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Recycled polycarbonate and polycarbonate/acrylonitrile butadiene styrene feedstocks for circular economy product applications with fused granular fabrication-based additive manufacturing

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Abstract

Distributed recycling and additive manufacturing (DRAM) holds enormous promise for enabling a circular economy. Most DRAM studies have focused on single thermoplastic waste stream. This study takes three paths forward from the previous literature: 1) expanding DRAM into high-performance polycarbonate/ acrylonitrile butadiene styrene (PC/ABS) blends, 2) extending PC/ABS blend research into both recycled materials and into direct fused granular fabrication (FGF) 3-D printing and 3) demonstrating the potential of using recycled PC/ABS feedstocks for new applications in circular economy contexts. A commercial open source large-format FGF 3-D printer was modified and used to assess the different printability and accuracy of recycled PC and PC/ABS. The mechanical properties (tensile and impact) following the ASTM D638 and D6110-18 standards were quantified. A weather simulation test (ASTM D5071-06) was performed to assess outdoor performance. Finally, two applications in sporting goods and furniture were demonstrated. In general, better printability was achieved with recycled PC/ABS compared to recycled PC, as well as good dimensional accuracy at printing speeds of 30 and 40 mm/s. Minimal qualitative differences and discoloration were visible on the samples after accelerated weather exposure, with results in accordance with the state-of-the-art. The rPC/ABS results from tensile tests show similar values to those of rPC for elastic modulus (2.1 ± 0.1 GPa), tensile strength (41.6 ± 6.3 MPA), and elongation at break (2.8 ± 0.9 %), which are also comparable with previous studied virgin 3-D printed filaments. Similarly, impact energy (115.78 ± 24.40 kJ/m^2) and resistance values (810.36 ± 165.77 J/m) are comparable in the two tested formulations, reaching similar results compared to FFF 3-D printed filaments, as well as virgin materials for injection molding. Finally, the two demonstration products in the sporting goods and furniture sectors were successfully fabricated with rPC/ABS, achieving complex patterns and good printing speeds for recycled feedstocks. It is concluded that rPC/ABS blends represent a potential high-performance feedstock for DRAM, validating its use in direct FGF 3-D printing systems and potential applications for a circular economy.

Keywords: 3-D printing; Recycling; Fused Filament Fabrication (FFF); Fused Granular Fabrication (FGF); Distributed Recycling for Additive Manufacturing (DRAM); Mechanical properties.

1. Introduction

The global plastic waste problem [1] has the potential to be aggravated by the rapid rise of 3-D printing waste products [2]. Although the first self-replicating rapid prototyper (RepRap) [3-5] released as an open source 3-D printer resulted in rapid innovation, cost reductions, and the democratization of additive manufacturing (AM) [6-8], a concerning amount of 3-D printing plastic waste is landfilled [9] as the global 3-D printing market is going to reach $7.7 billion by 2024 [10]. A superior approach to a linear material process, even if distributed, is to target a circular economy for AM [11,12]. This can be done by using the distributed recycling for additive manufacturing (DRAM) model [13-15]. In the DRAM model prosumers (producing consumers) save or make money when recycling with DRAM, as opposed to complete lack of personal financial incentive in traditional recycling models [13]. Prosumers following DRAM can use both AM waste and traditional plastic waste to produce 3-D printing feedstocks, which are generally worth $20/kg as opposed to $1-5/kg for plastic scrap. By producing either the feedstock or even more valuable 3-D printed products directly where they are consumed, there are substantive environmental benefits [16-18]. DRAM prosumers can generally save substantial money per product [19,20] and

obtain an extremely high return on investment [21]. The DRAM approach can be applied globally [22] and thus could have a major impact on global value chains [23].

DRAM research started and for some time has centered on the use of some type of recyclebot, which is a waste plastic extruder [24,25] that manufacturers filament to be used in fused filament fabrication (FFF)-based low-cost 3-D printers. Recyclebot-based DRAM has been demonstrated for a wide range of single polymer materials: acrylonitrile butadiene styrene (ABS) [22,26-29], polylactic acid (PLA) [25,30], high-density polyethylene (HDPE) [31,32], and polyethylene terephthalate (PET) [33]. Only a few studies, however, looked at the potential of making filament from composites or blends [35-38]. When using the recyclebot-to-3-D printing filament method for each cycle of materials, there are two melt and solidification sub-cycles. Each sub-cycle reduces the length of the polymer chains, which weakens the material until by approximately the fifth full cycle the material properties have degraded enough that an intervention is necessary [34]. Another approach, with more promise to extend the lifetime of recyclable polymers in the DRAM context is to use direct extrusion-based waste 3-D printing. This can be done via fused granular fabrication (FGF)/fused particle fabrication (FPF) for a number of plastics including polypropylene (PP) [39], PET [40]; PLA [41,42], and ABS [43]. There are a number of commercial large-format 3-D printers (GigabotXLT, Exabot, Terabot, The BoX, T3500, 400 series, BIG-meter, BigRep One, F1000) [44-52], waste hangprinters [53,54] that can be used in DRAM and the use of these printers for DRAM is gaining attention for the potential to foster new applications, i.e., furniture and building. A limited range of recycled feedstocks, however, is currently considered within this context.

FPF/FGF has also been demonstrated with the higher strength single polymers such as polycarbonate (PC), which also allows for molding of lower melting temperatures and the use of materials that are less easily processed by AM [55]. PC, however, has disadvantages, such as poor processability, hygroscopic behavior, low scratch resistance, and high production costs [56]. These disadvantages can be overcome using a blend of PC and ABS, improving the processability of PC and keeping good tensile, impact, and flexural properties [57,58]. PC/ABS blends usually show good heat resistance and high values of impact strength and toughness [59]. Focusing on FFF systems, parts made of PC/ABS filaments reach good mechanical properties, such as elastic moduli and tensile strength [60,61], despite the strong influence of interlayer adhesion and building orientation of 3-D printed samples [62]. Despite the increasing interest in recycling engineering polymers for new applications in 3-D printing, no previous works were found on the use of PC/ABS feedstock on FGF systems, as well as recycled blends on FFF/FGF 3-D printers. The processability of PC/ABS after multiple reprocessing cycles was previously assessed only for injection molding [63], showing a knowledge gap for FFF/FGF systems.

This study thus takes three paths forward from the previous literature. First it expands the DRAM work into high performance PC/ABS blends. Second, it extends the PC/ABS blend research into both recycled materials and direct FGF 3-D printing. Thus, this work investigates the use of recycled PC and PC/ABS blend with large-format open source FGF 3-D printing system. Finally, the aim is to demonstrate the potential of using recycled PC/ABS feedstocks for new applications in circular economy contexts, exploiting its use with large-format 3-D printers. A commercial open source large-format FGF 3-D printer was modified and used to assess the different printability and accuracy of recycled PC and recycled PC/ABS as a preliminary step. The mechanical properties (tensile and impact) following the ASTM D638 and D6110-18 standards were quantified. A weather simulation test (ASTM D5071-06) was then performed to assess the influence of weather to allow for outdoor potential applications. Finally, two application fields were selected for the development of two new products to demonstrate the feasibility of using recycled feedstock for real applications, i.e., protective sports equipment and customizable furniture. To this end, recycled PC and PC/ABS feedstock can potentially foster the design of new products by widening the range of possible application fields to more technical sectors and accessible products.

2. Methods

2.1 Materials

Recycled polycarbonate (rPC) feedstock was provided by Advanced Composites (Sidney, OH, US) in the form of shredded shards and particles. After a preliminary 3D printing test with an average particle size of 5 mm, the recycled feedstock was sorted with a 3 mm mesh size sieve to obtain a relatively uniform granulometry of the material, reaching the average particle size of commercial 3-D printing-graded pellets. Moreover, a maximum particle area of 22 mm^2 can be used as a feedstock for the GigabotX extruder chamber [39]. Recycled acrylonitrile butadiene styrene (rABS) from McDunnough (Fenton, MI, US) was used in the form of pellets as provided (3 mm of average particle size). Two different materials were tested within this work: 100% wt. of rPC feedstock (Fig. 1a) and a blend of rPC and rABS (Fig. 1b). In detail, the rPC/ABS blend was composed of 70% wt. of rPC feedstock and 30% wt. of rABS pellets. This percentage was selected by considering the state of the art in 3-D printed PC/ABS blends with FFF and FGF systems [57,64,65], as well as the aim to maximize the percentage of rPC feedstock of the blend and increase its mechanical properties, especially to reach high-performance application fields and technical equipment. The rPC/ABS ratio was then selected to obtain a blend with good mechanical properties and processability of the extrudate [57,58].

