Mechanical evaluation of recycled PETG filament for 3D printing

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Visual Abstract

Mechanical Evaluation of Recycled PETG Filament for 3D Printing

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INTRODUCTION

Plastic recycling has emerged as a critical strategy in addressing the environmental challenges posed by plastic waste worldwide. With plastics pervading nearly every aspect of modern life, from packaging to construction materials, the urgency for sustainable disposal and reuse solutions has never been more apparent. Despite ongoing efforts and growing awareness, achieving a circular economy for plastics remains a complex and multifaceted challenge. [1] The endeavours of recycling and data analysis in Europe, for example the E³UDRES² Alliance, focus on leveraging interdisciplinary research and collaboration to enhance waste management practices and promote sustainable regional development. [2]

Amidst this global backdrop, 3D printing has gained significant attention for its potential to transform manufacturing processes and product design. Offering unparalleled flexibility and customization capabilities, 3D printing has swiftly gained traction across industries ranging from aerospace to healthcare [2]. Various techniques have emerged in Additive Manufacturing, each shaped by distinct manufacturing processes. Fused Deposition Modelling (FDM) stands out among these methods. FDM entails the creation of a three-dimensional model through the extrusion of molten material, layer by layer, forming small beads that solidify to shape the model. Originating in the late 1980s, FDM was pioneered by Mr. S. Scott Crump and subsequently refined by Stratasys in 1990 [4]. The authors provided a comprehensive overview of the materials available for crafting 3D designs, discussing their strengths and weaknesses in practical terms. Gunaydin et al. [5]

also shared practical tips for improving print quality. They suggested simple precautions, like ensuring a draft-free environment and placing the printer on a stable surface. Additionally, they emphasized the importance of using high-quality filament material for better results. However, the successful implementation of 3D printing hinges on the availability of suitable materials with desired mechanical properties and performance characteristics. Polyethylene terephthalate glycol (PETG) has emerged as a leading contender among 3D printable materials due to its favourable balance of strength, flexibility, and printability. Derived from the same polymer family as PET (polyethylene terephthalate), PETG offers enhanced durability and impact strength, rendering it suitable for a wide array of applications [6]. Despite its favourable properties, PETG's sustainability is still a subject of scrutiny, primarily revolving around concerns regarding its end-of-life disposal and the efficacy of recycling processes. As the world grapples with the environmental consequences of plastic waste, researchers have intensified their efforts to address these challenges by shifting their focus towards the recycling of PETG and other materials used in 3D printing. This growing interest in recycling initiatives aims to not only mitigate the environmental impact of plastic waste but also to explore innovative solutions for the circular economy, where materials are reused, repurposed, and recycled to minimize resource consumption and waste generation [7].

Plastic materials offer various recycling options, and the ease of recycling varies depending on the type of polymer, package design, and product complexity. For example, rigid containers made of a single polymer are simpler and more cost-effective to recycle compared to multi-layer or multi-component packages. Thermoplastics such as PET, PE, and PP promise mechanical recycling, while thermosetting polymers like unsaturated polyester or epoxy resin cannot be mechanically recycled. However, they can potentially be repurposed as filler materials after size reduction. A significant challenge in producing recycled resins from plastic waste arises from the inherent immiscibility of different types of plastic at the molecular level and differences in processing requirements on a macro scale. For example, even a small amount of PVC contaminant in a PET recycling stream can degrade the recycled PET resin due to the evolution of hydrochloric acid gas from the PVC at the higher temperature required to melt and reprocess PET. Conversely, PET in a PVC recycling stream may form solid lumps of undispersed crystalline PET, significantly reducing the value of the recycled material. Therefore, it is often technically infeasible to incorporate recovered plastic into virgin polymer without compromising some quality attributes, such as colour, clarity, or mechanical properties like impact strength. As a result, most uses of recycled resin involve blending it with virgin resin, often in non-critical applications such as garbage bags or irrigation pipes. The ability to substitute recycled plastic for virgin polymer depends on the purity of the recovered plastic feed and the property requirements of the plastic product. Current recycling schemes for post-consumer waste generally focus on easily separable packages, such as PET bottles and HDPE milk bottles, which can be positively identified and sorted from waste streams. Conversely, there is limited recycling of multi-layer or multi-component articles due to contamination concerns between polymer types [8]. The recycling of PLA and PETG for additive manufacturing has been explored in several studies, yielding varied results. Zhao et al. [9] observed that PLA exhibited a significant decline in viscosity after just two recycling cycles, rendering it unprocessable. To mitigate this, recycled PLA was mixed with fresh granules, which successfully restored processability. Anderson [10] compared the mechanical properties of virgin and recycled PLA, finding that recycled PLA, after one cycle, showed a mechanical tension decrease of approximately 4 MPa compared to virgin material. Sanchez et al. [11,12] conducted two studies on PLA recycling over five cycles. In 2015, they noted no significant decrease in tensile strength but observed a 10% reduction in elongation at the breaking point after five cycles. However, their 2017 study found a 35% decrease in the mechanical properties of additively manufactured tensile specimens, highlighting inconsistency in the results for recycled PLA. In their study, Mats Bremmer et al. [13] investigated the recyclability of PETG, a PET-based plastic modified with glycol to enhance its 3D printing properties, within the context of additive manufacturing. The research specifically examined the use of 3D printing waste, such as misprints and support structures, to produce recycled filament on a laboratory scale. To assess the feasibility of filament production, Bremmer et al. tested three blends of recycled and virgin granulate. The quality of the recycled filament was evaluated based on filament diameter, the dimensional accuracy of printed test specimens, and their mechanical properties. Given that previous studies on PLA recycling have shown significant issues only after multiple recycling cycles, Bremmer et al. chose to reuse PETG plastic just once to minimize potential impacts on material properties. This approach aimed to provide initial insights into the feasibility of PETG recycling for additive manufacturing, addressing a gap in the literature, as no specific information previously existed for this application. This study demonstrated that PETG 3D printing waste can be effectively recycled into new filament. Uniform granulate shape and size improved filament diameter and mechanical properties. While recycled material showed slightly lower tensile strength, a 50/50 mix with virgin granules matched the tensile strength of purchased material, indicating significant potential for industrial recycling. Further research should explore the impact of multiple recycling cycles and optimize production parameters. [13]

