



# **Experimental Research on 3D Printing Recycled Poly- mer/Aluminum fraction of Beverage Cartons by Fused Granular Fabrication method**

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## ABSTRACT

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According to circular economy principles, material cycles of composite packages should be closed or narrowed to contribute to the EU green deal ambitions. This is achieved through the waste minimization, efficient recycling and deploying recycled materials extensively.

The circular challenge is to maximize the value creation of post-consumer beverage cartons, and at the same time narrow the material loop. However, non-fiber components of beverage carton (at least 30% of total), have been a problem for the recyclers, resulting in low recycling rate of Polymer/Aluminum (Poly-Al). The aim was to find a solution to improve circularity of Poly-Al, and also empowering recyclers by offering distributed recycling, 3D printing here to the current centralized recycling activity.

Experimental part of the study involved 3D printing tests with Poly-Al material made from recycling non-fiber fraction of beverage carton in a form of granule at lab-scale by using "Fused Granular Fabrication" (FGF) as part of the material extrusion technology. Optimal 3D printing parameters for this novel material were set up, and a tensile strength test was subsequently implemented to validate the assumptions.

As far as our knowledge goes, this study was the first to concentrate its attention on printability of Poly-Al material composed of recycled materials in large-scale niche 3D printing market by using Fused Granular Fabrication method.

This study showed that Poly-Al as a heterogeneous material mixed of various polymers, exhibited specific thermo-physical behavior during 3D printing process because of interaction between Polymer matrix and Aluminum particles. The material unexpectedly disclosed high viscosity, high co-efficient thermal expansion and low flow rate which relatively lessened the speed of processing.

To recap, in the event of speeding up the flow rate of Poly-Al material which makes sense a rapid prototyping, the material can be broadly used as a feedstock for large scale 3D printing purpose. Thus, not only will recycling rate of Poly-Al be improved, but also profitability will be yielded by manufacturing high-quality 3D printed end-products for the dedicated recyclers.

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Key words: 3D printing, beverage carton, Polymer, Aluminum, Fused Granular Fabrication (FGF), waste valorization, recycled feedstock.

## **PREFACE**

First and foremost, I would intend to appreciate Tetra Pak TIRCCA for such an exceptional opportunity that has enabled me to carry out my master thesis around this super fascinating topic. Special thanks to my supervisors, Silja Kostia and Tero Haapakoski and Jarmo Lehtonen (Mechanical laboratory's technician) for giving me academic guidance and support. I also thank my family for their endless support.

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## ABBREVIATIONS

IOT	Internet of Things
AI	Artificial Intelligence
M2M	Machine-to-Machine
MFI	Material Flow Index
RPM	Round per Minute
PMC	Polymer Matrix Composite
LDPE	Low Density Polyethylene
HDPE	High Density Polyethylene
FFF	Fused Filament Fabrication
FGF	Fused Granular Fabrication
LSAM	Large Scale Additive Manufacturing
PETG	Polyethylene Terephthalate Glycol
MaaS	Manufacturing as a Service
3DP	Three-Dimensional Printing
CAD	Computer-Aided Design
CTE	Co-efficient thermal Expansion
AC	Aluminum Can
rPP	Recycled Polypropylene
CF	Carbon Fiber
PCW	Post-Consumer Waste
PME	Pellet Material Extrusion
STL	Standard Triangle Language
TMA	Thermo-Mechanical Analysis
GHG	Green-House Gas
BF	Basalt Fiber
GF	Glass Fiber
AF	Aluminum Foil
AD	Aluminum Dross
PA	Polyamide
OS	Open Source

## 1 INTRODUCTION

The immoderate use of “synthetic materials” particularly materials which are often arduous to dispose of, has recently raised environmental concerns. Thus, the world’s attention has immensely been focused on “Circular Economy” (CE) concept and “sustainable materials” based on the recycling routs (Fico et al. 2022).

Beverage cartons are predominantly made from long, strong paper fibers (70% on average) that can be recycled several times. Additionally, the thin layer of polymer (25%) protects the product from moisture, and also Aluminum foil (5%) preserves the product from oxygen and light. In this respect, non-fiber fraction can be recycled through chemical and mechanical recycling methods. However, according to the annual reported data, recycling rate of Poly-Al is proportionately lower than expectations across the markets. To fill the gap in lower recycling rate, recyclability of non-fiber fraction (Polymer/Aluminum (Poly-Al<sup>1</sup>)) of beverage carton, can be viewed as a main challenge and a promising opportunity for emerging circular solutions simultaneously.

According to the observations in the market, the majority of the recyclers favour Aluminum-free cartons. The reason behind this is that the actual monetary value of Poly-Al material has still been remained undiscovered for them so far. So, Poly-Al waste should be seen as an asset in order to shape new ways of serving end-products market, whereby new flow of revenue will be obtained through circular initiatives.

The Alliance for Beverage Cartons have recently confirmed that new technologies are emerging in order to revolutionize recycling residual of polymer and Aluminum (Poly-Al) fraction remaining after fiber recovery process. The aim of this study was testing Poly-Al material for large scale direct extrusion from granule to 3D printed part to fully capture environmental and economic potential of recycled feedstock.

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<sup>1</sup> Wherever Poly-Al is used in this study, it implies non-fiber fraction of a beverage carton composed of Polymer and Aluminium.

## 1.1 Tetra Pak as a case company

Tetra Pak TIRCCA is a subsidiary of Tetra Pak global; it's considered as a key player in the food and beverage packaging industry. Aligned with Tetra Pak sustainability strategy, Tetra Pak TIRCCA also commits to contribute to the collection and recycling objectives. According to Tetra Pak sustainability report, "actual global recycling was 26% in 2021." On the flip side, "74% of beverage cartons are ended into disposal, landfill incineration. As an industry, European Union hopes to reach a 90% collection rate and a 70% effective recycling rate by 2030." (Tetra Pak Sustainability Report 2022).

In recent years, Tetra Pak TIRCCA has scaled up the recycling capacity including paper and Poly-Al materials through recycling partners. To unlock the potential of Poly-Al as a recyclable material, deploying industry 4.0 technology can empower the recyclers in creating a profitable, innovative circular business model. It exceptionally results in a sustainable competitiveness, do more with less for longer in the market.

In this regard, Tetra Pak Italiana signed a contract with Ethica to commercialize filament from granule made from recycling the polymer and Aluminum fraction of beverage cartons in 2020, and filament "AlfaPak" has been recently launched by FiloAlfa company. This 3 years collaboration has had some sustainability targets including strengthening the message of Poly-Al waste as a valuable material, producing filament as a bridgehead for new application of Poly-Al waste in the future.

In addition, Tetra Pak and Aectual (design brand based in Amsterdam) collaborated and developed a series of 3D printed unique interior objects entirely composed of Poly-Al material in 2022. This joint-project was part of workplace experience strategy to furnish Tetra Pak offices with these innovative objects. According to the statement of Actual co-founder, in contrast to their thought, Poly-Al is not a viable base material and Aluminum particles in the granule makes it difficult to work with. However, as he noted, "Poly-Al is a most impressive material, it's a bit clay-like and looks like concrete. But, it's really versatile. And the tiny chips of Aluminum give it depth and allow the color to mix well into the material. It has a

tactile finish” (Tetra Pak 2022). Therefore, prior to this study, transforming pallet to filament for large scale 3D printing purpose had been accomplished previously.

## **1.2 Research structure and limitations**

The contribution and primary focus of this experimental study was based on the following four research questions:

- How can waste minimization of beverage carton benefit from the implementation of 3D printing?
- Which methods of material extrusion is well-matched with Poly-Al material properties and corporation’s purpose?
- What are challenges in printability of Poly-Al material?
- What value propositions can be fully captured through Poly-Al material?

Finally, recommendations for material properties enhancement which could offer high quality of 3D printing, were presented.

The structure of the report is as follows. After introduction and theoretical framework in the form of literature review, the beverage carton structure and the common recycling method is introduced in chapter 3. The PolyAl material properties, and extrusion technology methods are described in chapter 4. Chapter 5 includes the experimental part namely tests on material by using selected 3D printing method. Chapters 6 concludes the findings of the study by answering the research questions, suggesting recommendations to improve material properties and performance of 3D printing.

Prior to this study, there were two limitations that should be taken into account. The material was composed of various polymers from heterogeneous waste streams, which was assumed that it made an uncertainty. Another limitation was “Fused Filament Fabrication” (FFF) is still the most long-established technique related to recycled feedstock. Most conducted scientific researches in recent years had been concentrated on FFF printing, and most popular materials e.g., Polylactic Acid (PLA), Polyethylene Terephthalate Glycol (PETG). Hence, a few relevant sources for the case study were available.

## **2. LITERATURE REVIEW**

### **2.1 A Global picture of solid waste generation**

The world yearly generates 2.01 billion tonnes of municipal solid waste and at least 33 percent of that waste is not managed in a way that is responsible towards the environment. Most waste is currently dumped or disposed of in some type of landfill around the world. It is projected that worldwide waste will reach 3.40 billion tonnes by 2050. Based on the volume of waste generated, its composition and how it is managed, it is estimated that 1.6 billion tonnes of carbon dioxide (CO<sub>2</sub>) equivalent greenhouse gas emissions were generated from solid waste treatment and disposal in 2016, mostly as a result of open dumping and disposal in landfills without landfill gas capture system. This accounts for around 5% of global emissions. Waste-related emissions are predicted to exceed 2.6 billion tonnes of CO<sub>2</sub> equivalent per year by 2050 (Kaza et al. 2018).

From the resource efficiency standpoint, the current global economy utilizes 100 billion tonnes of materials each year, with secondary materials accounting for a percentage of total consumption. The global economy is now just 7.2% circular (secondary materials). Owing to surge of material extraction and usage, it's getting worse year after year. This means that the worldwide waste is relatively predicted to be 35.26 Gt (Fraser et al. 2023).

It is crystal clear that the waste management is ideally positioned to recover materials from the technical cycle. The existing linear economic system urgently requires a transition to a more circular system and sustainable in which resources and goods are recovered and sustainably retained within the cycle as long as possible. "Circular Economy" is a manufacturing practice for waste management that allows for the reprocessing, remanufacturing and recycling of materials in the producing process. In comparison to landfilling and incineration, recycling not only eliminates waste pollution and harmful materials in the environment, but it may also fabricate advanced goods with fine physical and mechanical features (Jamnongkan et al. 2022).

By embracing circular economy, we can meet the people's needs using only 70% of the materials that we currently consume while safely remaining within the planet's boundaries. It's vital that we modify our relationship with materials in order to maximize advantages for business or people while putting less strain on the planet's life support system. Overconsumption, waste and prosperity are the true drivers of global material demand. A circular economy is a critical solution to address total societal collapse: increases in secondary material utilization beyond recycling, must be matched by a systemic approach. Smart material management enables substituting with regenerative managed materials, doing more with less, using for longer period of time (Fraser et al. 2023).

### **2.1.1 Packaging waste**

Packaging performs wide range of indispensable tasks, ranging from storage, protection and transportation functions to facets such as ease of use and the provision of commodity information. These services significantly contribute to sustainability. Because without packaging, food losses can occur and sensitive products can be damaged. To prevent negative public reputation, packaging as a consumer good can assist in a "sustainable economy". Furthermore, issues such as the generation of emissions, littering and the consumption of resources are being highlighted. The circularity level of packaging, in particular, is becoming even more urgent, as part of the EU circular economy package, is demanding a reduction in the resource use, and the use of recycled material as a "secondary raw material" as well as strikingly pushing higher material recycling quotas (World Packaging Organization 2020).

Looking at relevant regulations, the "Packaging and Packaging Waste Directive (PPWD)" is an EU directive that was first introduced in 1994. It sets recycling targets for packaging placed on the EU market, and for different packaging materials across the EU. The PPWD has been revised several times. The goal of revision is the lowering the amount of packaging waste and ensure that all packaging is recyclable by 2023. The EU is concerned because the amount of packaging waste has been increasing. There was a 9% growth in packaging waste in the EU between 2007-2017. All packaging shall be recyclable, meaning that it will need to be designed for recycling, sorted, collected and recycled at scale, 75%

of the EU population. By extension, packaging producers should concentrate on achieving high recycling rates for packaging which is more difficult to recycle (EU-ROOPEN 2016).

Furthermore, the European commission published a proposal in 2022 that is currently being reviewed in the European parliament. The directive will most likely be turned into a regulation, so it will become the Packaging and Packaging Waste Regulation (PPWR). According to Decision (EU) 2019/665, composite packaging should be reported per material contained in the packaging e.g., Polyethylene and Poly-Al, with a possible derogation for materials that constitute less than 5% of the total mass of the packaging unit. The new measurement guidelines aim to ensure that only waste that has achieved end of life status is used to calculate recycling target.

Spaltini et al. (2021) described that in general, throughout the product's value chain e.g., beverage carton, diverting the attention on the manufacturing processes that once the waste is accumulated to be transformed into new-born products, and be reacquainted into the loop once more. In this respect, recycling beverage cartons is not a complex process. Firstly, the fibers are separated from the polymers and aluminum. The fibers are recycled and utilized to make new paper products, while the residual aluminum and polymers can be used across a variety of applications.

Today, the highest share of recovered Poly-Al mix is used for energy recovery. However, there are more innovative projects such as our study in place to enable a progressive increase in Poly-Al recycling. In 2020, in relevant to the mentioned brand 7% of Poly-Al recovered from collected and recycled beverage cartons respectively. To put a well-operating internal market in place for secondary raw materials, "secondary raw materials" must be competitive enough in comparison with primary raw materials e.g., performance, availability and cost. According to Circular Economy Action Plan 2020, recent import restrictions on waste, have mobilized the recycling industry placed in each country to increase its capacity and squeeze out as much value as possible from the materials.

## 2.2 Industry 4.0

The industry 4.0 (14.0) revolution refers to the continuous and incremental automation of traditional manufacturing operations. The German government coined the phrase “industry 4.0” in 2011 as part of the country’s high-tech scheme to boost industrial potential through digitally controlled manufacturing. 14.0 contributes in the efficient management of sophisticated systems by assimilating the application of large-scale machine-to-machine (M2M), Robot, Additive Manufacturing (AM), Artificial Intelligence (AI), Big data, Internet of Things (IoT) digital platform, autonomous Drone, digital twins, cloud computing, automation and monitoring without the need for human intervention. 14.0 presents industrialization paradigm in which machines learning algorithms, humans and products interface with one another through both cybernetic and physical tools, potentially boosting performance and sustainability (Kagermann et al. 2013).

Javaid et al. (2021) presented that manufacturing processes and supply chain activities are improved by seamless integration of information technology systems in industry 4.0. For instance, a designer can identify the underlying cause of an issue while designing and developing a commodity. The manufacturing sector can improve productivity and minimize internal waste process. The scheme modifications are made rapidly, and the production team can fabricate appropriate components. By digitizing the manufacturing process, these revolutionary technologies support sustainable production.