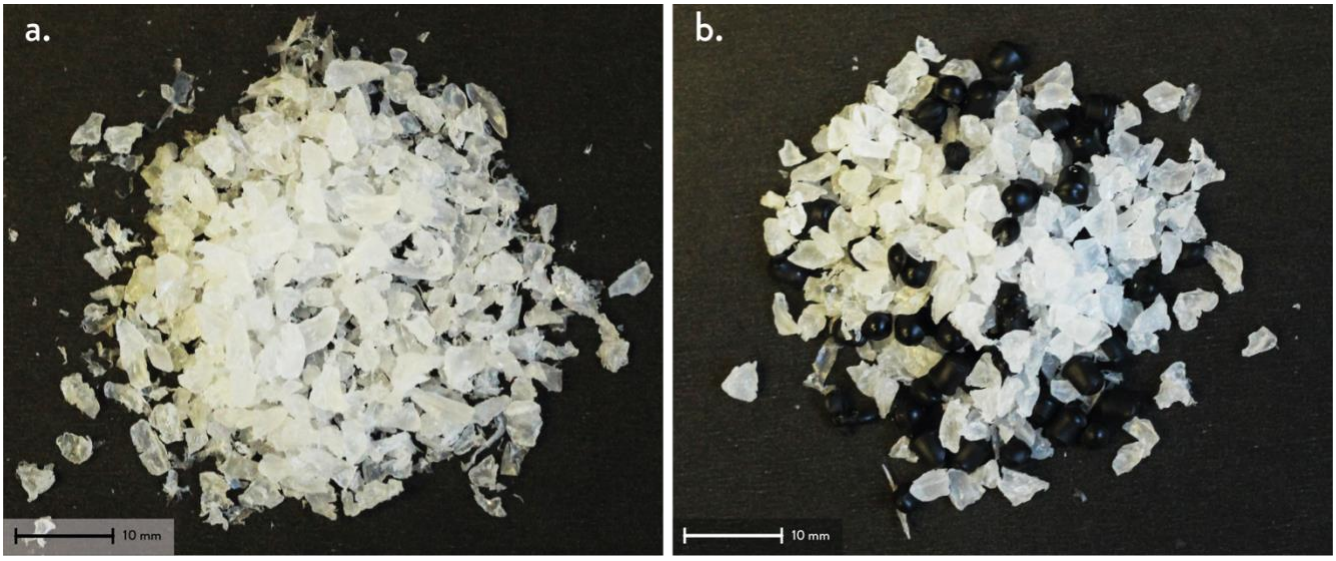


Fig. 1. Secondary raw materials used in this work: (a) 100% wt. of rPC feedstock and (b) rPC/rABS blend (70% wt. and 30% wt.).

2.2 Printability comparison tests

The accuracy of the FGF 3-D printing system was assessed through a comparison of different variations of a 3-D printed sample, designed to evaluate the printability and accuracy of the system with recycled feedstock (Fig. 2). This sample has a nominal dimension of 87x45x45 mm. Its shape comprises some perpendicular walls for wall thickness measurements and some overhang walls to verify the shape retention after the extrusion of each layer. The sample designs were sliced with the open source PrusaSlicer (Prusa Research, Prague, Czech Republic) [66], and all designs for this study are available on the Open Science Framework (OSF) under GNU General Public License (GPL) 3.0 [67]. The sample was sliced by combining different printing speeds (20, 30, and 40 mm/s) with a layer height of 0.4 mm, which means ½ of the nozzle diameter, obtaining a batch of three combinations for each material, six different sets of 3-D printed pieces in total. The other printing parameters were kept constant for each gcode and are summarized in Table 1 (rPC) and Table 2 (rPC/ABS). The 3-D printed samples were visually inspected and measured to obtain experimental values to compare with the nominal ones. The comparisons were performed on the weight, overall dimensions, and wall thickness. The nominal weight values were estimated by calculating the density of rPC (1.2 g/cm^3) and rPC/ABS (1.15 g/cm^3) and inserting the values in PrusaSlicer. The values were then used to assess the absolute error of the 3-D printer at different layer heights and printing speeds, resulting in two comparative matrixes. This sample was also used to preliminary evaluate the extrudability of rPC feedstock with 5 mm particle size.

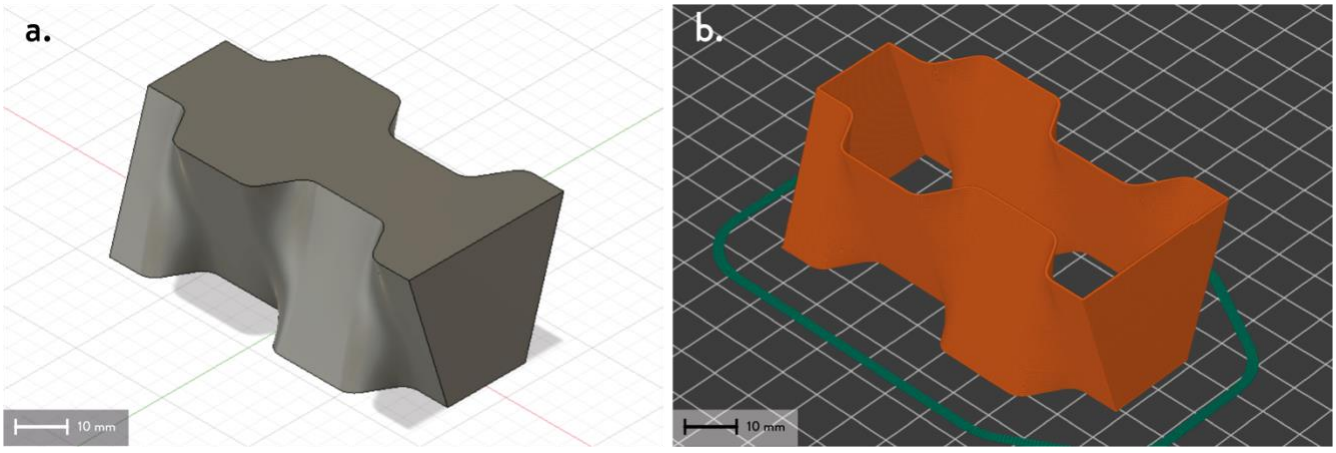


Fig. 2. Printability test design: (a) CAD preview of the STEP file and (b) gcode preview.

Table 1. Main 3-D printing parameters - recycled PC.

Parameters	Unit	Brick samples	Tensile tests	Impact tests	UV tests
Nozzle diameter	mm	0.8	0.8	0.8	0.8
Layer height	mm	0.2 – 0.4	0.3	0.3	0.3
Speed	mm/min	20, 30, 40	25	30	40
Wall line count – perimeters	//	1	2	2	2
Extrusion temperature	°C	250 (H1), 235 (H2), 220 (H3)	250 (H1), 235 (H2), 220 (H3)	250 (H1), 235 (H2), 220 (H3)	250 (H1), 235 (H2), 220 (H3)
Bed temperature	°C	110	110	110	110
Flow	%	90	80	70	60
Cooling fan	%	0	0	0	0

Table 2. Main 3-D printing parameters - recycled PC/ABS.

Parameters	Unit	Brick samples	Tensile tests	Impact tests	UV tests	Application (Sports)	Application (Furniture)
Nozzle diameter	mm	0.8	0.8	0.8	0.8	0.8	0.8
Layer height	mm	0.2 – 0.4	0.3	0.3	0.3	0.4	0.4
Speed	mm/min	20, 30, 40	30	20	30	30	40
Wall line count – perimeters	//	1	2	2	2	3	2
Infill percentage	%	0	100	100	100	0	10
Extrusion temperature	°C	240 (H1), 230 (H2), 215 (H3)	240 (H1), 230 (H2), 215 (H3)	240 (H1), 230 (H2), 215 (H3)	240 (H1), 230 (H2), 215 (H3)	240 (H1), 230 (H2), 215 (H3)	240 (H1), 230 (H2), 215 (H3)
Bed temperature	°C	110	110	110	110	110	110
Flow	%	80	60	40	50	60	60
Cooling fan	%	0	0	0	0	0	0

2.3 Weather simulation test

The possible photodegradation effects of weather on 3-D printed rPC and rPC/ABS were preliminarily evaluated through simulated weathering experiments, helping in narrowing down the possible range of applications of the tested materials. The simulation was done using the Xenon Arc apparatus Q-Sun Xenon Test Chamber Model Xe-3 (Q-Lab Corporation, Westlake, OH, USA) equipped with a sunlight simulation lamp and following the ASTM Standard D5071-06 [68]. Two different batches of five squared samples each (nominal dimensions: 70x70x3 mm) were 3-D printed using the printing parameters shown in Table 1 (rPC) and Table 2 (rPC/ABS). An additional sample of commercial injection-molded PC/ABS was used as a control material during the simulation. The surface of each sample was cleaned with methanol before positioning it in the testing chamber with some tape. The simulation was performed by following the parameters of Cycle 2 for 24h, simulating exposures with slight moisture stresses. The main cycle (2h in total) consists of a continuous light time of 102 min and 18 min of light and water spray conducted with a temperature of 63°C, relative humidity of 60%, and irradiance of 0.35 W/(m^2·nm) at 340 nm.

