

Sustainable 3D printing using recycled PLA: A comparative study based on the mechanical resistance

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Abstract: Nowadays, sustainability is one of the major objectives for manufacturing. The use of recycled materials can be a suitable strategy when economic, environmental and technical feasibility are verified. 3D printing is still a novel manufacturing process and the research on the topic is still limited, particularly when referring to sustainability issues. This paper attempts to evaluate the suitability of the substitution of virgin PLA by recycled PLA. To do that, it includes an experimental plan divided into three phases including ... to evaluate the technical feasibility based on mechanical resistance. The results showed that recycled PLA may be used due to the, though slightly lower, similar mechanical resistance than that of the virgin material. This reduction is limited to 13 % in the worst configuration (vertical). Besides, it was identified how the infill density and the orientation used for printing played a major role on the mechanical resistance, when others such as the infill pattern, printing speed and layer height had non-significant influence.

Keywords: 3D printing, mechanical resistance, PLA, polymers, recycled materials, sustainability

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26 1.INTRODUCTION

27 Additive manufacturing (also called 3D printing) is becoming a key technology for
28 a cross domains applications. The principle of manufacturing objects layer by layer
29 enable a higher freedoms degree in the product design phase [ref]. The technology
30 brings advantages such as customization of objects of complex geometries with a
31 great deal of detail, combination of different materials, no need for assembly and high
32 utilization rate of raw materials (Columbus, 2017; Jin *et al.* (2017); Xiao *et al.*, 2014).
33 Thus, the technology is receiving great attention by companies and public.

a supprimé: The manufacturing by layer addition, commonly known as 3D printing or additive manufacturing is a revolutionary technology with a wide range of applications

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34 3D printing has developed significantly over time. A great development is
35 expected in sectors such as product consumption, medical products and aerospace
36 components (Peng *et al.*, 2018).The rapid prototyping market reached \$7.3 billion US
37 dollars in 2018 according to Wohlers (Rodriguez-Hernández *et al.*, 2020), which also
38 forecasts the market to reach 23.9 and 35.6 billion dollars by 2022 and 2024,
39 respectively (Forbes, 2019). The industry has grown from \$ 295 million to 5.1 billion
40 over the past 25 years.

41 Nowadays, there is a need to find paths to reduce the ecological impact that
42 manufacturing processes (including 3D printing technology) have is an important topic
43 addressed by a large number of researchers [ref]. Research efforts to identify
44 opportunities of 3D printing on the circular economy paradigm start to be explored
45 [@Despeisse2016]. Plastic is one of the most used materials in the 3D printing
46 industry, and given their non-biodegradable nature, it is one the most abundant type
47 of waste produced and their impact is well document in the different ecosystems [ref].
48 Thus, reducing the consumption of plastics and/or the use of the plastics already
49 presented in the ecosystem is of great importance for the environment. To this

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a déplacé (et inséré) [1]

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57 purpose, distributed recycling via additive manufacturing approach methodology make
58 an emphasis in the technical steps to possible reuse plastic waste through the recycling
59 chains for material extrusion based techniques of 3D printing [CruzSanchez2020,
60 @Little2020]. In the DRAM methodology, consumers have an economic incentive to
61 recycle because they can use their waste as feedstock for a wide range of consumer
62 products that can be produced for a fraction of conventional costs of equivalent
63 products. Moreover, 3D printing is especially suited because it allows producing parts
64 with almost no waste (Sehdev *et al.*, 2017) and could reduce more than 40 % of the
65 waste related to the used material, reusing 95 % of the unused material (Petrovic *et al.*
66 2011). Nevertheless, one of the the systemic problem of plastic waste relies on
67 dependency of the indiscriminate disposal of plastics which carries multiple risks
68 because many plastic products contain additives that modify their physico-mechanical
69 properties difficulting their recycling/reuse [Wagner2020]. The use of biopolymers
70 of natural and renewable origin promising strategies to alleviate these problems (Zhao
71 *et al.*, 2018a) (Bhatia and Ramadurai, 2017) (Liu *et al.*, 2018).

72 However, the number of publications is limited when it comes to sustainable
73 manufacturing using 3D printing (Peng *et al.*, 2018). Although the technology offers
74 high efficiency in the use of material, the great democratization of this technology could
75 cause a feedback impact due to the increasing generation and disposal of huge
76 amounts of waste or polluting emissions to fabricated virgin feedstock.

77 Currently, most of the cost of 3D printing is associated with the cost of the filament
78 [ref]. By recycling raw materials such as polylactic acid (PLA), the emissions of carbon
79 dioxide can be reduced in the transport to landfills or shipping to customers offering
80 environmental benefits [ref]. The technical feasibility for recycling in laboratory

a supprimé: a technology

a supprimé: . According to Petrovic *et al.* (2011), 3D printing

a déplacé (et inséré) [2]

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a déplacé vers le haut [1]: Plastic, which is one of the most used materials by industry, is a non-biodegradable material and among the most abundant type of waste produced. Thus, reducing the consumption of plastics is of great importance for the environment. To this purpose, 3D printing is a technology especially suited because it allows producing parts with almost no waste (Sehdev *et al.*, 2017). According to Petrovic *et al.* (2011), 3D printing could reduce more than 40 % of the waste related to the used material, reusing 95 % of the unused material. However, the number of publications is limited when it

a supprimé: the high level of material consumption generates large amount of pollution, which constitutes a great threat to the environment and health. Minimizing the environmental impacts of manufacturing is an important topic addressed by a large number of researchers. However, these impacts are still difficult to mitigate, measure, and evaluate. Therefore, a suitable strategy is to reduce the use of raw materials, energy, transportation as well as manufacture parts with an optimized and efficient design.¶

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Commenté [FACS2]: Que quieren decir con 'sustainable manufacturing'? En términos de análisis de ciclo de vida? De consumo de energía?

