDPE in the blend. The observation of 240 cold crystallization peak in the blend but not in the individual polymers, however, can be 241 attributed to an interaction between both polymers, where the rHDPE might act as a nucle-242 ating agent. Table 2. list the thermal properties for rPET, rHDPE and rPET90//rHDPE10. The melting point for rHDPE and rPET are 131.7 °C and 249.9 °C, respectively and are similar to those found in the literature (Chen et al., 2015). It is observed that the melting 245 and crystallization temperature of rPET shifted to a higher value, while slightly decreasing 246 for rHDPE. In addition, the crystallization of rPET was found to be somewhat affected by 247

the addition of rHDPE as it was increased 4%. It is likely the addition of the rHDPE acted as germination point for crystallization (Vaucher et al., 2022). The slight changes in the rPET temperatures of fusion-crystallization and degree of crystallinity showed the interaction of both polymers.

252 Fig. 5 - DSC thermograms of recycled materials and blend.

Table.2 Thermal analysis of rPET, rHDPE and their blend. Sample Glass transition Melting Crystallization % Crystallinity Xc Tg (°C) Tm (°C) Δ Hm (J/g) Tc (°C) Δ Hc (J/g) Δ Hcc (J/g) (J/g) rPET 82 249.9 31.6 196.7 34.8 - 22.6

257 rHDPE - 133.8 172 118.7 158.5 - 58.7

 ${}^{258} \quad \text{rPET90/rHDPE10} \ 77 \ / \ - \ 254/131.7 \ 40.3/1.30 \ 210.6/117.4 \ 37.9/6.7 \ 6.8 \ 24.8 \ 18.4 \$

259 Rheology MFI

The melt flow index of the flakes was determined, enabling a fast and practical screening 260 of the viscosity of the material. Following the DSC results, the initial temperature to start 261 the MFI test was 250°C, however the material did not flow reliably so the temperature was 262 increased by 5°C to enable the melt flow index of the rPET90//rHDPE10 to be determined. 263 A temperature of 260°C was also tested, however, the material flowed rapidly and the measurement could not be reliably obtained. MFI tests were performed three times and the results of the rPET90//rHDPE10 was a medium MFI 39.4± 2.4 g/10min, which is roughly 266 consistent with values found in literature for rPET (Salminen?). This result suggests that 267 the low percentage addition of HDPE do not highly impact the MFI value of rPET. As the 268 material flowed at temperature of 255°C in the MFI, it provided the input temperature for 269 3-D printer parameters optimization. 270

271 Density

The density allows the estimation of cost, material use, time consumed and weight of the printing object in the slicer. This information is useful to find the accurate printing parame-

ters with the PSO experimenter as fitness is calculated as a function of the dimensional accuracy and weight of the printed object. Hence, density was useful to determine the weight of 275 the geometries. From calculations made after weight and measuring the rPET90//rHDPE10 276 injected object, the density of the material was found to be 1.13 g/cm³. From the results it 277 was observed that the theoretical density of PET ($\sim 1.38 \text{ g/cm}3$) was slightly decreased with 278 the presence of the HDPE, which has a theoretical density range from 0.93 to 0.97 g/cm³ 279 (Jonathan GUIDIGO1 et al., 2017). It normally occurs when a polymer is blended with lower 280 density polymer. A theoretical blend density could be calculated by equation @ mishra1989 281 _12=1/((W_1)/ _1 +W_2/ _2) . (3) Where, _12 is the density of the blend, W_(1) 282 and W_(2), the weight fractions of each polymer, _1 and _1, the theoretical density of 283 each polymer.

285 3. Case of study

286 3.1. Particle swarm optimization (PSO) Experimenter

Geometries were 3-D printed changing the parameters as expressed in the PSO Experi-287 menter software. The fitness function is defined by the weighted sum of the dimensional 288 measurements (length, width, height, and weight) of the printed object, where fitness below 289 0.1 was set as a desirable threshold. In the software five particles were established for one 290 iteration, thus in one iteration five different parameters combinations were printed. The first 291 geometry (line) was able to reach a fitness of < 0.1 after six iterations for a total of thirty 292 lines printed. The ideal parameters for this geometry are listed in column two of Table 3. 293 Afterwards, these parameters were used as initial guess for plane geometry, which reached 294 the desired fitness in the first iteration. Likewise, cubes were printed using the plane ideal 295 parameter as initial guess and optimal parameters, where found in the first iteration. Results showed a significant change in the printing speed, which lowers at higher geometry 297 complexity. Moreover, the cube geometry required a higher extrusion multiplier to fill gaps 298 and overcome under-extrusion problems. The optimization of the parameters for the three 299 geometries took around 10h reducing the experimental time, compared with conventional 300

