Multi-material distributed recycling via Material Extrusion: rHDPE and rPET case of study

The high volume of plastic waste and the extremely low recycling rate have created a serious challenge worldwide. Local distributed recycling coupled with additive manufacturing (DRAM) offers a solution by economically incentivizing local recycling. One DRAM technology capable of processing large quantities of plastic waste 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, the characterization and optimization of parameters for 3D printing were performed. Results showed the feasibility of printing a large object from rPET/rHDPE flakes, reducing the production cost by up to 88%.

| Acronym | Definition |
| --- | --- |
| ABS | Acrylonitrile Butadiene Styrene |
| AM | Additive Manufacturing |
| DRAM | Distributed recycling via additive manufacturing |
| DSC | Differential scanning calorimetry |
| FDM | Fused deposition modeling |
| FFF | Fused filament fabrication |
| FGF | Fused granular fabrication |
| FPF | Fused particle fabrication |
| FTIR | Fourier-transform infrared spectroscopy |
| HDPE | High-density polyethylene |
| MFI | Melt flow index |
| PC | Polycarbonate |
| PET | Poly(ethylene terephthalate) |
| PLA | Poly(lactic acid) |
| PP | Polypropylene |
| PSO | Particle swarm optimization |
| PS | Polystyrene |
| SEBS | Poly (styrene-block-ethene-co-butene-block-styrene) |
| Tg | Glass temperature |
| pBC | Printed Bottle-Cap |
| rHDPE | Recycled High-density Polyethylene |
| rPET90//rHDPE10 | Recycled Bottle-Cap (Cristaline bottle shredded without separation) |
| rPET | Recycled Poly(ethylene) terephthalate |
| vPET | Virgin or commercial Poly(ethylene terephthalate) |

# Introduction

The disposal of plastic waste is one of the most challenging current environmental concerns given its systemic complexity ([Evode et al., 2021](#ref-evode2021)). The mass of micro- / meso- plastics in the oceans is expected to exceed the mass of the global stock of fish by 2050 ([MacArthur, 2017](#ref-macarthur2017)). More critically, the global annual plastic production is expected to reach 1100 metric tons by the same year ([Geyer, 2020](#ref-geyer2020)). Societal awareness of plastic recycling has received substantial attention from scientists, policymakers, and the general public ([Soares et al., 2021](#ref-soares2021)). Unfortunately, the statistical analysis of the centralized recycling process proves that it has been largely ineffective ([Siltaloppi and Jähi, 2021](#ref-Siltaloppi2021)) with only 9% of the plastic produced since 1950 being recycled from the total stock ([Geyer et al., 2017](#ref-Geyer2017)). Therefore, it remains an open challenge to identify alternatives to valorize discarded plastic material.

Distributed recycling and additive manufacturing (DRAM) is an innovative technical approach to recycling plastic waste ([Cruz Sanchez et al., 2020](#ref-cruzsanchez2020); [Dertinger et al., 2020](#ref-dertinger2020)). DRAM was initially implemented using recyclebots, which are waste plastic extruders that produce filament for conventional fused filament-based 3-D printers ([Baechler et al., 2013](#ref-baechler2013); [Woern et al., 2018](#ref-woern2018); [Zhong and Pearce, 2018](#ref-zhong2018)). Previous studies have shown that distributed recycling aligns with the circular economy paradigm ([Despeisse et al., 2017](#ref-Despeisse2016); [Ford and Despeisse, 2016](#ref-Ford2016)). This approach allows consumers to directly recycle their own waste into consumer products using open-source designs, ranging from toys for children ([Petersen et al., 2017](#ref-Petersen2017)) to adaptive aids for individuals with arthritis ([Gallup et al., 2018](#ref-gallup2018)). Distributed manufacturing is now widely adopted ([Pearce and Qian, 2022](#ref-pearce2022)). In this way, DRAM-based recycling operates within a closed-loop supply chain network ([Santander et al., 2020](#ref-santander2020)). The primary goal of this type of recycling is to reduce the environmental impact by minimizing the transportation from the waste source to recycling facilities ([Kreiger et al., 2014](#ref-kreiger2014)). In that sense, it aims to propose innovative closed-loop strategies that utilize waste materials as raw resources ([Romani et al., 2021](#ref-romani2021)).