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a supprimé: use causes environmental problems such as

a déplacé vers le haut [2]: being recycling or the use of biopolymers of natural and renewable origin promising strategies to alleviate these problems (Zhao *et al.*, 2018a) (Bhatia and Ramadurai, 2017) (Liu *et al.*, 2018).

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112 conditions been proved for PLA [CruzSanchez2017], ABS [Vidakis2020], PET
 113 [Zander2018], that recycled plastics have a similar performance to their virgin
 114 counterparts and they have even been applied in the manufacture of high value
 115 products in some sectors such as the automobile (Zhao *et al.*, 2018a). However,
 116 studies on the technical acceptability of recycled materials as substitutes for
 117 conventional virgin materials are still limited for particular applications.

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Commenté [FACS3]: Esto es para mi, un research gap que hay hoy en la literatura. Si bien, la parte técnica ya se ha probado en laboratorio, no se sabe aun, si estas partes reciclada, funcionan para una determinada aplicacion

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118 The present study evaluates the mechanical properties of both conventional and
 119 recycled PLA specimens. The objective of the study is the assessment of the suitability
 120 of the recycled PLA as replacement to advance to more sustainable 3D printing
 121 processes. To do that, the study analyzed the influence of several printing parameters
 122 on the mechanical properties of the parts based on a comprehensive experimental
 123 program. Then, a printed parts was designed comparing with a virgin to evaluate the
 124 acceptability in ...

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125 2. LITERATURE REVIEW

126 According to the international ISO standard, there are seven types of additive
 127 manufacturing categories [ref]. From a feedstock material perspective In general,
 128 there are three main categories of materials for 3D printing; based on liquids, solids
 129 and dust. Each of these three categories also has different types of materials
 130 associated, including ceramics, compounds, metals and polymers. The state of the
 131 raw material (e.g., dust, sheet, wire or liquid) must be compatible with the printing
 132 process (Bourell *et al.*, 2017) (Sunpreet *et al.*, 2017) (Chua *et al.*, 2017) (Noorani,
 133 2018). One of the most common processes in 3D printing is the fused deposition
 134 modeling (FDM) process. The process is based on material extrusion, so the material
 135 is heated above the melting point of the material and then deposited onto a platform

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141 (Wolszczak *et al.*, 2018). In FDM, a variety of thermoplastic materials are commonly
142 used, such as acrylonitrile butadiene styrene (ABS), *polyvinylchloride* (PVS),
143 polycarbonate (PC), nylon, polifenilsulfona, *high density polyethylene* (HDPE), *low*
144 *density polyethylene* (LDPE), *polyethylene terephthalate* (PET), *high impact*
145 *polystyrene* (HIPS) and polylactic acid (PLA) (Chua *et al.*, 2017) (Zhao *et al.*, 2018b)
146 (Kumar and Singh, 2018).

147 The development of new materials such as polymers, elastomers and composites
148 in engineering plays a fundamental role in the advance sustainable manufacturing
149 (Ashby and Johnson 2010). The use of biopolymers of natural and renewable origin,
150 replacing synthetic polymers, is the cellulose, hemicellulose, lignin, starch, alginate,
151 chitosan and derivatives derived from them represent the most abundant bio-based
152 and renewable raw materials for different 3D printing technologies. In addition, these
153 biopolymers of natural origin include lignocellulosic materials, seaweed materials and
154 exoskeleton crustacean materials (Liu *et al.*, 2018). Liu *et al.* (2018) reviews a wide
155 range of biobased printing materials with the adequate printing/solidification approach
156 and printed structures and potential applications. Most of them find potential
157 applications in biomedicine such as Cellulose nanofibers (CNF)/alginate and
158 Methylcellulose alginate that have as potential applications cartilage tissue engineering
159 and regenerative therapy, respectively. To print biological matter, new 3D printing
160 methods, such as cryolithography, are appearing. These methods find applications in
161 tissue engineering and food engineering (Zawada *et al.* (2018)).

162 Polylactic acid is a type of natural biopolymer obtained from crops such as starch
163 or sugar cane. It is a biodegradable biopolymer constituted of lactic acid molecules and
164 it is one of the most used materials in 3D printing. In addition, PLA is a sustainable

165 alternative that shows a range of crystallinity and mechanical properties between
166 polystyrene and polyethylene terephthalate (Kumar *et al.*, 2018) (Zhao *et al.*, 2018b).