methods. According to (Oberloier et al., 2022a) this time of experimentation can be reduced 97%. Indeed, the efficacity of PSO in finding global optimum parameters is high, partic-302 ularly when there is a large or complex design space (Saad et al., 2019, p. selvam2020). 303 Additionally, PSO converge to optimum solutions with fewer iterations that DoE methods 304 (Zhang et al., 2015) while mixing PSO with other meta-heuristic methods has shown higher 305 ability of predict and optimize parameters (e.g. minimize surface roughness(Shirmohammadi 306 et al., 2021), compressive strength and porosity of scaffolds (Asadi-Eydivand et al., 2016), 307 mechanical properties (Raju et al., 2019)). However, the DoE methods are still widely used 308 as they provide insight into the effects of individual design parameters and their interactions 309 while the ability to find interaction between the variables is not possible using PSO. In the 310 beginning of a set of optimization experiments, the complete understanding of the process 311 technique as well as the function settings might be complex. The methodology used in this 312 study, however, was easy to implement and the software has the advantage of being free, 313 open source and user-friendly, lessening the initial difficulty. Thus, PSO demonstrated to be 314 an effective and high accuracy prediction technique able to find the initial optimum parame-315 ters for rPET90//rHDPE10 material for FGF/FPF. From the results it is observe that the 316 optimal parameters may change depending of the object printed and each parameter has its 317 own variation. One hypothesis is that geometry might play a role in parameter assignment 318 and probably could be more visible in larger prints, yet this hypothesis needs further inves-319 tigation. There are several physical mechanisms at play that would be expected to change optimal printing parameters based on the geometry and size of the object. For example, the 321 cooling time and temperature history of a voxel will depend on the geometry of the printed 322 object. Thus, to maintain the same thermal history the printing parameters must change 323 as the geometry changes. This same effect of thermal history can also have more subtle 324 impacts such as degree of crystallization even for PLA (Wijnen et al., 2018). In addition, 325 some physical effects of materials extrusion are magnified with scale. The most obvious is the 326 impacts of thermal expansion and contraction. Small changes in contraction as a part cools 327 may cause acceptable distortions for small prints, but these are magnified for large prints 328 (e.g. causing deformation and in the worst cases delamination or loss of bed adhesion)(Shah

et al., 2019). Although, (Roschli et al., 2019) showed the obstacles and possible solutions of the large-scale AM according to the way of the parts are designed the incidence of the geometry in the printing parameters needs far more detailed future studies. Specifically better models for mapping 3-D printing parameter optimizations of small printed objects to large volume objects is needed. Table 3. Ideal printing parameters for fused granule fabrication of waste PET and HDPE blend made from shredded whole plastic water bottles.

Variable Line value Planes value Cube value Δ Units T1 258 263 264 6 ± 3.2 °C Tb 86 82 84 4 ± 2 °C Ps 21 14 10 11 ± 5.6 mm/s E 1.07 0.87 1.32 0.5 ± 0.3 -

338 3.2. Functional object print

The ideal parameters found for the cube geometry were used as final parameters for print 339 the case study product, except the print speed for the optimized line speed was used to 340 decrease the printing time and delamination. This change was performed as according to 341 the PSO results the material is printable in a range of 10 to 20 mm/s. Additionally, the 342 faster the printing the lesser the time of cooling between the layers thus avoiding possible delamination (Roschli et al., 2019), which is exacerbated for larger objects. The Gigabot X successfully produced a piece of furniture from multi-material recycled water bottles that 345 included mixing HDPE and PET as shown in Fig. 6.a. The printing quality is acceptable 346 as a prototype, proven the machine capacity of printing large-scale functional objects where the chair was able to hold a child with a mass of 20 kg comfortably as shown in Fig.6 f. The material, in the other hand, needs further evaluation as the printed object showed 349 weak bond strength between the adjacent layers (delamination Fig. 6.b). This is probably 350 due to the difference in chemical properties of both materials or immiscibility (William et 351 al., 2021), high crystallinity (Verma et al., 2023) and the large volume of the object as 352 delamination issues were more visible at the time of print the chair that in the parameters 353 optimization process. Indeed, delamination presented in the printing of a larger object can 354 be attributed to the rapid cooling of the layers before the material is once again deposed 355 contrary of the cube printing where the small surface allowing the adhesion of layers before 356 there are completely cooled. This can even be an issue for more popular 3-D printing materials 357