Moreover, the industrial revolution “4.0” has a potential to improve effectiveness of “solid waste management” through machinery and digital applications, beneficially eliminating, recovering and repurposing waste. Several processes in solid waste management are reviewed as well as the use of technologies that assist sustainable development in terms of circular economy, such as AI, ML, IoT, system integration, cyber-physical systems and smart automation. These stages include waste sorting, composting, landfilling, energy conversion and recycling which convert recyclable solid wastes into valuable products (Cheah et al. 2022).

The perception is that this decade will be characterized by mainstream technological breakthroughs, will be resulting in fundamental transformation of waste

industry (Figure1). Decontamination processes in recycling industry will be transformed from low-cost material recovery solutions to specialized processes for higher value secondary materials. Mechanical sorting and recycling advancements will create new chances to better utilize resources by incorporating additional waste streams into creating a resource for new products and the recycling industry (Solid Waste Association 2021).



FIGURE 1. Waste management steps and the impact of Industry 4.0 (Solid Waste Association 2021)

In brief, when it comes to the environmental component of sustainable development, “industry 4.0 technologies” result in a drop in waste generation by streamlining productive recycling, production and remanufacturing activities (Javaid et al. 2021).

### 2.2.1 Additive manufacturing

Today, loads of materials and energy are required for modern manufacturing, resulting in gigantic waste and emissions. Sanchez et al. (2017) stated that Additive Manufacturing (AM) refers to the process of fabricating items from prototypes to finished commodity deploying methods that “deposit material” layer after layer. When compared to traditional manufacturing process, additive manufacturing technologies have numerous environmental benefits. Above and beyond all

considerations, there is reduced waste of raw materials, smart and innovative materials are used.

“Ecological efficiency” is a pillar of sustainability, and any diminution in massive quantity of materials and energy utilized absolutely result in less global warming. Additive Manufacturing emits no harmful pollutants into the environment, guaranteeing that water and air calibre are unaffected.



FIGURE 2. Sustainable additive manufacturing ecosystem (Javaid et al. 2021).

As illustrated in (Figure 2), it reflects different essential and well-substantiated strategies propels the conglomerate of technological platforms towards sustainability, offered by the “additive manufacturing ecosystem”. Here, along with environmental, societal and economic sustainability, the safety factor contributes to the whole sustainability of the ecosystem (Javaid et al. 2021).

The concept of economy-of-one becomes possible with this technology, as opposed to the notion of economy-of-scale. The precise supplement of material also reduces waste, resulting in a smaller ecological footprint (Spaltini et al. 2021).

According to Javaid et al. (2021), the best practice of AM is to generate commodities that no other manufacturing process can achieve. AM simplifies the engineering process by employing “cloud simulation” to evaluate fatigue, compute stress and so forth, eliminating the obligation for continual “physical prototyping”

and so saving resources, time, and waste. AM also enables the printing of futuristic designs with lesser waste and fewer components. In simple words, AM encourages customers to create only the parts they need rather than purchasing the entire assembly.

Rad et al. (2022) asserted that AM opens up the space for collaboration among various actors, including as designers and customers, and it can build up supply chain integration. By embracing AM, risks related to supplier, disposal, manufacturing and distributing can be reduced dramatically. Due to the closer place to the customer, additive manufacturing machines can make products. AM can reduce the number of suppliers required by cutting the quantity of components required to build a final commodity. This may imply fewer interactions with suppliers, fewer journeys to organize manufacturing, and lower production-related outflows. Furthermore, AM has the potential to cut the emissions generated during the manufacturing and transportation of products (Simonetto et al. 2022).

Producers optimize procedures in their supply chain by utilizing AM technology. Am minimizes the costs of supply chain through directly delivering commodities to the clients. 3D printers will produce a whole part that doesn't require assembly. Hence, it results in shortening the supply chain. This spectacular solution would permit manufacturers to minimize "operating costs" of manufacturing, increase productivity and customise services and parts to maximize efficiency of process (Javaid et al. 2021).

As a consequence, "Additive Manufacturing" functionally is more versatile than conventional "Additive Manufacturing". It can enable industries or enterprises to enhance their business model's profitability and speed by diminishing the expenses of resources and energy required to scale production without significant adjustment.

## **2.2.2 Sustainable Manufacturing**

"Sustainable Manufacturing" is characterized as a method that integrates process design and product problems with manufacturing challenges. With an increasing stress on "sustainable development" around the world, majority of customers are

eagerly pursuing corporations that use ecologically responsible manufacturing processes. Sustainable manufacturing techniques in operations will strengthen customer loyalty, boost the industry image and establish new leadership (Javaid et al. 2021).

### **2.2.3 Sustainability and Additive Manufacturing**

In the beginning, “Additive Manufacturing” was not basically envisioned as a “Green technology”. Since releasing in 1986, the goal has just been manufacturing geometries and parts that were tough to create, not to provide a fabricating solution to cut CO<sub>2</sub> outflows. Throughout three and half years, AM has experienced rapid advancement and progress.

At first glance, Additive Manufacturing appears to be the ideal technology for environmentally responsible manufacturing. The reality, on the other hand, may not live up expectations. Almost all Additive Manufacturing techniques necessitate the use of materials that have undertaken further processing phase. In addition, Additive Manufacturing fabrication is always pursued by one or more post-processing phases that enlarge the “ecological footprint” even further. Waste generated during resins and filaments preparation for use in the AM process, as well as testing novel materials and prototyping, should be considered. Overall, additive manufacturing is capable enough to become sustainable. To date, the AM industry has seldom compared LCA of current manufacturing to the traditional process or rarely published whole “Life Cycle Analysis (LCAs)” for AM parts. However, an equitable LCA is required to demonstrate whether AM truly has a lower “environmental impact” than a conventional part or not.

As displayed in (Figure 3) processing AM components results in an energy pitfall when comparing 1 kg of material in each process phase of the manufacturing chain. In the event of design phase is optimized, much “energy-intensive material” and production phases results in reduction of material needed by AM. Printed parts can then be upgraded by simplifying sophisticated designs as an energy-saving approach. It results in weight minimization whereas “mechanical properties” and operation of parts are sufficiently maintained. In other words, evaluating LCA validates that when AM comes to play during usage phase rather than

production and material phases, additional energy savings affects the whole “energy consumption” throughout product’s value chain (Langeled et al. 2022).

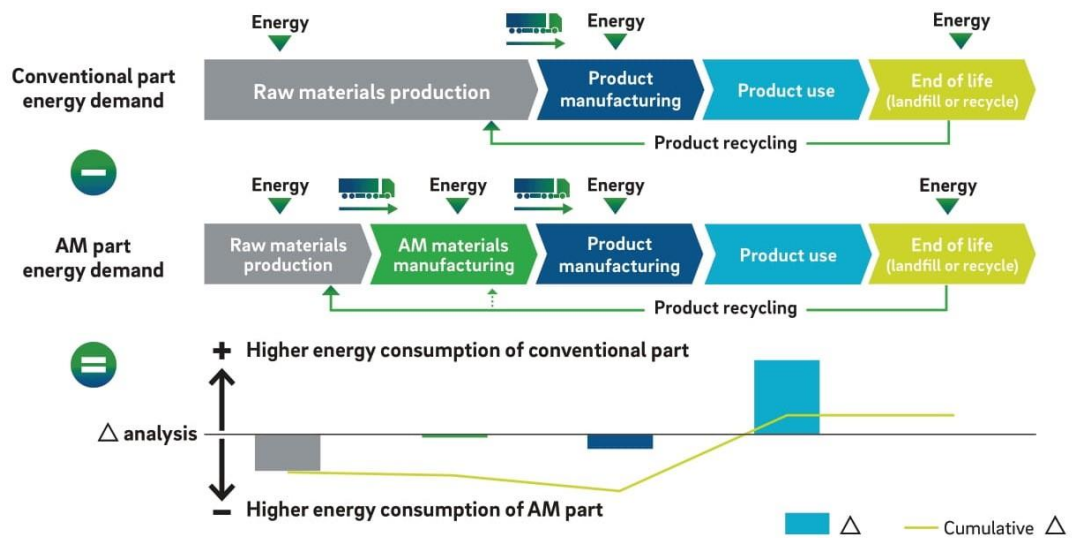


FIGURE 3. Life cycle analysis in Additive Manufacturing and Conventional Manufacturing (Langeled et al. 2022).

In short, an intelligent application of additive manufacturing potentials can reduce a component’s environmental footprint not only during the early phases of life cycle, but also during the downstream “product use” phase. Moreover, a long-time collective effort would involve Research and Development (R &D) surrounding suitability of material e.g., 100% recycled material for additive manufacturing in a product manufacturing phase could considerably cut waste and CO<sub>2</sub>.

#### 2.2.4 3D printing

The process of “accumulating materials to form items from 3-dimensional (3D) model data, usually layer upon layer”, is known as additive manufacturing. It is seen as a crucial component of the new paradigm 14.0 and is also as three-dimensional printing (Simonetto et al. 2022). 3D printers enable the automated production of personalized products that are exactly suited to the application (Democratization of manufacturing). 3D printing (3DP) provides new design flexibility, allowing manufacturers or producers to create revolutionary solutions to solve existing problem in the market (Javaid et al. 2021).

The “Waste Management” has two facets. The first is to reduce waste during the manufacturing process, and the second is to recycle used items into new ones. 3D printing technology is critical in dealing with this scenario. The key advantages of this new technology are boosting market speed, minimizing waste and cost-effectiveness (Figure 4). Because this technology takes shorter production time, the manufacturer can meet the client’s demand swiftly. Furthermore, a 3D printer can be used to reduce emissions and waste (Thomas & Mishra. 2022).

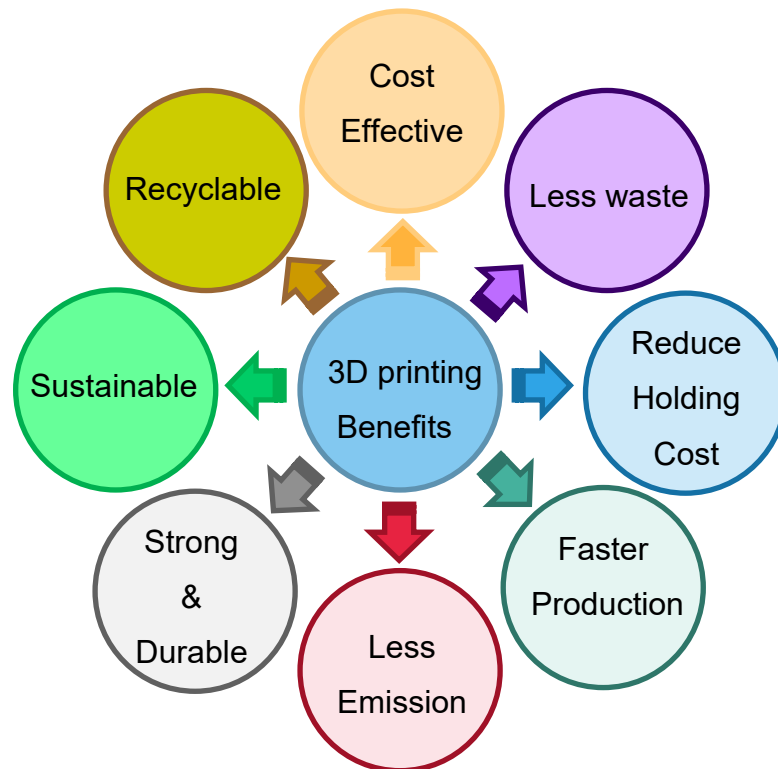


FIGURE 4. Benefits of 3D printing technology (Thomas & Mishra. 2022).

This technology assists businesses in better meeting demand while lowering the manufacturing costs and emissions throughout a product’s value chain (Simonetto et al. 2022). As a waste minimization technique, 3D printers can handle both aspects of waste management.

### 2.2.5 Distributed recycling 3D printing waste

The distributed manufacturing paradigm offers assets such as shorter supply chains, more concise lead times and reduced expenses, but it also causes significant waste. Throughout the 3DP process by the researcher, as depicted in (Picture 1), considerable amount of waste was created.



PICTURE 1. Generated waste during 3DP process

To tackle the technological challenges and benefits related to sustainability, the experiential evidence is required such as chances for “distributed recycling” of generated waste by 3D printing processes e.g., the left-over filament from previous printing, design-relevant supporting structures, additional “prototype printing”, direct waste extruders and misprints due to inappropriate designs or material uses (Peeters et al. 2019).

In case of distributed recycling 3D printed parts as a waste, Peeters et al. (2019) remarked that small-scale recycling is feasible. However, the quality of the material degrades, diameter variation escalates, and the tensile strength of printed parts shrinks with each recycling cycle, necessitating the addition of at least 50% virgin material to produce high quality prints.

Moreover, external contaminations such as dirt, grease, and water attach to the plastic or any other materials can be hinderance for re-recycling. The contamination eventually crystalizes and can negatively impact on the mechanical properties (e.g., brittleness, elasticity, tensile strength) and printability of recycled material.

In summary, concerning our study, using heterogeneous input materials for 3DP purpose at Large Scale Additive Manufacturing (LSAM) makes the recycling of

consumed prints, left-over materials and failed prints difficult. It affects the quality of materials unless we add virgin materials to further enrich noticeably.

### **2.2.6 3D printing and recycled material**

3DP is still a niche. Employing recycled materials for 3D printing is therefore a niche within a niche. “Digital fabrication” technologies as the most admired configuration of 3D printing enables circular production system in which waste materials as feedstock for 3D printing processes is smoothly recycled and reclaimed. In the field of sustainable 3D printing, the content used is probably the most differentiating factor.

One of the most noteworthy advantages of additive manufacturing is recyclability. The rapid prototyping is now used for complicated commodities. There is a stimulus for the producers or manufacturers to deal with chemical businesses to accelerate production of inventive materials. There have recently been much efforts to produce enough contents composed of recycled materials e.g., wide range of thermoplastics. This technology can be used to print any type of plastics and “green materials”.

Javaid et al. (2021) pointed out that the material is inexpensive and light-weight, which makes it a great candidate for additive manufacturing to replace plastics. Examples of inclusive polymers used in 3D printing include Polylactic Acid (PLA), Polyethylene Terephthalate Glycol (PETG), Acrylonitrile Butadiene Styrene (ABS), and Thermoplastic Poly Urethane (TPU). They can be made up of bio-based materials which are bio-degradable, sustainable and non-toxic, and also contain minimal waste. Due to their low melting temperature, they are suitable for a wide range of applications.