This paper aims to investigate the degeneration or evolution of mechanical behaviour exhibited by PETG throughout the recycling process within the context of a circular economy. Specifically, we seek to assess how the mechanical properties of

PETG specimens, initially printed from virgin material, are influenced as they undergo successive cycles of recycling and reutilization. Our study involves subjecting 3D printed PETG samples to mechanical testing, including tensile, compression, and impact tests, followed by re-milling and reusing the material to produce new filament for printing. By examining the mechanical behaviour of PETG in the context of recycling and circular economy principles, this research serves as a crucial step towards understanding the material's sustainable usage in 3D printing applications. Establishing a baseline of mechanical properties for PETG after the first cycle of recycling will elucidate the potential trade-offs between material sustainability and performance. Furthermore, insights gleaned from this study can inform future efforts aimed at optimizing recycling processes and advancing the development of eco-friendly 3D printing materials within the framework of a circular economy.

MATERIALS AND METHODS

Keeping in mind that the domain with which we concern ourselves is 3D printing, a plethora of materials are available for use and study, but we will be concerning ourselves with the polymer Polyethylene Terephthalate Glycol or PETG for short. This polymer is a widely known and very popular in the industry of 3D printing for its mechanical properties, transparency and impact strength. Being that is part of the thermoplastic families of polymers it has the advantage of being able to be moulded into different shapes or for our study, extruded into filament-based forms. An additional property to be taken into consideration is also the chemical resistance of such a copolymer, having very good behaviour when being acted upon by a large number of solvents, acids and alkaline substances [14]. Important to note is that the material is hygroscopic and therefore proper storage would be required to minimize the absorption of water from the environment.

From a mechanical characteristics point of view, our batch had the following properties, given by the technical spec sheets on this polymer:

1. Density: 1.23 g/cm^3
2. Melting point: 245-250°C
3. Elasticity modulus: E = 2-3 GPa
4. Poisson coefficient: 0.35-0.4
5. Tensile strength: 50-70 MPa
6. Flexural strength: 70-100 MPa
7. Hardness: between: 70 – 90 Shore D

The material used in our tests found itself in 2 distinct starting forms, that being filament (Fig. 1a) and pellets (Fig. 1b).

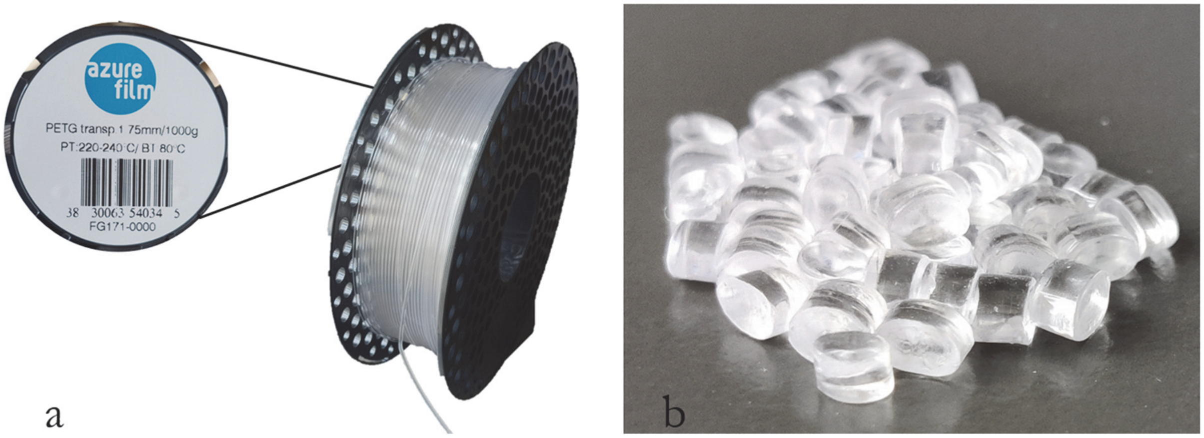

Figure 1: Filament based (a); Pellet based (b)

The main reasoning of starting with different forms of the same material is to see if there are any inherent differences between these two regarding the mechanical and printability characteristics of them. The comparison is also of importance because this would be a starting point for future recycling loops where the material will end up in the end as shredded pellets ready to be reformed into filaments, marking a new usage cycle. Having prepared the necessary material, we must turn our attention to the aforementioned cycles and which route the finished specimens would have taken in the study, as the recycling process is mechanical only, with no addition of other composites or additives. In the following Fig. 2 the routes are presented from the start as raw material to the end as shredded polymer.

