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Use of Recycled Poly Lactic Acid (PLA) Polymer in 3D Printing: A Review

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Abstract - *Plastics and polymers play a major role as filament materials in Industrial as well as open-source additive manufacturing, and thus with the increasing growth of AM, the demand for polymers has increased exponentially. Studies have shown that the use of plastic has increased to over 300 million metric tons in the recent years, however in the U.S., about 75 percent plastic waste end up in landfills and only 10% is recycled. The coupling of additive manufacturing with polymer recycling is an effective way to add value to waste, which used to be repudiated. This research article provides an overview on why and how Poly Lactic Acid (PLA), a biodegradable thermoplastic material can be recycled to manufacture 3D printed components. A literature survey of comparison of properties of recycled PLA with virgin PLA is carried out, and solutions are discussed with respect to variation in properties. Combination of virgin PLA with recycled PLA, as well as coating of polydopamine on recycled PLA pellets to improve its mechanical features have also been discussed.*

***Key Words*: Poly Lactic Acid (PLA), 3D Printing, closed-loop recycling, polydopamine, biodegradable, plastic**

1. INTRODUCTION

The additive manufacturing sector is an exponentially growing industry. Additive manufacturing generally involves producing components from raw materials bearing similar geometry to its 3D CAD design. The raw materials may be present as filament, powder or solid blocks. Polymers, due to their favorable mechanical properties (high strength-to-weight ratio, stiffness, ductility and durability), are widely used as 3D printing filament materials. The ever increasing use of plastics and polymers has also seen an increase of waste generation throughout the world. However, several companies and associations throughout the world have taken initiative to solve this problem, of which the most exciting and exemplary idea is the use of recycled plastics as filament material for the purpose of additive manufacturing. Although the production of plastics has increased to around 300 million metric tons, a very small fraction of it is actually recycled. Even in a fully developed country such as the U.S., about 75% of waste plastics end up in landfills, and only 10% is actually recycled. Scientists have estimated that around 5.25 trillion of plastic products float around the seas of the world. The durability of polymers becomes an issue when waste reduction is considered, as plastics take over 100 years to decompose. Thus, it becomes ideal to efficiently try to minimize the production of new plastics while simultaneously cater to the needs of additive manufacturing. Plastic materials such as Poly Lactic Acid (PLA) can be recycled to produce 3D printing filament. PLA being biodegradable can be recycled to produce 3D Printing filament. The following flow sheet can summarize the general process of PLA recycling to produce filament material.

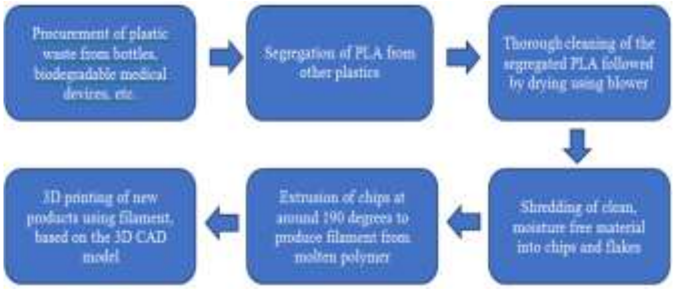

Fig -1: Process of producing recycled PLA filament

The role of Additive Manufacturing in reducing plastic pollution in the world is a huge one. Products which have been produced from recycled materials give a tangible example of how we can reduce plastic pollution in the world. It is estimated that 86% of the ocean debris is plastic, creating a toxic environment for the marine animals, that could get hurt by them or even eat them, causing plastics to enter the food chain. It's been found that aquatic species farmed for consumption contains micro-plastics. The potential implications of this are worrying. Our responsibility extends beyond the production line, and to encourage sustainable development, we have to understand the importance of recycled products with regard to our ecosystem. Additive Manufacturing plays a huge role in resolving this issue. As usage of recycled polymer in 3D printing continues to grow, perhaps this could be a way to change people's outlook on the value of what was previously considered waste to be discarded.

Thus, the use of recycled polymer as filament material in additive manufacturing helps in promoting the 3 R's: Reuse, reduce and recycle. In the following sections of the paper, the various plastics that can be used for recycling will be discussed and comparison between them is carried out.