The challenge would be the absence of global standards for the mechanical and thermal properties of material for additive manufacturing (Javaid et al. 2021). The circularity principle is in line with 3D printing process because low rejection rates necessitate high material purity. Product defects continue to be a major issue when impure or heterogenous material exists (Despeisse et al. 2017). To make

this technology affordable for mass, lean and sustainable production, more research and development is needed.

### **2.3 Circular economy strategies: closing/narrowing the loop**

Cheah et al. (2022), asserted that 75% of current resources is globally consumed by human population rather than the earth is yearly regenerating even with the advances in efficiency of extracting resources. Preceding the notion of recycling and reusing resources, the linear economy model was put into practice. This model simulates the extraction of natural resources, the production of commodities, consumption of those items, and disposing the product once it is out of use. “Waste mismanagement endangers human health and local ecosystems, while simultaneously exacerbating climate change and its negative effects. The World Bank projects global waste to increase by 70% from a 2018 baseline by 2050 if we continue our “Take-Make-Waste” linear economy model” (Tetra Pak Sustainability Report 2022).

Conversely, the focus of transition to the circular economy is must centre on getting more value out of fewer materials. As displayed in (Figure 4), Narrow strategy reduces material use. Currently, material consumption is incredibly inefficient and useless. Using less is a core tenet of the circular economy. The Slow strategy seeks to keep materials in use for as long as possible, for example, by designing for durability. This will reduce material demand in the long run, effectively narrowing resource flows. Regenerate strategy phase out hazardous toxic materials such as plastic. Cycle strategy strive to reuse materials and cycle at their highest value, maximizing the number of secondary materials re-entering the economy, and as a result limiting the requirement for virgin material inputs and narrowing flows (Fraser et al. 2023).

To embed the circular economy in practice systems, tangible actions must be considered for the circulation of material flows (Spaltini et al. 2021). In comparison with alternatives in traditional waste management such as landfill, energy recovery and centralized recycling, a pervasive execution of “distributed recycling” (3D printing) has been revealed to be economically and environmentally beneficial (Zhao et al. 2018).

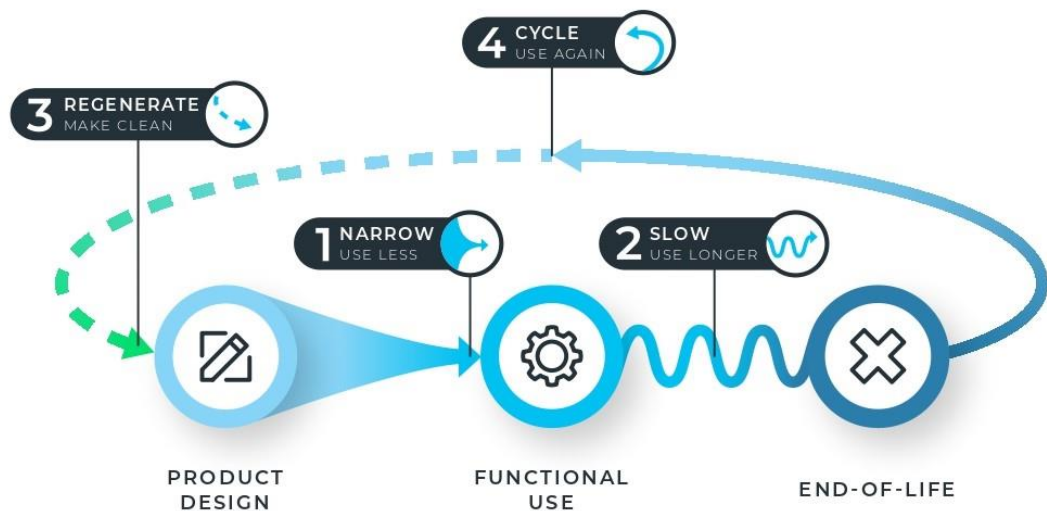


FIGURE 4. The four flows to achieve circular objectives: Narrow, Slow, Regenerate and Cycle (Fraser et al. 2023).

In summary, circular business models can potentially decrease negative impacts related to mismanagement and promote innovation, raise competitiveness and encourage business growth (Tetra Pak Sustainability Report 2022). In this respect, 3D printing with recycled materials can remarkably contribute use less, use longer and use again.

#### 2.4 Intersection of circular economy and industry 4.0

Last decade is recognized by two central mega trends, “digital transformation” and “sustainability”. More specifically, the present is described by “circular economy” and “industry 4.0” that will influence the future. Although researchers recognize an achievable alliance between these two topics, a practical integrated framework capable of achieving circular economy at systemic level is currently lacking.

Spaltini et al. (2021) framed that “industry 4.0” and “Circular Economy” are candidates to be two sides of the same coin”. As shown in detail in (Table1), a circular economy is built to be self-regenerating. Therefore, there is an interrelation between affirmation of the new technological progress and the growth of this paradigm: on the other side, industry 4.0 will promote a new business model centred

on environmental and social factors, leveraging the well-being of people and the environment, as a result of the growing interconnection of businesses and resources. The below table from Spaltini et al. indicates factors of connection between industry 4.0 and circular economy.

TABLE 1. Factors of connection between Industry 4.0 and Circular Economy (Spaltini et al. 2021).

Circular Economy and 14.0	IOT	Big Data	Cloud	AM
Process optimization	X	X	X	
Effective resources usage	X	X	X	X
Higher labour productivity	X		X	
Higher product quality	X	X	X	X
Lower time to market		X	X	X
Supply chain interconnection	X	X	X	
Demand and supply matching	X	X	X	X
Improved inventory management	X	X		X
Asset lifecycle extension	X	X		X
Product lifecycle extension	X	X		X
Reduce environmental emissions	X	X	X	X
Energy consumption optimization	X	X	X	
Productive cost reduction			X	X
Waste reduction	X			X
Remanufacturing support	X			X
Improved production flexibility			X	X

The focal point has been shifted towards these 4Rs (Reduce, Redesign, Recycling and Remanufacturing) since they have been considered the most affected by 14.0. The greatest enabling technologies for the application of industry 14.0 technologies have been identified as Cloud manufacturing (Maas) and AM. In fact, they enable the respective resource aggregation and waste reduction (Spaltini et al. 2021).

### 2.4.1 Circular 3D printing economy

Despeisse et al. (2017) argued that there is a lot of uncertainty surrounding the ambiguous question whether creating an alternative scenario in which requires customized goods with higher rate of obsolescence and less eco-efficient localized production, collectively increase resource consumption or the current path of 3D printing acquisition leading to more “circular material flows”. Before the adoption of 3D printing gets to a crucial inflection point, it’s imperative that “circular economy principles” be ingrained into the new production system.

An inquiry by Kreiger et al. (2014) disclosed that when employing readily recognizable single polymer types, a distributed 3D printing market procured by recycled materials led to reductions in “embodied energy” and carbon emissions. Conversely, “centralised recycling” market does not lead to such reduction. When more disseminated demands exist, 3D printing may bring the capability of material’s value into play by accounting on amounts of high-quality waste that are now unvalued in the recycling system due to limited aggregation. For instance, distributed recycling of plastics in order to produce filament for 3D printing purpose might assist increasing the recycling rate at a lower economic cost, whereas plastics are still only recycled at low rates in centralized recycling facilities.

Additionally, circular material flows may be made possible by tracking and tracing data about materials. A lack of supply chain transparency may contribute to a highly consolidated market structure and shortage of necessary information to convert waste materials into 3DP feedstock<sup>2</sup>. Design software and distribution platform as well as new models of 3D printers, material and “material processing” technologies, have all been invented by entrepreneurs. Three areas of activities have served as the centre of attention for entrepreneurial activity in the 3DP-CE thus far:

- 1) Application of 3DP for remanufacturing and re
- 2) Manufacturing 3DP filament, commercializing filament with recycled materials content

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<sup>2</sup> 3DP feedstock relates to how the raw material is manufactured, and determines the different processes for the production of feedstock (Sanchez et al. 2017).

3) Local recycling network to make filament (Despeisse et al. 2017).

Circular ecosystem and interconnected business models that seek to extract value from waste are also rising, and the accessibility of versatile technologies such as 3D printing may facilitate discovering advanced applications for waste material. The idea of industrial metabolism, now commonly referred to as industrial ecology, cultivated in reaction to the resource scarcity and the consequent rise in material cost. Therefore, proliferating efficiency of manufacturing can be achieved through minimizing material waste in process which is obvious benefit of 3DP technology. Industrial ecology stimulates companies across industrial sectors to systemically consider waste as a frequent, free and local resource.

#### **2.4.2 Centralized and Distributed Recycling**

Centralized recycling (conventional) is an acclaimed technique to cut the damaging effects of solid waste. Recyclable material such as plastic, paper, glass is collected, and is cycled back into the loop as reusable material through centralized recycling. This recycling technique contributes energy-savings, reduced contaminations and protects natural resources. The aim of this circular economy's strategy is reducing amount of waste that is to be fully incinerated or discarded in a landfill (Cheah et al. 2022).

Diverse approaches to managing plastic waste have been studied in the industrial ecology of polymers, from recycling (Chemical and mechanical feedstock) and re-utilize to "thermolysis/Recovery processes". When centralized recycling is compared to landfilling or incineration as alternatives for managing plastic waste, research has shown that mechanical recycling is the most effective method for managing relatively clean and homogenous plastic waste streams. To reuse plastic solid waste, mechanical recycling makes direct recovery of plastic and manufacturing new plastic possible. The applied technologies in this process include those for sorting out based on type of polymer, decontamination, size reduction, remelting and manufacturing new products from plastic waste.

Distributed recycling is a new method in which distributed sources of waste or recycled materials are 3D printed and used as products or filament feedstock, back to the value cycle. To exemplify, a novel viewpoint on polymer recycling to create feedstock for 3D printers has recently been investigated by combining “open-source (OS) 3D printers” which have domestic waste plastic extruders. Given the enormous increase in open-source 3D printing use and the current low recycling rates, a distributed polymer recycling solution may be extremely important (Sanchez et al, 2017).

### 3 BEVERAGE CARTON MATERIAL STRUCTURE

Typically, packaging comprises of multiple components. These may be further separated into packaging aids and packaging materials, and they are made up of various of packaging materials. A “packaging material” is perceived to be the component which shapes the central part of packaging and holds together the packaged contents. This is the foundation of a composite beverage carton. Components called packaging aids allow for supplementary functions such as fastening, tagging. These include staples, closures, sticky binding and sealing foils. In tandem, packaging aids and basic packaging form the packaging.

Paper, cardboard and carton are materials that primarily consist of fiber layers and are further processed throughout number of phases (impregnating, laminating, coating etc.). A wide variety of paper grades, cardboard and carton can be distinguished based on the nature of the fibers (bleached or unbleached), thickness of the fiber layers, structure (composite, corrugated board etc.) and any additional fillers (World Packaging Organization 2020). According to Kreplin, Schabel and Putz, “thanks to low losses throughout recycling, fibers from corrugated boxes can be recycled 25 times without experiencing signs of a Recycling Collapse. Depending on the fiber type and the corresponding recycling processes and losses during recycling, the average number of cycles can be lower.” (4everGreen 2022).

In wet processing, so-called papers are created by subjecting the paper fibers to mechanical stress in an aqueous solution. This stage also involves the removal undesired external substances e.g., metal clips, adhesive foils and particles. Printing ink is also separated via de-inking process depending on the intended application. There is often no de-inking process employed in the recycling of packaging papers. The produced secondary fibers can be utilized again as reuse to produce paper. However, due to the fact that the fiber quality (fiber length) declines, there are technological restrictions. Paper that has been coated on both sides, as well as paper that has been impregnated with wax or paraffin on one side or both sides, is regarded as a composite material and is not suitable for use as paper packaging. There is specialized recycling mechanism for particular fractions such as composite beverage cartons, which often have layers of PE-paper-

PE-Aluminum-PE or PE-paper-PE (World Packaging Organization 2020). According to the German Packaging Act, composite materials often known as multi-layered packaging, are a collection of packaging materials, none of which has a mass proportion of more than 95%, and cannot be manually separated.

The right mix of materials prevents food loss by shielding the contents from degradation and nutritional loss while ensuring food safety. As evidenced in (Figure 5), these barrier layers in the aseptic beverage cartons consists of a combination of polymers and Aluminum which enables the product be distributed and stored at room temperature. It should be highlighted that essential quantities of each material can ensure food safety and package functionality. The quest for a circular economy should provide a compliance of materials in beverage carton's structure with enhanced recycling processes in dedicated paper mills and encouraged solutions for the Poly-Al, as is the case with other packaging materials (Alliance for Beverage Cartons 2022).

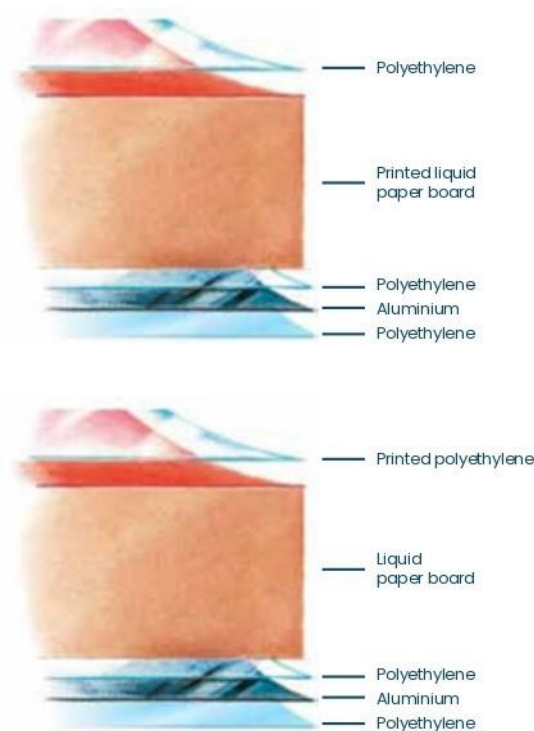


FIGURE 5. Typical material structure of aseptic beverage cartons (Alliance for Beverage Cartons 2022)

### 3.1 Polymer fraction

Typically, a Low-Density Polyethylene (LDPE) added by “extrusion coating” in aseptic beverage cartons provide a moisture barrier for the board, allowing the flaps of package to be sealed and can protect a printed surface.

“Beverage cartons” may incorporate an easy opening, closures (Tethered caps are widely used in Tetra Pak package profile due to assimilate those to the recycling mainstream aligned with EU anti-littering regulations) which impede spillages, facilitate the action of pouring and permit re-closure of the package. Even after being opened and thrown away, tethered caps or non-detachable caps remain attached to the beverage cartons. The most common materials used to manufacture opening and closure features are low density polyethylene (LDPE) or Polypropylene (PP) or High-Density Polyethylene (HDPE), a mix of these materials. Prior to the Poly-Al recycling process, minor parts such as cutting rings, along with carton tops and necks are frequently removed from the film fraction using a wind-shifter ability to differentiate between various material densities and or thicknesses (Alliance for Beverage Cartons 2022).