167 Recycling (mixing of virgin material with used material) of raw material is a
168 method of special interest to contribute to manufacture in a sustainable way (Zhao *et al.*, 2018a). However, it is important to evaluate the properties of the recycled materials
169 before substituting virgin for recycled materials. In this sense, Kumar *et al.* (2018)
170 compared the elongation at break, load at break, flow index, Young's module and
171 breaking stress of recycled ABS, high impact polystyrene (HIPS) and PLA. The PLA
172 showed the highest elongation at break along with the ABS. In addition, the PLA had
173 a higher breaking load and breaking stress, although a smaller Young's modulus. Other
174 authors such as Gu *et al.* (2016) identified the suitability of using recycled
175 polypropylene instead of virgin polypropylene based on mechanical properties.
176 Specifically, they found that the use of fillers (talc and glass fibre) improved the
177 mechanical properties. Babagowda *et al.* (2018) studied the influence of the
178 percentage of recycled PLA used in the filament (i.e., 10 to 50 %) showing that the
179 smaller the percentage the higher the ultimate tensile strength. Pinho *et al.* (2020)
180 obtained higher values of tensile stress for recycled PLA when comparing to the virgin
181 one. Nur-A-Tomal *et al.* (2020) presented a paper to evaluate the reusing of waste
182 children's toys as raw material for 3D printing.
183

184 As Suárez and Domínguez (2020) pointed out, the use of recycled materials is
185 still uncertain because the potential changes in the material properties then recycling.
186 Zhao *et al.* (2018a) studied the cycles of printing that is able to withstand the PLA until
187 it loses much of its properties. Thus, they showed that PLA withstands two printing
188 cycles, since in a third cycle the mechanical properties and viscosity decreased

189 considerably. The increase in crystallinity and melting enthalpy and the decrease of
190 cold crystallization enthalpy are attributed to the 3D printing process, not to the
191 extrusion recycling process. Table 1 shows the comparison of the properties between
192 the virgin material and the same material subjected to another printing cycle, being
193 possible to appreciate how properties such as the tensile strength and modulus are
194 close for the two materials. Similarly, other authors such as Lanzotti *et al.* (2019) have
195 proved how recycling PLA provides comparable mechanical properties as the virgin
196 material only after a second recycling process.

197

Table 1: Comparison of the mechanical properties of PLA virgin and PLA in their second cycle of printing (Based in Zhao *et al.* (2018a)).

	Tensile modulus (MPa)	Tensile strength (MPa)	Elongation at break (%)	Yield strength (MPa)
Virgin PLA	1572.43 ± 27.16	30.21 ± 0.89	2.74 ± 0.53	27.64 ± 0.77
Recycled PLA	1566.54 ± 45.61	29.47 ± 1.21	2.45 ± 0.47	27.65 ± 1.09

The recycling of PLA has certain limitations because of reducing molecular weight with its reuse, resulting in degradation and decrease of mechanical properties. For instance, the addition of polidopamine (PDA) on the surface allows improving these properties. Viscosity is also reduced with each printing cycle but, it could be corrected by adding virgin plastic (Zhao *et al.*, 2018a) (Zhao *et al.*, 2018b). When recycling, there is a decrease in the properties of the material as a result of the presence of carbonyl groups and superficial pitting due to thermomechanical degradation during the process of new melting process occurring during 3D printing (Zhao *et al.*, 2018a).

The mechanical properties of the parts are critical for engineering parts, particularly, for 3D printed parts. Several studies evaluated the mechanical properties of 3D printed parts for various materials; Popescu *et al.* (2018) reviewed some of them in their study. The printing conditions used to manufacture the parts have an important role on the obtained results. So, it is important to identify the most important parameters among all of the available to carry out the process that may affect the response variable and their expected influence based on the scientific literature (Sheoran and Kumar, 2020). For instance, Popescu *et al.* (2018) determined that the

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key parameters that influence the mechanical properties in 3D printing are porosity, layer height, filling density, printing direction and part orientation. In addition, for low values of layer height, as well as the thickness of the deposited or printed filament, the tensile strength of the material is improved. In addition, by orienting the printing direction towards the direction in which the tensile load will be applied during tensile strength, the property can be also maximized. The importance of the printing orientation was also identified by Yao *et al.* (2019). According to Alafaghani and Qattawi (2018), a higher extrusion temperature and an optimized layer thickness, a triangular filling pattern and a higher filling level maximize the strength of the parts. Regarding the printing speed, it is identified that higher printing speed with higher layer thickness result in lower part strength. Among others, Altan *et al.* (2018) also identified the influence of the layer height on the mechanical resistance.

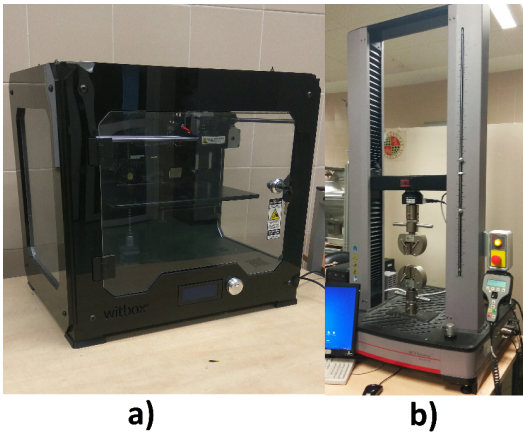
3. EXPERIMENTAL PROCEDURE

3.1 Materials and equipment

The printing materials used in the study were virgin and recycled PLA characterized by data listed in Table S1. Both materials were commercial ones, so the recycling was not done during the research. The recycled PLA contained 10 % of virgin PLA in the blend.