2. CHARACTERISTICS OF POLY LACTIC ACID

Poly lactic Acid (PLA) is a thermoplastic. It is different than most other thermoplastic as it is derived from renewable resources like corn starch or sugar cane. Most plastics, by contrast, are derived as a result of distillation and polymerization of nonrenewable petroleum reserves. PLA is a form of "bio-plastic", a term given for plastics that are derived from biomass. Poly lactic Acid is biodegradable and has characteristics similar to polypropylene (PP), polyethylene (PE), or polystyrene (PS). It can be produced from already existing manufacturing equipment. This makes it relatively cost efficient to produce. The chemistry of PLA involves the processing and polymerization of lactic acid monomer. Lactic acid HOCH3CHCOOH exists as 2 enatiomers: L-lactic and D-lactic acid.

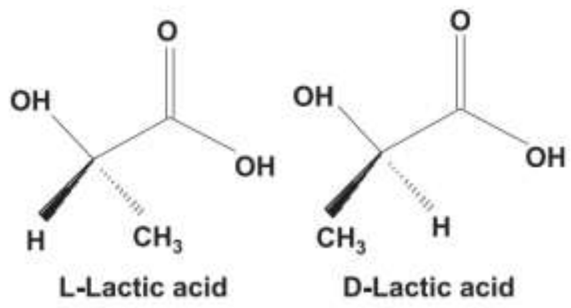

Fig -2: Optical isomers of lactic acid [1]

Poly lactic acid has several unique features: it is biodegradable, can be easily processed compared to other thermoplastics and is eco-friendly [1]. This makes it a viable option as raw material for potential applications such as commodity plastics, as in packaging, agricultural products and disposable materials. On the other hand, the polymer also has medical applications in sutures, pharmaceutical products, implants and tissue engineering.

3. CLOSED LOOP RECYCLING OF POLY LACTIC ACID

Poly Lactic acid has a specific density of 1.24 g/cm^3 and is frequently applied as raw material of 3D printing filament. A study was carried out that made used of a closed loop recycling process to evaluate the potential of recycled PLA from perspectives of material properties and environmental performance [2].

Initially, drying was carried out at 80°C to remove moisture content and filament was made using extruder. Dumbbell-shaped specimens were produced using an FDM 3D printer. Mechanical properties like tensile modulus, tensile strength, yield strength, flexural modulus and Flexural strength were analyzed. Thermal properties such as Glass transition temperature (T_g), crystallization temperature (T_c) and melting temperature (T_m) were obtained.

Following 3D printing and property characterization, apart from some samples, the rest were shredded by plastic mill to be dried again, hence forming a loop. This cycle was repeated twice as the reprocessed material could no longer be further processed. The results were tabulated for the same.

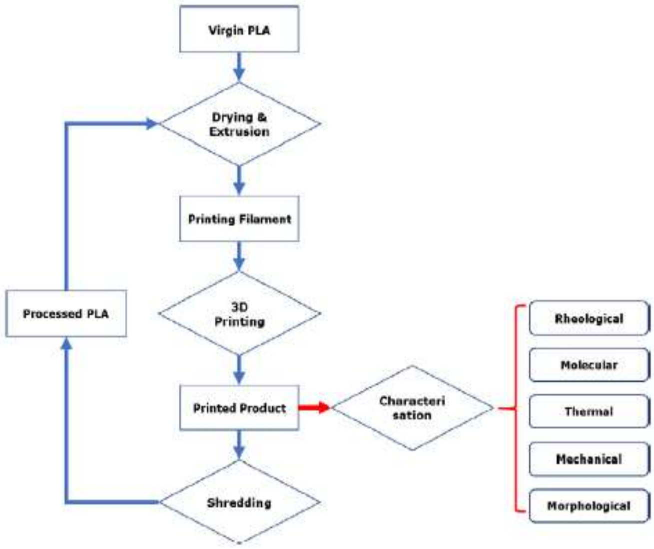

Fig -3: Scheme of close-looped recycling of PLA that used in 3D printing [2]

The mechanical, i.e. tensile and flexural properties exhibited very little deterioration after one cycle, in which two extrusion processes and one 3D printing process of FDM took place. The variations in properties are shown below.

Table -1: Variation in mechanical properties of PLA after closed loop recycling [2]

Property	Virgin PLA	After recycling once
Tensile Modulus (MPa)	1572.43±27.16	1566.54±45.61
Tensile strength (MPa)	30.21±0.89	29.47±1.21
Yield Strength (MPa)	27.69±0.77	27.65±1.09
Flexural modulus (MPa)	2423.73±56.42	2234.70±70.68
Flexural strength (MPa)	64.48±2.49	57.55±1.97

PLA becomes stiffer and more brittle with increase in recycling cycles.