Fused filament fabrication (FFF, which is also known as Fused Deposition Modelling –FDM©-) is the most widespread and established extrusion-based AM technology. It has gained popularity due to the open-source proliferation from the self-replicating rapid prototyper (RepRap) project ([Bowyer, 2014](#ref-bowyer2014); [Jones et al., 2011](#ref-jones2011); [Sells et al., 2009](#ref-sells2009)). FFF is favored for its simplicity, versatility, low cost, and ability to construct complex geometric objects in the industrial and prosumer domains ([Romani et al., 2021](#ref-romani2021)). Indeed, the open-source approach for 3-D printing has facilitated significant advancements in manufacturing and prototyping adding value to the recycled material ([Cruz Sanchez et al., 2020](#ref-cruzsanchez2020)). Efforts are being made to identify sustainable feedstocks for 3-D printing Pakkanen et al. ([2017a](#ref-Pakkanen2017)). Several studies have expanded the range of recycled filament materials including PLA ([Anderson, 2017](#ref-anderson2017); [Cruz Sanchez et al., 2017](#ref-cruzsanchez2017)), ABS ([Mohammed et al., 2017b](#ref-mohammed2017a), [2017a](#ref-mohammed2017)), PET ([Vaucher et al., 2022](#ref-vaucher2022); [Zander et al., 2018](#ref-zander2018)), HDPE ([Baechler et al., 2013](#ref-baechler2013); [Chong et al., 2017](#ref-chong2017); [Mohammed et al., 2017b](#ref-mohammed2017a)), and PC ([Gaikwad et al., 2018](#ref-gaikwad2018)). In fact, Kreiger et al. ([2014](#ref-kreiger2014)) conducted a comparative life cycle assessment in a low-density population case study in Michigan (USA) and estimated that a distributed approach could save approximately 100 billion MJ of energy per year from the recycling of 984 million pounds of HDPE. There is substantial evidence that DRAM can contribute to reducing energy consumption and greenhouse emissions in manufacturing processes.

Most DRAM studies have used mono-materials for the fabrication of feedstock for FFF. There are, however, several examples of mixed materials including wood waste and recycled plastic ([Löschke et al., 2019](#ref-loschke2019); [Pringle et al., 2018](#ref-pringle2018)) and textile fibers and recycled plastic ([Carrete et al., 2021](#ref-carrete2021)). Recently, Zander et al. ([2019](#ref-Zander2019)) reported the manufacturing of composite filament from recycled PET/PP and PS/PP blending through a compatibilizer 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 at a local level and in isolated areas contexts (e.g. during humanitarian crises ([Corsini et al., 2022](#ref-corsini2022); [Lipsky et al., 2019](#ref-lipsky2019); [Savonen et al., 2018](#ref-savonen2018)), supply chain disruptions ([Attaran, 2020](#ref-attaran2020); [Choong et al., 2020](#ref-choong2020) ; [Novak and Loy, 2020](#ref-novak2020); [Salmi et al., 2020](#ref-salmi2020) ) and/or isolated off-grid situations using solar-powered 3-D printers ([Gwamuri et al., 2016](#ref-gwamuri2016); [King et al., 2014](#ref-king2014); [Mohammed et al., 2018](#ref-Mohammed2018); [Wong, 2015](#ref-wong2015))). Likewise, Vaucher et al. ([2022](#ref-vaucher2022)) 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 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 was 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 successful in FFF, a new approach to DRAM is fused granular fabrication (FGF) or fused particle fabrication (FPF), where the material-extrusion AM systems print directly from pellets, granules, flakes, shreds or grinder material ([Fontana et al., 2022](#ref-fontana2022); [Woern et al., 2018](#ref-woern2018)). 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., 2017](#ref-cruzsanchez2017)). The FGF technique opens up the potential to use recycled materials as well as print large-scale objects either with a conventional cartesian 3-D printer ([Woern et al., 2018](#ref-woern2018)), delta 3-D printer ([Grassi et al., 2019](#ref-grassi2019)) or hangprinter ([Petsiuk et al., 2022](#ref-petsiuk2022); [Rattan et al., 2023](#ref-rattan2023)). Research groups have corroborated that plastic waste can be used as feedstock materials for FGF/FPF. Alexandre et al. ([2020](#ref-alexandre2020)) assessed the technical and economical dimensions of virgin and shredded PLA printed in a self-modified FGF machine and compared it with FFF. The investigation showed that the use of FGF reduced printing costs, time and its mechanical performance was comparable to that obtained using the traditional FFF technique. Likewise, Woern et al. ([2018](#ref-woern2018)) found comparable properties between PLA, ABS, PP, and PET recycled and virgin materials. Later publications demonstrated the technical and economic feasibility through the printing of complex objects validating the possibility of recycling plastic with FGF in both conventional and common FFF materials ([Byard et al., 2019](#ref-byard2019)), but also recycling PC ([Reich et al., 2019](#ref-reich2019b)) and rPET ([Little et al., 2020](#ref-little2020)). Few researchers, however, have addressed the problem of directly printing 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 of two immiscible polymers commonly used in the beverage sector through a distributed recycling process for its easy implementation operation at the local level. To demonstrate the feasibility of the process, the most commonly used plastic for bottled water in France, which consists of roughly 90% PET (body of the bottle) and 10% HDPE (cap) now referred to as *rPET90//rHDPE10*, is used as a test material. The experimental process of collection, characterization, and printing of the recycled material is described, and the results are discussed in the context of widespread DRAM adoption at the community-based level.