A longitudinal seal strip must be placed inside to roll-fed carton material to avoid product contact with the Aluminum and the liquid packaging board edges. The core layer of strip is composed of PolyAmide (PA) or Polyethylene Terephthalate (PET) that is coated with either LDPE or LLDPE (Alliance for Beverage Cartons 2022)

In terms of chemistry, Polyamid is kind of plastic based on peptide bonds and is comparable to protein molecules. It's characterized by a high level of good barrier properties strength as well as toughness. One of the most applicable polymers, Polyethylene is resistant against greases, alcohols and oils as well as diluted alkalis and acids. Additionally, it may be welded and is extremely cold resistant. It's also produced in a variety of qualities (HDPE, LDPE, MDPE). Polyethylene terephthalate is a transparent, stable plastic, and has good “barrier properties”. PET occasionally has a good lipid resistance and high aroma density. Polypropylene is a plastic similar to polyethylene chemically, but is more temperature-resistant and stronger. It is effective in blocking lipids and moisture.

Since they are designed specifically for the processing of composite beverage cartons, single-sided and double-sided plastic coatings don't pose any issues during the recycling process. Starch, Kaolin, Talc, Calcium carbonate, titanium oxide, and other industry-standard additives in paper content can be utilized without any problems, however they proportionately diminish the fiber yield after recycling (World Packaging Organization 2020).

### **3.2 Aluminum fraction**

Aluminum Oxide ( $\text{Al}_2\text{O}_3$ ) is used to varnish plastics in order to improve the barrier properties. Aluminum is a vapor-resistant extreme thin layer, deposited onto the substrate for this purpose. This is applicable to flexible or rigid packaging, including film packaging.

To secure the liquid packaging board to the Aluminum foil in aseptic beverage cartons, the inner side of the board is commonly blanketed with LDPE. Aluminum foil is very thin (measured in  $\mu\text{m}$ ), constituting 5% of the carton's total weight, and its thickness varies based on the barrier properties that are required. The Aluminum foil acts as a barrier against light, odour and gas. The usage of an LDPE layer with a low percentage of an adhesive co-polymer like EAA (polyethylene-co-acrylic-acid) or EMAA (polyethylene-co-methacrylic acid) content is often required to adhere the Aluminum foil to the innermost plastic layer. These co-polymers have thermo-mechanical properties similar to those of LDPE (Alliance for Beverage cartons 2022).

### **3.3 Poly-Al fraction recycling**

Following the pulping process to remove the paper fibers, a residue of polymer and Aluminum (Poly-Al) with a moisture content of approximately 20-40% is left behind. The majority of Poly-Al fraction consists of polyolefin (mainly propylene with Polyethylene) with a tiny quantity of residual fibers and 20% Aluminum particles in various shapes and sizes. Pre-cleaning minimizes the amount of residual

fibers content and other contamination and can be done at the paper mill or Poly-Al recycling facility. It's customary to separate the hard plastic fraction (caps, closures etc.) from the foil fraction (LDPE and Aluminum barrier layers), which is then separately processed into the flakes and integrated to a polyolefin stream. As illustrated in (Figure 6) the composite material is compacted either as an extruded granulate or as a pressed granulate for the operations where the plastic and Aluminum foil fraction are treated simultaneously. Injection moulding is one of the applications that this material is directly used for that (Alliance for Beverage Cartons 2022).

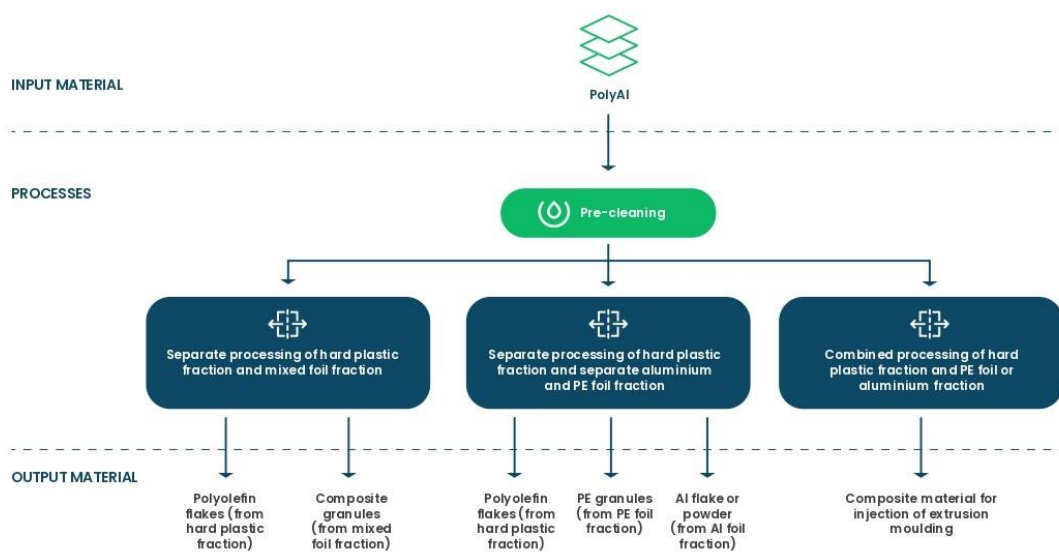


FIGURE 6. Generic routes for reprocessing Poly-Al fraction (Alliance for Beverage Cartons 2022).

There is other two other generic routes for reprocessing Poly-Al fraction. But they are out of this study's scope along with chemical recycling of Poly-Al fraction.

Plastic drinking straws and protective wrappers constitutes of Polyethylene (PE) and Polypropylene (PP) that were glued to beverage cartons should be removed from the beverage cartons during the fiber recovery process, and would become part of the Poly-Al stream, compatible with Poly-Al recycling process.

Adhesive barrier film is more challenging than extrusion barrier solution in which thermoplastics is placed on one side or two sides coated, the polymer will intensely perforate into the fiber and have an effect on the fiber yield. In addition,

4everGreen noted that “Aluminum foil may affect induction-based flowmeters and lead to metal being detected in the finished product”. (4everGreen 2022). Hence, pre-treatment and more cleaning steps should be given top priorities.

The Poly-Al fraction is currently being recycled by applying a number of technologies. After the polymer has undergone mechanical pre-treatment (Such as shredding, washing, cleaning and drying), “mechanical recycling” preserves the molecular polymer’s structure before re-melting it into granulate.

As described earlier, non fiber fractions of composite beverage carton have often times been problematic and been viewed as substantial challenge on the way of recycling. Hence, aligned with global regulations and commitment, transition from multi-material to mono-material beverage carton, some developments have been specified by brand owners and package producers as millstones e.g., removing Aluminum foil and replace paper barrier instead or using plant-based Polyethylene instead of fossil-based one. Hence, until fulfilling these milestones, deploying circular practices can mitigate environmental effects of Poly-Al waste.

## **4 MATERIAL and METHODS**

Gomes et al. (2022) presented that the waste source plays a role of differentiating point in the relevance of effectiveness and modification techniques. However, the source of this multiple polymers and Aluminum in the granules are from post-consumer packaging waste and is not considered as non-specified.

World Packaging Organization 2020 described that “post-consumer material is material from household, commercial or industrial establishments or institutes (which are the final consumers of the product) that can no longer be used for the intended purpose. It includes material recycled from the supply chain (also known as Post-Consumer Recycled PCR or Post-Consumer Waste PCW).” (World Packaging Organization 2020).

The specific material knowledge in this part can contribute us to predict thermo-physical behavior of Poly-Al made from recycled material, and find the suitability of material for use in engineering materials applications. Firstly, the study delves into material qualifications of recycled Polymer and then recycled Aluminum elaborately.

### **4.1 Poly-Al material qualification**

The rapid advancement of material engineering throughout time has made it possible to develop innovative materials that are replacing plastics, conventional woods in many industrial fields. Composite materials consist of at least two materials with distinctive properties that the matrix results in a new material with enhanced performance. Polymer composites stand out from the other composite materials in terms of their properties, indicating variety of industrial applications (Fico et al. 2022).

Composite materials favour recycling processes and offer parts that have excellent quality and long-life service. They also have low weight, less maintenance and high strength (Colorado et al. 2020). When working with products that have been mechanically recycled, quality is a major concern from a technical

standpoint. Heterogeneity and compatibility issues of the polymers are crucial factors to take into account in this process (Sanchez et al. 2017).

Gomes et al. (2022) highlighted that “in the case of thermoplastics e.g., PE, PP, PA6, a new recycling route is emerging, made possible by Material Extrusion Additive Manufacturing (MEX): distributed recycling. However, similar to what happens with mainstream recycling of these materials, the thermo-mechanical processes involved, as well as other factors such as exposure to UV-light, result in waste streams with degraded properties.” (Gomes et al. 2022). In this regard, the number of feasible life-cycles, potential application and functionality of the polymers are all extremely degraded. Therefore, the methods for modifying the properties of polymers, offsetting the degradation becomes substantial (Gomes et al. 2022).

When a material is treated, especially when high temperature and mere forces cause chain chemical reactions, thermo-mechanical degradation occurs. “On the other hand, thermo oxidative degradation produce physical and chemical changes in the polymeric structure due to exposure of specific conditions during service life. Thermo-mechanical and thermo oxidative degradations are responsible for changes in structural and morphological characteristics of the polymers such as mechanical-rheological-thermal properties, degree of crystallinity, viscosity and molecular composition.” (Peeters et al. 2019). The mechanical properties and diameter of nozzle can both be negatively impacted by heating and contamination, and the quality can be impacted by combining different thermoplastics (Peeters et al. 2019). Material degradation can be defined in terms of these parameters as follow (Figure 7):

- Degree of Degradation (DD): measures polymer degradation caused by processing and service life.
- Degree of Mixing (DM): refers to presence of impure polymers as a consequence of poor separation from waste streams in recycling plant.
- Low Molecular Weight Compounds (LMWC): related to the presence of additives, contaminants (Sanchez et al. 2017).

Zhao et al. (2018) asserted that when different circular economy strategies are adopted by manufacturers, the polymer degradation is consequently more common. And then, materials are re-introduced to the production value chain as a feedstock throughout multiple production cycles.

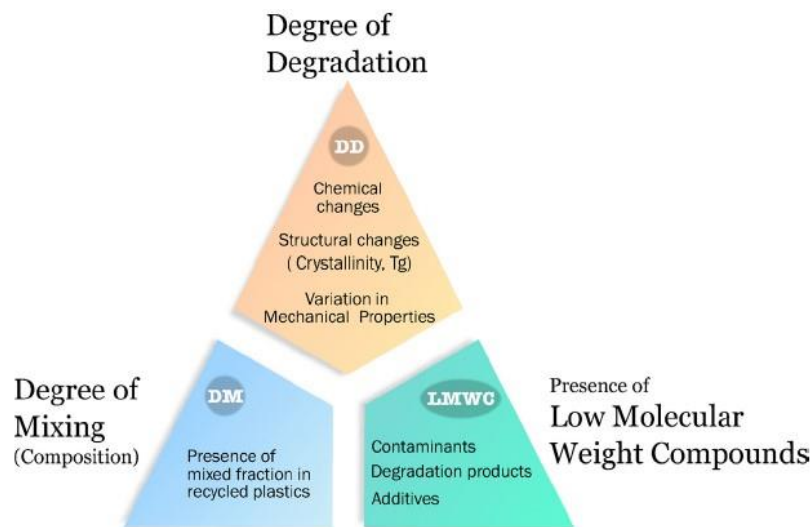


FIGURE 7. Key properties for quality assessment of recycled polymer (Sanchez et al. 2017)

Additional cycles can be accomplished without the polymer degradation. For instance, Polyethylene Terephthalate- Glycol PETG eventually reaches this point after six cycles of reprocessing. The melt flow index (MFI) is one attribute, traditionally required in the plastic industry (plastic processor or producer) to determine if same process may be utilized regardless of whether it employs virgin or recycled polymers (Sanchez et al. 2017). For instance, since some of deformation issues may be addressed by blending recycled thermoplastics e.g., Polyethylene reinforced with recycled thermosetting polymer e.g., Polyamide 6 (PA6). The MFI of virgin PA6 was observed to be 23.30 g/10 min, whereas the MFI of recycled PA6 was 19.01/10 min. The decline in MFI of recycled PA6 demonstrated a material's decreased ability to flow after its recycling (Vinay et al. 2022).

Jin et al. have researched the impacts of mechanical recycling process on low density poly ethylene (LDPE) and discovered that LDPE can be extruded up to 40 times without noticeably altering the material's processability or long-term mechanical properties (Jin et al. 2012). According to Tominaga et al. (2015),

molecular weight of recycled Polypropylene doesn't dramatically change during the recycling process (Tominaga et al. 2015).

In general, material degradation depends on the processing cycles e.g., much degradation is seen in the Fused Fabrication Filament (FFF) method due to the extra extrusion or heating phase for example, variations in degradation, crystallinity changes and viscosity reduction were reported about recycled polyethylene terephthalate (PET) (Zander et al. 2018). As a tangible example, the recycled material Poly-Al, which was deployed in 3DP fabrication, had been just exposed to heating or extrusion for two cycles in total.

Figure 8. represents the multiple processing approach (Material, Process, Evaluation and Recycling) for evaluation of morphological and structural changes throughout degradation. In this sense, the recyclability of polymer materials is assessed through reliable method of multiple extrusion throughout simulated extended life cycle which can identify the progress level of degradation. To avoid further degradation, there is a possibility to optimize the processing parameters such as altering the temperature and adding reinforcements in the course of mechanical recycling. (Sanchez et al. 2017).

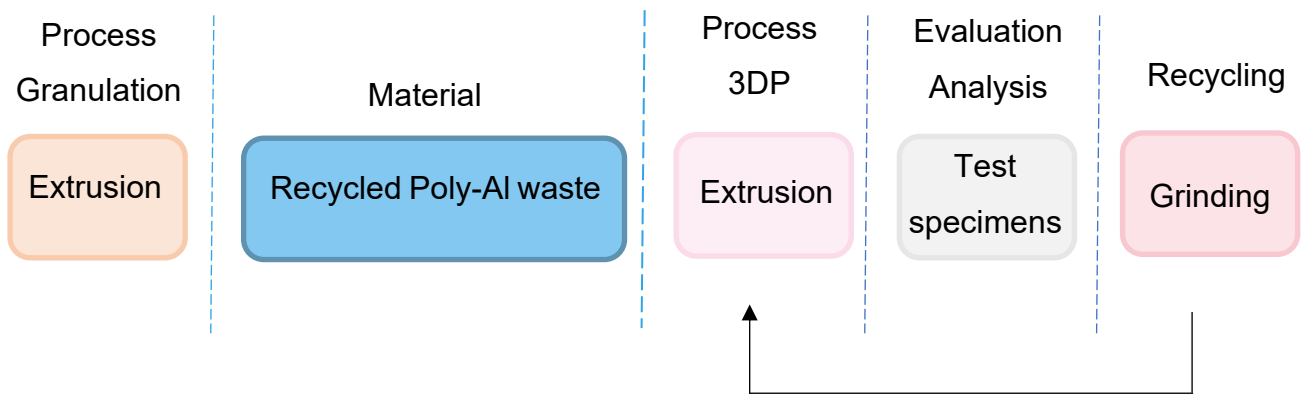


FIGURE 8. Multiple processing approach to evaluate thermo-mechanical degradation

Additionally, it is imperative to describe the qualitative parameters which are assessed in the evaluation phase as presented in (Figure 8). By ensuring the thermo-mechanical performance of recycled materials in the second market

applications, the needs of manufacturers and consumers become satisfied (Sanchez et al. 2017).