To detect some physical changes due to the simulation, a colorimetry test was conducted on the specimens before and after their exposure in the Xenon Arc apparatus by means of a spectrophotometer (Spectro2guide from BYK Additives & Instruments, Wesel, Germany), following the ASTM Standard D2244-21 [69]. Each sample was cleaned with methanol before measuring the L*a*b* coordinates of the CIELAB color space, where L* means lightness, and a* and b* the color coordinates. Five different points for each sample were measured, obtaining the mean values and standard deviations of the L*a*b* coordinates before and after the weather simulation test. The direction of the color difference ΔL*, Δa*, Δb* and the discoloration ΔE* were then calculated by using equations 1-4,

$$\Delta L^* = L^*_B - L^*_S \quad \text{Eq. 1}$$

$$\Delta a^* = a^*_B - a^*_S \quad \text{Eq. 2}$$

$$\Delta b^* = b^*_B - b^*_S \quad \text{Eq. 3}$$

$$\Delta E^* = \sqrt{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2} \quad \text{Eq. 4}$$

where L^*_B, a^*_B, and b^*_B are the coordinates after the test, and L^*_S, a^*_S, and b^*_S before the test. The signs of ΔL*, Δa*, Δb* give an overall idea of the difference in colors according to the following path: +ΔL* lighter, -ΔL* darker, +Δa* redder, -Δa* greener, +Δb* yellower, -Δb* bluer. The spectrophotometer was also used to measure the gloss of the sample surfaces before and after the weather simulation test with a 60° geometry, expressing the results in Gloss Units (GU). Five different measurements were performed on each sample, allowing to calculate the mean values and standard deviations.

2.4 Tensile tests

Tensile tests were carried out using a Zwick Roell Z010 testing machine (ZwickRoell GmbH & Co. KG, Ulm, Germany) equipped with a 10 kN cell load and an extensometer, following the ASTM standard D638-22 [70]. Tests were performed at a speed of 1 mm/min. Two batches of ten Type IV samples (33 mm of gauge length, 4 mm of thickness, and 6 mm of width) were fabricated following the parameters shown in Table 1 (rPC) and Table 2 (rPC/ABS) [71,72]. To avoid interlayer voids, the specimens were printed with a 100% rectilinear infill. In case of asperities on the top layers, they were manually removed by sanding to reach a constant cross-section in the gauge length. The actual thickness and width were then measured with a caliper. After testing, the mean values and standard errors of stress, strain, and elastic modulus were calculated from the stress-strain curves obtained in the tests.

2.5 Impact tests

Impact tests were performed with a Charpy impact tester (Instron, Norwood, MA, USA) following the ASTM standard D6110-18 [73] with a span of 40 mm. A pendulum of 30 kg with a capacity of 252 J was used for the tests (length of 1.3 m and angle of 70°). Two batches of ten notched samples were 3-D printed using the parameters of Table 1 (rPC) and Table 2 (rPC/ABS). Their overall dimensions are 12.7x63.5x6.5 mm. The notch has a maximum nominal depth of 2.6 mm and an angle of 45°. As for tensile bars, the samples were printed with a 100% rectilinear infill, and the main asperities were manually sanded when interfering with the notch geometry. The actual widths and depths under the notch were measured with a caliper for each specimen. The machine was calibrated before conducting the tests by raising and releasing the pendulum without loading any sample. The mean net breaking energy and impact energy values, as well as their standard deviations, were calculated starting from the experimental results of the test.

2.6 Fracture analysis

The morphology of the fracture cross-section surface was evaluated by using microscope micrographs performed with a Keyence VHX-6000 Digital Microscope (Keyence, Mississauga, ON, Canada). Two batches of fracture cross-sections were considered for the analysis, which means the surfaces from impact specimens of rPC and rPC/ABS. At least two fracture surfaces for each sample batch were observed. The samples were observed at 100 x magnification.

2.7 Application case studies

Some applications were chosen after the printability and material characterization tests were completed. The aim was to demonstrate the use of rPC and rPC/ABS, as well as the FGF 3-D printer system, for real applications, reducing the gap between experimental studies and practical implementations. Two different case studies were selected, which correspond to different application fields: protective sports equipment and customizable furniture, resulting in two products with functional purposes. They were mainly selected to exploit the properties of PC/ABS blends in a consolidated field for 3-D printing and a potential new area, i.e., furniture and sports. The specific products were designed to include some 3-D printing features challenging for large-scale AM, e.g., using infill, layers with small cross-sections, travel paths, and complex cross-sectional patterns [54]. The 3-D models of the protective equipment product were designed with Grasshopper plugin for Rhinoceros 7 (Robert McNeel & Associates, Seattle, WA, USA), which helped in optimizing the shape of the models by considering the possible extrusion path of the 3-D printing hot end. The 3-D model of the customizable furniture (coat hanger joint) was previously designed and used to test large-scale cable-robot-based hangprinting systems by Rattan et al. [54]. More details about the selection criteria of the two fields, the design constraints, and the rationale behind the development process are in the Results and Discussion section. The gcode files to fabricate the two products were done using PrusaSlicer [66], and the

main parameters for both applications are visible in Table 1 (rPC) and Table 2 (rPC/ABS). The open-source designs in native CAD format, STEP and STL are available in the OSF repository [67].

2.8 3-D Printing setup redesign and parameters

The 3-D printing system used in this work is a Gigabot XLT from re:3D Inc (Houston, TX, USA). This machine is a gantry FGF open-source 3-D printer controlled with Marlin Firmware [74], equipped with a pellet extruder heated by three different heating zones, hereinafter called H1, H2, and H3, and a stainless-steel conical nozzle with a diameter of 0.8 mm [40]. The building volume of Gigabot XLT is 590x760x900 mm. This means that it can be considered a large-format 3-D printer since its maximum printing volume reaches almost 1 m^3 [75]. To improve the feeding of recycled feedstock and the overall performance of the system, a new version of the extruder was developed by modifying the previous design [40]. In detail, the latest version (Fig. 3a) aims to fix four main issues:

- Facilitating the feeding of recycled feedstock by redesigning the inner shape of the 3-D printed feeder, avoiding sharp angles, and removing the crammer (Fig. 3b).
- Fixing the thermocouples on H1, H2, and H3 by using accessible spare parts such as metric inserts, screws, and ring terminals, improving the reliability of the temperatures measured from the three heating zones (Fig. 3b).
- Redesigning the 3-D printed motor spacer to improve the torque resistance of the part during the extrusion (Fig. 3d).
- Improving the insulation and protection of the heating zones from feedstock with a new 3-D printed barrel cover (Fig. 3c).

The parts were designed with Fusion 360, sliced with PrusaSlicer [66], and fabricated with an open-source Prusa i3 MK3S FFF-based 3-D printer with a 0.4 mm nozzle (Prusa Research, Prague, Czech Republic). PETG and ASA were selected to manufacture the parts with a 100% infill and a layer height of 0.2 mm. The BOM and stl files of the new extruder version are available in the OSF repository [67].
The main 3-D printing parameters for the Gigabot X and recycled feedstock used in this work are listed in Table 1 (rPC) and Table 2 (rPC/ABS). In addition, to improve the adhesion of rPC and rPC/ABS, some adhesive was applied with a glue stick on the heated bed. The Prusa slicer profiles with the main parameters can be found in the OSF repository [67].



Fig. 23 New version of the GigabotX extruder: (a) preview of the assembly without insulation; (b) insight on the feeding system and thermocouple fixing; (c) preview of the assembly with insulation; and (d) insight on the motor support and insulation cover.

3. Results and Discussion

3.1 Printability comparison tests

After a preliminary 3-D printing test with 5 mm size particles, feedstock with 3 mm size was selected for the experimentation. As a matter of fact, the quality of the extrudate by manually moving the extruder step motor and performing a 3-D printing test of the sample (Fig. 2) was not satisfactory, leading to frequent clogging and obstructions of the screw [40].
Six different sets of 3-D printed samples (Fig. 4) were fabricated to evaluate and compare the dimensional accuracy of the extruder using recycled PC (top) and recycled PC/ABS feedstocks (bottom). After a qualitative visual inspection, some general consideration may be done. In general, the overall extrusion consistency is quite accurate, especially considering using a 100% recycled PC and a PC/ABS blend with 70% recycled PC. In other words, the variation in the average feedstock granulometry size and shape linked to mechanical recycling processes is not affecting the quality of the extruded material. Consistent extrusion flows for rPC and rPC/ABS were obtained by choosing particle sizes similar to commercial 3-D printing graded pellets of more common materials, i.e., PLA. It should be noted, however, over-extrusion occurred in both cases. Focusing on recycled PC, the samples show a change in the overall quality of the extrusion according to the printing speed. Although the 3-D printed parts were successfully fabricated for each speed value, different extrusion line widths were obtained for each batch, resulting in thicker paths at lower temperatures. Furthermore, deformation occurred at lower speeds, i.e., 20 mm/s, and at higher speeds, such as 40 mm/s. The first deformation type is clearly visible from the lower accuracy in the z-axis direction and the low layer height consistency, whereas the second is linked to the slightly inclined perpendicular walls of the samples. These results may be linked to the shear rate of PC at different printing speeds, as well as the decrease in its die swelling at higher speeds after the extrusion, thus in thinner line widths [76,77]. Moreover, longer cooling times are required when using bigger nozzle diameters, affecting the overall deformation of the samples fabricated at higher printing speeds [78,79]. At the same time, the consistency of the extruded material decreased by increasing the printing speed, i.e., 40 mm/s, resulting in thinner walls and delamination between layers close to the sharp angles. This last issue may be linked to the lower interlayer adhesion at higher speeds, which means smaller bonding areas due to the thinner walls [80].

