Comparative property analysis of fused filament fabrication PLA using fresh and recycled feedstocks

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Abstract

Manufacturing industries generate much waste, which can be converted into raw materials for other allied industries. Thermoplastic wastes can be recycled in many cases. Recently these recycled plastics have been used to manufacture the input filament necessary for material extrusion additive manufacturing (MEAM). Polylactic acid (PLA) is one of the most common and potentially useful engineering plastics that can be additively manufactured, but the effect of recycling on the effective properties is still not well known. In this study, the mechanical properties of MEAM-processed PLA were compared for virgin and recycled feedstock. The collected data were analyzed using a full-factorial design of experiments method, providing insight into the effects of layer thickness and infill density on the performance of the material before and after recycling. The results showed a notable degradation in properties after recycling, but this effect was moderated by modifying the studied parameters. This work and its conclusions will serve as a screening study to guide future efforts in this area and promote the wider use of recycled materials in additive manufacturing.

List of symbols and abbreviations

| S.no. | Short form | Full form |
|-------|------------|-----------|
| 1     | PLA        | Polylactic acid |
| 2     | rPLA       | Recycled Polylactic acid |
| 3     | mm         | Millimeter |
| 4     | ISO        | International Organization for Standardization |
| 5     | KN         | Kilo Newton |
| 6     | MPa        | Mega Pascal |
| 7     | UTM        | Universal Testing Machine |

1. Introduction

The rapid advancement of technology and growing industries has resulted in many advancements in the human standard of living throughout the world; however, this has also fed the growing problem of industrial waste. Many sources of this waste exist, from manufacturing industries, electrical device production, construction, and other sectors, while treatments and sustainability efforts are often lagging. Throughout the world, more than
80% of this waste is dumped or stored instead of recycled, increasing pollution levels and threatening ecosystems and human health. A very large percentage of this waste are various thermoplastics [Hapuwatte et al 2016, Marks et al 2020, Neo et al 2021]. This is one of the most important and urgent thrusts toward more sustainable manufacturing and production methods to minimize the negative impact of production while maintaining and increasing technological innovation and advancement [Jambeck et al 2017, Mohammed et al 2018, Ghimire et al 2021, Lee et al 2021, Taghavi et al 2021]. This issue is becoming even more urgent due to the growing scarcity of resources in the past few years because of natural resource depletion, rapid lifestyle changes, growing demand, and growing socioeconomic problems throughout the world. The development and refinement of 3D printing processes in recent years offers much toward more sustainable production, as it is a fundamentally low-waste manufacturing approach also easily handles recycled materials, unlike many traditional thermoplastic manufacturing processes [Gebler et al 2014, Neumüller et al 2014, Faludi et al 2015, Al-Meslemi et al 2018, Ntousia & Fudos 2019, Shahrubudin et al 2019, Hossain & Liao 2020, Khalid & Peng 2021, Ruckstuhl et al 2020, Dharnidharka et al 2021, Chadha et al 2022]. Many materials which would otherwise not be cost-effective to recycle can be easily used with these processes in the form of plastic powder (using a basic chopper or grinder) and extruded filament [McAlister & Wood 2014, Ford & Despeisse 2016, Lundbäck, Lindgren 2017, Yang et al 2017, Colorado et al 2020, Shuaib et al 2021].

The most commonly used (and by far lowest cost) 3D printing technology is based on material extrusion and is often known as fused filament fabrication (FFF) (or, alternatively, fused deposition modeling (FDM)). For this process, waste thermoplastic materials can be chopped up and extruded as filaments and used directly. In many cases, especially if the resulting part does not need to be made from a specific material, the waste plastics can be mixed, reducing the cost and labor involved in recycling the waste. Additives such as dyes, powder, chopped fibers, and other things can also easily be added, increasing the usefulness or value of recycled plastic. One of the most useful and promising materials commonly processed using FFF is polylactic acid (PLA), a biodegradable thermoplastic with excellent mechanical properties made from biomass such as corn and sugar cane. However, PLA does have one major disadvantage: It has been observed to degrade quickly for some applications [Nampoothiri et al 2010, Elsawy et al 2017, Kakanuru & Pochiraju 2020, Kumar and Kumar 2021]. The question of how much and in what ways it degrades is still open and how severe the effect is is not known. If it can be successfully recycled with little or no degradation in the needed properties, it would expand its usefulness as a standard industrial engineering plastic that can be used within an effective circular production economy [Jambeck et al 2018, Burkhard & Aurich 2015, Freitas et al 2016, Tang et al 2016, Liu et al 2016, Jiang et al 2019, Niaki et al 2019, Wang et al 2020, Pasricha & Greeninger 2018] (figure 1).