The variations in thermal properties were:

Table -2: Variation in thermal properties of PLA after closed loop recycling [2]

Property	Virgin PLA	After recycling once	After recycling twice
T_g (°C)	59.86	59.47	59.11
T_c (°C)	123.69	104.34	98.32
T_m (°C)	165.64	168.39	167.55

Major reduction is observed in T_c due to the increase in mobility of the polymer chains, as the molecular weight reduces after recycling.

A similar study was carried out by Antonio Lanzotti et al. to assess the change in short-beam strength of recycled PLA [3].

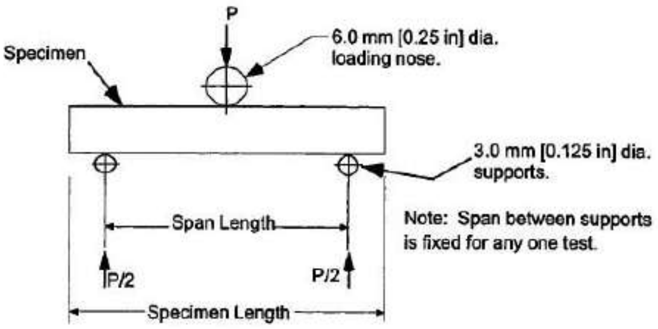

Fig -4: Short-beam test experimental setup [3]

PLA specimens were manufactured at 200°C and mechanically tested. They were then ground up and re-extruded. This was done for 3 cycles. Test specimens were loaded in three-point bending. They were center-loaded and the ends rested on two supports which allowed lateral motion. The loading nose was directly centered on the midpoint of the specimen. Short-beam strength was calculated using the formula:

$$F^{sbs} = 0.75 \times \frac{P_m}{b \times h}$$

Where, P_m was the max load measured b and h was the specimen width and thickness, respectively. The results were found to be as follows:

Table 3: Variation short beam strength of PLA after closed loop recycling [3]

Type of filament	**Short-beam strength F^{sbs} (MPa)**
Virgin	119.1±6.6
One time-recycled	106.8±9.0
Two times-recycled	108.5±9.9
Three times-recycled	75±16.2

Thus, Close-looped recycling can be a feasible solution of make used of processed PLA to produce filament for FDM 3D printing processes. Yet, the number of reprocessing cycles is limited due to significant thermo mechanical deteriorations, but it can be remediated by adding virgin PLA.

4. COMBINING VIRGIN PLA MATERIAL WITH RECYCLED PLA

4.1 Combining virgin PLA with recycled PLA pellets

Babagowda et al. carried out a study [4] that checked the characteristics of recycled PLA that was combined with virgin PLA filament, by combining virgin PLA with recycled PLA pellets. Used PLA materials are recycled and converted in to small pallets. Pallets are made by compacting the liquefied recycled plastic, which are ready for re-use. These pallets are then mixed homogeneously with virgin PLA before extracting the build filaments. Filament was extruded using a single screw extruder. A drawback of this process was the potential agglomeration of the disparately sized pellets. The percentages of recycled PLA can vary and mechanical properties vary according to the amount added. Varying the layer thickness can also affect the Ultimate Tensile Strength. For layer thickness of 0.1mm, material having 10% recycled PLA exhibited an Ultimate tensile strength of 37.829 MPa. In comparison, material having 50% recycled PLA for the same layer thickness exhibited an Ultimate tensile strength of 33.829 MPa. For the same compositions, material of 0.3 mm layer thickness exhibited Ultimate tensile strengths of 37.486 MPa and 30.27 MPa respectively. The optimized process parameters for accomplishing larger tensile strength are 0.1mm layer thickness, 10% additives in PLA. This helps us conclude that by varying the thickness and composition of recycled PLA appropriately we can form 3D printing filament.

4.1 Combining virgin PLA pellets with recycled PLA

Another study carried out by Daniel Tanney et al. was on Combination of virgin PLA pellets with recycled PLA filament [5]. Different compositions of virgin PLA pellets were mixed with recycled PLA to strengthen the recycled material. A three-stage pellet forming, extrusion, and printing process was used to study the recycling of PLA material from cast-off build material.