The 3D printer used to manufacture the specimens was a BQ's Witbox, shown in Fig1a. The software used to generate the printing code was the Ultimaker Cura 3.2.1. To perform the destructive test, the machine used was the MTS Criterion 43 universal testing machine (MTS, 2020) (Fig 1b) with a maximum load of 50 kN, being the maximum load supported by the LPS 104 cell of 10 kN. The clamping system was the

239 Instron 2716-015 system with a maximum supported load of 30 kN. The selected strain
240 rate was 0.5 mm/min.



241 **a)**
242 **b)**
242 **Figure 1: Equipment used in the study: a) 3D printer, b) Universal testing machine.**

243 In order to save material and time to carry out the experimental plan, the
244 geometry of the specimen was reduced in size from the one recommended by the UNE
245 116005:2012 standard (UNE, 2012), which define the length of the specimen at no
246 less than 120 and 150 mm depending on the orientation. However, in this study the
247 specimens are manufactured according to the dimensions proposed by Lin *et al.*
248 (2018) in which the length of the specimen was 75 mm. The dimensions of the
249 specimen are the ones depicted in Fig. 2.

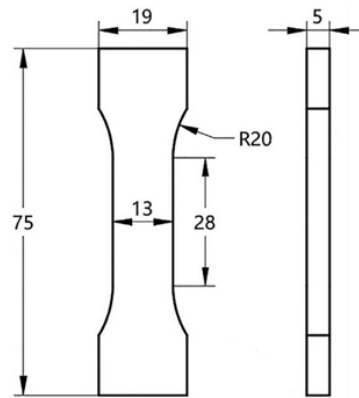


Figure 2: Geometry of the specimen to be 3D printed (Based in Lin *et al.* (2018)).

3.2 Methodology

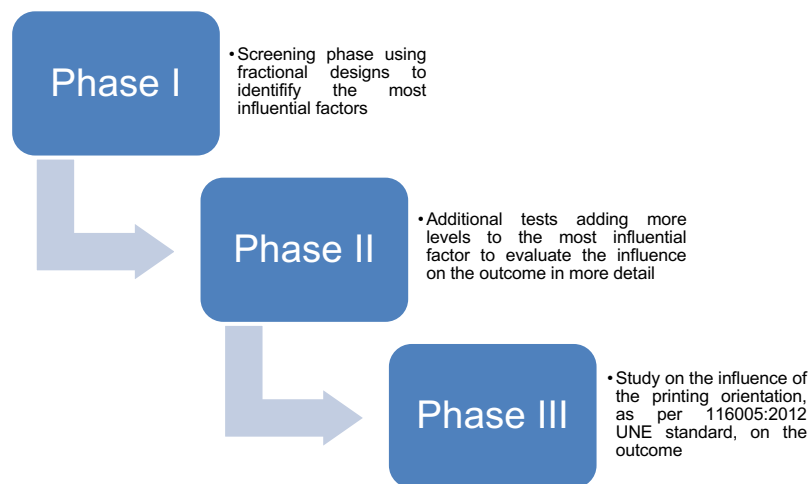
The aim of the study is identifying the most critical factors affecting the mechanical properties in 3D printing, evaluating their influence on the mechanical properties in 3D printing, particularly for both virgin and recycled PLA. Thus, based on the results, it is expected to gain a better understanding on the suitability of using recycled materials in 3D printing and how to properly select the printing conditions to guarantee sufficient mechanical resistance. In order to do that, an experimental plan comprising three phases will be developed. Based on the literature research, the critical parameters for the study are the layer height and infill pattern. In addition, taking into account the goal of sustainable manufacturing (i.e., trying to optimize the consumption of material), but also productivity (i.e., trying to minimize printing times), infill density and printing speed were considered (Singh *et al.*, 2018; Tanveer *et al.*, 2019). The response variable chosen was the maximum load attained during the testing of the specimen ((Kumar *et al.*, 2018), (Chacon *et al.*, 2017); (Letcher *et al.*, 2015)).

Commenté [FACS5]: Methodology, I think needs to be presented as section 3.1

Commenté [FACS6]: Literature on this:
DOI 10.1016/j.procir.2019.02.030
DOI 10.1016/j.addma.2017.05.013
DOI 10.1089/3dp.2016.0054
DOI 10.1115/IMECE2014-39379
DOI 10.1089/3dp.2019.0195

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267 The experimental plan included three different phases (Figure 2) to carry out a
268 comprehensive study with a limited number of tests not compromising the reliability of
269 the results. In Phase I, the objective was to identify which were the most important
270 factors that have influence on the response variable. The Phase II allowed studying in
271 more detail the influence of the most influential factors according to phase I. Finally,
272 the Phase III aimed at evaluating the influence of the anisotropy of the specimens
273 depending on the printing orientation, which may notably affect the mechanical
274 resistance of the specimen. Table S2 summarizes the tests of three phases.



275

276 **Figure 3: Summary of the three phases of the experimental plan.**

277 In Phase I, the factors analyzed were those identified in previous sections as
278 critical. Thus, layer height, infill pattern, infill density and printing speed were selected
279 using two levels for each of them, with large ranges, especially for the infill density.
280 Thus, the factors and their levels used were: layer height (0.15 and 0.3 mm), infill
281 pattern (tri-hexagonal and grid) , infill density (60 and 100 %) and printing speed (40

282 and 80 mm/s). The printing temperature chosen was 210 °C, which was the
283 recommended one for these materials (filament2print.com/es/).

284 Fractional designs are useful for reducing the number of tests reducing time and
285 money (Montgomery, 2001), being use as screening designs. So, in order to identify
286 and discard factors depending on their influence with a limited number of tests, phase
287 I recurred to fractional designs. The design included only specimen printed in the
288 horizontal orientation for each of the combinations, not evaluating the influence of the
289 orientation, in order to save time and material. The use of random order allowed
290 guaranteeing that the hypothesis that the errors are independently distributed random
291 variables was fulfilled (Montgomery, 2001).