According to study by Mohamad et al. Aluminum is produced by extracting bauxite. In 2015, yearly output of Aluminium was 270 million metric tons in comparison to year 2006 when it was 183 million metric tons. This phenomenon results in an increased Aluminum waste in landfills, leading to environmental hazard. Hence, a recycling strategy is demanded to lessen the adverse environmental ramifications of Aluminum waste. Due to the fact that the Aluminum industry generates the major part of metallurgical waste, which is challenging to recycle, this phenomenon raises awareness of the value in Aluminum waste (Mohamad et al. 2022).

Raabe et al. (2022) asserted that by 2050, it is anticipated that the amount of Aluminum that can be recycled will have doubled. 30-55% of the Aluminum used today consists of flat rolled product (foil, sheet and plate) which differs by company, market and region. A “green Aluminum” or “low-carbon Aluminum” trends may lead to limit unsustainable materials in future products, e.g., replacing Aluminum foil with paper barrier. A challenge is large amounts of Aluminum in different forms of waste are post-consumer waste possessing high level of contaminants.

$\text{Al}_2\text{O}_3$  is one of the valuable materials that may be recovered from Aluminum scraps and waste. Studies show that cost-effective recycling process of extracting of  $\text{Al}_2\text{O}_3$  from Aluminum waste only needs 5% of the net energy. The differences between physiochemical properties of  $\text{Al}_2\text{O}_3$  from various Aluminum waste (e.g., Aluminum Foil (AF)) have not been addressed, despite the fact that the extraction of  $\text{Al}_2\text{O}_3$  has been studied in depth (Mohamad et al. 2022).

According to Mohamad et al. (2022)  $\text{Al}_2\text{O}_3$  extracted from Aluminum Dross (AD) is higher than the same extracted from Aluminum Can (AC) and AF. Hence, processing methods such as stage of recovery and raw material composition influence the content of  $\text{Al}_2\text{O}_3$  in these wastes. As illustrated in (Figure 9), the formation of Aluminum hydroxide from AF shows less yield of  $\text{Al}_2\text{O}_3$ .

By extension, the progress in getting a higher yield of  $\text{Al}_2\text{O}_3$  during extraction phase is dependent on the presence of impurities. This specifically affects the transformation and granulometry of  $\text{Al}_2\text{O}_3$ . As a result, the rate of impurity presence in Aluminum foil in combination with LDPE might be a potential agent in seeing a bad performance of whole material as a granule feedstock throughout 3D printing process. It's worth noting that mechanical properties of Aluminum are reduced after 8 cycles extrusion cycle (Raabe et al. 2022).

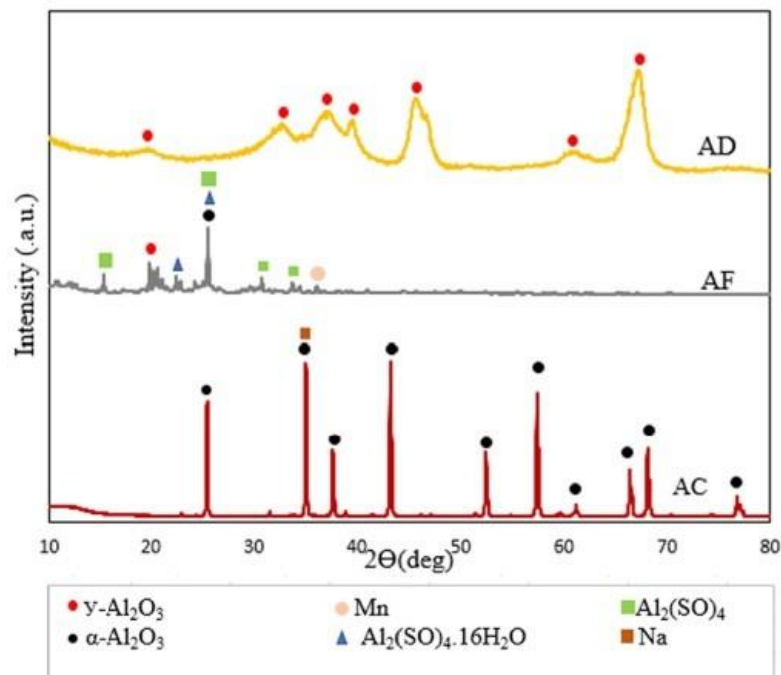


FIGURE 9. Content of  $\text{Al}_2\text{O}_3$  in various Aluminum waste, AD, AC, AF. AF is considered as a feedstock with the lowest yield of  $\text{Al}_2\text{O}_3$  with traces of impurities (Mohamad et al. 2022).

To further enrich the research result and discover whether presence of Aluminum particles could play a pivotal role as a reinforcement in this polymer matrix or not. In this sense, specific research about interaction between rLDPE and Aluminum was not found. So, in this part, we sufficed interaction between rPP, rHDPE and Aluminum, which due to similar mechanical performance of all thermoplastics, enabling us to give enough understanding about the materials.

The total mechanical performance of the Polymer Matrix Composite (PMC) is greatly influenced by composition of reinforcement phases. High specific strength

of composite can be reached through the design of proper fiber architecture, rather than light metal reinforcements (Tang et al. 2023). Functional properties such as electrical and high impact energy absorbing ability are present in the metal particles that make up the composite. However, a weak interfacial bonding between metals and polymers might restrict their applicability since it would weaken the performance.

In addition, Singh et al. (2018) confirmed that presence of reinforcement ( $\text{Al}_2\text{O}_3/\text{SiC}$ )10,20,30% in rHDPE matrix doesn't influence the thermal properties of the material since there was almost no modification in melting range of materials. Also, same mixer of reinforcement increases material flow of rHDPE from 9 g in to 11g in 10 min. The study revealed that augmenting Aluminum-Silicon and Iron powder to rPP/HDPE matrix, led to better thermal stability and yield strength by up to 17%. One explanation could be that the powders made of metal worked as filling agent, improving the interactions with polymers (Pan et al. 2016).

In summary, the occurrence of defects in components made of Aluminum using additively manufactured still constraints the application of AM methods to high-strength Aluminum alloy. Notable research has been directed over the years to minimize or eliminate these defects, which has expanded the number of applications for AM in high strength Aluminum alloys (Altiparmak et al. 2021).

## **4.2 Poly-Al granulation process**

Our sample for 3DP was supplied by KMK recycling plant with the capacity of 3000 tonnes granule production per year. So, it's so crucial to understand the process of granulation that is usually done in this factory. As showed in (Picture 2), the input material of this recycling line contains Polyethylene, PET, polyamide 6 and polypropylene, Poly-Al. Poly-Al waste recycling line includes cleaning and granulation lines. The first line is cleaning line where purity of Poly-Al waste to be yielded.



PICTURE 2. Input material of Trommel screen at the beginning of cleaning line

In what follows, some contamination e.g., caps, closures (hard plastic fraction) are separated from foil fraction in the Trommel screen and on continuous moving magnetic drying sieve (Picture3). Throughout the Trommel's spinning, material loses a part of moisture as well. Moreover, the worker separates this contamination e.g., even plastic bottles mixed with Poly-Al waste manually.



PICTURE 3. Output material of Trommel screen in the cleaning line

To eliminate fiber contamination, Poly-Al waste goes into 3 cleaning machines where it is de-dusted and sticky additional fiber can be wholly recovered by 100%. According to Alliance for Beverage Cartons, approximately 4% of the fiber that is adhered to the Poly-Al, is lost to the fiber recycling process. The reason behind existing 3 cleaning machines (Picture 4) is that in terms of a general rule, the

quality and efficiency of the output material improves, and overall yield increases, as there is less residual fibers in the Poly-Al stream.



PICTURE 4. Three cleaning machines, recovered fiber from Poly-Al waste

In the following phase, as illustrated in (Picture 5) Poly-Al waste is crushed and downsizing to 60 mm by shredder machine. It is kept in the silo machine in adjustment with the frequency of line's input material.



PICTURE 5. Downsizing Poly-Al waste by shredder (left), Stocking in the silo machine (right)

Afterward, the downsized Poly-Al waste moves on the conveyor slightly. It is transferred into the extrusion machine to turned into the form of granule as showed in (Picture 6).



PICTURE 6. The Conveyor (left) and Extruder machine (right)

The temperature on extruder's setting board is variable in terms of dirtiness, moisture, quality level of incoming waste to the extrusion machine as input. It worth noting that the temperature of extrusion machine is usually set in terms of quality of incoming waste. The temperature can be adjusted within the 165 °C to 205 °C range by machine operator (Picture7).



PICTURE 7. Input material into extruder machine (left) and extruder temperature profile (right)

As you may see in the temperature setting board above, there are four segments that include compactor, extruder, filter and horizontal cut. In each section, some parameters are displayed e.g., speed (with a screw speed of 75-950 rpm, Round

per Minute), weight (%), boiler temperature ( $^{\circ}\text{C}$ ), pressure (105-148 bar), melting temperature ( $195^{\circ}\text{C}$ ), water temperature in the horizontal cut sector ( $43^{\circ}\text{C}$ ). The granulation method is hot cutting and after cutting in a round-shape nominally 4 mm long, granules are cooled down in a water bath (Picture 8). Granules lose their heat  $200^{\circ}\text{C}$  by water gradually and the temperature exponentially reaches into  $25^{\circ}\text{C}$ .



PICTURE 8. Cooling round-shape granule by cold water (left) and sieving, separating granules from each other (right)

As evidenced in (Picture 9), 500 kg granules are often produced per hour. It is stored in the moisture-barrier bags, and sold to plastic industry manufacturers as an end-product in the market.



PICTURE 9. Granule bagging station (left) granule as an end-product (right)

### 4.3 Poly-Al granule properties

The selected material for 3D printing test was Poly-Al granule that had been supplied by KMK plant. The characterization shared by the supplier had to be considered to define initial “processing parameters” in 3D printing. This material was intended for fabrication of 3D printer feedstock according to the manufacturer’s specification, and enumerated in detail in (Table2).

It should be noted that chemical and mechanical properties vary in different types of thermoplastic. Moreover, the properties within the same thermoplastics (e.g., PLA, ABS, PET) are different based on suppliers and additives, making separation more complex (Peeters et al. 2019).

TABLE 2. Initial characterization of Poly-Al granule material

Primary information	Unit of measurement	Indicative value
Granule diameter	Mm	4
Filter size in production	Micron	840 (20 Mesh)
Moisture content	%	≥0.15
Density	g/cm <sup>3</sup> - ISO1183	1.07
Source of feedstock	Origin descriptive	Recycled beverage cartons from paper-mill
Material composition	%	rLDPE with traces of PP, PET, PA6 <sup>3</sup> , Aluminum
Inorganic content (TAG)	%	No data
Smell/odour	Score (0-no odour to 3 strong odour)	1* *Measured at 190°C
MFI (190°C, 2.16 kg)	g/10 min- ISO rt	3.345
Impact strength	KJ/m <sup>2</sup> , Notched Izod at 23°C- ISO 180 A	69.4
Tensile strength	MPa-ISO 527-3	11.5
Elongation at break	%- ISO 527-3	37

<sup>3</sup> “To create permanent wet strength urea resins, polyamidoamine epichlorohydrin (PAAE or PEA) and melamine resins are commonly used” (Alliance for Beverage cartons 2022)

Primary information	Unit of measurement	Indicative value
Flexural modulus	MPa-ISO 178	232
Recommended processing Temperature	Degrees Celsius (°C)	200

#### 4.4 Material Extrusion Technology

The chief objective here, is to represent generic methodology to evaluate the opportunity, level of interest and methods for granulating thermoplastic and Aluminum waste to utilize as a feedstock for “open-source 3D printers”. Invented in 1989, a technique called “Material Extrusion” (ME) was successfully commercialized. In the process, a thermoplastic material is selectively deposited through a nozzle. Similar to how extrusion and injection moulding function when making thermoplastics traditionally, ME processes requires the development of tailored material (Sanchez et al. 2022).

One such plastic route is the distributed recycling associated with the distributed manufacturing enabled by AM. Recent developments in distributed recycling of thermoplastic materials have been primarily applied through Material extrusion (MEX). Material extrusion is classified into Fused Filament Fabrication (FFF), and Fused Granular Fabrication (FGF) as illustrated in (Figure 10) (Gomes et al. 2022).

Sanchez et al. (2020) identified that closed-loop distributed recycling using ME involves a number of steps including recovery, feedstock, printing and quality testing. This maintains the material in the value chain as long as feasible, and application range must be maximised to enable its use in the producing functional parts that have high embedded value.

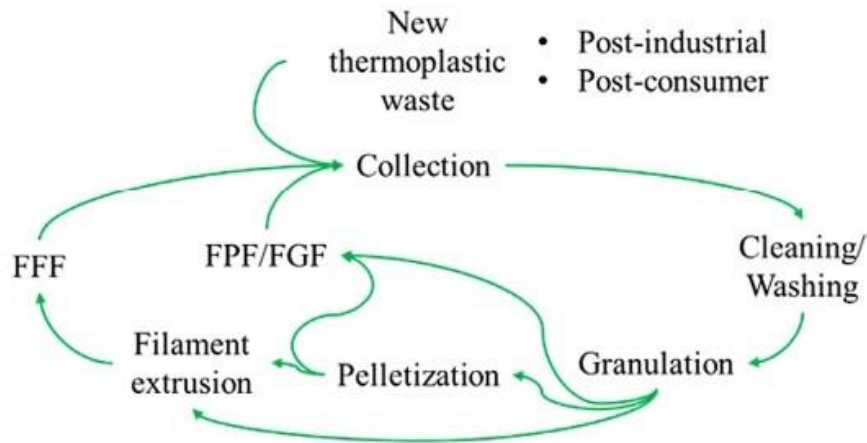


FIGURE 10. Typical mechanical recycling approach used in material extrusion (MEX) (Gomes et al. 2022)

#### 4.4.1 Fused Filament Fabrication

One of the most commonly used three-dimensional (3D) printing techniques is Fused Filament Fabrication (FFF) which is simple and relatively low cost (Katalagarianakis et al, 2022). Filament (plastic string) is the feed stock material used in printers operating on FFF technology. The filament is forced through a hot-end (heating column) and a nozzle which distributes the “semi-molten plastic” on to a bed. When “semi-molten plastic” adheres to the layer, is laid down and gently cools down, a 3D item is amazingly created. Polylactic Acid (PLA), Polyethylene Terephthalate (PET) are the only two of the thermoplastics used in the process (Peeters et al. 2019).