There is little previous work in the literature on the comparative properties of PLA and PLA-based polymer matrix composites (PMCs), which focuses on basic characterization and not on studying the influence of parameters. Lanzotti et al 2019 found that the bending strength of short beams (under 3-point loading) decreased based on the number of times the PLA was recycled. The printing parameters were not varied in that study. Completing a conceptually similar study using a traditional extrusion process, [Peinado et al 2015] asked how the recycled PLA maintained performance and stability over 20 recycling runs. While this study does not use AM, its results are certainly relevant to the questions of the present study. Anderson [Anderson 2017] tested the tensile and shear strength of PLA before and after recycling, with no modification of the printing parameters or infill pattern. The results showed that the tensile strength reduced slightly after recycling but the shear strength tended to increase slightly. Unlike the study by Lanzotti, the change in performance was small after recycling, showing that the recycled PLA material was still suitable for engineering applications. Finally, [Sasse et al 2022] looked at the tensile performance of FFF filament created by mixing virgin and recycled PLA, varying from 0% to 60% recycled material. It was observed that the case with 48% recycled PLA behaved the worst and had the worst surface finish, while 60% was far better. The results suggested an optimal mixture percentage when mixing the virgin and recycled materials together.

In order to expand the use of recycled PLA, the present study takes the lessons of the works by Lanzotti et al, Peinado et al, Anderson, and Sasse et al and expands upon them by varying the layer thickness and infill density. None of the previously referenced studies looked at the effects of the printing parameters nor used less-than-thoroughly-dense experimental samples. This work serves as a screening study to help direct future research directions and collect some initial data on the material properties. In addition to studying previously unreported printing combinations, this study presents a detailed statistical analysis of the results to explore interactions and effects of the input parameters. This analysis provided details about the materials and parameters and information about how stable these effects are after PLA is recycled. This article is divided into several sections, beginning with a detailed overview of the experimental setup in section 2. This is followed by a presentation of the results in section 3 and an analysis and discussion in section 4. Finally, section 5 provides conclusions and needed future work directions for this problem.
2. Materials and methods

2.1. Goals and research questions
The major goal of this screening study was to expand knowledge about the effects of using virgin versus recycled PLA while varying the layer height and infill density. This work will help to guide and direct future, larger-scale studies on the usefulness of recycled PLA. To this end, three research questions were formulated:

1. What is the effect of recycling on the PLA properties relative to the layer thickness and infill density for tensile loading?
2. What is the effect of recycling on the PLA properties relative to the layer thickness and infill density for flexure loading?
3. Do the effects of the layer thickness and infill density on the tensile and flexure properties change or remain constant after recycling the PLA material?

2.2. Parameters and responses
The important parameters in this study were the feedstock choice (recycled versus virgin PLA), the layer thickness used, and the infill density of the samples. The responses for the tensile and flexure tests were the ultimate tensile strength (MPa) and flexure strength (MPa), respectively. The tests were done according to ISO 178 and ISO 527 standards. Table I shows the factor levels for each run of the experiment.

2.3. Material selection and sourcing
The material was taken from two sources. The virgin PLA was obtained from Creality (the manufacturer of the FFF machine used in this study) as their standard PLA filament. The recycled PLA is made by Relow from PLA food containers collected in the Benelux region of Europe (Netherlands, Belgium, and Luxembourg). They were recycled one time into the filament and originally manufactured as flat sheets processed via thermoforming. From the manufacturer’s information, the drying is 50 °C in a hot air or vacuum oven for 5 to 8 h. The filament was used as it came from the factory and was sealed up until use. Both the virgin PLA and the recycled PLA are made from corn starch and both are specified to be engineering-grade by the manufacturer. The virgin PLA was manufactured from corn starch by the Creality 3D in Shenzhen, China.
2.4. Experimental design

This study was done using a designed experiment based on a full factorial design with two factors of three levels each, providing $3^2 = 9$-factor combinations. This design was used for four different experiments based on the PLA status (virgin versus recycled) and the test completed. These combinations are shown in table 2, while table 3 shows the factorial array used for each of them. A total of 36 samples were tested during this screening study. Each run of the experiments was done once; adjustments of the factors and replications are planned as part of future work on this problem.

2.5. Sample design and fabrication

The sample geometry was taken from the ISO 178 and ISO 527 standards. Figure 2(a) shows the dimensions of the flexure testing samples, while figure 2(b) gives the dimensions for the tensile testing samples. Representative printed samples are shown in figure 2(e). The CAD models used to model the samples were created using Autodesk Fusion 360 and preprocessed using Ultimaker Cura. Given that this is a screening study and not a full characterization of the materials, only the flat printing orientation was used with a line-based infill pattern. The printing temperature was 200 °C, the print bed temperature was 60 °C, and the printing speed was set at 100 mm s$^{-1}$ using a Creality CR-10 V2 FFF machine. The temperature and humidity during the printing and tests were noted as 25 °C and 60%, respectively.

2.6. Testing fixture design and setup

Mechanical testing was done using a universal testing machine (figures 2(c) and (d)) made by Instron with a capacity of 100 KN. Testing speed was 1 mm min$^{-1}$. A total of 36 samples were tested in different configurations, as shown in table 2.

