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Investigating Material Degradation through the Recycling of PLA in Additively Manufactured Parts

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ABSTRACT

The field of additive manufacturing (AM) has been expanding rapidly with the decreasing cost of desktop-scale material extrusion AM systems. As the cost of AM systems decreases, more users are investing in the technology, including universities, which have turned to AM as an option for providing wide-scale access to prototyping technology. However, this type of wide-access printing generates significant waste due to cast-off support material as well as failed prints from inexperienced users. This paper investigates the feasibility of recycling this cast-off material through the relationship between the mechanical properties of recycled PLA and the number of lifecycles it has experienced on a desktop material extrusion machine. A three-stage pelletizing, extrusion, and printing process is used to investigate recycling of PLA material from cast-off build material. Additionally, the research investigates how adding virgin pellets to pellets of the recycled material in various ratios can affect tensile properties.

1. INTRODUCTION AND MOTIVATION

As additive manufacturing (AM, or 3D printing) has continued to grow, the cost of desktop AM systems has fallen rapidly, while their capabilities have increased exponentially. This, combined with desktop systems' relative ease of use, has led to increased popularity across various educational levels. These systems can be used not only in engineering, but also across disciplines (e.g., art, architecture, science, etc.) to promote interest in design, making, prototyping, and manufacturing. While there are numerous case studies in literature exemplifying the use of printers in instructor-guided activities, such situations present potential issues with instructor and student training, safety, and availability of course resources [1]. To counter these limitations and offer wide access, many universities have turned to so-called "MakerBot Innovation Centers" [2]. These spaces are often characterized by numerous desktop-scale material extrusion systems (upwards of 30-50 printers) located in a central space, such as a library [3,4]. By offering this form of wide access, programs strive to engage as many students as possible, from across departments, to participate in AM. As an alternate approach to open-access AM, several universities have experimented with AM vending machines, several printers are located within a vending machine-like enclosure and made available to the students [5,6]. These machines allow for easier student observation of the printing process, while still minimizing safety concerns.

While this form of large-scale availability is essential to the continued growth of AM and the closing of the AM skills gap, use of the systems in an academic setting still constitutes a learning experience for participating students. This necessitates a certain "freedom to fail," whereby students can experience the potential of AM and begin to learn its nuances without fear of academic or financial punishment for failure. Encouragement of this experiential form of learning can result in significant amounts of waste, and thus added financial burden, on the host institution. This waste is generated not only from failed prints, but also from the support material and cast-off raft material needed to ensure successful prints. Material recycling and respooling is one possible avenue to counteract this waste generation inherent in large-scale access to AM. However, to date there has been little research to identify the impacts that material recycling has on the quality of printed parts.

To quantify the quality of printed parts created via filament recycling, this paper investigates changes in the tensile properties of material extrusion parts through multiple lifecycles. Additionally, the impact of combining recycled filament with virgin pellets in examined as a possible method to strengthen recycled material. This allows the authors to offer recommendations regarding the suitability of recycled filament to support large-scale printing access. Section 2 will discuss existing research in the field of recycled polymeric materials. Section 3 will then detail the authors' design of experiments approach, along with specific extrusion, printing, and testing parameters. Section 4 will quantify and discuss the implications of any statistically significant differences in tensile properties due to changes in filament lifecycles and composition. Finally, Section 5 will offer conclusions and discuss avenues for future research and implementation of material recycling.

2. OVERVIEW OF EXISTING FILAMENT RECYCLING CONSIDERATIONS

Related research relevant to this work has been divided into three sections. Section 2.1 discusses the existing plastics recycling process and how filament extruders fit into this, Section 2.2 discusses the impact of multiple extrusions on material properties of plastics, and Section 2.3 discusses material properties in printed parts.

2.1 Filament Extrusion as a Plastics Recycling Process

As discussed in Section 1, AM has the potential to produce a significant amount of waste due to support material, rafts and failed prints. While these elements are common for advanced AM users, the learning curve that novice users must undergo may result in even more waste generated. For material extrusion printers, all of this waste will be thermoplastic material, primarily PLA (Polylactic Acid) or ABS (Acrylonitrile butadiene styrene) depending on the chosen build material. As these plastics are not chemically altered during the printing process, there is the potential to convert all waste material back into usable raw materials for the printers by simply melting and reforming as a filament. This has given rise to numerous desktop-scale filament extruders aimed at the hobbyist community; example systems include the Filabot (https://www.filabot.com/), Filastruder (https://www.filastruder.com/), or Recyclebot (http://reprap.org/wiki/Recyclebot). While these systems aim to convert shredded waste plastic into usable filament, they often suffer problems with inconsistent filament diameter due to input material geometry or processing parameters [7].