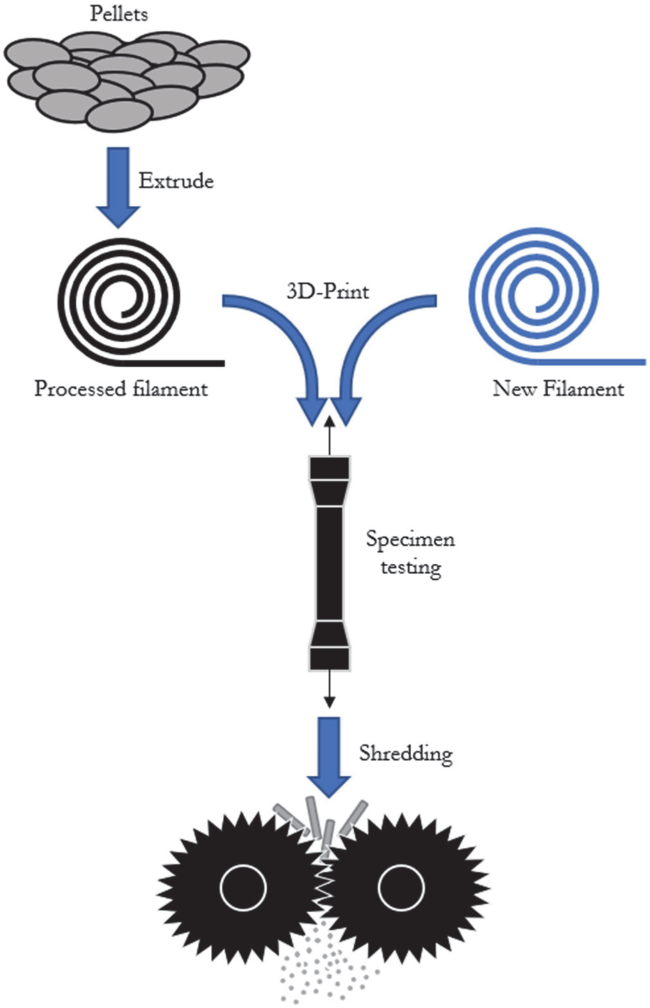

Figure 2: Flow of new and/or recycled material as a complete cycle of use

The 3D printing was carried on the Prusa i3 MK3S printer (Fig. 3a), mainly being its ease of use, very intuitive controls and overall adaptability. The extruding process was fulfilled by the Felfil -EVO- being a manually top loading heated screw extruder with adjustable feed speed (Fig. 3b). After the testing trials the parts are shredded in the Felfil Shredder.

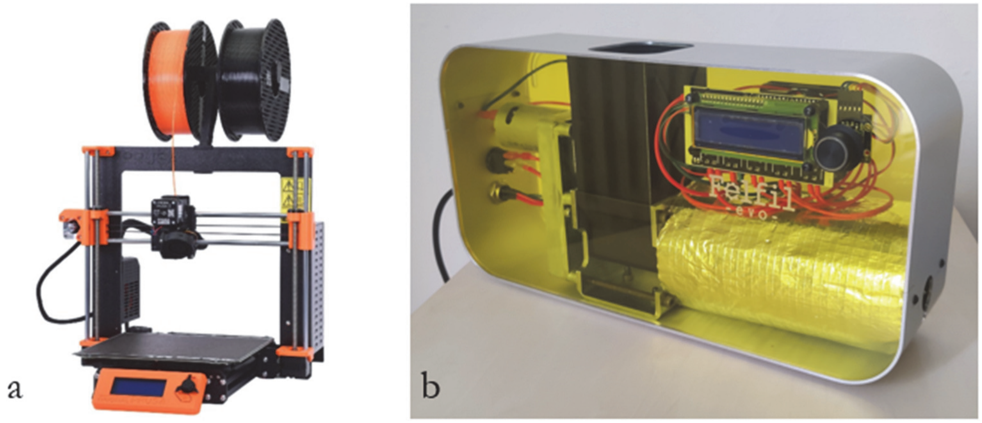

Figure 3: Prusa i3 MK3S (a); FelFIl -EVO- (b)

The aforementioned devices are the main part of the fabrication and reusing cycle (Fig. 4). The Evo extruder is heated up to 195°C with a set speed of 7 revolutions per minute. A higher temperature can be used, but of note is that above 350°C PETG starts to lose density. For the time being the spooling of the filament has to be done manually, with future endeavours including an automatic spooling mechanism for a higher yield of filament.