292 Phase II depended on the results of the previous phase. In this phase, the most
293 influential parameter of the ones studied in Phase I (note: the factor resulted to be the
294 infill density) was evaluated in more detail. The selection of levels for the other three
295 factors aimed at minimizing the printing time. Thus, the selection of the factors was
296 layer height of 0.3 mm, infill pattern tri-hexagonal and printing speed of 80 mm/s with
297 an estimated printing time of 20 min. Regarding the infill density, five levels were
298 chosen ranging from 40 to 100 % to evaluate the evolution of the maximum load versus
299 the infill density for both virgin and recycled PLA. The specific levels selected were 40,
300 55, 70, 85 and 100 %.

301 Phase III aimed at analyzing the influence of the orientation. Because of the
302 anisotropy, the UNE 116005:2012 (UNE, 2012) standard requires printing specimens
303 in three different orientations: edgewise (E), horizontal (H) and vertical (V), testing five
304 samples in each orientation. The selected printing conditions were infill density of 50
305 %, printing speed of 80 mm/s, tri-hexagonal infill pattern and layer height of 0.3 mm,

306 with the objective of limiting the use of material and the time required for printing. In
307 total, the phase included the printing of 15 specimens of each virgin and recycled PLA.

308 4.RESULTS AND DISCUSIONS

309 Table S3 lists all the maximum load results attained during testing.

310 4.1. Phase I: Screening phase

311 In general, shortly after attaining the maximum load, the fracture of the specimen
312 occurred. However, the nature of the fracture was not homogenous as shown in Fig 4.
313 Thus, in most cases, the specimens showed a fragile behavior and the fracture, either
314 horizontally or with a lower inclination angle, is clean. However, for the recycled
315 material, the specimens showed a ductile behavior and, properly, the fracture did not
316 occur after the maximum load was attained, canceling the tests minutes after the
317 maximum load was attained. The breakage in these cases occurred at a 45° angle
318 and, in the cases of the RE-2 specimen, two parallel fracture lines can be clearly seen.
319 The printing conditions did not allow seen a clear relation to the fracture of the
320 specimens. This behavior may relate to that explained by Yao *et al.* (2020). The
321 authors identified two different types of fracture: in-layer and interlayer. In general, the
322 fracture occurs at the interface of two layers when printing in vertical position, even
323 when varying the printing orientation up to 45° from the vertical position. In-layer
324 fracture is more likely when the specimen is printed using an edgewise position (or,
325 inclined up to 45° from that position). In this second case, the printing direction is the
326 same as the tensile stress direction, which also happens when the horizontal
327 orientation is used. In these cases, the material layer is not intact after the fracture.

328 Thus, it is likely that both modes coexist in this study, which may explain the
329 heterogeneity of the different fractures.



Figure 4: Specimens tested to traction (phase I).

332 The analysis of variance (ANOVA) performed using R software allowed
333 identifying the influential factors on the response variables. As criterion, critical factors
334 for the response variable were those with p-values lower than 0.05 (Pérez *et al.*, 2018).
335 Shapiro-Wilk normality tests allowed verifying the normality of the residuals. Table 2
336 lists the results of the ANOVAs carried out for both virgin and recycled PLA.

Table 2: Results of ANOVA in phase I.

Virgin PLA					
Factor	Degrees of freedom	Sum of squares	Mean squares	F-Valor	Pr(>F)
Layer height	1	32026	32026	9.87540	0.05156
Infill pattern	1	5	5	0.00160	0.97059
Infill density	1	4575597	4575597	1410.90320	4.151e-05
Printing speed	1	102	102	0.03160	0.87030
Residuals	3	9729	3243	-	-
Total	7	4617459			
Recycled PLA					
Factor	Degrees of freedom	Sum of squares	Mean squares	F-Valor	Pr(>F)
Layer height	1	49	49	0.0058	0.943986
Infill pattern	1	1349	1349	0.1598	0.716091
Infill density	1	3378909	3378909	400.1721	0.000273
Printing speed	1	3388	3388	0.4012	0.571455
Residuals	3	25331	8444	-	-
Total	7	3409026			

339 From Table 2, it can be clearly identified how only the infill density was a
340 statistically significant factor for the maximum load for both materials (p-value lower
341 than 0.001). When evaluating the contribution of each of the factors to the variability

explained by the model, there were calculated values of 99.30 % and 99.85 % for virgin and recycled PLA, respectively. Thus, when manufacturing new parts or specimens, infill density is a key factor for guarantying adequate mechanical properties of the specimens. A new ANOVA allowed evaluating the influence of the material on the maximum load, maintaining the sources of variation previously analyzed. Table S4 shows the obtained results. When including the material in the ANOVA, infill density is still the most influential factor. However, in this case, the type of material is also a significant factor for the response variable, being non-significant the rest of the factors. Though significant, when assessing the contribution of the material to the variability of the model, it only accounted for 1.25% of this variability, as shown in Figure S1.

4.2. Phase II: Evaluation of the infill density influence

Figure S2 shows the fracture of the specimens tested in phase II. Regarding the fracture, the results were similar as those of the phase I (i.e., more ductile behavior for the recycled PLA specimens). Figure 6 displays the maximum load results for both virgin and recycled PLA.