Fig -5: Filament pellets (left) vs. virgin pellets (right) [5]

The studies provide data that for a double recycle stage, material with 25% virgin PLA exhibited an Ultimate tensile strength of 32.69 MPa, compared to material with 75% virgin PLA which exhibited an Ultimate Tensile Strength of 32.92 MPa. This research shows that the recycling is feasible if producers are willing to accept a modest loss of part strength.

5. COATING RECYCLED PLA FILAMENT WITH POLYDOPAMINE

Dopamine readily adsorbs onto almost all kinds of surfaces and develops cohesive strength through self-polymerization; hence, aqueous solutions of dopamine can be used as adhesives. Xing Guan Zhao et al. exploited this characteristic of dopamine to produce polydopamine coated PLA pellets to reduce the degradation in the mechanical properties of recycled PLA that was initially used to fabricate parts by 3D printing [6]. A custom recycling system consisting of a shredder, an extruder, a spooler, a sensor, and a controller was used. Shredder was used to process the broken PLA parts fabricated by 3D printing. In order to maximize the PDA coating surface area, the pellets were shredded to the smallest size possible within the permissible range. Deposition of a thin adherent polymer film on PLA pellets was achieved by immersing the pellets in a dilute aqueous solution of dopamine. Recycled PLA and PDA/PLA filaments were also extruded from pellets used in the recycling system. Specimens were produced using an FDM 3D printer. These components were subjected to tensile tests. Stress-strain curves were plotted for components made purely with recycled PLA and components made with polydopamine coated recycled PLA.

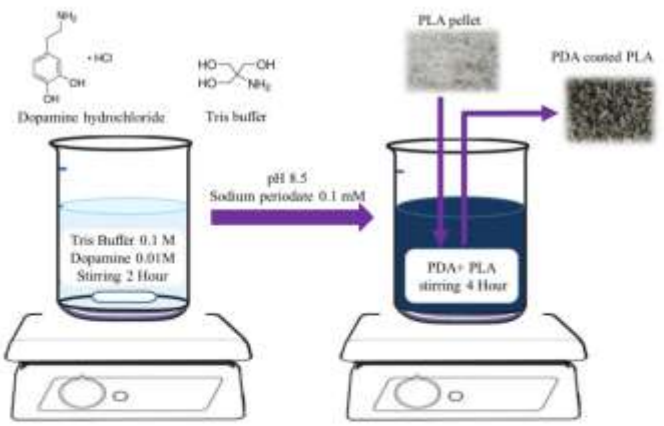

Fig -6: Way of coating PLA pellets with polydopamine [6]

Recycled PLA and PDA/PLA filaments were also extruded from pellets used in the recycling system. Specimens were produced using an FDM 3D printer. These components were subjected to tensile tests. Stress-strain curves were plotted for components made purely with recycled PLA and components made with polydopamine coated recycled PLA. Out of 5 components produced, the average Ultimate tensile stress for components made purely with recycled PLA was 46.35 N/mm^2 and Strain at break was 9.05%. For components made with polydopamine coated recycled PLA, the average Ultimate tensile stress was found to be 53.24 N/mm^2 and Strain at break was 12.79%. This shows that the presence of PDA in PLA improved the tensile strength of the fabricated specimens by enhancing the bonding strength. This finding could also be useful in injection molding applications because PDA coated plastic pellets can be used in 3D printing as well as injection molding process.

3. CONCLUSIONS

This paper provides insight on methods by which virgin PLA is mixed with recycled PLA to improve its properties. One way of doing this is by combining virgin PLA with recycled PLA pellets. A drawback of this process is the potential agglomeration of the disparately sized pellets. Various compositions of recycled PLA in virgin PLA were considered. Compositions with higher recycled PLA pellets concentration show lesser tensile strength. Another way of combination is by combination of virgin PLA pellets with recycled PLA filament. This research shows that the recycling is feasible if producers are willing to accept a modest loss of part strength.

Another way of compensating for property degradation of recycled PLA is by coating polydopamine on recycled PLA pellets. Deposition of a thin adherent polymer film on PLA pellets was achieved by immersing the pellets in a dilute aqueous solution of dopamine. It results in improved tensile strength of the fabricated specimens by enhancement of the bonding strength.

The growing Additive manufacturing industry requires a lot of plastic material. Thus, Recycled plastic like PLA provides a cheaper and more eco-friendly way to optimize future use of plastics and polymers. Moreover, as it is a bio-polymer, it has several unique features: it is biodegradable, can be easily processed compared to other thermoplastics and is eco-friendly, thus making it a good option to be recycled and reused in Additive manufacturing industry.

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