# Materials and Methods

The methodology presented in [Figure 1](#fig-1) outlines the approach adopted to develop the study. The three stages, namely *Material obtention*, *Printing process*, and *Evaluation* were thoroughly studied to control the major process steps and the technical characterization methods. In the following subsections, each step is explained.

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| Figure 1: Global framework of the study |

## Raw material obtention

The goal of the material stage is to collect and prepare post-consumer plastic sources. In this study, water bottles coming from the French brand Cristaline were used as feedstock. The process steps used are shown in [Figure 2](#fig-fig1) a/b. Post-consumer bottles were collected from receptacles placed in partnership schools in Lorraine, France. To convert the complete water bottles including their caps into 3DP feedstock material, the labels were removed before shredding in a cutting mill (Retsch MS300) using a 3 grid. After shredding, the obtained flakes were sifted with a 1.5 , 3 , and 5 sifters for further analysis. Next, the flakes were cleaned with hot water in an ultrasonic machine at 60°C for 1 hour to remove contaminants. Lastly, they were dried in a conventional oven overnight at 80°C ([Taghavi et al., 2018](#ref-taghavi2018); [Van de Voorde et al., 2022](#ref-vandevoorde2022)) 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 2.c.

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| Figure 2: Process steps to prepare the collected material |

The material composition was calculated as a function of the mass of the bottles and caps separately. The percentage (%) of bottle-cap was found to be ~90%rPET (bottle) and ~10% rHDPE (cap). The complete bottle was shredded without separation of both materials thus this percentage is constant for all the samples.

## Material preparation and characterization

### Material particle size analysis -Granulometry-

In order to ensure the particle size suitable for printing, the granulate particles were characterized using the open-source ImageJ software ([ImageJ, 2023](#ref-imagej2023)). The size characteristics of the particles were evaluated in four different samples: vPET (used as a reference) and the raw material sifted into three different sizes: , , and .

### Fourier-transform infrared spectroscopy –FTIR-

FTIR spectroscopy was conducted to determine the composition of the bottle and identify any impurities, plasticizers, or additives. The analysis involving testing separate samples of rPET and rHDPE. Additionally, a printed sample of both materials was examined to identify any potential chemical bonding. Each sample was measured at two different points, with three measurements taken at each point. The resulting curves were then normalized and analyzed using Origin Pro 8. The Fourier transform infrared spectra were recorded in the range of to with a resolution of using a Bruker IFS 66V spectrophotometer.

### Differential scanning calorimetry –DSC-

Differential scanning calorimetry analysis was performed using a DSC-1 Mettler Toledo with STARe software operating under nitrogen atmosphere at heating rate and cooling rate of . The samples investigated were rPET, rHDPE, and rPET90//rHDPE10. Three cycles were conducted: the first involved heating from 20°C to 270°C, cooling to 20°C and reheating to 270°C. The rHDPE sample was analyzed using similar cycles but with the maximum temperature set at 250°C and the blend was tested at temperatures ranging from -20 to 270°C. The glass transition temperature (Tg) of rPET was determined during the first heating cycle, while the Tg of rPET90//rHDPE10 was determined during the second heating cycle, along with the melting point of all materials. The crystallization temperature (Tc) was determined during the cooling cycle for each material. The degree of crystallinity (Xc) was calculated from the second cycle for recycled materials and the first cycle for the blend, as expressed in equation (1) ([Pan et al., 2020](#ref-pan2020); [Taghavi et al., 2018](#ref-taghavi2018)):

Where, is the latent heat of melt, is the weight percentage of polymer in the blend, and is the reference heat of 100% crystalline PET () and HDPE (), respectively, provided in the literature ([Kratofil et al., 2006](#ref-kratofil2006); [Pan et al., 2020](#ref-pan2020)).