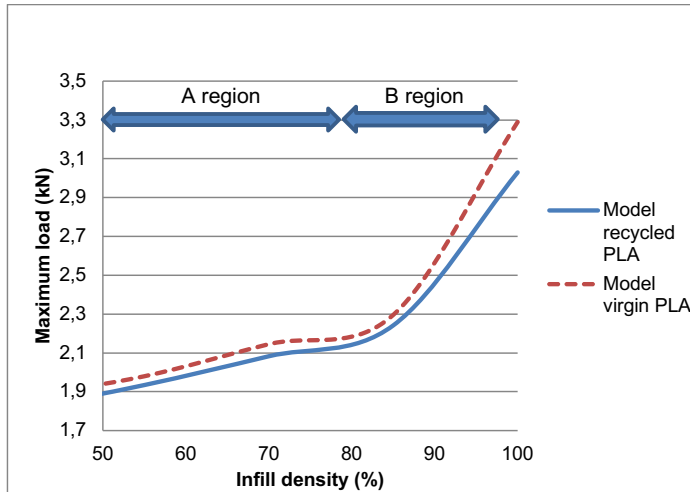


Figure 6: Maximum load versus infill density for both virgin and recycled PLA.

From the analysis of the Figure 6, it is possible to appreciate that there are two clearly different regions. Therefore, in the A region, comprised between infill densities from 40 to 85 %, the slope of the curve grows slowly with a lineal trend. From 70 to 80 % the maximum load remained almost constant. Thus, increasing the infill density did not provide an increase in the mechanical properties. In B region, the slope of the curve grows largely. Thus, with a small increase of infill density, the maximum load notably grows. Regarding the type of material, it is clear that virgin PLA outperforms recycled PLA, but a reduced difference between them. However, the difference notably increased as the infill density approached 100 %. The obtained results agree well with those presented by Wang *et al.* (2020). In their study, the authors studied infill density of 20, 40, 60, 80 and 100% and the evolution of the tensile strength is similar to the one shown in Figure 6.

Although the number of measured points is reduced, it is possible to model the relation between the maximum load (y) versus the infill density (x) for the two regions

374 and tested materials by means of linear regression. Thus, for the virgin material the
375 models are: $y = 0.009577243x + 1.474545487$ (A region) and $y = 0.066247363x -$
376 3.335335637 (B region). In the case of the recycled material, the models are: $y =$
377 $0.009666116x + 1.412545407$ (A region) and $y = 0.052543783x - 2.224621187$ (B
378 region). The models may help to anticipate the mechanical resistance of a part based
379 on the selection of the infill density.

380 Based on the developed models, it is possible to highlight that recycled PLA is a
381 suitable substitute for virgin PLA guarantying similar mechanical resistance. Moreover,
382 by developing models for the mechanical properties, it is possible to minimize the
383 material consumption for both virgin and recycled materials satisfying the mechanical
384 resistance requirements. Thus, by accurately knowing the influence of the printing
385 conditions on the mechanical resistance, it is possible to advance towards sustainable
386 manufacturing.

387 4.3. Phase III: Study on the printing orientation

388 In this experimental phase, testing included the three different orientations
389 established by the UNE 116005:2012 (UNE, 2012) standard (i.e., five specimens for
390 each of the orientations for both virgin and recycled PLA). Fig S3 and S4 show the
391 images of the tested specimens observing the same type of fracture as in the first two
392 phases.

393 It is interesting to evaluate the reduction in the maximum load depending on the
394 type of material and orientation in which the specimens were printed. Thus, in Table
395 S5, the mean values for the five specimens at each orientation are shown and the
396 maximum load reduction between the two materials is calculated. Based on Table S5,

397 it is clear that the horizontal orientation is the one that provided the higher mechanical
398 resistance, followed by the edgewise orientation. The vertical orientation provided the
399 worse results due to the deposition of the layers was perpendicular to the tensile
400 direction. These results are in good agreement with those by Corapi *et al.* (2019) and
401 Wang *et al.* (2020). For the recycled material, there is a slight decrease in the maximum
402 load attained from 3 to 13 % depending on the orientation. Particularly, the biggest
403 reduction of the load happens in the vertical orientation with a 12.97 %. However, the
404 other two orientations are more adequate for substituting the virgin material for the
405 recycled material with a limited reduction in mechanical resistance (3 to 8 %).

406 5.CONCLUSIONS

407 The present study includes a comprehensive experimental program to analyze
408 the FDM process, based on mechanical resistance, by using virgin PLA and recycled
409 PLA. The paper aims at improving the sustainability of 3D printing process, assessing
410 the technical feasibility of the substitution. The main conclusions of the study are the
411 following:

412 The printing conditions determined in a great manner the mechanical resistance
413 of the specimens. Specifically, the most influential factor on the maximum load for both
414 virgin and recycled PLA was the infill density. In addition, non-statistically significant
415 factors were the layer height, infill pattern and printing speed.

416 The fracture for the virgin material corresponded to that of a fragile material, while
417 the fracture of the recycled material showed a more ductile behavior.

418 The influence of the infill density on the maximum load allowed identifying two
419 different regions: A, from 40 to 85 %, linear behavior with a slight slope and, B, from
420 85 to 100 %, the maximum load increases notably with a much higher slope.