The recycled PC/ABS samples show less visible changes in the overall dimensions according to the printing speed, as well as in the extrusion line width. No signs of delamination are visible between the layers for each batch of samples, although thicker extrusion lines can be observed in the samples printed at 20 mm/s. The main reason may be the use of rABS pellet in the blend, which reduced the variability in feedstock granulometry in terms of shape, thus improving the consistency of the extrusion flow. Furthermore, the use of rABS in this PC-rich PC/ABS blend may help in improving its printability by reducing the viscosity of PC [81]. The former aspect reduces the risk of clogging and obstructions of the screw, leading to more homogeneous extrudate and lower variation in the flow consistency, improving the accuracy and reducing the presence of voids and under-extrusion [39,55]. The selected feedstock has a particle size (~3 mm) corresponding to an area lower than 22 mm^2, which has been previously found to be the limit to fit the extruder chamber of the GigabotX [39]. The latter aspect can also reduce the presence of voids and under-extrusion in the 3D printed part, reducing the flow resistance at the nozzle [57,81].

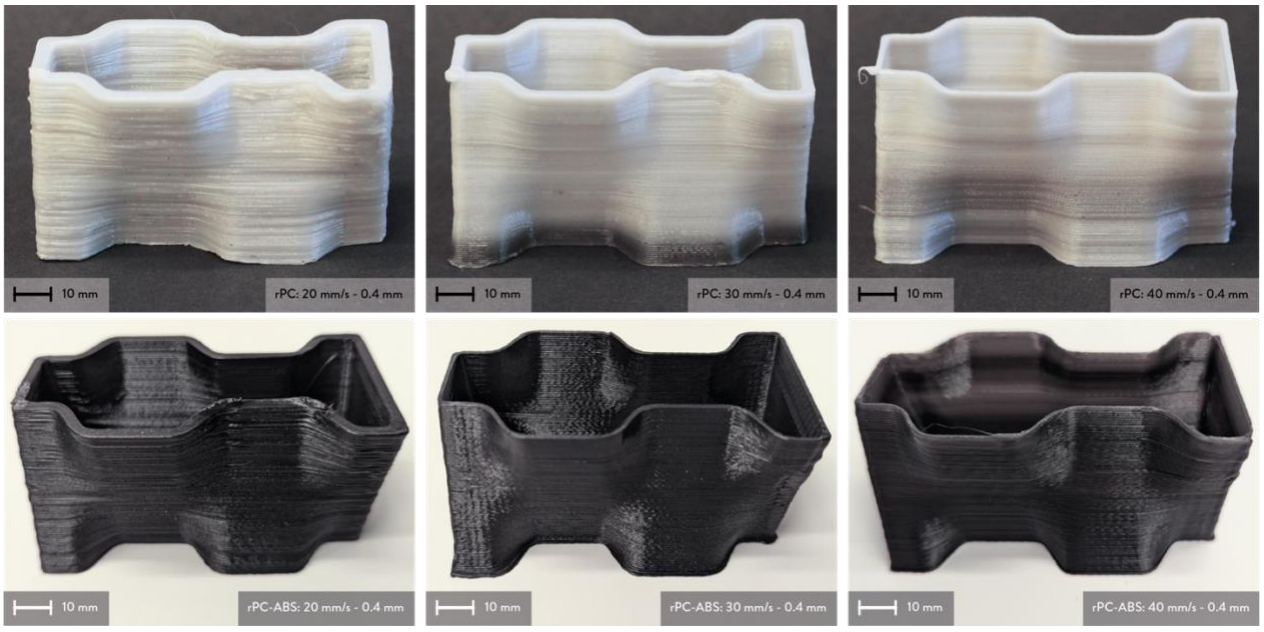


Fig. 4. 3-D printed brick samples on large-format FGF 3-D printer (layer height: 0.2 and 0.4 mm; printing speed: 20, 30, and 40 mm/s;): recycled PC (top) and recycled PC/ABS (bottom)

The overall dimensions and weight of the different batches were then measured to compare with the nominal values given by PrusaSlicer. The results of the measurements are resumed in Fig. 5, confirming the visual inspection and qualitative analysis. Less accurate measurements were obtained for samples printed at lower speeds using both materials, i.e., 20 mm/s, whereas more precise values can be seen at higher speeds.

rPC, however, exhibited higher absolute errors compared to rPC/ABS, especially considering weight and line width. The values of weight and line width at 20 mm/s of rPC were more than 200% higher than the nominal values, indicating critical over-extrusion problems. These high values in absolute error decreased by increasing the printing speed, although some over-extrusion remains at 40 mm/s. A similar trend can be seen by measuring the overall dimensions of the rPC samples despite the lower absolute and percent errors, i.e., less than 6%. Hence, the samples printed at 20 mm/s have bigger dimensions, especially length, whereas more accurate measurements were reached at 40 mm/s.

Considering rPC/ABS, more accurate values were obtained from the samples, especially at 30 and 40 mm/s, reaching lower absolute and percent errors, i.e., less than 1%. These values confirm the considerations made from the qualitative assessment, showing better printability and more consistent extrusion flow when dealing with rPC/ABS. The results also confirmed the influence of ABS pellets on the extrusion consistency. Considering the qualitative assessment and the dimensional analysis, the best compromise is reached at 30 mm/s, which provides a more consistent extrusion flow and homogeneity in the overall sample [78,79]. The recommended settings for PrusaSlicer can be found in the OSF repository [67].

	20 mm/s - 0.4 mm					30 mm/s - 0.4 mm					40 mm/s - 0.4 mm				
rPC	weight (g)	line width (mm)	length (mm)	width (mm)	height (mm)	weight (g)	line width (mm)	length (mm)	width (mm)	height (mm)	weight (g)	line width (mm)	length (mm)	width (mm)	height (mm)
Nominal values	14.35	1.2	85	45	45	14.35	1.2	85	45	45	14.35	1.2	85	45	45
Mean values	44.08	3.80	89.71	46.42	46.02	37.60	3.17	88.64	45.62	46.35	23.33	1.81	87.28	44.95	46.64
Standard Deviation	0.10	0.08	0.54	1.38	0.17	0.18	0.10	0.06	0.40	0.23	0.13	0.03	0.22	0.38	0.93
Absolute error	29.73	2.60	4.71	1.42	1.02	23.25	1.97	3.64	0.62	1.35	8.98	0.61	2.27	0.05	1.64
% error	207.2	216.7	5.5	3.1	2.3	162.2	164.2	4.3	1.4	3	62.5	51.1	2.7	-0.1	3.6
rPC-ABS	weight (g)	line width (mm)	length (mm)	width (mm)	height (mm)	weight (g)	line width (mm)	length (mm)	width (mm)	height (mm)	weight (g)	line width (mm)	length (mm)	width (mm)	height (mm)
Nominal values	13.75	1.2	85	45	45	13.75	1.2	85	45	45	13.75	1.2	85	45	45
Mean values	33.85	2.71	88.26	45.32	46.41	20.98	1.73	87.14	45.84	46.59	16.80	1.65	86.56	43.92	45.19
Standard Deviation	0.24	0.08	0.29	0.28	0.45	0.10	0.12	0.10	1.72	0.56	0.08	0.40	0.18	0.12	0.70
Absolute error	20.10	1.51	3.26	0.32	1.41	7.23	0.53	2.14	0.84	1.59	3.05	0.45	1.56	1.08	0.19
% error	146.2	126	3.8	0.7	3.1	52.5	43.8	2.5	1.9	3.5	22.2	37.1	1.8	-2.4	0.4

Fig. 5. Absolute error variation (weight, overall dimensions, line width) of recycled PC (top) and recycled PC/ABS (bottom). Red indicates high errors, orange indicates moderate errors, yellow indicates low errors and green indicates very low errors.

3.2 Weather simulation test

The photodegradation effects on rPC and rPC/ABS samples from their exposition to weather conditions were assessed with weather simulation tests performed according to ASTM D5071-06. Thanks to the constant temperature of the chamber, the wavelengths close to UVA, and the settings described in Section 2.3, the test can be considered as performed in accelerated sunlight weather conditions of a factor between 10 and 30 times, as shown by previous works [82,83]. Therefore, the 24-hour test of this work corresponds to a range between 10 and 30 days of exposure to natural sunlight weather conditions.