#### 4.4.2 Fused Granular Fabrication

One method of overcoming cost barrier of FFF thanks to additional melt-solidify cycle is to leave out the whole process of fusing filament into a 3D printed item by printing directly from granules or pellets. Any size or shape of polymer feed-stock are being produced through Fused Granular fabrication (FGF) with various designs and commercialized printers. To date, only a tiny portion of the studies address thermoplastic materials printed deploying this technique (Woern et al. 2018).

In this method, granules are transferred from a dryer to a hopper. Then gravity makes them move inside of the screw. During the rotation of the screw, granules are passed through a barrel with several heating zones, which enables the material to heat up and melt. Then pressure within the extruder forces melted material in a form of large nozzle along the digitally defined tool path.

Sanchez et al. (2022) accurately asserted that the material should preferably have a low “Coefficient of Thermal Expansion (CTE)”, a sufficient “melt flow index” and acceptable mechanical properties. This will help facilitate “manufacturing process” for Pellet Material Extrusion (PME) purpose.

The studies show that when recyclable feedstock used as a distributed recycling system employing, “open source industrial FFF/FGF 3D printers” offer remarkable economic potential. Some products have an outstanding profit margin and return on investment (Byard et al, 2019). Additionally, from GHG emission perspectives, large plastic products with a long lifecycle (e.g., building components or furniture) offer a long-life time carbon sink and can potentially use high amount of plastic waste (Rattan et al. 2023).

Regarding large-scale 3D printing, high-volume material extrusion coupled with advanced machining may lead to a higher chance of using additive manufacturing in 3D printing, particularly when dealing with high performance polymer materials. Jalava et al. (2023) pointed out that as a result, potential material flow rates are not the most suitable for large-scale manufacturing, leading to lengthy print durations due to the print failures. With the increase in nozzle sizes and layer heights, accuracy in shape was inevitably omitted. The possible chance of attaching an extruder or robot makes it feasible to print in virtually any desired volume, and only be limited in the extruder material flows, and hence the print times.

For comparison purposes, FGF has advantages e.g., lower cost, higher production speed and higher availability of materials. Although, prior to FFF, filament must be processed (filament extrusion), resulting in time-consumption. The comparison between FGF and FFF methods is presented in (Table 3).

TABLE 3. Comparison between FGF and FFF material extrusion methods

Aspects	Fused Filament Fabrication (FFF)	Fused Granular Fabrication (FGF)
Feedstock	Strand	Granule/pellet
Throughput (lbs/hour)	0.005- 0.25 lbs/hr	0.5-20 lbs/hr
Resolution	0.15-0.4 mm 0.3-1.00 mm	1-5 mm 2-10 mm
Print dimension	Small parts	Large parts
Tool path	Complex geometries	Simpler geometries
Temperature	Get heat into the material	Get the heat out of material
Cost <sup>4</sup>	Higher	10x Lower per part
Portfolio	Wider choice	Wider choice
Overhangs	Much support material	Less support material

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<sup>4</sup> Thanks to less additional manufacturing and heating cycle, the process of Fused Granular Fabrication is cheaper than Fused Filament Fabrication.

## 5 EXPERIMENTAL PART

This chapter of study introduces an experimental part of the thesis. In the beginning, a required quantity of material for the test was estimated. Afterwards, the samples of recycled Poly-Al in the form of granule including two sacks 11, 14 kg were shipped from the KMK factory (granulation line) in Turkiye to the TAMK's Fieldlab in Finland. The laboratory work commenced with a preparation of Poly-Al material, which was the target material. For the next step, various parameters were examined to find out material-specific optimal parameters for 3D printing process. Prior to carrying out 3D printing process, 3D printed parts were simulated to get assurance about selected parameters. Ultimately, 3D printing process were monitored and analyzed quantitatively and qualitatively. In the end, the chapter ends up with tensile strength test and obtained results.

### 5.1 3D printing test by FGF

The preparation of materials was carried out for the 3D printing test. With refence to the shared information including material properties by the supplier, the moisture content of granule equates to less than 0.15%. Although, polymer is considered as non-hygroscopic materials. It means that they don't absorb moisture internally and might absorb through their surface.



PICTURE 10. Dehydrating of Poly-Al granule in a drier's tank prior to 3D printing processing

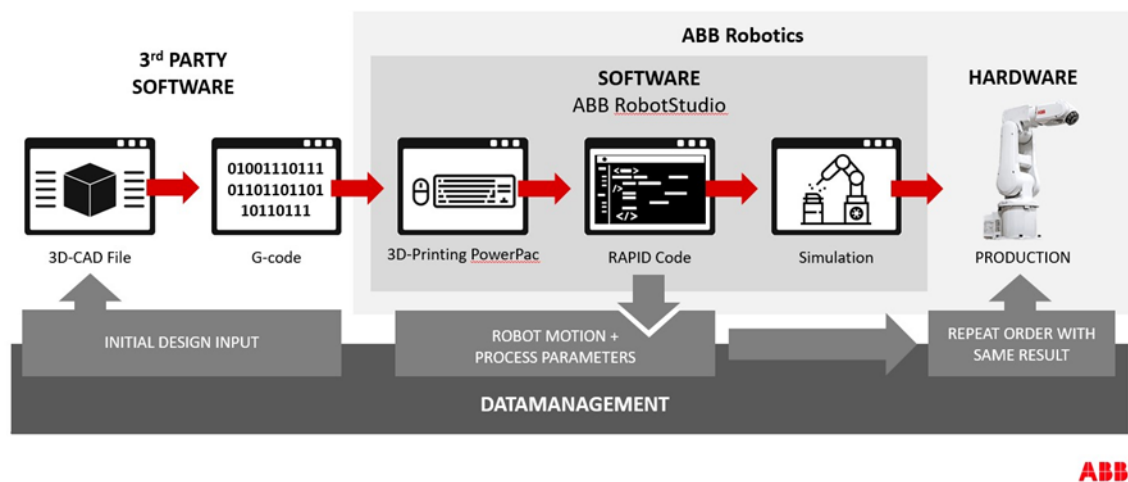
Despite this, prior to 3DP processing, there is a preference to uniformly dry those granules in a humidifier where they are kept at temperature of 50°C for one night in a dryer with the purpose of withdrawing humidity as much as possible (Picture 10). The reason behind pre-treatment and dehydration is that moisture makes discoloration and lowers mechanical properties of the material.

### 5.1.1 Printer specifications

The CEAD large-scale AM printer was a printer that was used for this study. The output of the printer equates to 12 kg per hour and maximum nozzle temperature is 400 °C. Moreover, there is a dryer and hopper which are connected with the single-screw extruder and robotic arm. The granule feeder is efficiently done automatically.

#### 3D Printing

Additive Manufacturing Process Flow



PICTURE 11. The process of 3DP from Computer Aided Design (CAD) file to production (CEAD n.d.)

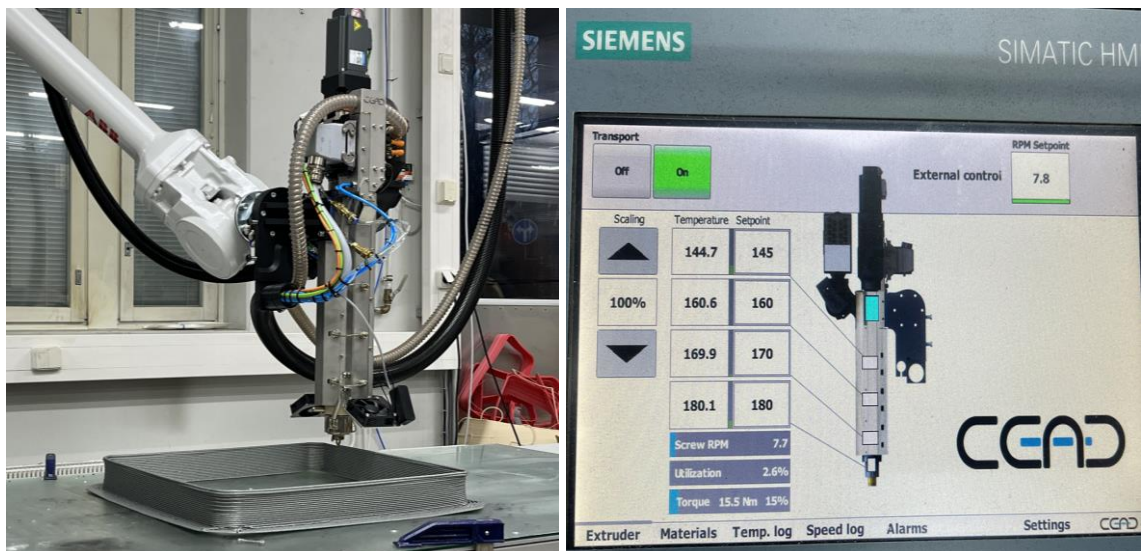
The CEAD LSAM has its own slicer software (Simplify 3D) to convert the Standard Triangle Language (STL) file into the corresponding G-code. As depicted in software-printer compliance (Picture 11), to completely utilise handy design options in AM, “automated design tools” should be used to assist the designer in producing effective frameworks. Essentially, a “design process” must include simulation within an optimization loop (Aboulkhair et al. 2019).

In the context of extrusion-based system, the dried Poly-Al granules are automatically sucked into the extrusion where materials were subjected to 4 setpoints of temperature in terms of witnessed experimental outcome. These temperature setpoints (heat zones) are controlled through the extruder control menu which was the main menu of the robot extruder interface (Picture 12). The control menu allows us to control granule transport to the extruder, heaters of the extruder, extrusion (individual four heat zones), Round Per Minute (RPM) setpoint of the screw in the extruder which is calculated through a formula below, current speed of screw in the extruder, status of cooling, material collectively. Hence, there are several variables that to be shifted and will affect our experiment with the Poly-Al material.

Setpoint formula (CEAD n.d.):

$$\text{Screw rpm setpoint} = \frac{(\text{Movement speed mm/min} * \text{Layer height (mm)} * \text{layer width (mm)})}{\text{material constant}^5 (\text{Typical between 1000 to 2000})}$$

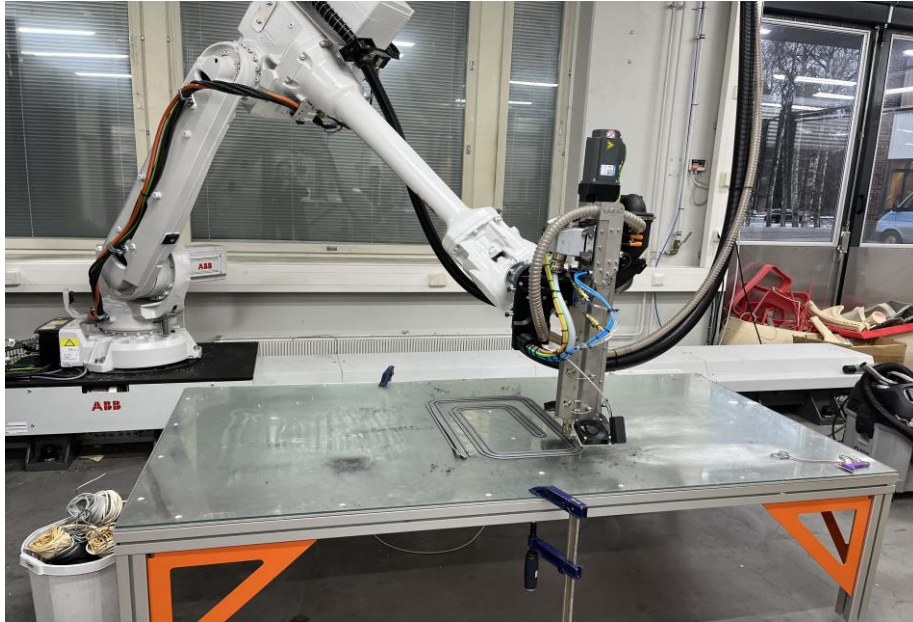
It should be noted that other settings must be selected in terms of material properties. In case of Poly-Al material, the name of material was selected as LDPE in the materials menu due to undefined Poly-Al and also high percentage of polymers in the matrix.



PICTURE 12. Extruder control menu and direct HMI control screen

<sup>5</sup> Basic PP should have a material constant of 1500. Notably, material constant is also lightly dependent on nozzle size (CEAD n.d.)

One challenge was observed throughout the 3DP Poly-Al granule would be that the material exhibited a poor adhesion with printing table/bed (Size of 2000\*1000\*1500) made of pure glass on top layer (Picture 13). To solve this, the Ply wood was tested but didn't get a decent result.



PICTURE 13. Poor adhesion between Poly-Al 3D printed part and the Glass

To improve adherence of 3D printed part and bed, as evidenced in (Picture 14) different adhesive tapes and fixing spray were used and tested on the printing table prior to 3D printing to achieve the best result and discovered which one makes much stickiness pertinent to this distinctive Poly-Al material. There was a speculation that this might be attributed to Aluminum particle, thereby causing weak adhesion here.



PICTURE 14. Different kinds of adhesive tapes and fixing sprays applied in 3DP

### 5.1.2 Poly-Al 3D printing profile

There are two major objectives for “3D printing process”: 1) to describe the “open-source 3D printer”, and 2) to determine the production specifications of the samples. A survey of relevant literature on the chosen features in the context of AM might provide some basic insight into crucial factors to consider (Sanchez et al. 2017).

One of the challenges in 3D printing composite materials is discovering how different materials respond to various parameters during both individual and combined processing. Katalagarianakis et al. (2022) highlighted that the quality of the printed composite is influenced by the printer settings. For instance, controlling the cooling conditions in printing is necessary to provide sufficient bond quality between neighbouring paths. This will be visible in the mechanical properties of printed segments.

In the following step of investigation, 3DP test on recycled Poly-Al material was carried out by CEAD 3D printer and an initial 3D printing profile was established. The effects of varying nozzle diameter and temperature control, turning off/on fan, print speed, layer time and height were studied on the recycled Poly-Al material, and by quantitatively, optimal printing parameters were determined. Table 4 enumerates optimal printing parameters, variables and observed effect of each variable on the quality of 3D printed parts. Owing to these crucial factors, extrusion of this material is a very sensitive process. The machine temperature and the process of cooling down must be kept consistent and superlative. Thus, the set up should be done step by step.