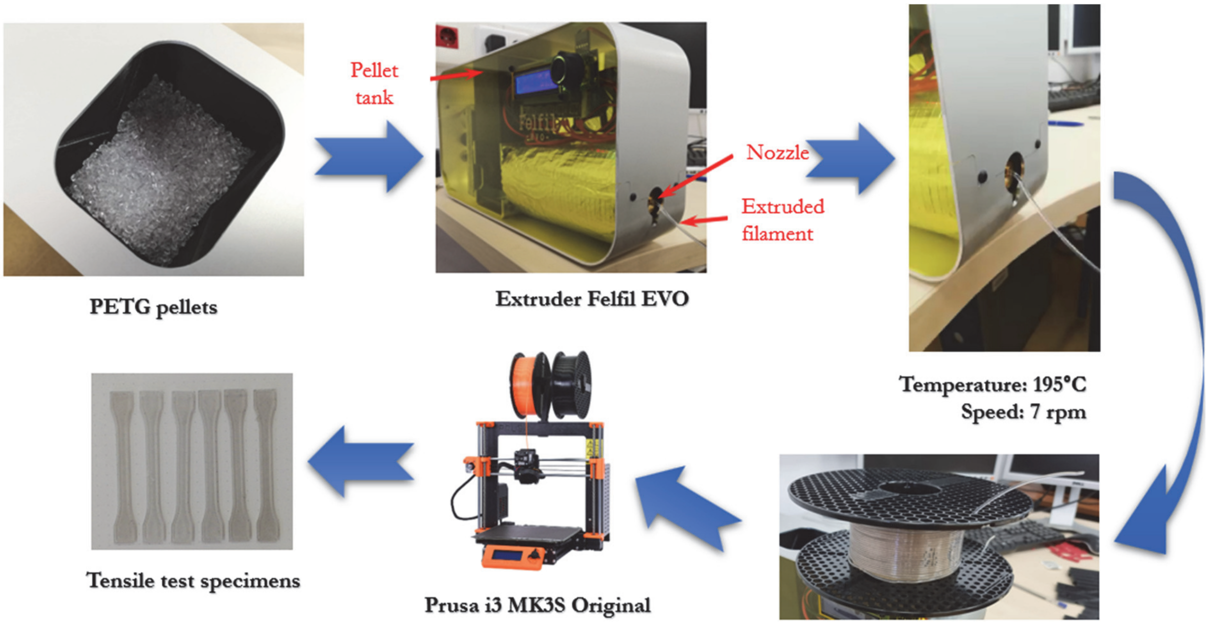

Figure 4: Workflow of raw material from pellets to finished samples.

For each of the planned tests the printing of 2 distinct sets of specimens was required in order to compare the behaviour of new filament and the processed filament from the pellets. Therefore, samples were numbered from 1 to 6 for the new filament, and for the processed filament an extra asterisk/dot was added. Dimension wise, both sets of specimens were in the same value range, without noticeable variations in tolerance range. Being that there are no noticeable differences between new and processed filament specimens. Each sample has been measured at 3 points on the relevant area and averaged in order to have an overall view of the value. No other post processes have been done to the samples after the printing phase, other than the removal of the hair like strands of material, where applicable, after the process finished.

Tensile test setup

Tensile specimens are printed according to the ISO 527 – 2:2012 for extruded and injected polymers (Fig. 5).

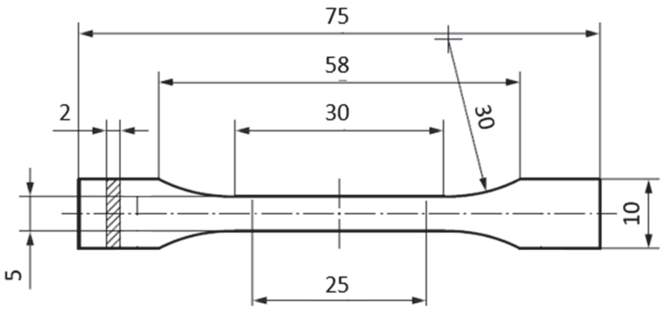

Figure 5: Specimen dimensions according to ISO 527-2:2012 [15]

After the printing process each specimen was measured and recorded in order to assure that there was no inherent fault with the process that could negatively influence the results of the tests. The average thickness and width of the specimens is 2.10 mm and 5.05 mm respectively with a measurement error of ± 0.05 mm. A slight challenge has appeared when printing with the processed filament, because of its air inclusions into its mass. Such issues need to be troubleshooted and the printing process supervised as it could potentially decalibrate the printing process, resulting in either thinner specimens or with defects.

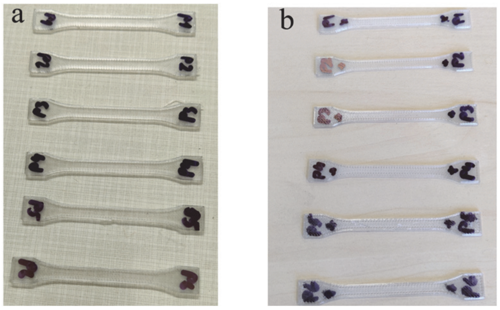

Figure 6: New filament (a); processed filament (b)

For the effective testing of said specimens the equipment used was a Zwick/Roell Z005 universal testing machine (Fig. 7), with a maximum force reading of 5 kN, precision class 0.5 according to DIN EN ISO 7500-1, ASTM E4, and a speed between 0.0005 to 1500 mm/min. Measurement of the elongation was done using an electronic extensometer with an initial state of 30 mm gap between the measuring points. Reading of the required data is done by the extensometer and such it is imperative that the breaking point would have been between the 2 anchor points. The speed with which the test was carried out is 5 mm/min at room temperature and normal atmospheric pressure.