The samples before and after the weather simulation tests are visible in Fig. 6. Since no significant visible changes were qualitatively assessed, a colorimetry test was done to assess the change in color due to photodegradation from a quantitative point of view. The results resumed in Table 3 show minimal differences before and after the weather simulation tests. Slight discoloration (ΔE^*) happened to both samples, with a greater difference observed from the rPC samples. Even if the values of Δb^* have a negative sign, no significant trends were detected in the direction of the color difference. Only a slight decrease of lightness was noticed by analyzing the results, expressed by the negative sign of ΔL^* for both batches. This aspect may be linked to some minor photooxidation phenomena that occurred on the surface of the samples. Considering gloss measurements, similar values of GU can be seen from Table 5 before and after weather tests, showing no significant differences linked to exposure.

From the literature, yellowing phenomena, especially for ABS, were noticed after accelerated weathering tests [84,85], as well as a decrease in gloss [86], even after weeks of continuous exposures. According to the small differences detected from colorimetry, no further characterization tests were conducted on the batches after weather simulation tests. No remarkable changes in the mechanical properties of PC and PC/ABS were noticed after longer weather exposures in previous studies [86,87]. As investigated by Santos et al., significant modifications in the mechanical behavior of ABS were found only after more than 1200h of exposure in Xenon Arc apparatuses, which means more than one year of continuous exposure to sunlight [83]. Even if ABS generally shows photooxidative effects from sunlight exposure [84,85], the content of ABS represents the minor part of the blend analyzed in this work (30% wt.), hence the impact of its photodegradation on the mechanical properties is limited [86].

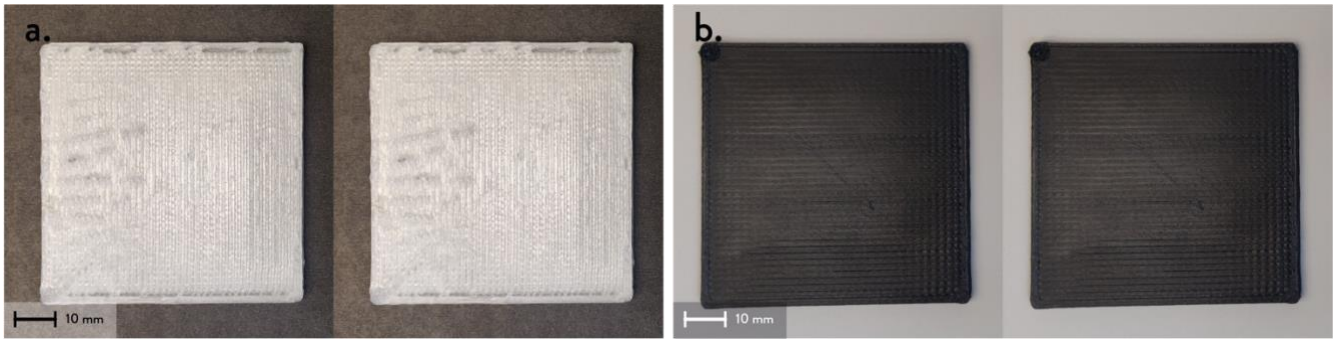


Fig. 6. Weather simulation specimens of (a) recycled PC and (b) recycled PC/ABS before (left) and after (right) conducting the test.

Table 3. Mean CIELAB L*a*b* values, ΔL*, Δ*a, Δ*b (direction of the color difference), ΔE* (discoloration), and gloss values at 60° of rPC and rPC/ABS before and after weather simulation test.

Batch		L*	a*	b*	ΔL*	Δa*	Δb*	ΔE	GU (60°)
PC/ABS (Control material)	Ref.	8.95 ± 0.13	-0.37 ± 0.05	-2.45 ± 0.28	//	//	//	//	//
rPC	Before	59.67 ± 1.86	-0.75 ± 0.02	1.33 ± 0.16	-0.50	0.07	-0.81	1.62	8.91 ± 2.14
	After	59.26 ± 1.16	-0.68 ± 0.01	0.53 ± 0.12					9.24 ± 0.65
rPC/ABS	Before	19.64 ± 0.43	-0.23 ± 0.07	-1.77 ± 0.04	-0.69	-0.01	-0.16	0.71	9.36 ± 1.05
	After	18.95 ± 0.43	-0.24 ± 0.01	-1.92 ± 0.02					9.86 ± 1.03

3.3 Tensile tests

According to the experimental tests, rPC and rPC/ABS specimens (Fig. 7a and b) exhibited brittle failure and similar mechanical behaviors (Fig. 8a). As visible in Table 4, rPC and rPC/ABS have the same elastic modulus values. In addition, tensile strength and elongation at break have comparable values, although rPC/ABS demonstrated a lower variability in results with respect to rPC (Fig. 8b and c). This result may be linked to the use of 30% wt. of rABS pellets for rPC/ABS, increasing the granulometry homogeneity of the feedstock. Hence, specimens with higher 3-D printing quality and better replicability can be printed, reducing the presence of possible interlayer voids where premature failures can occur. The mean stress-strain curves of rPC and rPC/ABS confirm the comparability of the results from the tests, especially in the first part of the curve. In this case, the low percentage of ABS in the blend, hence its rubber component, has a low influence on the decrease in the elastic modulus value [58] with similar decreasing trends of tensile strength and elongation at break [57].

Similar values of tensile strength and elongation at break were obtained for 3-D printed virgin PC/ABS blends on small-format FFF systems were observed in the literature. For example, Zhou et al. obtained tensile strength values of ~40 MPa [64]. Kannan and Ramamoorthy, and Yap et al. reached slightly higher ranges, i.e., ~43 MPa [60] and ~45 MPa [61]. This trend is also confirmed by considering the values of elongation at break, with values ranging from ~1.5% to ~4% [61,62]. Focusing on the elastic modulus, similar values were reached by Yap et al., 2.2 GPa [61], whereas lower values were shown by Kannan and Ramamoorthy and Rivet et al., i.e., ~1.7 GPa [60,62].

In general, the values with the greatest divergence were reached using a different ratio between PC and ABS, decreasing the percentage of PC, or for the high presence of interlayer porosity from the extrusion process. As a result, the over-extrusion trends from the FGF large-scale 3-D printing system may help in decreasing the interlayer porosity, hence improving the adhesion between different layers [88,78]. These values are also comparable with recycled PC/ABS blends processed with conventional manufacturing processes, i.e., injection molding, especially considering elastic modulus values [89,63]. In short,

this comparison confirms the good interlayer adhesion of the samples, hence the viability of using recycled feedstock for new applications relying on manufacturing with FGF systems.

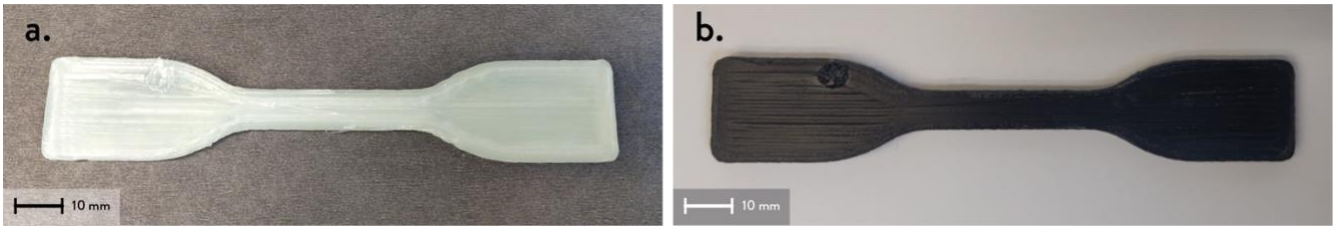


Fig. 7. Tensile specimens of (a) recycled PC and (b) recycled PC/ABS.

Table 4. Main mechanical tensile properties of 3-D printed recycled PC and recycled PC/ABS.

Batch	Elastic Modulus (GPa)	Tensile strength (MPa)	Elongation at Break (%)
Recycled PC	2.1 ± 0.1	47.0 ± 14.6	4.2 ± 1.9
Recycled PC/ABS	2.1 ± 0.1	41.6 ± 6.3	2.8 ± 0.9