Additionally, just as AM enables distributed manufacturing, it likewise enables distributed recycling. As only roughly 6.5% of plastics are recycled through centralized recycling systems in the United States [8], any plastics placed into centralized recycling have only a small chance of being reused. Furthermore, the energy demands of locally recycling waste plastics are lower than recycling through centralized recycling facilities and much lower than creating virgin materials [7,8]. The localized recycling process has the potential to save not only materials, but also energy. This savings is even more pronounced in rural or developing areas where recycling costs are higher and new raw materials are harder to come by [9]. These advantages of localized recycling provide further justification for polymer filament recycling.

2.2 The Effects of Multiple Extrusions on Plastic Properties

The goal of this research is to investigate the impact of multiple heating and extrusion cycles on the material properties of printed plastics. Though there is no literature on the effect of multiple extrusion cycles on the properties of printed parts, there is literature on the effect of multiple extrusions on the material properties of both injection molded PLA [10] and ABS [11]. In both of these studies, the plastics were heated and reformed a number of times using an injection molding process. After injecting molding various testing specimens, the specimens were tested to identify material properties as they related to the number of extrusion cycles they went through.

For PLA, the material properties remained relatively stable over a total of ten extrusion cycles [10]. The tensile strength of the material decreased by only 5.2% while the elongation at failure and the modulus of elasticity remained constant. However, impact strength did significantly decrease by 20.2%. This indicates that the material became more brittle with repeated extrusions. However, the largest difference was with respect to the melt flow rate, which increased by 236% over the ten cycles. Though bulk material properties remain largely unchanged, this change in melt flow rate has the potential to alter the printing process, which will affect print quality, which in turn can affect the strength of printed parts.

For ABS, the bulk material properties also remained relatively constant over multiple extrusion cycles [11]. Tensile strength and modulus of elasticity showed no significant change over five extrusion cycles, while elongation to failure showed a small decrease. As with PLA though, the melt flow rate of ABS does increase with repeated extrusions, which may influence print quality and thus printed part strength as previously mentioned.

2.3 Material Properties in Material Extrusion Parts

The authors hypothesize that the use of recycled filament will amplify existing concerns and considerations regarding the material properties of polymer AM parts. The inherent layer-by-layer process of additive manufacturing leads to final parts that require special attention to their mechanical properties. For example, as with all AM processes, parts produced via material extrusion in particular will exhibit anisotropic behavior due to poor layer-to-layer adhesion [12]. This anisotropy can also appear in the XY build plane, but depends on the chosen infill percentage and raster pattern [13]. Additionally, small voids in each layer can cause localized stress concentrations that further weaken parts [12]. Because of this, bulk material properties will not entirely predict printed part properties. The exact printer settings and characteristics will play a vital role in determining part strength. In a study looking at a variety of desktop material extrusion systems, Tymrak and colleagues [14] found large variations in tensile strength and other properties from one device to the next and even from one filament to the next, though they were all nominally the same process and material. This may indicate that even small variations, such as the small changes to material properties that may occur over multiple extrusions may lead to much larger changes in printed part mechanical properties.

3. EXPERIMENTAL APPROACH

While there are researchers investigating the use of scrap polymer material in the creation of recycled filament [15], there are currently no studies that compare the mechanical properties of printed parts made with recycled plastic filaments to similar parts made with virgin plastic filament. This work seeks to identify the changes in the material properties of printed parts as a result of multiple extrusion cycles and to investigate the use of mixed virgin and recycled material as a method for maintaining material integrity. Section 3.1 discusses the selection of independent variables as well as the design of experiments, including the mixtures for the sample groups and the rationale driving our selection of certain ratios. Section 3.2 discusses the pelletizing and filament creation processes used to mimic recycling, and Section 3.3 discusses detailed printing and testing parameters and procedures.

3.1 Design of Experiments

A design of experiments approach is used to examine both the number of times filament can be recycled without impact on material properties, as well as the effects of supplanting specific quantities of recycled stock with virgin material. The selection of these as crucial independent variables is as follows:

Number of Times Recycled: Often, cast-off waste material from printing is disposed of and new filament is used for subsequent prints. However, other AM process types (specifically powder bed fusion and binder jetting) have shown that loose powder material can be reused, though it suffers degradation of material properties. While reuse of unsintered powder is conceptually different from breakdown and reuse of previously heated filament, it nonetheless emphasizes the need to understand the impact that the number of lifecycles has on material properties.