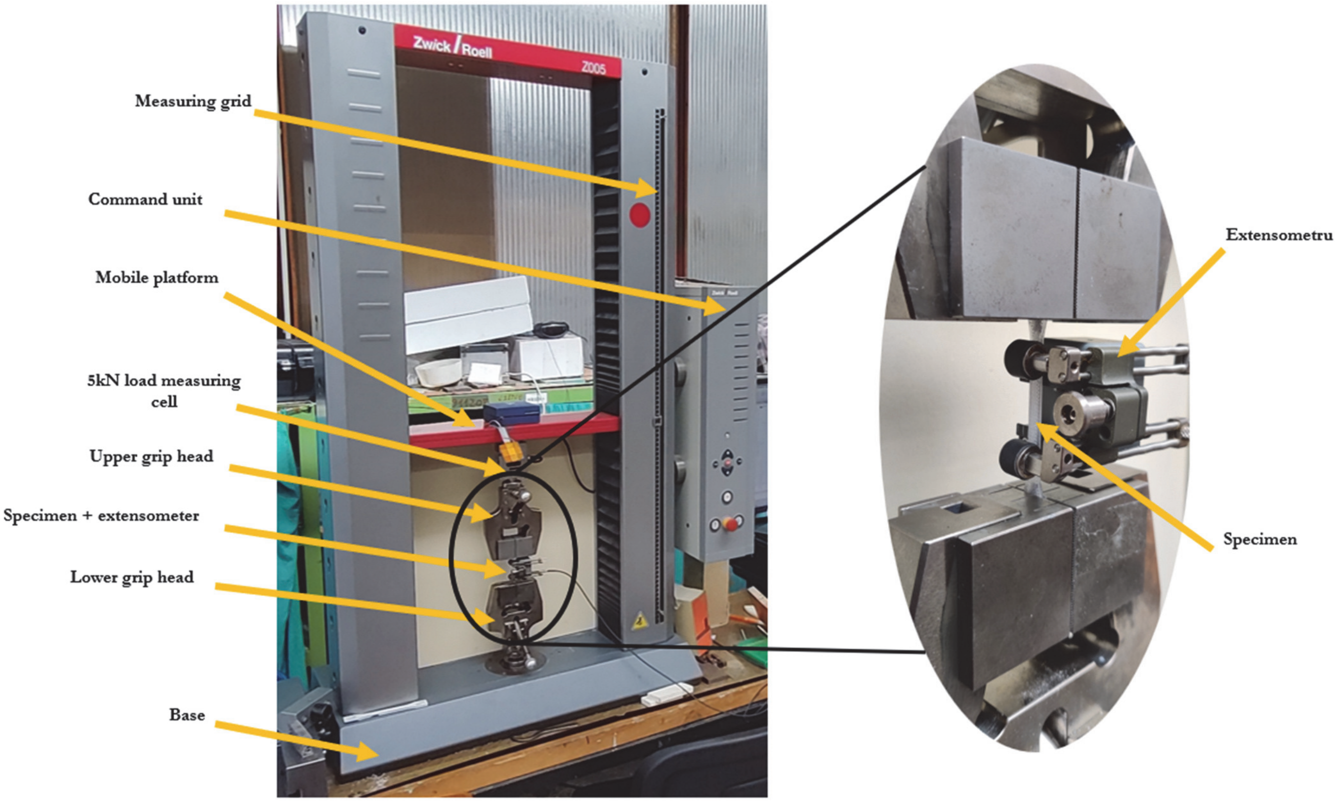

Figure 7: New filament (a); processed filament (b)

Compression test setup

The specimens are cylinder shaped (Fig. 8), with an average height of 25.3 mm and a diameter of 12.5 mm with a measuring error of 0.05mm.

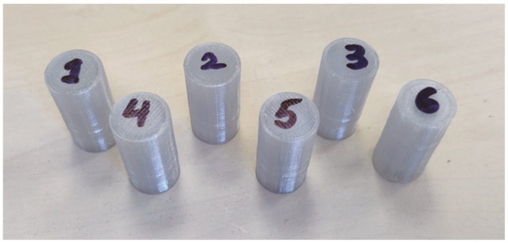

Figure 8: Compression test specimens.

The equipment used for the compression test is LBG A0009-TC100 06N/1 series (Fig. 9), with a maximum force of 100 kN

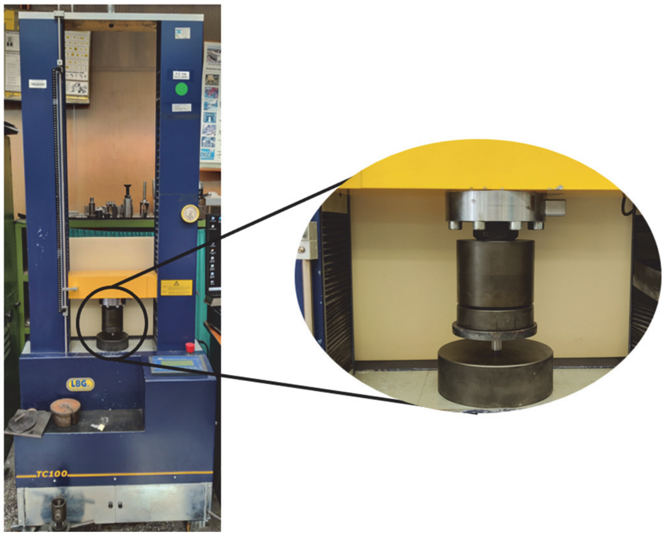

Figure 9: LBG A0009-TC100 universal testing machine

Impact test setup

The specimens are printed according to ISO 179 - 2, and Fig. 10, with a 2 mm deep notch, along with the impact setup consists of an INSTRON: CEAST 9050 Pendulum Impact System. Being that the notch is present in the specimens a 5 J potential energy impact hammer was chosen.