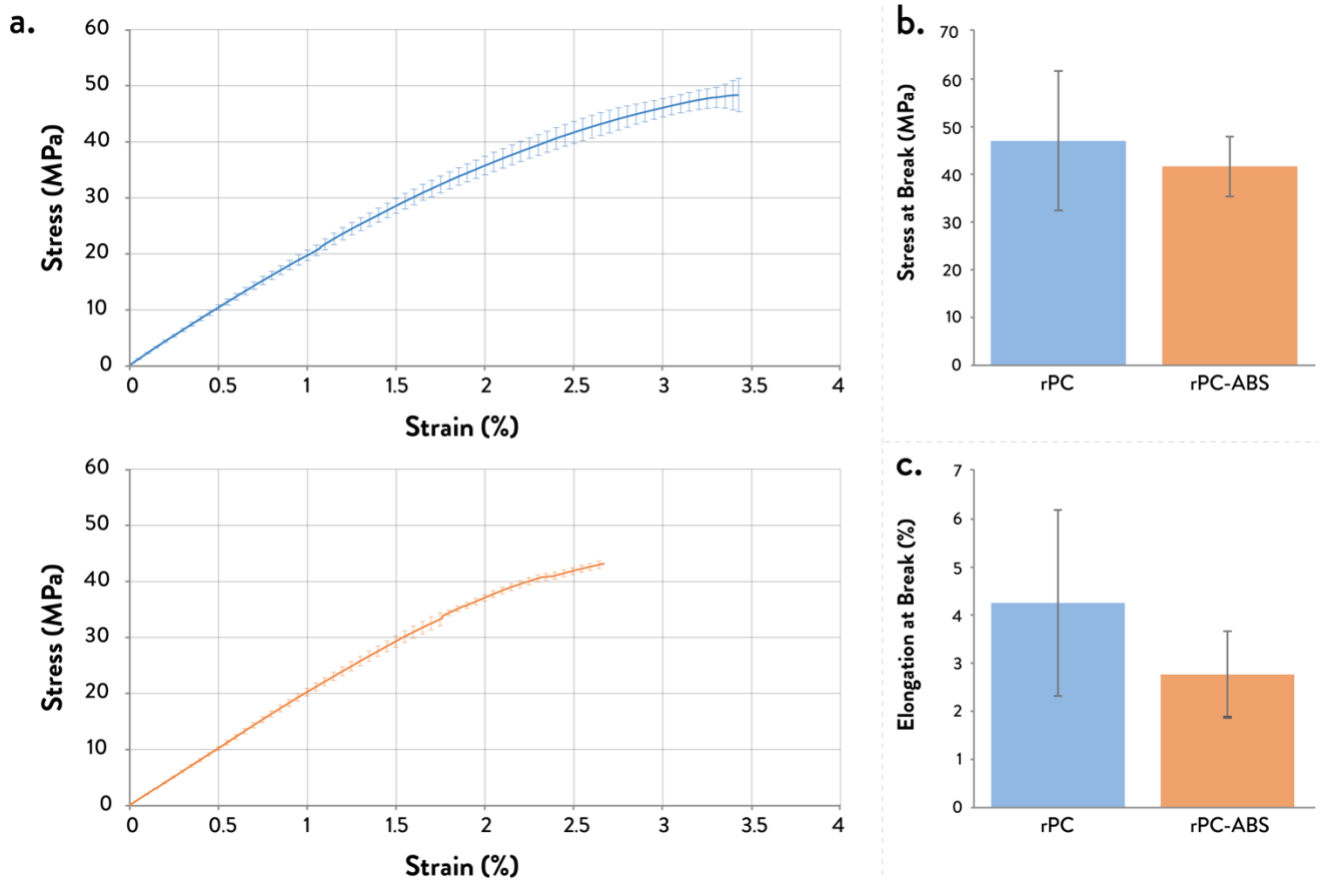


Fig. 8. Tensile tests: (a) mean stress-strain curves of 3-D printed recycled PC (top) and recycled PC/ABS (bottom); (b) stress at break and (c) elongation at break of recycled PC and recycled PC/ABS.

3.4 Impact tests

Samples for impact tests (Fig. 9a and b) confirmed the brittle failure mode of rPC and rPC/ABS formulations. The fracture surfaces showed almost no deformations or irregularities of the samples, maintaining their original rectangular cross-section shape. According to Table 5, Fig 9c and 9d, the impact resistance and impact energy of rPC/ABS remain comparable to rPC. The accuracy of the measurements for rPC/ABS increases, resulting in more reliable values. This fact may be linked to the use of 30% wt. of rABS pellet for the rPC/ABS formulation, decreasing the variability of recycled feedstock in terms of

particle granulometry and shape. Accordingly, a consistent extrusion flow during the 3-D printing process results in fewer interlayer voids in the final part and the concomitant possible defects for cracking and premature failures.

Although Charpy tests may result in higher impact resistance and energy [90], lower values were obtained in previous studies with PC/ABS and FFF or FGF 3-D printing systems. For example, Verma and Banerjee reached ~15 kJ/m^2 of impact energy [91], whereas impact resistance values of ~350 J/m and ~200 J/m were obtained from Kumar et al. and Peng et al. [92,93]. The percentage of ABS can mainly influence these differences in PC/ABS blends characterized in these studies, i.e., 75% wt., and from the different scales of the 3-D printing process, mainly desktop-size small-format apparatuses. As a matter of fact, using FGF large-scale 3-D printers generally means using thicker layer heights and bigger nozzle diameters, causing over-extrusion trends during the process, longer cooling times, and bigger compressive deformations of the layers [88,78]. This fact may help in reducing the dimension and frequency of cross-sectional interlayer voids, hence improving the fusion between layers and, consequently, the mechanical properties of the parts [91,64]. Furthermore, these values are comparable with virgin PC/ABS from injection molding. Chiu et al. reached impact energy values of ~90 kJ/m^2 for virgin PC/ABS [63], whereas Seo et al. obtained an impact resistance of ~150 J/m [57]. Although shape strongly influences the impact behavior of PC/ABS blends with high percentages of PC [57], results confirm a good interlayer adhesion of the samples, showing the potential of recycled feedstock and FGF systems in high-performance fields of application, i.e., technical equipment and sports.

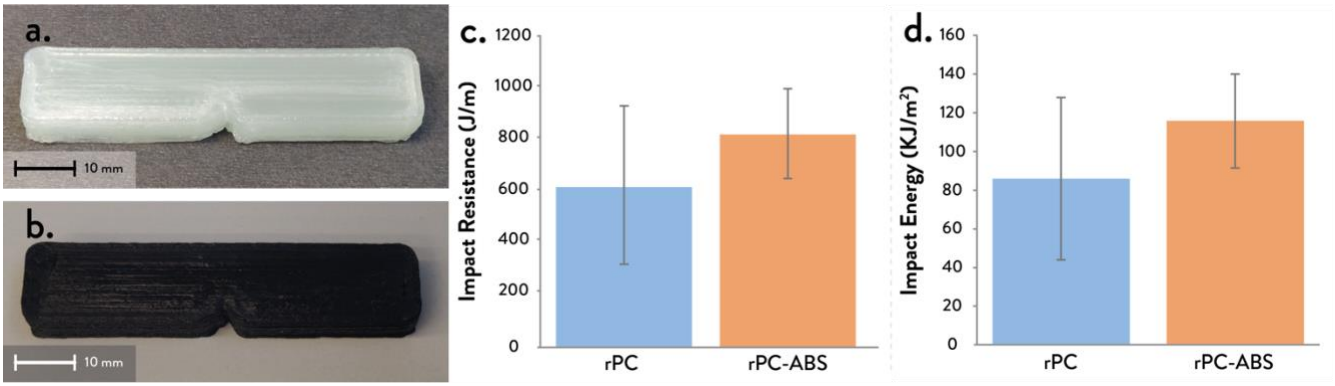


Fig. 9. Impact tests: specimens of (a) recycled PC and (b) recycled PC/ABS; (c) impact resistance and (d) impact energy of recycled PC and recycled PC/ABS.

Table 5. Experimental values from impact tests of 3-D printed recycled PC and recycled PC/ABS.

Material	**Neat Breaking Energy (J)**	**Impact Resistance *IR* (J/m)**	**Impact Energy *IE* (kJ/m^2)**
Recycled PC	9 ± 4.45	609.66 ± 330.04	85.98 ± 42.02
Recycled PC/ABS	11.43 ± 2.15	810.36 ± 165.77	115.78 ± 24.40

3.5 Fracture analysis

Micrograph images on the fracture cross-section surfaces were made to evaluate the 3-D printing quality of the tested materials. In general, 3-D printed paths are less noticeable than those observed in FFF systems [60,61]. In the case of rPC (Fig. 10a), the fracture surface is quite homogeneous without visible voids in the cross-section. The direction of the brittle fracture is visible thanks to the whitened cracks close to the notch, which means on the left side of the cross-section. Moreover, few marks from the 3-D printing process are visible, i.e., division of the extrusion paths. This confirms the good quality of the extrusion flow and its homogeneous consistency, also influenced by the over-extrusion noticed in the comparison tests of Section 3.1. As a result, some over-extrusion may help in improving the interlayer adhesion by reducing the presence of voids typical of the FFF and FGF processes, achieving higher mechanical properties [64].

For the rPC/ABS samples (Fig. 10b), more visible voids can be seen in the cross-section of the samples. The direction of the fracture is not clearly noticeable, and only some cracks can be seen close to the notch. Considering the 3-D printing process, the extrusion path is quite noticeable in some points, i.e., the external perimeter of the sample on the right side, as well as the different layers and the building orientation of the sample. This fact can result from the reduced over-extrusion compared to rPC, although some points show better interlayer adhesion, i.e., the right bottom corner of the cross-section. In short, consistent extrusion lines can be reached with both materials, especially with rPC, and tuning the 3-D printing parameters related to the extrusion flow may help in obtaining parts with higher bonding and fewer voids.