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Table 1. Factor and levels.

| Factors               | Levels |
|-----------------------|--------|
|                       | −1     | 0  | +1 |
| A: Layer Thickness (mm) | 0.1    | 0.2| 0.3 |
| B: Infill Density (%)  | 5      | 50 | 100 |

Table 2. Experimental runs and samples.

| Experiment | Test  | Material     | Samples |
|------------|-------|--------------|---------|
| 1          | Tensile | Virgin PLA  | 9       |
| 2          | Tensile | Recycled PLA| 9       |
| 3          | Flexure | Virgin PLA  | 9       |
| 4          | Flexure | Recycled PLA| 9       |

Table 3. $3^2$ full factorial design.

| Runs | Parameters |
|------|------------|
|      | A: Layer Thickness | B: Infill Density |
| 1    | −1          | −1        |
| 2    | −1          | 0         |
| 3    | −1          | +1        |
| 4    | 0           | −1        |
| 5    | 0           | 0         |
| 6    | 0           | +1        |
| 7    | +1          | −1        |
| 8    | +1          | 0         |
| 9    | +1          | +1        |
3. Experimental results

The experimental results are shown in table 4 (virgin PLA) and table 5 (recycled PLA) below and in figure 3.

4. Analysis and discussion

This article describes a screening study completed to study the effects of layer thickness and infill density and observe if the factor significance changes once the PLA material is recycled. To this end, a statistical analysis was done on the collected dataset. Analysis of Variance (ANOVA) was done for each experiment combination, with the Anderson-Darling statistic checked for each case to ensure that the ANOVA conclusions were valid. The results of the analyses are shown in table 6.

At a typical level of significance, $\alpha = 0.05$, the layer thickness was not found to be a significant factor for any of the cases. Infill percentage was a significant factor for tensile tests and for the virgin PLA flexure tests, but was not significant for the recycled PLA. This indicates that the recycled PLA was significantly more brittle than the virgin material, as the material’s lower infill and recycled nature have equivalent effects on the flexure properties. To further study this, the main effects plots and interactions plots for each case were created and are shown in figure 4.
Analysis of the main effects and interactions (figure 4) shows the conclusions as the ANOVA, but provides additional detail. It was noted that for the standard PLA, the 0.2 mm layer thickness had the highest tensile and flexure strength, which is consistent with other studies in the literature. However, the recycled PLA showed a linear relationship between the layers, as expected from the basic mechanical analysis. For the virgin and recycled PLA, there was a linear relationship between the tensile strength and the infill percentage, while for flexure strength, the 100% dense samples had significantly different properties. These properties were consistent between virgin and recycled PLA. Based on this analysis, while the raw properties of the PLA degraded after recycling, the relationship and influence of factors remain mostly consistent. This is a useful finding, showing that recycled PLA can be used with the same design conditions and assumptions as the newly produced PLA.

5. Conclusions and future work directions

This short screening study examined the difference in tensile and flexure properties between virgin and recycled PLA. It was concluded that the PLA properties degrade upon recycling (as seen in the reviewed studies discussed

### Table 5. Experimental results (tensile and flexure) for recycled PLA.

| Runs | Layer thickness (mm) | Infill percentage (%) | Ultimate tensile strength (MPa) | Flexural strength (MPa) |
|------|-----------------------|-----------------------|---------------------------------|------------------------|
| 1    | 0.1                   | 5                     | 14                              | 76                     |
| 2    | 0.1                   | 50                    | 29                              | 60                     |
| 3    | 0.1                   | 100                   | 46                              | 76                     |
| 4    | 0.2                   | 5                     | 16                              | 51                     |
| 5    | 0.2                   | 50                    | 27                              | 58                     |
| 6    | 0.2                   | 100                   | 37                              | 71                     |
| 7    | 0.3                   | 5                     | 19                              | 46                     |
| 8    | 0.3                   | 50                    | 27                              | 54                     |
| 9    | 0.3                   | 100                   | 27                              | 54                     |

### Table 6. Results of statistical analysis.

| Experiment Combination | AD Statistic | P-Values | AD | Layer T | Infill % |
|------------------------|--------------|----------|----|---------|----------|
| Virgin PLA Tensile     | 0.172        | 0.898    | 0.362 | 0.005   |
| Virgin PLA Flexure     | 0.163        | 0.941    | 0.269 | 0.028   |
| Recycled PLA Tensile   | 0.214        | 0.785    | 0.610 | 0.039   |
| Recycled PLA Flexure   | 0.193        | 0.850    | 0.089 | 0.324   |
in section 1) but the relationship between the properties remains mostly consistent. Therefore, the information and lessons from this project can be used to drive and guide further, more comprehensive studies on this problem. At the current time, recycling PLA is usually more expensive than buying it new, but this should improve over time as the material becomes more widely used as an engineering plastic. More development on this question will help to improve acceptance of PLA and aid in efficiently recycling it, helping to improve the sustainability of products made from thermoplastics by better integrating them into a circular production economy. Future work in this area should focus on factors such as infill pattern, sample size (to study size effects), and recycling methods. This short paper provides a starting point for this and some useful information about the influence of factors and test types to aid this further work.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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A Dash et al

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