Percent of Virgin Pellets Injected Before Recycling: The authors hypothesize that, as the PLA filament undergoes additional recycling stages, its material properties will begin to degrade. In order to counteract this anticipated degradation, a set mass percentage of virgin PLA pellets will be added to the recycled pellets prior to each recycle process. The authors hypothesize that as the mass percentage of virgin material increases in each recycle stage, the recycled filament will be able to maintain its mechanical properties over a larger number of lifecycles.

Table 1 denotes the different variable combinations used for testing. The first set of prints consisted of five test samples printed material from a new, unopened 2 pound reel of light gray filament direct from the supplier, MakerBot. This first set serves as the control and provides a baseline for comparison. Subsequent recycling groups were printed with varying ratios of virgin and recycled material, as is detailed in Table 1; each recycling group was printed in three sets of 0% virgin, 25% virgin, and 75% virgin material. Each material ratio was printed in a set of five samples to control for slight mixture variations within the heated chamber of the filament recycler. In total there were seven sets tested, for a final count of thirty-five specimens.

Table 1: Independent Variable Combinations Used for Experimentation

Set Number	Number of Times Recycled	Percent Virgin Material Introduced to Mix During Recycling
1	0	-
2	1	0
3		25
4		75
5	2	0
6		25
7		75

3.2 Filament Creation

In order to mimic the recycling process that would be used for cast-off waste material, the authors utilized a three-stage process, which includes a pelletizing phase, a filament creation phase, and a printing phase. This section discusses the first two stages necessary to create recycled filament, while Section 3.3 will discuss parameters used for final printing of the recycled filament.

Pelletizing Process

In order to test the strength of the recycled filament, pellets were made using a simple pelletizing process established in-house (see Figure 1). In this process, filament is fed into a chamber with a standard wood routing bit controlled via a drill press. This filament can be either virgin filament provided by a manufacturer or previously recycled filament created via the extrusion process discussed in the next subsection. By maintaining a constant filament feed rate into the chamber and a constant rotational speed for the routing bit, pellets of relatively consistent length could be created. Note that this approach does not fully replicate the recycling process as it would occur with naturally cast-off material from a printer; waste material from a printer will take a wide variety of geometries, which makes the pelletizing process challenging. By performing this initial experimentation based entirely from pellets created from spooled recycled filament, it is possible to recreate the heating and cooling processes that natural waste material would undergo, without the difficulty of addressing irregular waste geometries at this time.

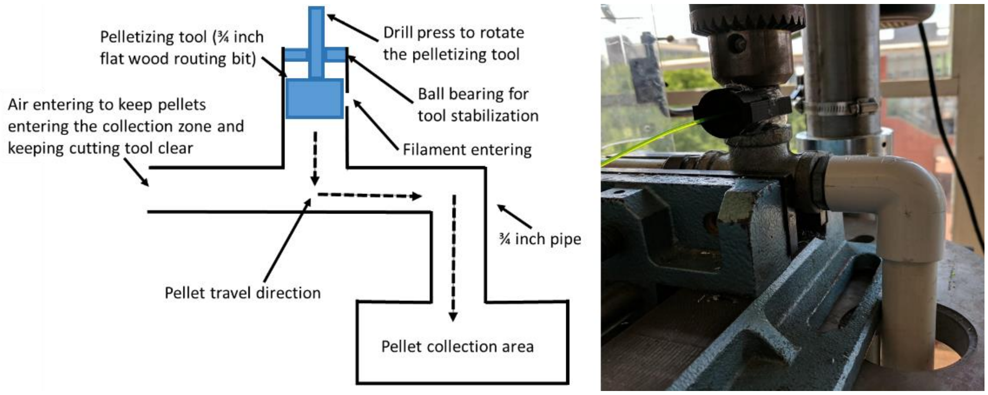


Figure 1: Filament pelletizing system diagram (left) and filament pelletizing embodiment (right)

The pellets of recycled material that exit the pelletizing system were cylindrical in shape with an approximate diameter of 1.7mm and height of 5mm. In contrast, the virgin pellets (IC3D Natural PLA Fine Plastic Resin Pellets) were ellipsoidal with a minor axis diameter of 3mm and a major axis diameter of 5mm. An example of each pellet form is shown in Figure 2. While the recycled and virgin pellets would ideally possess the same geometry, the difference shown in Figure 2 is likely more similar to what would be encountered in a true recycling environment. In the case of large-scale, open-access AM environments, it is likely that it would be difficult to secure virgin pellets that are identical in material properties and geometry as the pre-spooled print material used in the material extrusion systems.