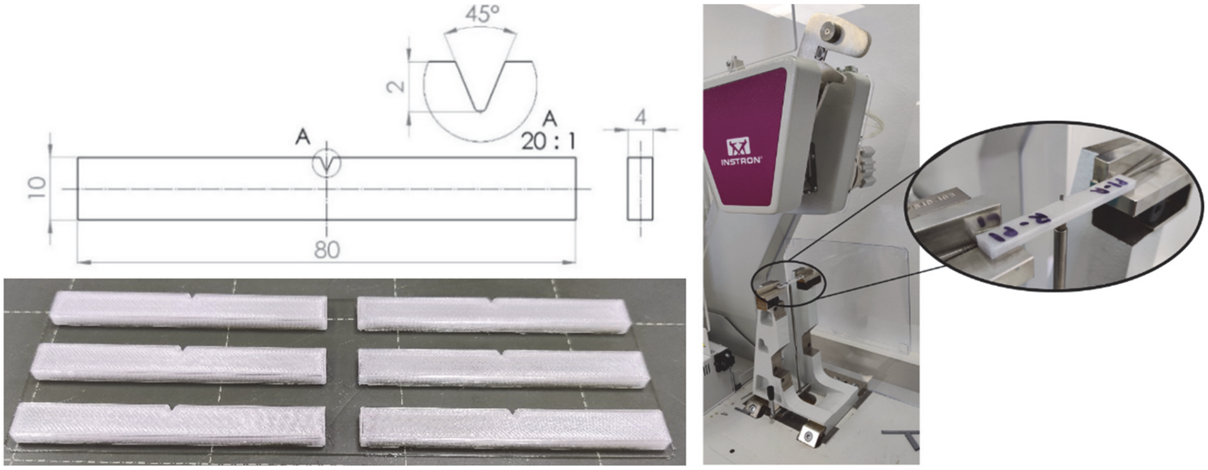

Figure 10: Specimens (left); Pendulum impact system (right).

Below in Tab. 1 are the parameters and characteristics of the impact hammer used for this test along with the measured values during the calibration period.

Nominal Parameters	
Hammer type	Charpy
Potential energy	5 [J]
Impact speed	2.9 [m/s]
Starting angle	150°
Length	229.7 [mm]
Measured parameters	
Hammer at 90°	11.637 [N]
Oscillation time	0.962 [s]
Reduced length	0.22977 [m]
Impact length	0.2297 [m]
Energy loss	0.009 [J]
Vertical offset at impact	-0.1 [mm]
Impact surface	
Angle	30°
Radius	2.04 [mm]

Table 1: Charpy impact hammer characteristics

RESULTS AND DISCUSSION

Tensile Tests

Even at this step of material processing, signs of different characteristics are beginning to show (Fig. 11), regarding to the Young Modulus, and maximum stress. The new filament has an average E = 2 GPa compared to the processed one at E = 2.28 GPa. Together with the increase in elasticity there is also a difference in the value of the maximum tensile strength, σ_{max} = 48.31 MPa for processed compared to σ_{max} = 46.47 MPa for the new filament. This increase could be attributed to swings in material quality of the processed PETG.

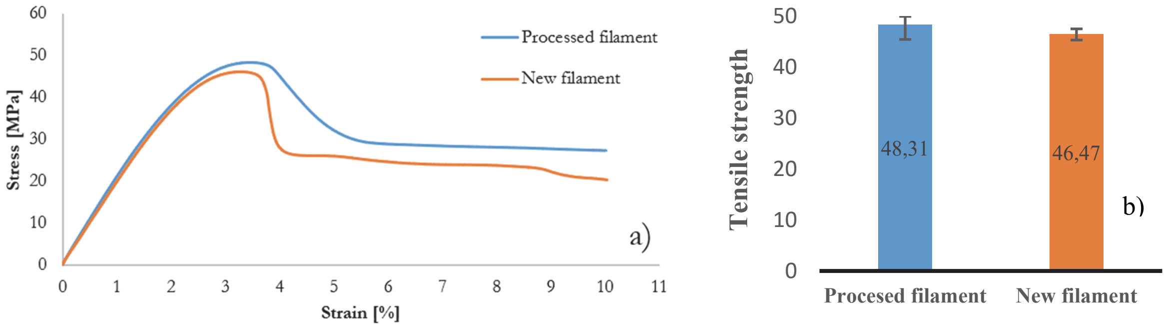

Figure 11: Typical stress - strain curves (a); Tensile strength (b).

During the testing, the processed material although presenting marginally better results, it was more prone to instability of the region where the fracture took place, having a bigger swing in the position where the rupture took place. Possible cause is the presence of small defects from the printing process, either inside the samples or on the surface from the very thin threads of plastic that are creating during passes. From the graph it can be also observed that the slope present in the elastic zone for the processed material is indeed steeper compared to the new one. One exception would be in the area of the interval where stress abruptly drops, almost vertical for the new material, compared to the processed one which seems like it's somewhat delayed, not such a fragile drop. As for the maximum force required, even is the new filament performed slightly worse, the spread of its values is more contained compared to the processed one where the values are distributed in a wider interval, that implies that the stability of the material can be taken into question, but not forgetting that the printing process and faults are also an important variable.