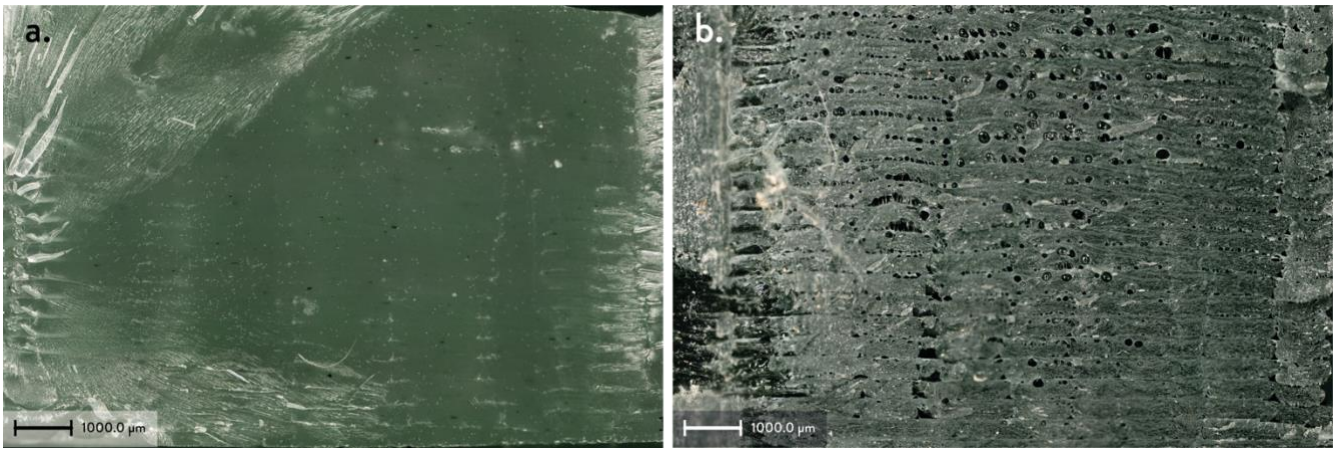


Fig. 10. Microscope micrographs of the fracture cross-sections from the impact test: (a) recycled PC and (b) recycled PC/ABS (100 x magnification).

3.6 Fostering new product applications: case studies

After the previous characterization tests, some plausible application case studies were analyzed to demonstrate the use of rPC and rPC/ABS for real applications, aiming to reduce the gap between experimental studies and exploitation in real contexts. The literature indicates that AM technologies show great potential in fostering sustainable and circular design practices in practical contexts, i.e., in industrial contexts or design projects [94,23]. Furthermore, a growing number of projects and initiatives have been focusing on using recycled materials for new products, demonstrating an interest in exploring new applications and paths toward sustainable models of production and consumption [95,96]. As summarized in Table 6, some real applications were found in the last five years considering recycled PC or recycled ABS and FFF/FGF processes. In most cases, the application field is related to technical equipment [11,55] or furniture elements [97,98], dealing both with small and large-scale 3-D printing systems. No real applications, however, were found with recycled PC/ABS blends, although these materials offer good mechanical and technical properties to be exploited in new products, especially for high-performance applications that require better processability, thermal stability, or good impact resistance [60,99]. Furthermore, medium- and large-scale 3D printing of recycled plastics are mainly used for furniture, furnishing, or decorative applications, scarcely considering other technical fields [94]. Nevertheless, combining high-performance blends from secondary raw materials and large-scale systems can be beneficial to foster new distributed production and consumption models.

Table 6. Resume of the main application-driven works and design projects dealing with FFF or FGF 3-D printing of PC and ABS adapted from [96].

Material	Source	Year	3-D printing system	Application fields and products
rPC/ABS	//	//	//	No applications found
rPC	[55]	2019	Large-format FGF (GigabotX)	Household appliances and tooling - Molds, part replacement, car accessories
	[97]	2018	Large-format FFF	Furniture – indoor cabinet
rABS	[100]	2018	Small-format FFF	Gardening – Piping connectors
	[11]	2018	Small-format FFF	Photography equipment – Camera tripod and hood
	[43]	2019	Large-format FGF	Sports goods – Skateboard, kayak paddle, snowshoes
	[98]	2020	Small-format FFF	Urban furniture – outdoor modules
	[101]	2022	Small-format FFF	Medical technology – Patient sensor

Considering the results achieved in this work, customized protective sports equipment represents a good case study for the exploitation of recycled PC/ABS blends for FGF systems. These two specific applications were defined by choosing one emerging field and one consolidated application for recycled plastics in 3D printing [94] to demonstrate the flexibility of using similar AM blends. Moreover, the products were designed by considering some challenging features for large-scale FGF systems, such as (i) infill, which usually requires travel and can cause extruder leakages due to the difficulties in using retraction with bigger nozzles [102]; (ii) layers with small cross-sections, where shape deformations can occur due to the longer cooling times and bigger compressive deformations between the layers [88,78]; (iii) travel paths, which can lead to extruder leakages [102]; and (iv) complex cross-sectional patterns, which should avoid sharp edges for good accuracy with big nozzles. These features were mainly considered in processing virgin and recycled PC/ABS blends, also according to the printability comparison tests, i.e., interlayer adhesion and detachment of the extrusion paths, curling of corners and small geometrical details on broad cross-sections, and detachment of the part from the building plate. Accordingly, protective shin

guards and customizable furniture elements were selected as a proof-of-concept to be designed and fabricated with rPC/ABS and GigabotX.

The first demo product, the protective shin guard (Fig. 11a), is made of a main external structural part in rPC/ABS combined with a flexible internal shape made of thermoplastic polyurethane (TPU). The two parts can be customized according to the user by modifying their overall dimensions and pattern, i.e., the pattern fill percentage, adapting the technical properties of the product according to the use and the specific sports activity.
Focusing on rPC/ABS, the external structure was designed to be 3-D printed as a flat component to reduce material waste during the fabrication and printing times. The overall geometry was developed by considering the possible extrusion path of the 3-D printing hot end, as visible in the gcode preview of Fig. 11b. After its fabrication, the external structural part can be easily adapted with an accessible heat source, i.e., a hot gun that can be used to seal pores [103] and reshape, to reach the 3-D profile of legs and ankles. The demo shin guard was designed by following these key principles [104, 105]:

- The pattern was created by defining a single curve, representing the path followed by the extrusion hot end, limiting the travel paths to be done, hence retraction-related issues with bigger nozzles.
- A longer path was created to allow each layer to cool down properly before the following layer, especially in case of over-extrusion and/or thicker layer heights.
- Printing a flat customized component helps in reducing the printing times and waste of material, i.e., for support generation, and in improving the adhesion of the whole part by increasing the area touching the build plate.
  The thickness of the pattern is influenced by the nozzle dimension, which was also considered a variable in customizing the shape of the final design.
- The pattern part was also designed to test some common issues in 3-D printing PC/ABS feedstock, mainly linked to warping. The overall shape was designed to check the occurrence of curling corners and warping, as well as the partial or complete detachment of the part from the building plate during the fabrication.

Fig. 11c shows the 3-D printed part, whereas the result after manual thermoforming is visible in Fig. 11d. The overall 3-D printing quality was satisfactory from a functional point of view, although the part required post-processing to remove some exceeding material, i.e., from stringing. However, the part has not detached from the building plate, and curling corners were not detected. This result validates the potential use of rPC/ABS and FGF systems with complex layer patterns for customizable technical applications, such as in the sports goods sector (Fig. 11e).