Figure 2: Filament pellets (left) vs. virgin pellets (right)

<u>Filament Creation Process</u>

For this experiment, the filament was recycled using a Filabot Original filament extruder (Figure 3). The Filabot was set to extrude at a temperature of 175°C and fans were used near the extrusion nozzle in order to rapidly cool the material once it left the nozzle. The recycled filament was allowed to coil onto the ground where it was manually spooled post extrusion. The consistent height of the extruder causing a constant tension on the new filament, the nozzle temperature, and the rapid cooling with the fan were all tuned to ensure that the filament diameter remained as close to the desired diameter of 1.75 mm as possible.

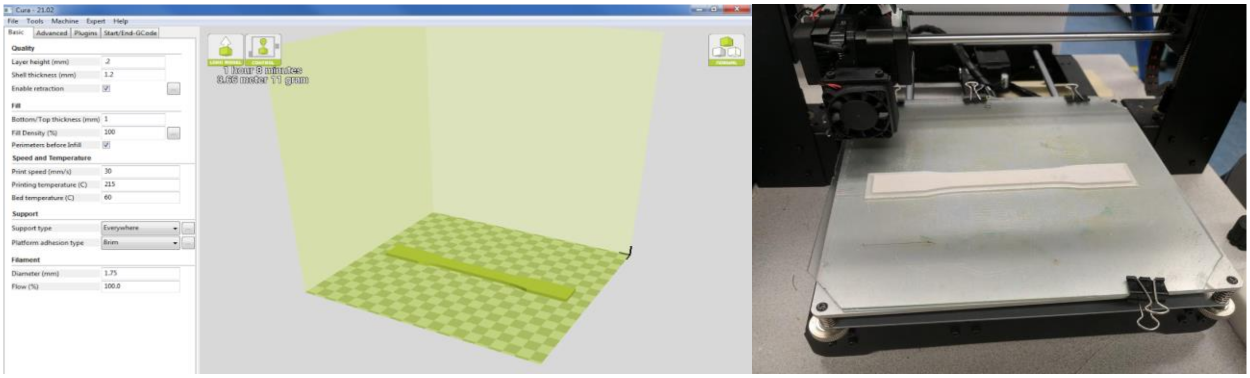
All filaments that had a percentage of virgin pellets introduced were created by weighing specific amounts of material in the desired ratio and thoroughly mixing the recycled and virgin pellets in a container. While mixing was performed in an attempt to evenly distribute the recycled and virgin pellets, agglomeration of the disparately sized pellets is still a potential noise factor. Despite the slight different in geometry between the recycled and virgin pellets, the pellets mixed well and still resulted in filament consistent enough that there were no problems printing with it. The resulting filament was approximately 1.75mm +/- 0.05mm in diameter; no obvious warping or ovalling was seen in the filament’s cross-section.

Figure 3: Filabot extruder used in the filament recycling process

3.3 Detailed Printing and Testing Parameters

For tensile testing, a specimen was modeled according to the ASTM D638 Type I standard [16]. This standard is used to test tensile properties of unreinforced and reinforced plastics in the form of dogbone-shaped specimens. This model was exported as an STL file and imported into the Cura *Lulzbot Edition* slicing engine (Figure 4). The G-code for the specimen was generated with a brim to prevent any warping that potentially could have occurred, as shown on the printed specimen in Figure 4. This brim was removed after the print had been removed from the print bed and allowed to completely cool.

All specimens were printed on a desktop-scale Monoprice Maker Select V2. The Monoprice system was chosen as a typical printer representative of the AM technology commonly found in large-scale, open-access printing environments. All specimens were printed with a cross-hatched 45°/-45° infill pattern at 100% density in order to closely mimic the properties of homogeneous material. The deposition rate was 30mm/s with a travel speed of 150mm/s, as recommended for PLA material by the system manufacturer. The nozzle temperature was set at 215°C and bed temperature was held at a constant 60°C.