421 The selected orientation for printing the specimens is of great importance for the
422 maximum load because of the anisotropy. In this sense, the horizontal orientation
423 allowed attaining a higher maximum load, while the vertical orientation provided the
424 lower value due to the fact that no layers were deposited in the tensile direction.

425 The substitution of virgin PLA for recycled PLA is possible based on the
426 mechanical resistance advancing towards sustainable manufacturing. Despite
427 recycled PLA offers a slightly lower mechanical resistance, when possible, by properly
428 selecting the printing conditions (mainly, by the infill density and orientation) it could be
429 approximate to that of the virgin PLA. Particularly, when using edgewise and horizontal
430 orientations it is possible to obtain maximum loads close to that of the virgin material
431 (from 3 to 8 % lower).

432 6.ACKNOWLEDGMENTS

433 The authors would like to thank the "Mechanical and Energy Engineering" TEP 250
434 research group.

435 7.REFERENCES

436 Alafaghani A., Qattawi A. (2018). Investigating the effect of fused deposition modeling
437 processing parameters using Taguchi design of experiment method. Journal of Manufacturing
438 Processes 36, 165-174.

439 Altan M., Eryildiz M., Gumus B., Kahraman Y. (2018). Effects of process parameters on the
440 quality of PLA products fabricated by fused deposition modeling (FDM): surface roughness
441 and tensile strength. *Materials Testing* 60, 471-477.

442 Ashby M., Johnson K. (2010). Material profiles. En *Materials and design* Oxford: Butterworth-
443 Heinemann. Second ed., págs. 194-249.

444 Babagowda, R.S. Kadadevara Math, Goutham R., Srinivas Prasad K.R. (2018) Study of effects
445 on mechanical properties of PLA filament which is blended with recycled PLA materials, in *IOP*
446 *Conference Series: Materials Science and Engineering* 310:1

447 Bhatia S., Ramadurai K. (2017). *3D Printing and Bio-Based Materials in Global Health* Springer
448 International Publishing AG, Switzerland.

449 Bourell D., Kruth J. P., Leu M., Levy G., Rosen D., Beese A. M., Clare A. (2017). Materials for
450 3D printing. *CIRP Annals – Manufacturing Technology* 66, 659–681.

451 Chacon J., Caminero M., Garcia-Plaza E., Nunez P. (2017). 3D printing of PLA structures
452 using fused deposition modelling: effect of process parameters on mechanical properties and
453 their optimal selection. *Materials & Design* 124, 143–157.

454 Chua C., Wong C., Yeong W. (2017). Standards, Quality Control, and Measurement Sciences
455 in 3D Printing and 3D printing. Chapter One-Introduction to 3D Printing or 3D printing. Pages
456 1-29.

457 Columbus L. (2017). The-state-of-3d-printing. *Forbes.com*, website, last visit 18/06/2019.

458 Corapi, D., Morettini, G., Pascoletti, G., Zitelli, C. Characterization of a Polylactic Acid (PLA)
459 produced by fused deposition modeling (FDM) technology. *Procedia Structural Integrity* 24
460 (2019) 289–295

461 Filament2print (2020). Webpage: <https://filament2print.com/> Last time accessed 18/03/2020.

462 Forbes (2019) Significant 3D Printing Forecast Surges To \$35.6 Billion.
 463 [https://www.forbes.com/sites/timccue/2019/03/27/wohlers-report-2019-forecasts-35-6-billion-](https://www.forbes.com/sites/timccue/2019/03/27/wohlers-report-2019-forecasts-35-6-billion-in-3d-printing-industry-growth-by-2024/)
 464 [in-3d-printing-industry-growth-by-2024/](https://www.forbes.com/sites/timccue/2019/03/27/wohlers-report-2019-forecasts-35-6-billion-in-3d-printing-industry-growth-by-2024/) Accessed 17 October 2020

465 Gu, F., Hall, P., Miles, M.J. Performance evaluation for composites based on recycled
 466 polypropylene using principal component analysis and cluster analysis. J. Clean. Prod., 115
 467 (2016), pp. 343-353

468 Jin, Y, Wan, Y., Zhang, B., Liu, Z. Modeling of the chemical finishing process for polylactic acid
 469 parts infused deposition modeling and investigation of its tensile properties. Journal of
 470 Materials Processing Technology 240 (2017) 233–239

471 Kumar R., Singh R. (2018). Prospect of Recycling of Plastic Product to Minimize Environmental
 472 Pollution. DOI: 10.1016/B978-0-12-803581-8.11302-5.

473 Kumar R., Singh R., Farina I. (2018). On the 3Dprinting of recycled ABS, PLA and HIPS
 474 thermoplastics for structural applications. PSU Research Review Vol. 2, 115- 137.

475 Lanzotti, A., Martorelli, M., Maietta, S., Gerbino, S., Penta, F., Gloria, A. A comparison between
 476 mechanical properties of specimens 3D printed with virgin and recycled PLA. Procedia 79
 477 (2019) 143-146.

478 Lechter T., Rankouhi B., Javadpour S. (2015). Experimental study of mechanical properties of
 479 additively Manufactured ABS plastic as a function of layer parameters.

480 Lin W., Shen H., Xu G., Zhang L., Fu J., Deng X. (2018). Single-layers temperature- adjusting
 481 transition method to improve the bond strength of 3D-printed PLC/PLA parts. Composites Part
 482 A, 115, 22-30.