like PLA from the print surface (Wijnen et al., 2018). The addition of agents that reduce the interphase tension between polymers might help to solve the delamination present and 359 enhance the properties of the material (Kramer et al., 1994, @ dai1997, @ inoya2012) as 360 interfacial bond can be enhanced by polymer modification (Gao et al., 2021) and viscosity 361 decrement (Ko et al., 2019). Additionally, while printing warping problems were observed, 362 (Fig. 6.c) which are likely caused by the high crystallization rates of HDPE (Schirmeister 363 et al., 2019) suggesting that the incorporation of this plastic even in a low portions have 364 affected the printing negatively. The use of Magigoo (https://magigoo.com?/) and the 365 addition of a brim was tested in order to enable bed adhesion, vet this solutions did not 366 completely solve the problem. A previous study showed that the use of a building plate 367 made of thermoplastic elastomer SEBS allowed the adhesion of the plastic and enables facile detachments of the printing object without breaking or damaging (Schirmeister et al., 2019), 369 suggesting a solution that needs further evaluation in future work. Another visible issue 370 present in the close angles of the printed object was the shrinkage (Fig. 6.d) which occurs 371 during solidification and especially upon polymer crystallization. Moreover, as is well-know, 372 PET has hygroscopic tendencies and easily absorbs atmosphere moisture making it difficult to extrude (Bustos Seibert et al., 2022) thus is likely to break down in the presence of water, 374 lowering the quality of the print. Before the chair printing some samples showed brittle 375 behavior and voids formation therefore, the material was constantly dried and the hopper 376 was closed in order to avoid moisture coming from the environment helping to have a more printable material. Visually it can be observed some vibration and ringing problems (Fig. 378 6.e) caused by the machine upgrading, both acceleration and jerk (maximum value of the 379 instantaneous speed change) needs finer tuning. 380

Fig. 6 a to e) Finished children chair and printing issues, f) Usability test.

3.2.1. Cost and environmental impact

The printing time took 10 h and the printed object has a mass of 840 g. Due to the found optimized speed being low, the printing rate (grams per hour) is low considering the machine that pellet printers have a typical throughput of 220 g to 9 kg per hour. The

printing time could be improved by upgrading the extruder motor to a more powerful one. Besides, the energy required for 10 hr of 3-D printing was found to be 6 kW-hr resulting in 387 a production cost of $\sim 1.2 \in$ in function of the electricity cost in France, without considering 388 the material cost as bottles were obtained from post-consumers waste. When labor costs 389 are not included the price was significative reduced (~88%) compared with those low cost 390 found in the market [@ https://www.ikea.com/fr/fr/p/mammut-tabouret-enfant-interieur-391 exterieur-jaune-20382324. The economics of fabricating the case study product remained 392 competitive even if recycled plastic pellets or shreds are used, which can be found on the 393 market from 1-10 €/kg. However, labor, maintenance and machine devaluation were not con-394 sidered in the final price and are needed in future work for a complete economic evaluation. 395 Regarding the environmental impact, although this study does not evaluate the entire life cycle of the printed object, various scientific studies have already shown feasibility of distributed 397 recycling (Santander et al., 2020, p. kerdlap2022), the comparation between conventional 398 and distributed manufacturing in terms of energy consumption and emissions (Kreiger and 399 Pearce, 2013), environmental performance of AM (Colorado et al., 2020) and the appearance 400 of DRAM as a source of raw material for diverse 3-D printers coming from post-consumer 401 plastic waste in the form of either filament (Mohammed et al., 2017b) or granules (Alexandre 402 et al., 2020). Additionally, (Caceres-Mendoza et al., 2023) has developed a complete life of 403 cycle assessment of a DRAM system based on the production of PLA filament comparing 404 virgin and recycled materials. Their environmental results showed a reduction of the impacts 405 of production (climate change, fossil depletion, water depletion and potential eutrophication) 406 of ~97% compared to virgin filament. These results, however, are dependent on the energy 407 supply and can vary depending on location. 408

409 Conclusion and future work

This study analyzed the feasibility of using mixed post-consumer waste as feedstock material for direct 3-D printing without compatibilization. The results showed the potential of mixing solid waste plastics (PET/HDPE) for its use as feedstock material by printing a water bottle with two incompatible polymers from the cap and body of the bottle. Additionally, the results

showed that a large-scale FGF 3-D printer was capable of producing a cost-effective functional object from these mixed waste PET/HDPE plastics. Further research is needed in the analysis 415 of mechanical properties of the material as well as the possibilities of using compatibilizers 416 capable of enhancing the interphase tension between plastics and lower their crystallinity, 417 which might help improve performance and enhance material and 3-D printed part properties. 418 These factors increase in importance as the scale of the 3-D printed part increases. The improvement of the material science of the approach can also offer an opportunity to improve 420 the quality of the printing printing time, lowering the energy consumption of the machine 421 and thus improving the economic viability of DRAM with mixed plastic waste. 422 In addition, future work could assess the different types of combinations or blends between 423 commodity plastics using or not compatibilizer towards its printability, bringing out the possibility of selection/sorting process elimination. In the same way, the development of 425 a methodology that allows the reproducibility of the process even in areas with limited 426 infrastructure opens up the potential of plastic revalorization using DRAM. 427

4. Declaration of competing

The authors declare that they have no known competing financial interest or personal relationships that could have appeared to influence the work reported in this paper. Acknowledgements

The authors thank the LUE program for the financing of the thesis, the LF2L platform,
INEDIT European project and the Thompson endowment. References

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