Quality
Layer height (mm): .2
Shell thickness (mm): 1.2
Enable retraction: ☑

Fill
Bottom/Top thickness (mm): 1
Fill Density (%): 100
Perimeters before Infill: ☑

Speed and Temperature
Print speed (mm/s): 30
Printing temperature (C): 215
Bed temperature (C): 60

Support
Support type: Everywhere
Platform adhesion type: Brim

Filament
Diameter (mm): 1.75
Flow (%): 100.0

Figure 4: Build Tray Preparation in Cura (left) and Printed Tensile Specimen (right)

An Instron Material Testing System 810 was used for the tensile tests. This system uses a laser extensometer to record gauge length and elongation of the samples. The load applied by the system on the test specimen was measured via a 3 kip load cell. Wedge serrated-faced grips were used to secure the samples. The gauge length, width, and thickness of each sample was recorded in millimeters prior to testing. During testing, the tensile stress and tensile strain were measured every 0.2 seconds. Testing was concluded when the specimen fractured, and the final tensile strength and elongation at failure were recorded.

4. RESULTS AND DISCUSSION

This section will focus on data analysis made a Two-Way ANOVA approach to determine statistical significance. Section 4.1 will discuss the results from the tensile testing. Section 4.2 will discuss the impact this data could have on a material extrusion desktop AM when considering the use of recycled filament.

4.1 Results from Tensile Testing

Table 2 collects the data gathered from tensile testing, including the mean and standard deviation values for each dependent variable. As previously mentioned, ultimate tensile strength, strain at fracture, and material toughness were recorded in order to characterize the general behavior of the recycled specimens as they underwent additional recycling stages with added virgin material. Note that, due to errors with data collection, three of the seven sample sets contain data for only 4 measured specimens, rather than the intended 5.

Table 2: Results from the tensile testing

Recycle Stage	% Virgin Pellets	N	Ultimate Tensile Strength (MPa)		Strain		Tensile Toughness (MJ m^{-3})	
			Mean	**Std_Dev**	**Mean**	**Std_Dev**	**Mean**	**Std_Dev**
0	0	4	45.61	0.83	0.05	0.00	2.02	0.13
1	0	5	42.31	5.69	0.04	0.02	1.38	0.82
	25	5	36.57	2.77	0.03	0.01	1.00	0.57
	75	4	47.70	2.09	0.04	0.02	1.47	0.69
2	0	5	37.35	6.67	0.03	0.01	0.72	0.24
	25	5	32.69	4.01	0.02	0.01	0.50	0.24
	75	4	32.92	6.85	0.03	0.00	0.64	0.20

Ultimate Tensile Strength

A Two-Way ANOVA was chosen as the statistical test to determine both the main and interaction effects of the two independent variables (recycling stage and the percentage of virgin pellets) on ultimate tensile strength (UTS). The Shapiro Wilk test confirmed that the data for each set was normally distributed. The Lavene's test was also conducted and indicated homogeneity of variances in the data between all groups. Adherence to these assumptions validates the use of ANOVA.

As Figure 5 shows, the recycling state had a statistically significant effect on the ultimate tensile strength of measured specimens. As the recycled filament underwent additional recycling stages, strength decreased significantly ($p < 0.0005$). After 2 recycling phases, the average UTS had decreased by approximately 22%. This behavior is as anticipated by the authors; it demonstrates that, if the recycled filament is being utilized in a load-bearing capacity, the designer must account for the decrease in tensile strength, or their product may be under-designed.

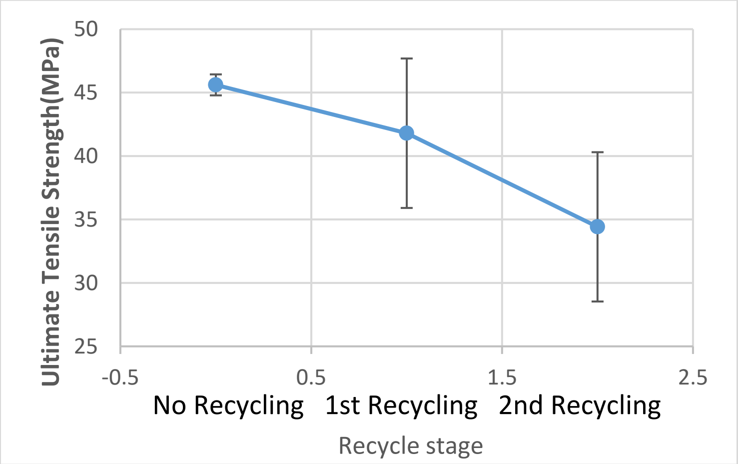


Figure 5: Ultimate tensile strength with respect to the main effects of Recycling Stage