### Melt Flow Index –MFI-

The melt-flow index (MFI) of rPET90//rHDPE10 flakes was determined using an Instron CEAST MF20. The analysis was performed using three samples of ~5 g at a temperature of 255 °C with a 2.16 kg weight following the ASTM D1238 standard. The process was repeated three times. The average value of the three results was reported in units of .

### Density

The material’s density was calculated as follows: first, the volume was found by measuring the dimensions of a solid cubic geometry fabricated by injecting rPET90//rHDPE10 flakes into a square mold with a known volume using an open-source desktop injection machine(Holipress, Holimaker, France). Then, the model was weighed, and the mass was obtained. Finally, the density was calculated as expressed in [Equation 2](#eq-density). To ensure the accuracy of the test it was performed twice and the average value was reported in .

Where, is the density, is the volume, and the mass.

Afterwards, experimental results were compared with the theoretical blend density which could be calculated by [Equation 3](#eq-density_theory).

Where, is the density of the blend, and , the weight fractions of each polymer, and , the theoretical density of each polymer for PET () and HDPE 0.93 to 0.97 ([Jonathan GUIDIGO1 et al., 2017](#ref-jonathanguidigo12017)).

## Printing process

### Establishing optimal parameters

Establishing the optimal combinations of parameters is essential for improve the quality and mechanical properties of printed parts ([Jaisingh Sheoran and Kumar, 2020](#ref-jaisinghsheoran2020)). According to Oberloier et al. ([2022a](#ref-oberloier2022)), particle swarm optimization (PSO) is an accurate and time-effective method for achiving this goal. To optimize the 3-D printing parameters for the rPET90//rHDPE10 material in the GigabotX we utilized the open-source PSO Experimenter platform which is available for Linux. The methodology developed by Oberloier et al. ([2022a](#ref-oberloier2022)) was followed during the optimization. For benchmarking purposes, three artifacts were printed: a line, a plane, and a cube. These artifacts were modeled in CAD software Onshape CAD v1.150 and sliced using Prusaslicer v2.52.0. [Figure 3](#fig-cad) presents the geometry models and dimensions of the artifacts.

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| Figure 3: Dimensions and CAD models of the geometries used for parameters optimization. |

Four parameters were assessed: 1) nozzle temperature, 2) bed temperature, 3) printing speed and 4) extrusion multiplier ([Oberloier et al., 2022b](#ref-oberloier2022a)). The initial parameters for the line are presented in Table 1a while additional parameters were obtained from preliminary experimental work shown in Table 1.b. Finally, the PSO tuning parameters were found in the previous PSO work ([Oberloier et al., 2022a](#ref-oberloier2022)) Table 1.c.

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| (a) Line optimization initial parameters   | Variable | Min | Max | Guess | True/False | Description | | --- | --- | --- | --- | --- | --- | | T1 | 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 | |

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|  |  | (b) Fixed parameters to perform printing parameters optimization based on PSO   | Parameters | Value | Units | | --- | --- | --- | | Layer height | 0.5 | mm | | Width | 2 | mm | | T2 | 230 | °C | | T3 | 220 | °C | | Cooling | 0 | % | | Infill density | 2 | % | |

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|  |  | (c) Recommended parameters for PSO tuning   | Variable | Value | Description | | --- | --- | --- | | Kv | 0.5 | The emphasis given to the velocity component | | Kp | 1.0 | The emphasis given to a particle's personal best position | | Kg | 2.0 | The emphasis given to the swarm's group's best position | |

Table 1: table 1

### Fused Granular Fabrication –FGF-

To print the obtained raw material, a modified open-source printer with three heat zones (Gigabot XL re:3D, Houston, TX, USA) was utilized as illustrated in [Figure 4](#fig-gigabot). The machine is a single screw extrusion-based 3-D printer capable of direct printing pellets, flakes, or granules, with a nozzle size of . For this study, a chair was printed to evaluate the material’s ability to be 3-D printed and the printer’s capability to produce large objectslike furniture. The ideal parameters determined for the cube geometry were employed to print the final part.

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| Figure 4: Fused granular fabrication printer Gigabot |

# Results and discussion

## Material characterization

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

### Material particle size analysis (granulometry)

Previous studies demonstrated that particles with areas smaller than were optimal for printing without experiencing jamming or under-extrusion problems ([Woern et al., 2018](#ref-woern2018)). However, our experiments revealed that particles with areas exceeding caused clogging in the feeding system and auger screw of the machine. As a result, granulometry analysis was performed using three different mesh sizes.