TABLE 4. Extrusion process parameters for 3D printing Poly-Al granule

Nozzle Diameter	Parameter	Variables	Effect
4 mm	Cooling system	On	Warping, high speed
	Cooling system	Off	No warping, low speed
	Temperature (°C)	145-160-170-180	Tensile strength reduction
	Temperature (°C)	145-165-200-210	Increased Tensile strength

	Print speed (mm/s)	20-25/50-100	Collapse at high speed
	Infill pattern	Yes	Heat dissipation
	Infill pattern	No	Deformation
	Layer height (mm)	2.4	High flexural strength
	Layer height (mm)	3.2	low flexural strength
	Screw extruder's speed (rpm)	12	Low flow
	Screw extruder's speed (rpm)	16	High flow
6 mm	Cooling system	On	Warping, high speed
	Cooling system	Off	No warping, low speed
	Temperature (°C)	145-165-170-180	Tensile strength reduction
	Temperature (°C)	145-165-200-210	Increasing tensile strength
	Print speed (mm/s)	20-25-50	Collapse at high speed
	Infill pattern	Yes	Heat dissipation
	Infill pattern	No	Deformation
	Layer height (mm)	2.4	High flexural strength
	Layer height (mm)	3.2	low flexural strength
	Screw extruder's speed (rpm)	12	Low flow
Screw extruder's speed (rpm)	16	High flow	
8 mm	Cooling system	On	Warping, high speed
	Cooling system	Off	No warping, low speed
	Temperature (°C)	145-165-170-180	Tensile strength reduction
	Temperature (°C)	145-165-200-210	Increasing tensile strength
	Print speed (mm/s)	10-25	Collapse at high speed

	Infill pattern	Yes	Heat dissipation
	Infill pattern	No	Deformation
	Layer height (mm)	4.8	High flexural strength
	Screw extruder's speed (rpm)	10	Low flow

By qualitatively, the 3D printing process was repeated until the material performance reached acceptable level, and observing a good alignment (intra-bonding) of 3D printed layers without surface defects.

Coupled with this, 3D printing from a simple geometry to complex geometry was implemented to observe the results. In this respect, there was a sweet spot when geometry with support structure or inner filling (even not visible but necessary) was opted. There was a sweet spot for a constant temperature from the feed throat to the die somewhere between 180°C/210°C and a screw speed somewhere between 8-16 rpm respectively. To complete the test, The nozzle diameters were in a range of 4-8 mm respectively.

It should be noted that shot size on control menu is defined in terms of material's density. For instance, the density of Poly-Al granule is 1.07 g/cm<sup>3</sup> (ISO1183). So, the shot side was set up to 8.0 (s).

To interpret the result of this experimental 3DP with Poly-Al (Recycled materials), Sanchez et al. (2017) asserted that in the printing process, the ultimate shape will depend on the viscosity of melt and surface where the layer is laid on. Also, the printed material will spread into an oblong shape based on functional parameters (e.g., nozzle speed, nozzle diameter, thickness of layer).

A flaw that influences the structural quality of printed parts is the voids created due to the rounded-shape of the deposited material. In layer alignment, the point where printing of each layer is started and stopped could be another considerable parameter for defect creation (Sanchez et al. 2017). Mostly, in case of 3D printed parts made of Poly-Al, each 3D printed layer was perfectly aligned and didn't

demonstrate any deformation including detachment, swelling, or warping, when we chose geometries with support structure.

The reason behind this phenomenon is that regarding thick bed grid, support structures are the most crucial factors when manufacturing 3D printed parts. "Support structure" can contribute preventing collapse while be printing because the elements of printed parts are subjected to collapse if they are not championed. "Support structures" also act as "heat dissipators" in manufacturing processes that include high temperatures, for instance, "metal 3D printing". To avoid deformation during printing, 3D-printed parts also require supports which must be considered when deciding how sustainable the process is (Javaid et al. 2021).

Aboulkhair et al. (2019) pointed out that adding support structure multiplies time and material consumption and cost. Despite this, reducing the use of "support structures" can cause negative impact on the performance functionality of the part. Therefore, the expenses related to support material and its overall consequences on the design should be carefully weighed by the designer.

When various "heating zones" are unstable or there are adverse environmental circumstances (such as cold wind through a window), parts of first extruded print may be below quality standards (Peeters et al. 2019). The bonding quality is significantly influenced by the thermal history of interfaces. Due to intrinsic nature of printing process, uneven heating and cooling cycles results in stress concentration in the built part, which is a chief cause of weak bonding and therefore impacts the robustness. For that reason, the toolpath and build part orientation are dependent on the mechanical properties (Sanchez et al. 2017).

Concerning effect of cooling on/off on quality of 3D printed parts, turning on cooling fan induces warping distortion which was concentratedly observed in the corners of 3D printed parts. Despite this, it influentially increases the layer speed.

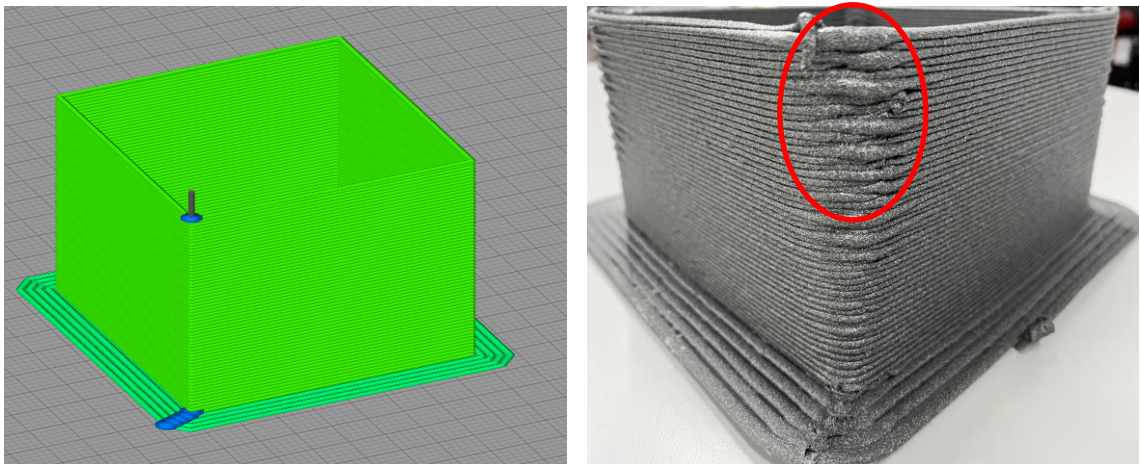
Regarding 3DP speed and temperature, there is a correlation between these two factors, and high nozzle diameter can exacerbate the effect, causing longer cooling time, thick walls. When both temperature and speed were high, Poly-Al which has intrinsically had high thermal expansion (Aluminum particle is commonly

recognized as a key material with high thermal conductivity), take longer time means lower cooling rate to dynamically solidify, resulting in numerous defects and parts with poor structural integrity.

In consequence, the experimental results seem to indicate that overall Poly-Al 3D printed parts exhibited a remarkable layer interfacial adhesion. It is mechanically characterized by high specific resistance, yielding enhanced mechanical properties might be attributed to Aluminum particles. But downside is high thermal expansion (CTE), high viscosity which decreases the material flow (MFI), thereby causing lower printing speed. So, if Poly-Al will be improved for better material process-ability, receiving significant attention of industries.

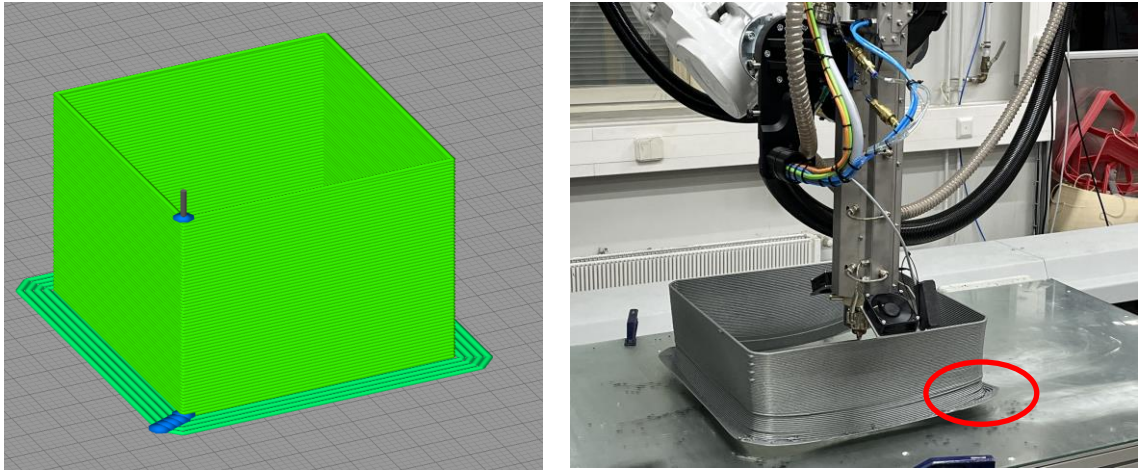
### 5.1.3 3D printing test simulation

In order to validate experimental 3DP test, main defects which were observed during 3DP Poly-Al material are highlighted in the photos. As described earlier, optimal parameters were set up to discover that Poly-Al could be a good option for use in engineering material applications.



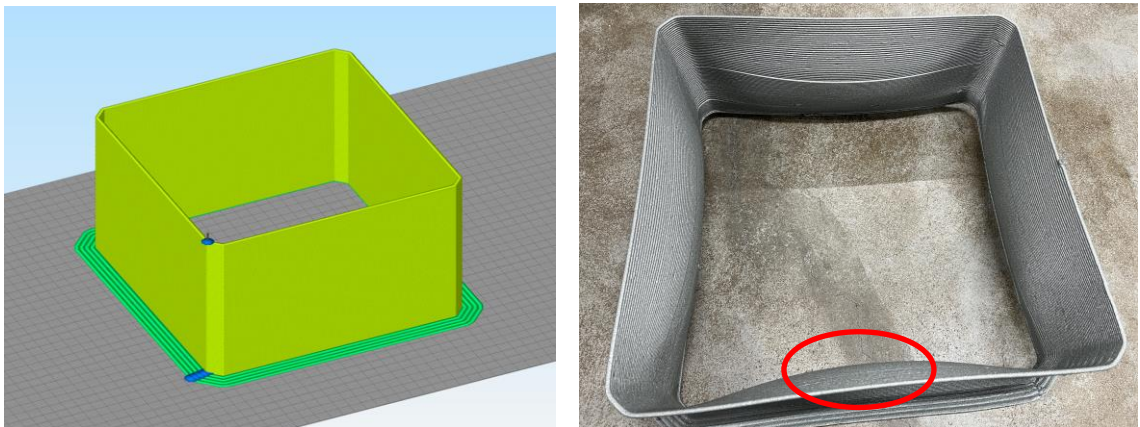
PICTURE 15. Simulation and 3D printed cube with nozzle 4 mm layer height of 2.4 mm

As depicted in (Picture 15), consequence of geometry effects without support structure and infill pattern, causing warping in one corner was evidently clear. Main defects are highlighted in the Picture series.



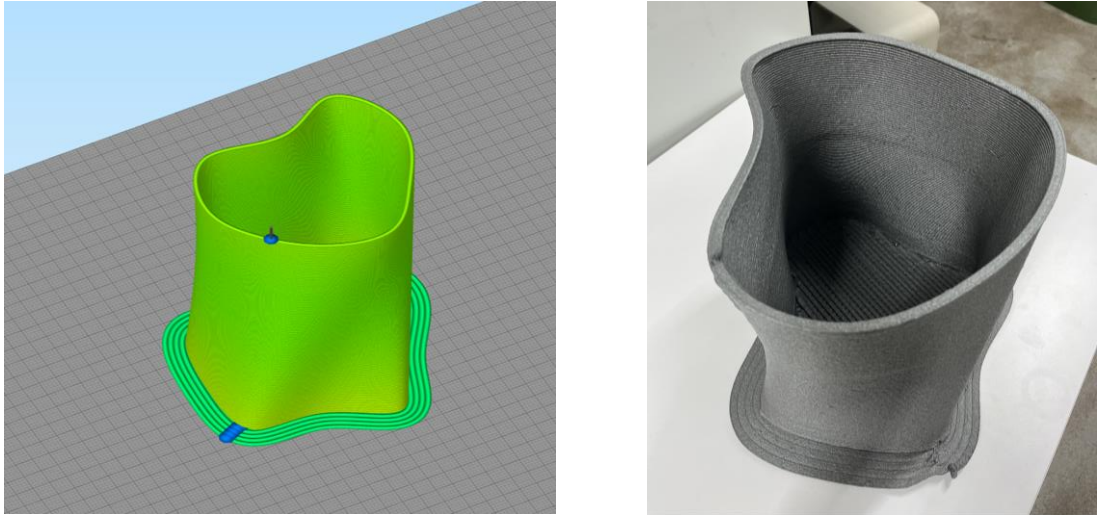
PICTURE 16. Simulation and 3D printed cube with 4 mm nozzle, layer height of 3.2 mm

As illustrated in (Picture 16), consequence of geometry effects without support structure and infill pattern, causing warping in more than one corners besides detachment of layer from the printing bed was perceived.



PICTURE 17. Simulation and 3D printed cube with 8 mm nozzle with layer height of 4.8 mm

As evidenced in (Picture 17), consequence of geometry effects without support structure and infill pattern, causing intense bending, layer misalignment on four sides was remarkably demonstrated. The effect of room temperature on material deformation was obvious.



PICTURE 18. Simulation and 3D printed vase-shaped with 6 mm nozzle, layer height of 3.2 mm

As shown in (Picture 18), To avoid shrinkage and warpage, the vase-shaped geometry with infill line pattern and 3 layers bottoms solid layers, defect-free part was fabricated by CEAD LSAM.