Analysis also shows a statistically significant main effect on UTS as virgin pellets are added to the recycled filament ($p < 0.026$). However, as shown in Figure 6, pellet addition does not appear to constitute a consistent increase as initially hypothesized. A pairwise comparison was conducted and results indicated that there was a statistically significant decrease between 0% virgin pellets added and 25% virgin pellets added ($p < 0.04$) along with a just short of significant increase between the 25% virgin pellets added and the 75% virgin pellets added ($p < 0.054$). However, no significant difference was found between the 75% virgin pellets added and the 0% virgin pellets added. It is possible that the reduction of strength in the 25% of virgin pellets added can be attributed to the quality of the produced samples due to minor inconsistencies in the filament production process. For example, there is the possibility that an uneven distribution of virgin pellets and recycled pellets is causing inconsistent filament properties. As a second option, the packing density of the input stock to the Filabot may be influencing filament quality. Another option would be differences in the melt flow rate of the mixed raw materials might be manifesting in unexpected ways. As a final possibility, differences between the original filament and the pelletized material in terms of additives, moisture content, or other factors may be affecting the print quality or material properties in unknown ways. In the end, the addition of virgin pellets to the mix clearly has an impact, though additional testing is needed better understand this relationship.

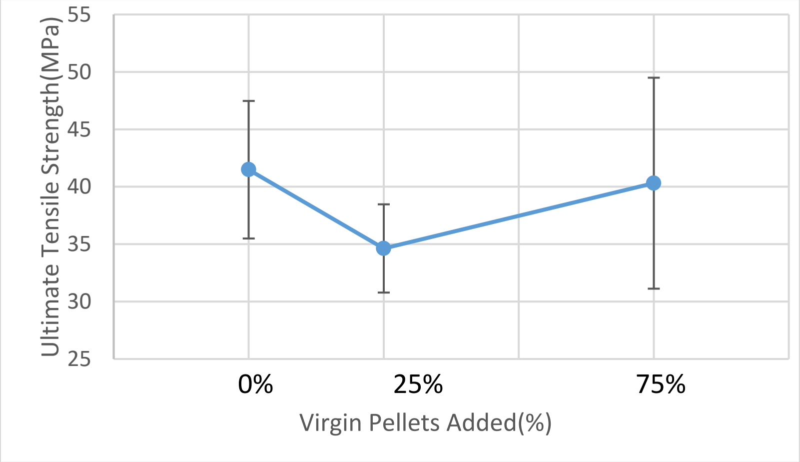


Figure 6: Ultimate tensile strength with respect to the main effects of % Virgin Pellets added

The data in Table 2 also showed that there was a significant interaction effect between both the recycling stage and the percentage of virgin pellets added ($p < 0.047$). As shown in Figure 7, the lines for the first and second recycling stages run parallel from 0% virgin pellets added to 25% virgin pellets added. However, these lines skew beginning at the 25% virgin pellets added to the 75% virgin pellets added on the graph. This trend signifies that the difference in strength values at 25% virgin pellets added and 75% virgin pellets added are significant at different recycling stages. In other words, the strength of the object appears to depend on the recycle stage and the effect of the percentage of virgin pellets added (for added percentages greater than 25%). It is worth noting that the addition of 75% virgin pellets during the 1st recycle stage results in a higher tensile strength than the base filament. The authors hypothesize that this is due to the difference in base material properties between base Makerbot PLA filament and virgin PLA pellets; the Makerbot PLA contains additives which may decrease its tensile strength when compared with true virgin PLA.

Based on the collected data, it appears that the second recycling stage is not affected by adding between 25% and 75% of virgin pellets. As stated earlier, a number of factors might be at play to explain these results, though there is no one obvious explanation. Also observed in Figure 7, the standard error bars for virgin pellets added in the second recycling stage is high, when compared to the first recycled stage. This likely leads to the conclusion that the recycled filament is less consistent than commercial filament, as extruders are sensitive to small variations in filament.