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| |  | | --- | | Figure 5: Granulometry analysis | |  |  |

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| |  | | --- | | Figure 6: Gigabot feeding system | |  |

[Figure 5](#fig-granulometry) presents the obtained results, indicating that particles sifted at exhibited an average area similar to the reference. There are, however, particles with areas exceeding caused blockages in the feeding and extrusion section. Particles sifted to displayed a distribution ranging from 0 to approximately , which was deemed too small for printing purposes. The presence of these small particles can lead to their complete melting in the initial heat zone, thereby impeding the smooth flow of other particles and preventing the necessary pressure for extruding the melted particles further down the screw. Although flakes measuring 3 mm exhibited a more dispersed distribution and slightly smaller area compared to the reference, they were found to be optimal for printing.

The final objects, however, still showed under-extrusion issues. To address this problem, a crammer was implemented ([Little et al., 2020](#ref-little2020)) as presented in [Figure 6](#fig-crammer). The crammer physically pushes particles towards the auger, facilitating their transfer from the feeding tube to the extruder. After the crammer implementation the under-extrusion issues were greatly reduced. It was concluded that flakes with areas ranging from to were the most suitable for printing when using a crammer to assist the feeding system.

### Chemical analysis from FTIR

Chemical structure information of the materials was obtained using FTIR spectroscopy, which allowed the analysis of the characteristic spectral bands of the polymers.

In the case of rPET (bottle) four distinct bands can be observed in [Figure 7](#fig-FTIR). The first band, located at represent the double bond. The second band, at , corresponds to the single bond ester. The third band, at , is associated with band the methylene group and vibrations of the ester bond. Lastly, a band at which represents the CH2 rocking bending vibration. Similar results were reported in the literature for PET derived from recycled water bottles, soda bottles, and food containers ([Zander et al., 2018](#ref-zander2018)).

Regarding rHDPE (caps), four characteristic peaks were identified: the C-H functional group bond at and , the primary bending mode of the -CH2 at and the CH2 rocking bending vibration at . The results obtained confirmed the chemical structures of the starting materials. Additionally, no other indicative resonances, apart from those associated with the polymer structures were detected. This leads to the conclusion that there were no significant amounts of additives or plasticizers present in either of the samples. Moreover, the spectrum of the printed blend (rPET90//rHDPE10) exhibited identical characteristic peaks to those observed in the bottle, thus confirming the predominant presence of PET. There are, however, noticeable differences between and as well as in the C-H bond ( and peaks), which confirm the presence of HDPE (cap). The observed shift can be attributed to interactions between the two materials.

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| Figure 7: FTIR spectra of rPET, rHDPE, and their blend |

### Thermal analysis DSC

The thermal properties of both recycled materials and their blend were characterized using DSC to establish a baseline for optimizing process parameters of 3-D printing.

Two distinct endothermic peaks are observed in the representative heating and cooling thermograms shown in [Figure 8](#fig-DCS), for the printed blend sample. These peaks are associated with the fusion of the crystalline fractions of rHDPE and rPET, providing confirmation of the immiscibility of both materials. Moreover, the enthalpy of fusion and crystallization of the rHDPE in the blend is significantly reduced, which can be attributed to the low percentage of HDPE present in the blend. Furthermore, the presence of a cold crystallization peak in the blend, but not in the individual polymers,suggests an interaction between the two polymers. It is possible that the rHDPE acts as a nucleating agent in this interaction. Table 2.lists the thermal properties of rPET, rHDPE and rPET90//rHDPE10. The melting points of rHDPE and rPET are 131.7 °C and 249.9 °C, respectively, which align with previous findings in the literature ([Chen et al., 2015](#ref-chen2015); [Lei et al., 2009](#ref-lei2009); [Vaucher et al., 2022](#ref-vaucher2022)). It is observed that the melting and crystallization temperature of rPET increased, while that of rHDPE slightly decreased. Furthermore, the crystallization of rPET was found to be somewhat affected by the presence of rHDPE, resulting in a 3.5% increase in degree of crystallization.This can be attributed to the rHDPE acting as a germination point for crystallization ([Vaucher et al., 2022](#ref-vaucher2022)). The slight changesin the fusion-crystallization temperatures and degree of crystallinity of rPET indicate an interaction of both polymers.