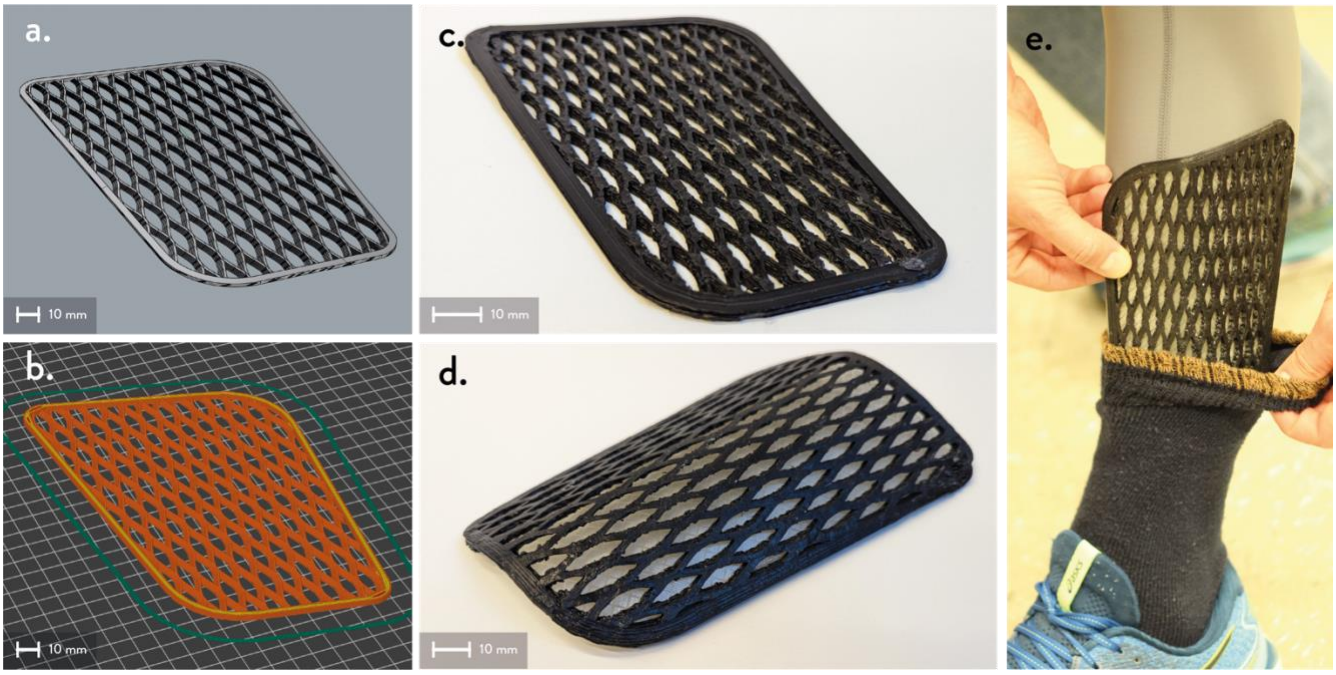


Fig. 11. Application case study from sporting goods sector (protective equipment – shin-guard): (a) 3-D model; (b) gcode preview; (c) 3-D printed part (flat design); (d) 3-D printed part after manual thermoforming; (e) result showing athlete preparing for soccer (football) match.

Further validation comes from the 3-D printing of a second demo product from the furniture sector. The selected 3-D model is a joint for a customizable coat hanger rack and can be used to evaluate the potentials and limitations of GigabotX in terms of achievable features and shapes (Fig. 12a) [54]. To sum up, the part was designed and sliced to assess the feasibility of the following features: (i) incremental overhangs to evaluate eventual limits in shape retention and overhang fidelity; (ii) different cross-sectional shapes and dimensions to test stringing and shape accuracy with short cooling times and retraction; (iii) no support material, reducing material waste and avoiding removal issues; (iv) 10% of gyroid infill to check the extrudate consistency during the 3-D printing, especially after travels; and (v) higher printing speed to test the extrusion path

consistency, shortening the 3-D printing times to fabricate new products in real-world context. This part was also assessed by considering the main issues in 3-D printing PC/ABS, i.e., curling corners, warping, and detachments.
Fig. 12b shows the gcode preview of the part sliced with a printing speed of 40 mm/s, according to the results from the first printing test (Section 3.1). Hence, the part can be obtained in less than 1h 30m, allowing fast replication of small batches of customized joints. The result is visible in Fig. 12c, which validates the use of recycled PC/ABS feedstock to fabricate more complex 3-D shapes, i.e., furniture elements. The main features of the model were successfully achieved, i.e., incremental overhangs, different cross-section shapes, and assembly tolerances. In detail, the part does not present visible voids on the layers nor significant differences in the extrusion flow, confirming the overall quality of the extrusion with a higher 3-D printing speed. The overhangs were successfully obtained without leakages on the external wall. The upper part of the piece reached a satisfactory accuracy despite the small cross-sectional shapes and the use of travels during the 3-D printing process, also in the last layers of the model. The infill was also successfully fabricated, showing no voids on the external walls due to flow inconsistencies after travel leakages. Moreover, no deformation or delamination was detected in the 3-D printed part, as well as curling corners and warping of the bottom layers.
In general, the quality of the 3-D printed parts can be compared to other commercial FGF systems or virgin material feedstock, showing the potential of using recycled materials for new applications in DRAM contexts [55,97,106,107]. Furthermore, the achieved quality can help in facilitating the acceptance of recycled polymers for the end-users, reducing the gap between virgin and recycled feedstock in terms of perceived quality for accessible products [108-110].

The next step in future work can consider the design and production of large-scale components, i.e., large-size furniture elements, or more complex geometries for technical applications, i.e., freeform customized protective helmets, elbow protectors, full-scale 3-D printed furniture, and automotive components [111]. Moreover, similar application fields may use digital imaging technologies to customize the final design, i.e., photogrammetry or x-ray imaging, taking advantage of distributed AM of high-performance recycled feedstock.

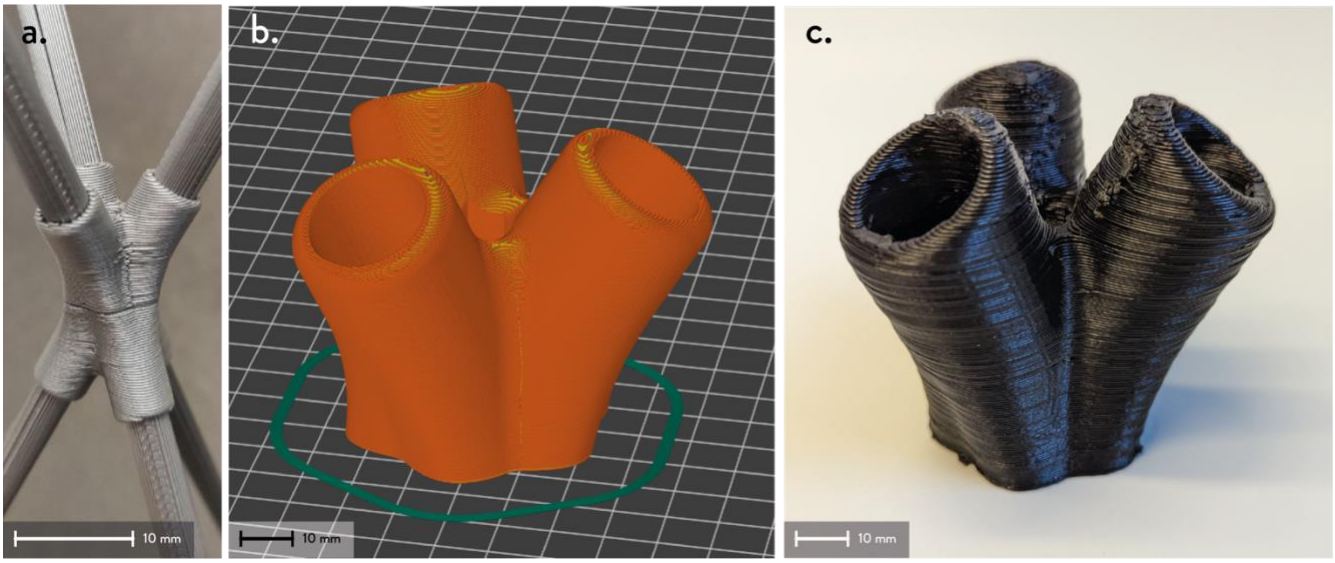


Fig. 12. Application case study from furniture sector (coat hanger joint) [54]: (a) prototype of the assembled product (1:4 scale), (b) gcode preview, and (c) 3-D printed part.

4. Conclusions

This work demonstrated the potential of using recycled PC/ABS feedstock with large-format FGF 3-D printing systems. After modifying a commercial open source FGF 3-D printer, the accuracy and printability of the two feedstocks were assessed with 3-D printed samples. Tensile and impact tests were performed to better understand the mechanical behavior of 3-D printed specimens, as well as to analyze their fracture surfaces. The influence of weather on the samples was assessed through a weather simulation test. The feasibility of using recycled feedstock for real applications was finally evaluated by choosing two application fields and fabricating two different demo products, i.e., customizable protective shin guards and furniture joints.

In general, better printability was achieved with recycled PC/ABS compared to recycled PC, as well as good dimensional accuracy, especially at 30 and 40 mm/s. These results demonstrate the influence of rABS pellets in improving the printability of recycled PC shard-based feedstock. Minimal qualitative differences and discoloration were visible on the samples after accelerated weather exposure of 24h, with results in accordance with the state-of-the-art. These results indicate the materials

may be used for outdoor applications. The results from tensile tests show similar values of elastic modulus, tensile strength, and elongation at break for rPC and rPC/ABS, comparable with previously studied virgin 3-D printed filaments. Similarly, impact energy and resistance values are comparable in the two tested formulations, reaching similar results compared to FFF 3-D printed filaments, as well as virgin materials for injection molding. Although some voids and interlayer gaps are visible from the fracture surfaces, these results confirm the possible use of rPC and rPC/ABS in functional applications. Finally, the two demonstration products in the sporting goods and furniture sectors were successfully fabricated with rPC/ABS, achieving complex patterns and good printing speeds for recycled feedstocks.

In summary, rPC/ABS blends represent a potential high-performance feedstock for DRAM, validating its use in direct FGF 3-D printing systems. In addition, new application fields can be capitalized on in circular economy contexts following DRAM principles, including technical sectors. Further research is needed in future work to assess the mechanical properties of 3-D printed rPC/ABS blends, such as flexural and bending tests, and other characterizations can be performed to deepen the knowledge of these blends, i.e., other percent mixtures and thermal characterization. Future work should also focus on larger and more complex parts to fabricate while also considering a broader range of application fields. Finally, full environmental and economic life cycle analyses should be run on this new recycled blend material applications of DRAM.

Acknowledgements

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