Type of filament	New filament						Processed filament					
Specimen no.	1	2	3	4	5	6	1	2	3	4	5	6
Peak Force [N]	460.22	446.15	467	466.5	465.55	458	489	495.65	482.5	489.42	492.67	446.34
Peak Stress [MPa]	46.23	45.33	46.83	47.54	46.53	46.34	49.39	49.81	48.34	48.89	49.99	45.44
Strain at peak Stress [%]	3.27	3.16	3.29	3.15	3.32	3.21	3.5	3.51	3.45	3.46	3.4	3.22
Young's Modulus [GPa]	2.02	2.28	2.25	2.79	2.11	2.21	2.19	2.02	2.19	2.47	1.9	2.25

Table 2: Tensile test results.

Compression tests

The compression test follows a similar process to those that were done for tensile, having a batch of 6 samples from new and processed filament and seeing their respective behaviour and if there are things to note. The speed with which the compression test was performed was set at 6 mm/min. As it can be observed in Fig. 12, the overall form and values are comparable, with almost a perfect match between them.

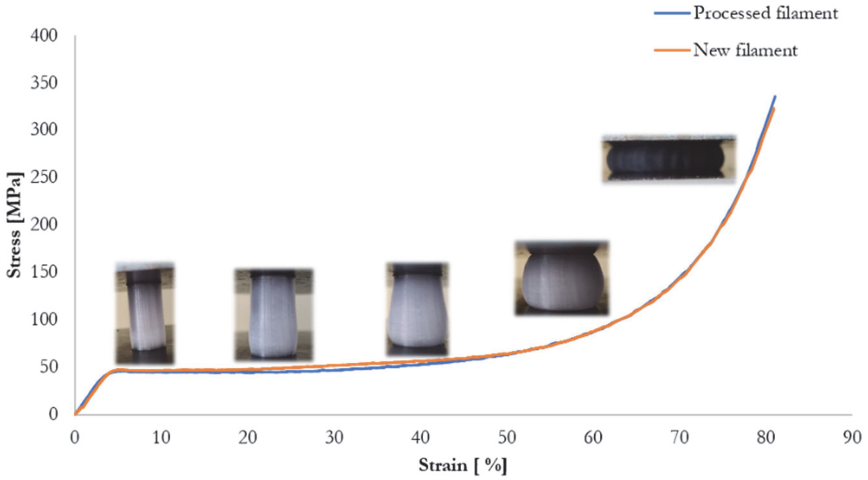

Figure 12: Stress – strain diagram with sample behaviour.

Type of filament	New filament						Processed filament					
Specimen no.	1	2	3	4	5	6	1	2	3	4	5	6
Compression modulus [GPa]	1.05	1.15	0.91	0.96	1.02	1.09	1.06	1.01	1.26	1.27	1.31	1.25
Plateau stress [MPa]	48.21	48.45	47.66	45.72	45.38	46.4	46.65	47.16	44.62	44.67	45.37	44.07
Densification strain [%]	46.8	47.26	45.36	44.46	45.62	45.94	46.40	52.4	45.65	45.53	45.88	44.78

Table 3: Compression test results

During testing the specimens were stable, not presenting brittle behaviour. At the end of the process, the samples have been compressed to a height that is 20.5 mm shorter than their starting value. This was picked as an endpoint due to the fact that more pressure wouldn't bring any other discernible results, having already been necessary 37 kN of force to bring them into this state. The overall difference in compression-modulus is insignificant, approximately 0.15 GPa between all samples, Tab.

3. The average plateau stress, corresponding to 10% deformation is 46.97 MPa for new filament, respectively 45.42 MPa for processed filament. Regarding the densification strain, defined the value of the strain at 30% stress increased from plateau stress, the value for new filament is 45.90 %, respectively for processed filament 46.77%.

Impact tests

The impact test results display a trend that closely mirrors previous findings, showing a tight distribution of outcomes between the processed filament and the new one. For clarity, the data points that most accurately represent the average results for each type of filament were selected and illustrated in Fig. 13.

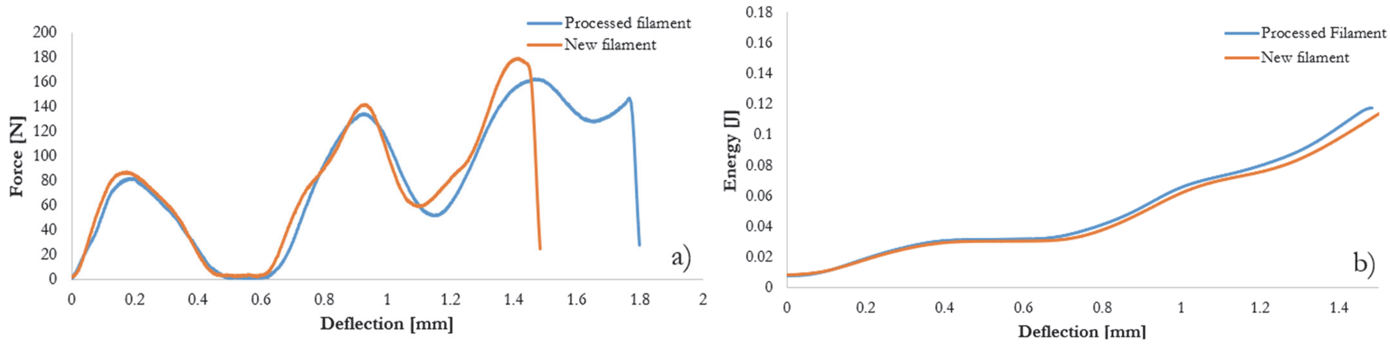

Figure 13: Specimen behaviour during testing load-deflection (a); Energy–deflection (b).