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| |  |  |  | | --- | --- | --- | | |  | | --- | | (a) Heating curves | |  |  |  |  | | --- | --- | | |  | | --- | | (b) Cooling curve | |   Figure 8: DSC thermograms of recycled materials and blends |

Thermal analysis of rPET, rHDPE, and their blend

|  | Glass transition | Melting | | Crystallization | | | % Crystallinity |
| --- | --- | --- | --- | --- | --- | --- | --- |
| Sample | Tg (°C) | Tm (°C ) | ΔHm (J/g) | Tc (°C ) | ΔHc (J/g) | ΔHcc (J/g) | Xc |
| rPET | 82 | 249.9 | 32.3 | 196.7 | 33.3 | - | 23.1 |
| rHDPE | - | 133.8 | 172 | 118.7 | 158.2 | - | 58.7 |
| rPET90/rHDPE10 | 77 / - | 254/131.7 | 40.3/1.30 | 210.6/117.4 | 37.9/6.7 | 6.8 | 26.6 / 18.8 |

### Rheology MFI

The melt flow index of the flakes was determined, enabling a fast and practical screening of the viscosity of the material. Based on the DSC results, the initial temperature for the MFI test was 250°C. However, the material did not flow reliably at this temperature, so it was increased by 5°C to enable the determinationof the melt flow index of the rPET90//rHDPE10 blend. A temperature of 260°C was also tested, however, the material flowed too rapidly, making difficult to obtain reliable measurements. The MFI tests were performed three times and the results for the rPET90//rHDPE10 blend showed medium MFI of 39.4± 2.4 g/10min. This value is consistent with similar values reported in the literature for rPET ([Bustos Seibert et al., 2022](#ref-bustosseibert2022); [Langer et al., 2020](#ref-Langer2020); [Nofar and Oğuz, 2019](#ref-nofar2019)). This result suggest that addition of low percentage of HDPE does not significantly impact the MFI value of rPET. Since the material flowed at a temperature of 255°C in the MFI test, this temperature was used as the input temperature for optimizing the parameters of the 3-D printer.

### Density

The density provides valuable information for estimating the cost, material usage, time consumption, and weight of the printed object in the slicer. This information is useful to determine the accurate printing parameters using the PSO experimenter, as the fitness of the object is calculated based on its dimensional accuracy and weight. Hence, density plays a significant role in determining the weight of the geometries.

After conducting calculations and measuring the rPET90//rHDPE10 injected object, it was determined that the density of the material is 1.13 . The inclusion of HDPE in the matrix polymer resulted in a slight decrease in density, which is a common occurrence when a polymer is mixed with a lower-density polymer. However, if we consider a PET/HDPE blend with a mass ratio of 90/10, the calculated theoretical density would be 1.32 . The observed decrease of 14% in the results could be attributed to factors, such as experimental conditions and manual measurements.

## Particle swarm optimization (PSO) Experimenter

Geometries were 3-D printed by adjusting the parameters using the PSO Experimenter software. The fitness function is defined by the weighted sum of the dimensional measurements (length, width, height, and weight) of the printed object. A fitness value below 0.1 was consider desirable. In the software five particles were established for each iteration, resulting in five different parameter combinations being printed in each iteration.

After six iterations and a total of thirty lines printed, the first geometry (line) achieved a fitness value of less than 0.1. The optimal parameters for this geometry are listed in column two of [Table 2](#tbl-table3) and images of the resulting geometries are illustrated in [Figure 9](#fig-geometries) .

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| Figure 9: Images of the resulting geometries a) line, b) plane, c) cube |

Afterwards, these parameters were used as initial guesses for plane geometry, which achieved the desired fitness in the first iteration. Similarly, cubes were printed using the plane ideal parameter as the initial guess, and optimal parameters, were found in the first iteration. The results showed a significant decrease in printing speed, as the geometry complexity increased. Moreover, the cube geometry required a higher extrusion multiplier to fill gaps and overcome under-extrusion problems. The optimization of parameters for the three geometries took approximately 10h reducing the experimental time, compared to conventional methods. According to Oberloier et al. ([2022a](#ref-oberloier2022)), this experimentation time can be reduced by 97%. Indeed, the effectiveness of PSO in finding global optimum parameters is high, especially in cases with a large or complex design space ([Saad et al., 2019](#ref-saad2019a); [Selvam et al., 2020](#ref-selvam2020)).

Additionally, PSO converge to optimum solutions with fewer iterations than DoE methods ([Zhang et al., 2015](#ref-zhang2015)). Combining PSO with other meta-heuristic methods has demostrated higher ability to predict and optimize parameters (e.g. minimize surface roughness([Shirmohammadi et al., 2021](#ref-shirmohammadi2021)), compressive strength and porosity of scaffolds ([Asadi-Eydivand et al., 2016](#ref-asadi-eydivand2016)),and mechanical properties([Raju et al., 2019](#ref-raju2019))). However, DoE methods are still widely used as they provide insight into the effects of individual design parameters and their interactions while the ability to find interaction between the variables is not possible using PSO. In the beginning of optimization experiments, the understanding the process technique and function settings might be complex. The methodology used in this study, however, was easy to implement and the software used was free, open source, and user-friendly, which reduced the initial difficulty. Therefore, PSO was demonstrated to be an effective and highly accurate prediction technique for finding the initial optimum parameters for rPET90//rHDPE10 material for FGF/FPF.