483 Liu J., Sun L., Xu W., Wang., Yu S., Sun J. (2018). Current advances and future perspectives
 484 of 3D printing natural-derived biopolymers. Carbohydrate Polymers 207, 297-316.

a mis en forme : Français

485 Montgomery D. C. (2001). Design and analysis of experiments. Editorial John Wiley&Sons,
486 Inc.

487 MTS (2020). Webpage: [Mts.com/cs/groups/public/documents/library/mts_006225.pdf](https://mts.com/cs/groups/public/documents/library/mts_006225.pdf) Last
488 time accessed 18/3/2020.

489 Noorani (2018). 3D Printing. Editorial CRC Press.

490 Nur-A-Tomal M.D., Pahlevani F., Sahajwalla V. (2020) Direct transformation of waste
491 children's toys to high quality products using 3D printing: A waste-to-wealth and sustainable
492 approach. Journal of Cleaner Production 267, 122188

493 Peng T., Kellens K., Tang R., Chen C., Chen G. (2018). Sustainability of 3D printing: An
494 overview on its energy demand and environmental impact. 3D printing 21, 694–704.

495 Pérez, M., Medina-Sánchez, G., García-Collado, A. Gupta, M. Carou, D. Surface Quality
496 Enhancement of Fused Deposition Modeling (FDM) Printed Samples Based on the Selection
497 of Critical Printing Parameters. Materials 2018, 11, 1382

498 Petrovic, V., Gonzales, J.V.H., Ferrado, O.J., Gordillo, J.D., Puchades, J.R.B., Ginan, L.P.
499 Additive layered manufacturing: sectors of industrial application shown through case studies
500 Int. J. Prod. Res., 49 (4) (2011), pp. 1071-1079

501 Ana C. Pinho, Ana M. Amaro, Ana P. Piedade (2020) 3D printing goes greener: Study of the
502 properties of post-consumer recycled polymers for the manufacturing of engineering
503 components. Waste Management 118, 426-434

504 Popescu D., Zapciu A., Amza C., Baci F., Marinescu R. (2018). FDM process parameters
505 influence over the mechanical properties of polymer specimens: A review. Polymer Testing 69,
506 157-166.

507 A. G. Rodríguez-Hernández, Angelica Chiodoni, Sergio Bocchini, Rafael Vazquez-Duhalt
508 (2020) 3D printer waste, a new source of nanoplastic pollutants. *Environmental Pollution* 267,
509 115609

510 Sehdev M., Thapliyal H., Sharma A. (2017). 3D Printing for manufacturing of biodegradable
511 products. Department of Mechanical Engineering.

512 Sheoran, A.J., Kumar, H. Fused Deposition modeling process parameters optimization and
513 effect on mechanical properties and part quality: Review and reflection on present research.
514 *Materials Today: Proceedings* 21 (2020) 1659–1672

515 Singh R., Singh H., Farina I., Colangelo F., Fraternali F. (2019). On the 3D printing of an energy
516 storage device from recycled material. *Composites Part B* 156, 259–265.

517 Suárez L, Domínguez M (2020) Sustainability and environmental impact of fused deposition
518 modelling (FDM) technologies. *The International Journal of Advanced Manufacturing*
519 *Technology* (2020) 106:1267–1279

520 Sunpreet S., Seeram R., Rupinder S. (2017). Material issues in 3D printing: A review. *Journal*
521 *of Manufacturing Processes* 25, 185-200.

522 Md. Qamar Tanveer, Abid Haleem, Mohd Suhaib (2019) Effect of variable infill density
523 on mechanical behaviour of 3-D printed PLA specimen: an experimental investigation. *SN*
524 *Applied Sciences* 1:1701

525 UNE, 2012. UNE 116005 (UNE 116005:2012): Fabricación por adición de capas en materiales
526 plásticos. Fabricación aditiva. Preparación de probetas.

527 Wang, S., Ma, Y., Deng, Z., Zhang, S., Cai, J. Effects of fused deposition modeling process
528 parameters on tensile, dynamic mechanical properties of 3D printed polylactic acid materials.
529 *Polymer Testing* 86 (2020) 106483

530 Wolszczak, P., Lygas, K., Paszko, M. and Wach, R.A. (2018). Heat distribution in material
531 during fused deposition modelling. *Rapid Prototyping Journal*, 24(3), 615-622

532 Xiao, K., Zardawi, F., van Noort, R. Yates, J.M. Developing a 3D colour image reproduction
533 system for 3D printing of facial prostheses. *Int J Adv Manuf Technol* (2014) 70:2043–2049

534 Yao, T., Deng, Z., Zhang, K., Li, S. A method to predict the ultimate tensile strength of 3D
535 printing polylactic acid (PLA) materials with different printing orientations. *Composites part B*
536 163 (2019) 393-402.

537 Zawada, B., Ukpai, G., Powell-Palm, M.J., Rubinsky, B. Multi-layer cryolithography for 3D
538 printing. *Progress in 3D printing* (2018) 3:245–255

539 Zhao P., Rao C., Gu F., Sharmin N., Fu J. (2018a). Close-looped recycling of polylactic acid
540 used in 3D printing: An experimental investigation and life cycle assessment. *Journal of*
541 *Cleaner Production* 197, 1046-1055.

542 Zhao X., Hwang K., Lee D., Kim T., Kim N. (2018b). Enhanced mechanical properties of self-
543 polymerized polydopamine-coated recycled PLA filament used in 3D printing. *Applied Surface*
544 *Science* 441, 381-387.