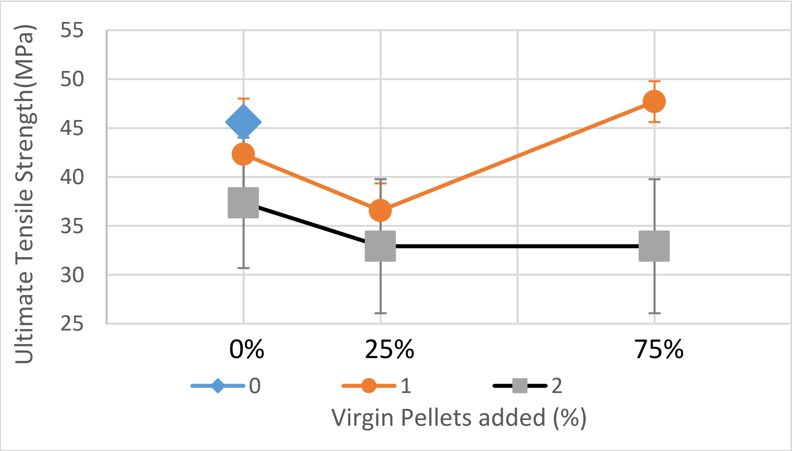


Figure 7: Interaction effect between Recycling Stage and % Virgin Pellets added

Maximum Strain

The Shapiro Wilk test was also performed for the maximum strain data; it confirmed that the data for each set was not normally distributed for the maximum strain values. The Lavene's test similarly showed that the data did not show homogeneity in the variances between all groups. While this denotes that ANOVA may over or underestimate statistical significance (despite its relative robustness to violations of these assumptions), the authors can still extract relevant trends from the collected data. As Figure 8 shows, the recycling phase appears to have a significant effect on the maximum strain, as it did for UTS. As the filament undergoes additional recycling phases, the specimens are able to withstand less extension before ultimately failing. This decrease is especially pronounced when comparing the 2nd phase of recycling against the base material. While statistical analysis suggests that there is no significant impact due to virgin pellet addition or its interaction with recycle stage, additional sample testing is needed to confirm this, due to the initial violation of ANOVA assumptions. Note that the large variation seen the 1st recycle stage in Figure 8 is due to the effects of varying virgin pellet addition; pellets introduced in this stage caused changes in the maximum strain, but not enough to be statistically significant.

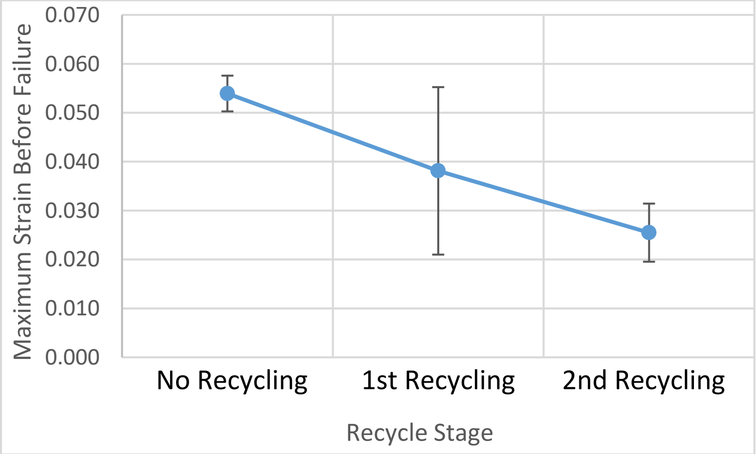


Figure 8: Maximum Strain with respect to the main effects of Recycling Stage

Tensile Toughness

Similar to that of the maximum strain values, measurements for toughness values violated both the assumptions of normality and homogeneous variances. Due to these violations, as with strain, additional specimens may be required before conclusive recommendations regarding statistical significance can be made. However, as Figure 9 shows, there does appear to be a noticeable decrease in the average toughness of printed specimens as they undergo recycling, with the difference between the 2nd recycling phase and the base material being particularly striking. Once again, the data does not suggest any significant effects due to the addition of virgin pellets or interaction effects. As with the strain data, the variation in the 1st recycle stage is due to the effects of varying virgin pellet addition.