Based on the result, it is evident that the optimal parameters for printing may vary depending on the object and each parameter has its own variation. One possible hypothesis is that the geometry of the object could influence the assignment of parameters and this effect might be more noticeable in large printings, yet further investigation is required to confirm this hypothesis. There are several physical mechanisms at play that are expected to alter the optimal printing parameters based on size and geometry of the object. For example, the cooling time and temperature history of a voxel will depend on the geometry of the printed object ([Cleeman et al., 2022](#ref-cleeman2022)). Thus, to maintain a consisten thermal history the printing parameters must be ajusted as the geometry changes. This thermal history can also have more subtle effects, such asimpacting the degree of crystallization even in the case of PLA ([Wijnen et al., 2018](#ref-wijnen2018)).

In addition, the effects of material extrusion are magnified with scale, including the impact of thermal expansion and contraction. Small changes in contraction during cooling may cause acceptable distortions for small prints, but these are magnified for larger prints (e.g. causing deformation and in the worst cases delamination or loss of bed adhesion)([Shah et al., 2019](#ref-shah2019)). Although, Roschli et al. ([2019](#ref-roschli2019)) showed the obstacles and possible solutions of the large-scale AM according to the way 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 optimization of small printed objects to large-volume objects are needed.

Table 2: 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 | - |

## Functional object print

The final parameters for print the case study product were determined based on the ideal paraeters found for the cube geometry.However, the print speed was ajusted to decrease the printing time and prevent delamination. This adjustment was made in accordance with the PSO results, which indicated that the material can be printed at a speed range of 10 to 20 . Increasing the printing speed reduces the cooling time between the layers,thereby minimizing the risk of delamination ([Roschli et al., 2019](#ref-roschli2019)), This is particularly important for larger objects, as delamination tends to be more pronounced in such cases.

The Gigabot X successfully produced a piece of furniture from multi-material recycled water bottles that included mixing HDPE and PET as shown in [Figure 10](#fig-child) a.

The printing quality is acceptable as a prototype, proving the machine’s capacity to print large-scale functional objects. The chair was able to comfortably hold a child with a mass of 20 kg, as shown in [Figure 10](#fig-child) f. However, further evaluation is needed for the material used in the printing process. the printed object showed weak bond strength between the adjacent layers resulting in delamination, as seen in [Figure 10](#fig-child) b . This could be attributed to the difference in chemical properties of the materials, their immiscibility ([Chu et al., 2022](#ref-chu2022); [William et al., 2021](#ref-william2021)), high crystallinity ([Verma et al., 2023](#ref-verma2023)) and the large volume of the object as delamination issues were more prominent during the printing of the chair compared to the parameters optimization process. The delamination observed in larger objects can be attributed to the rapid cooling of the layers before the material is once again deposited. This is in contrast to cube printing, where the smaller surface area allows better layer adhesion before complete cooling. Even popular 3-D printing materials like PLA can be affected by this issue, as observed from the print surface ([Wijnen et al., 2018](#ref-wijnen2018)). To address the delamination problem and improve material properties, the addition of agents that reduce could be benificial ([Dai et al., 1997](#ref-dai1997); [Inoya et al., 2012](#ref-inoya2012); [Kramer et al., 1994](#ref-kramer1994)). This can enhance interfacial bonds through polymer modification ([Gao et al., 2021](#ref-gao2021)) and viscosity reduction ([Ko et al., 2019](#ref-ko2019)). Additionally,we observed printing warping problems ([Figure 10](#fig-child) c), which are likely caused by the high crystallization rates of HDPE ([Schirmeister et al., 2019](#ref-schirmeister2019)). we tested the use of Magigoo adhesive (Thougth3D Ltd., Paola, Malta) and the addition of a brim to improve bed adhesion, yet these solutions did not completely resolve the problem. A previous study showed that the use of a building plate made of thermoplastic elastomer SEBS allowed the adhesion of the plastic and facilitated easy detachment of the printed object without any breakage or damage [([**schirmeister2019.This?**](#ref-schirmeister2019.This)) suggests a potencial solution that should be further evaluated in future work. Another visible issue present in the close angles of the printed object was the shrinkage ([Figure 10](#fig-child) d) which occurs during solidification and particularly upon polymer crystallization. Moreover, it is well-known that PET has hygroscopic tendencies and easily absorbs moisture from the temperature, which makes it difficult to extrude ([Bustos Seibert et al., 2022](#ref-bustosseibert2022)). As result, it is likely to break down in the presence of water, lowering the quality of the print. Prior to printing the chair some samples exhibited brittle behavior and void formation therefore, the material was consistently dried and the hopper was kept closed to prevent moisture from entering the environment.These measures helped to ensure a more suitable material for printing.Additionally, there are visible vibration and ringing problems ([Figure 10](#fig-child) e) caused by the machine upgrades.Both acceleration and jerk (the maximum value of instantaneous speed change) require finer tuning to resolve these issues.