The specimens although presenting similar values, do show a slight difference in the failure mechanism, that being the processed ones have a clearer cut fracture as opposed to the new filament ones, which show less brittle behaviour. All the specimens do present, from each category, this behaviour which points to a stiffening of the processed material, at least for the impact test. What is also to note is the Charpy impact strength of the specimens Fig. 14, having similar values calculated with Eqn. (1), but slightly lower for the processed specimens as opposed to the new filament. To note is the tighter spread of values for the new filament as opposed to the processed one where the amplitude of results is higher.

$$a_{cN} = \frac{W_c}{h \times b_N} \times 10^3 \qquad (1)$$

where:

W_c is the corrected energy absorbed by breaking the test specimen in [J];

h is the thickness of the test specimen, in [mm];

b_N is the remaining width in the notched section of the test specimen in [mm].

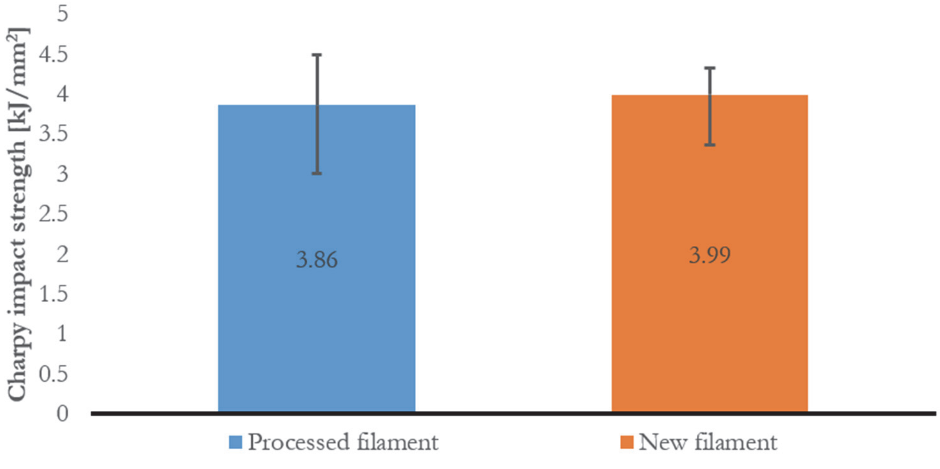

Figure 14: Average Charpy impact strength.

Type of filament	New filament						Processed filament					
Specimen no.	1	2	3	4	5	6	1	2	3	4	5	6
Surface area [mm^2]	31.69	31.7	31.78	31.34	31.72	31.17	31.5	31.42	31.56	31.73	31.21	31.46
Peak Force [N]	158	180.71	195.82	179.29	152.06	153.48	167.81	162.13	176.33	168.74	170.9	145.22
Total Energy [J]	0.13	0.10	0.13	0.11	0.12	0.13	0.09	0.13	0.14	0.13	0.09	0.12
Charpy impact strength [kJ/mm^2]	4.33	3.35	4.29	3.75	3.91	4.31	3.11	4.25	4.49	4.07	4.21	3.01

Table 4: Charpy test results.

This study acknowledges some limiting factors that may influence the generalizability of the findings. Firstly, the relatively small sample size could limit the statistical power of the findings, making it difficult to draw broader conclusions. Additionally, the focus on only one recycling cycle restricts the ability to assess how material properties might change across multiple cycles, which is important for understanding long-term durability and performance.

CONCLUSIONS

In conclusion, this study examined the behaviour of PETG when comparing specimens printed from new material versus recycled material through compression, tensile, and impact tests. The preliminary findings indicate a marginal difference in stiffness, with recycled material exhibiting slightly higher stiffness. Additionally, there is a small alteration in mechanical characteristics, notably an increase in brittleness. However, it is important to exercise caution in drawing definitive conclusions based solely on this initial data. Upon closer examination, the data reveals that the new material demonstrates greater stability, particularly in its predictable performance in tensile strength tests. In contrast, the recycled material, while occasionally stronger, displays greater variability in its results. This variability suggests a degree of instability, possibly due to variations in material composition or inconsistencies in filament formation during the recycling process. Thus, while recycled PETG shows promise in certain aspects of mechanical performance, such as strength, its variability necessitates further investigation and scrutiny. The potential impact of these limitations on the generalization of the findings is significant. The small sample size and single recycling cycle may result in conclusions that are not fully representative of real-world scenarios, where materials often undergo multiple recycling processes. These constraints highlight the need for caution when applying these findings to broader contexts. Future research should aim to address these limitations by increasing the sample size and examining material properties over multiple recycling cycles. Such studies would provide a more comprehensive understanding of the effects of recycling on material performance and enhance the generalization of the results. This will allow for a better understanding of the trade-offs and implications of utilizing recycled PETG in 3D printing applications, ultimately advancing sustainable manufacturing practices while ensuring consistent and reliable performance.

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