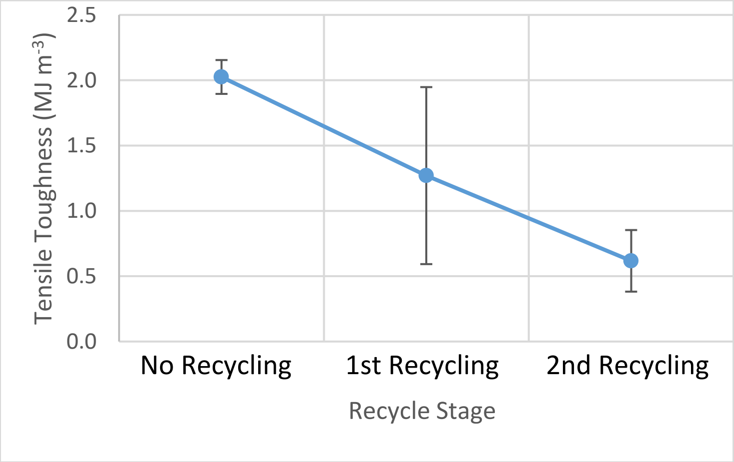


Figure 9: Tensile Toughness with respect to the main effects of Recycling Stage

4.2 Discussion and Implications for Desktop AM

This analysis shows that increasing recycling stages causes significant reduction in tensile strength, strain at failure, and toughness for material extrusion parts. Adding virgin pellets does impact the tensile strength values, however the nature of this impact was found to be highly dependent on the percentage of virgin pellets added, as well as the recycling stages. A 25% addition significantly reduced part strength in both the recycling stages, which may be attributed to inconsistencies when producing filament. A 75% addition improved the part strength but only for the first recycling stage. Strain at failure and toughness were unaffected by the addition of virgin pellets. A better understanding of feed parameters like size, density, and the extruder parameters with respect to filament quality is required in order to pinpoint the cause for the strength reduction on adding 25% of virgin pellets. The results also suggest that the first recycling stage showed an improvement in strength with 75% addition of virgin pellets, which shows that recycling can be performed while retaining the material properties. However, a more detailed observation of filament quality with added virgin pellets is required to ultimately find the most optimized ratio of virgin to recycled material in the feed, so as to achieve an acceptable level of material properties with maximum recycled waste.

The promise of recycling to reduce waste and therefore the cost incurred by desktop systems in terms of raw materials can be met if makers are willing to accept some modest loss of part strength. For likes of MakerBot Innovation Centers, where students often rely on 3D printers for just prototyping purposes, and do not require high material strength and toughness, the research shows that recycling is feasible. Additionally, as is shown in Figure 10, the visible print quality of the specimens changed little with recycling stage or virgin material content. As visible print quality is often the primary concern for the parts over part strength, this shows that using recycled content is a highly viable option.

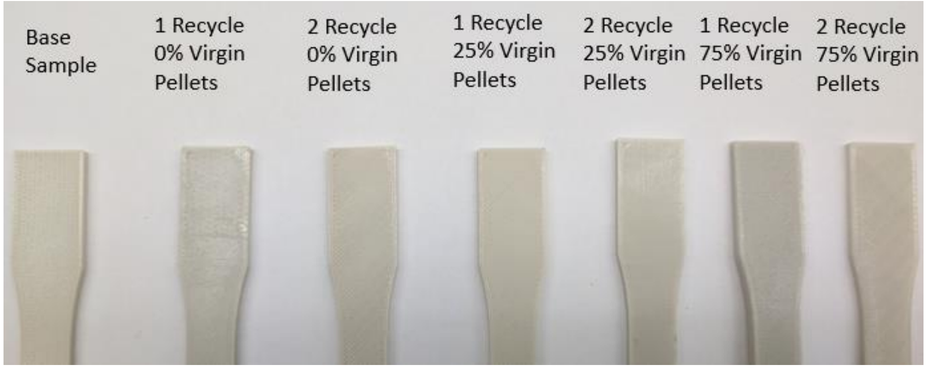


Figure 10: Printed samples from testing to demonstrate relative consistency of build quality

5. CONCLUSIONS AND FUTURE WORK

The goal of this research was to investigate the impact of material recycling of PLA 3D printed parts on their tensile strength values. The specimens were prepared with multiple recycling stages and different ratios of virgin pellets. It was observed that:

- Multiple heat cycles significantly reduce material strength, maximum strain, and toughness properties.
- The amount of virgin pellets added can increase the tensile strength of the filament under controlled ratio and at the first recycling stage.
- The ratio of added virgin pellets with recycled filament is crucial in impacting filament properties and requires further investigation

More control on the feed production is required to find the factors influencing the filament quality. For future work, higher sample sizes will be taken to enable a more a robust analysis of the data. Additionally, the impact of size, shape, and unevenness of the pelletized feedstock will be studied to find the reason of inconsistencies observed in this research. Finally, the impact of heat cycles of the filament on the 3D printer performance will be observed to confirm the feasibility of recycling processes as more and more recycling cycles are added.

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