|  |
| --- |
| Figure 10: Finished children’s chair and printing issues |

### Cost and environmental impact

The printing process took 10 hours and the printed object weighs 840 grams. 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. To improve the printing time, upgrading the the extruder motor to a more powerful would be benefitial. Besides, the energy required for 10 hours of 3-D printing was found to be 6 kW-hr resulting in a production cost of ~1.2 € in function of the electricity cost in France, and does not include the material cost, as the bottles used were obtained from post-consumer waste. When labor costs are not included, the price was significant reduced (~88%) compared to the low-cost options available in the market.

The economics of fabricating the case study product remained competitive even when using recycled plastic pellets or shreds, which are avalable on the market for prices ranging from 1-10 €/kg. However, it is important to note that labor, maintenance, and machine devaluation were not considered in the final price. These factors should be considered in future work to ensure a comprehensive economic evaluation.

Regarding the environmental impact, this study does not evaluate the entire life cycle of the printed object. However, various scientific studies have already shown the feasibility of distributed recycling ([Kerdlap et al., 2022](#ref-kerdlap2022); [Santander et al., 2020](#ref-santander2020)). A comparison between conventional and distributed manufacturing in terms of energy consumption and emissions has been conducted ([Kreiger and Pearce, 2013](#ref-Kreiger2013)). Other studies have examined the environmental performance of AM ([Colorado et al., 2020](#ref-colorado2020a); [Garcia et al., 2018](#ref-garcia2018)) and the appearance of DRAM as a source of raw material for diverse 3-D printers coming from post-consumer plastic waste in the form of either filament ([Hart et al., 2018](#ref-hart2018); [Mikula et al., 2021](#ref-mikula2021); [Mohammed et al., 2017b](#ref-mohammed2017a); [Pakkanen et al., 2017b](#ref-pakkanen2017)) or granules ([Alexandre et al., 2020](#ref-alexandre2020)).

Additionally, Caceres-Mendoza et al. ([2023](#ref-caceres-mendoza2023)) have developed a comprehensive life cycle assessment of a DRAM system focusing on the production of PLA filament, comparing virgin and recycled materials. The findings of their environmental analysis revealed a analysis revealed a reduction of approximately 97% in the production impacts, including climate change, fossil depletion, water depletion, and potential eutrophication, when using recycled filament as opposed to virgin filament. It is important to note that these results are subject to the energy supply and might vary depending on the geographical location.

# Conclusion and future work

This study examined the feasibility of using mixed post-consumer waste as a feedstock material for direct 3-D printing without the need of compatibilization. The results demostrated the potential of mixing solid waste plastics (PET/HDPE) to be used as feedstock material, as evidenced by successfully printing a water bottle using two incompatible polymers from the cap and body of the bottle. Additionally, the results found that a large-scale FGF 3-D printer was capable of producing cost-effective functional object using these mixed waste PET/HDPE plastics. However, further research is necessary to analyze the mechanical properties of the material and explore the use of compatibilizers that can enhance the interphase tension between plastics and reduce their crystallinity. These measures could potentially improve and enhance the properties of both the material and the 3-D printed parts.

These considerations become increasingly important as the size of the 3-D printed part increases. The improvement of the material science of this approach can also offer an opportunity to improve the quality of the printing time, reduce energy consumption of the machine, and improve the economic viability of DRAM using mixed plastic waste.

In addition, future work could assess the different combinations or blends of commodity plastics with or without the use of compatibilizers, to determine their printability. This investigation could lead ti the elimination of the selection/sorting process. In the same way, the development of a methodology that ensure process reproducibility, even in areas with limited infrastructure opens up the potential for plastic revalorization using DRAM.

# Declaration of competing

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

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