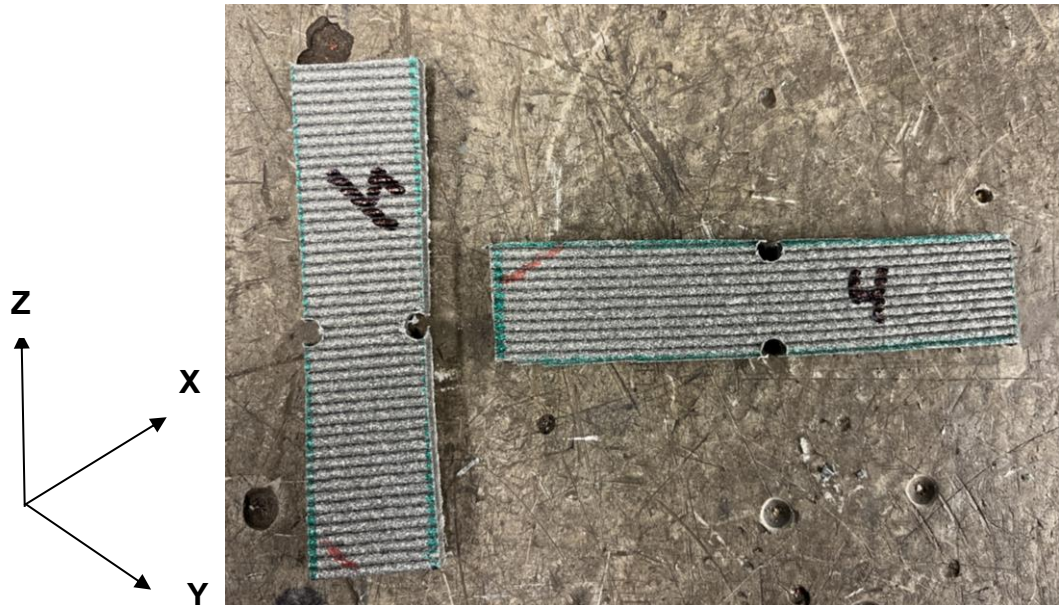
## 5.2 Tensile Strength Test

To measure tensile strength of 3D printed parts made of recycled Poly-Al, the test was conducted by tensile strength equipment (Picture 19), one specimen was tested for each nozzle diameter of samples produced. The measurement of the tensile properties of the printed parts was set according to the UNEEN ISO 527. All the specimens were fabricated to a dog-bone shape 100 mm in length and 20 mm in width.



PICTURE 19. Tensile strength test equipment at TAMK lab

First, tensile strength of 4mm nozzle diameter with the layer height of 2.4 mm was measured in a specific speed (10 mm/min) and maximum load of 200 Newton. Picture 20 illustrates two horizontal (as labelled XY) and vertical (as labelled XZ) cross-sections of 3D printed part with 4 mm nozzle diameter, 2.4 mm layer height and central dimensions of  $14.7 * 4.4 \text{ mm}^2$ .



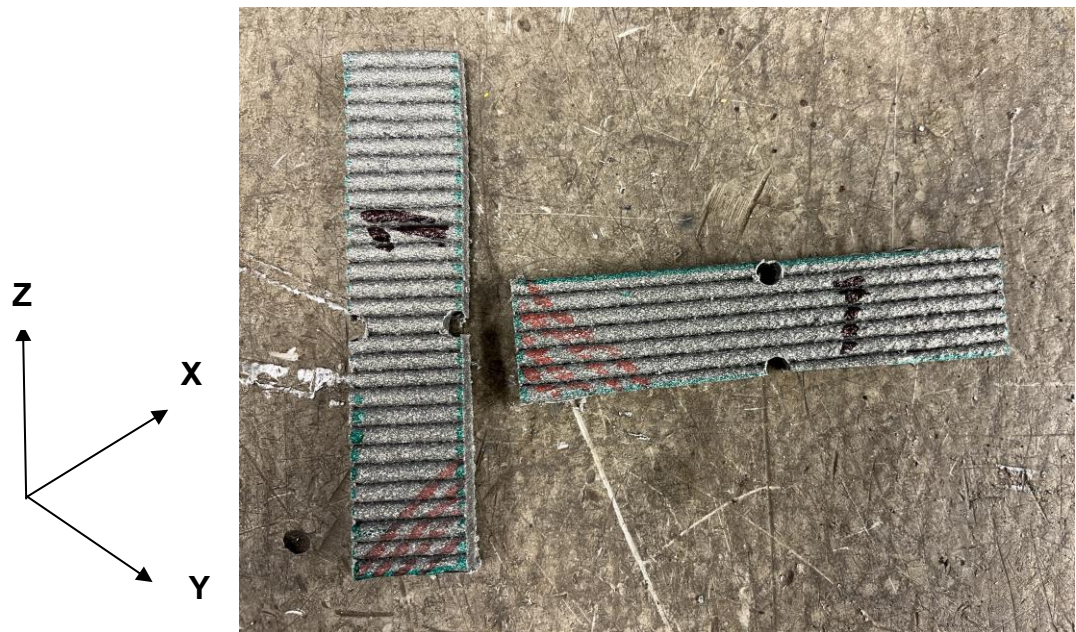
PICTURE 20. Cross-sections of specimen, 4mm diameter, 2.4 mm layer height

As Picture 21 shows two horizontal (as labelled XY) and vertical (as labelled XZ) cross-sections of 3D printed part with 4 mm nozzle diameter, 3.2 mm layer height and central dimensions of  $14.7 * 3.3 \text{ mm}^2$ .



PICTURE 21. Cross-sections of specimen, 4mm diameter, 3.2 mm layer height

As Picture 22 demonstrates two horizontal (as labelled XY) and vertical (as labelled XZ) cross-sections of 3D printed part with 6 mm nozzle diameter, 3.6 mm layer height and central dimensions of  $14.7 \times 3.7 \text{ mm}^2$ .



PICTURE 22. Cross-sections of specimen, 6mm diameter, 3.6 mm layer height

Picture 23 displays two horizontal (as labelled XY) and vertical (as labelled XZ) cross-sections of 3D printed part with 8 mm nozzle diameter, 4.8 mm layer height and central dimensions of  $14.7 \times 5.5 \text{ mm}^2$ .



PICTURE 23. Cross-sections of specimen, 8mm diameter, 4.8 mm layer height

In this part, the results obtained for the tensile strength and strain test in the case of specimens from 3D printed parts will be presented. In the diagram below, there is a curve in accordance with two axis of diagram stress-strain (Load-Extension). According to Sanchez et al.:

“The value can be measured by linear regression between load (Newton) and Extension (mm).

Tensile Strength (MPa): Maximum stress sustained by the test specimen.

Tensile strain (mm/mm): Strain at the moment of maximum stress.

Tensile strength at break (MPa): Stress at which it occurs the rupture.

Nominal strain at break (mm/mm): Strain at the moment of rupture”. (Sanchez et al. 2017).

To measure, the specimens were placed between two clamps which applied a maximum pressure of 1000 N load to prevent slipping. Then, the crosshead was displaced with a speed of 10 mm/min until the Poly-Al specimens failed. The initial gauge length of the specimen and the crosshead displacement determined the calculation of the strain. The tensile strength and tensile strength at break were calculated for each horizontal (as labelled XY) and vertical (as labelled XZ) specimen.

As (Figure 11) represents, when the stress of 200 Newton load was put on horizontal (as labelled XY) cross-section of 4 mm nozzle 3D printed specimen with 3.2 mm layer height, the deviation on extension axis occurred at 1.4 mm while the specimen was tolerating maximum load 168.8 N until it was broken at 2.4 mm. Consequently, the stiffness of this specimen was equated to 342k N/m.

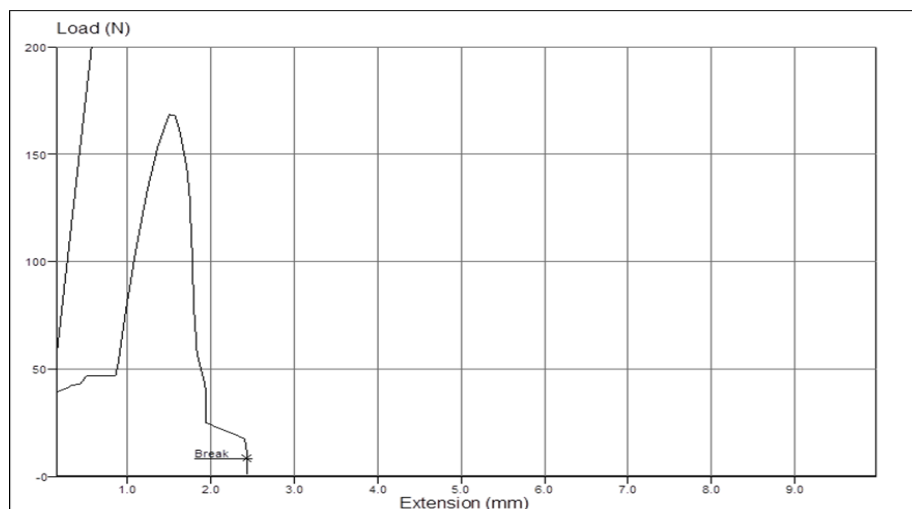


FIGURE 11. Tensile strength test, 4 mm diameter nozzle, 3.2 mm layer height (Horizontal)

Figure 12 demonstrates the moment when the stress of 1000 Newton load was applied on vertical (as labelled XZ) cross-section of 4 mm nozzle 3D printed specimen with 3.2 mm layer height, the deviation on extension axis happened at 5.1 mm while the specimen was tolerating maximum load 691.7 N until it was broken at 8.8 mm. By doing so, the stiffness of this specimen was estimated 550k N/m.

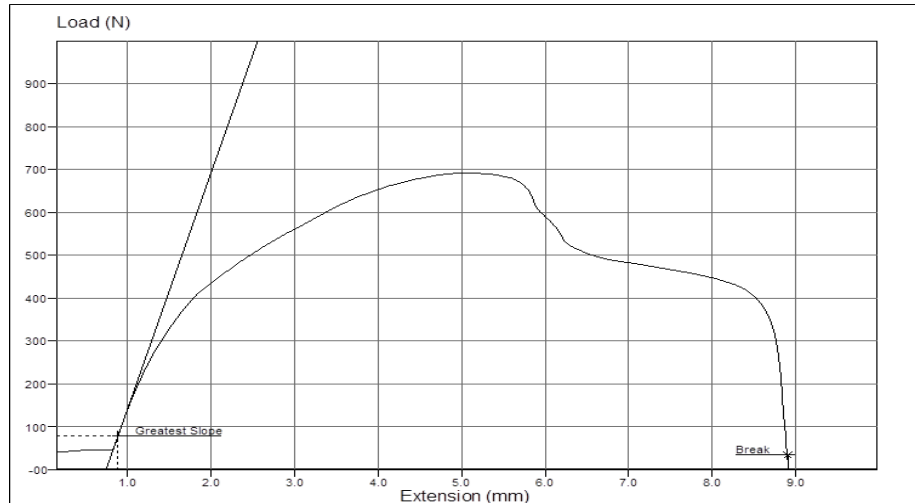


FIGURE 12. Tensile strength test, 4 mm diameter nozzle, 3.2 mm layer height (Vertical)

As (Figure13) exhibits when the stress of 500 Newton load was put on horizontal (as labelled XY) cross-section of 4 mm nozzle 3D printed specimen with 2.4 mm layer height, the deviation on extension axis took place at 1.4 mm while the specimen was tolerating maximum load 243.2 N until it was broken at 1.6 mm. Consequently, the stiffness of this specimen was equated to 428k N/m.

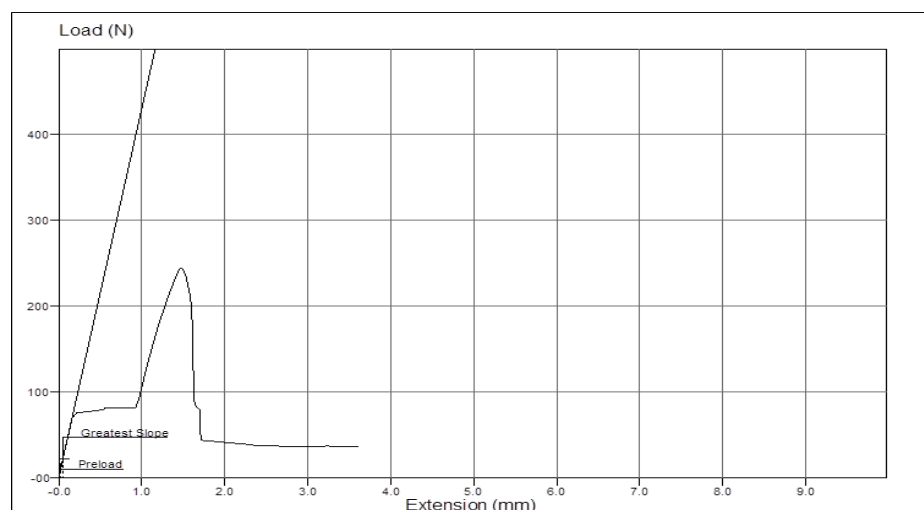


FIGURE 13. Tensile strength test, 4 mm diameter nozzle, 2.4 mm layer height (Horizontal)

Figure 14 presents the moment when the stress of 1000 Newton load was exerted on vertical (as labelled XZ) cross-section of 4 mm nozzle 3D printed specimen with 2.4 mm layer height, the deviation on extension axis occurred at 4.7 mm while the specimen was tolerating maximum load 789.0 N until it was broken at 7.0 mm. As a result, the stiffness of this specimen was estimated 646k N/m.

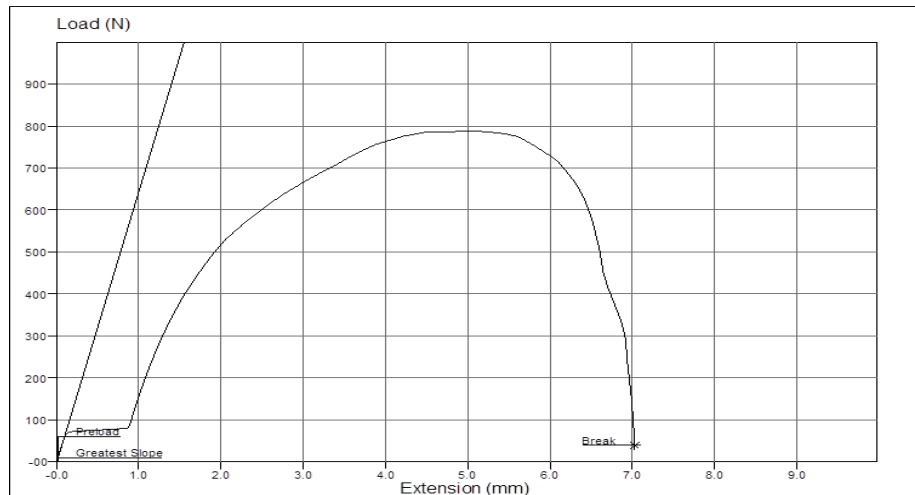


FIGURE 14. Tensile strength test, 4 mm diameter nozzle, 2.4 mm layer height (Vertical)

As (Figure15) describes when the stress of 500 Newton load was deployed on horizontal (as labelled XY) cross-section of 6 mm nozzle 3D printed specimen with 3.6 mm layer height, the deviation on extension axis emerged at 1.8 mm while the specimen was tolerating maximum load 288.4 N until it was broken at 1.9 mm. By doing so, the stiffness of this specimen was equated to 492k N/m.

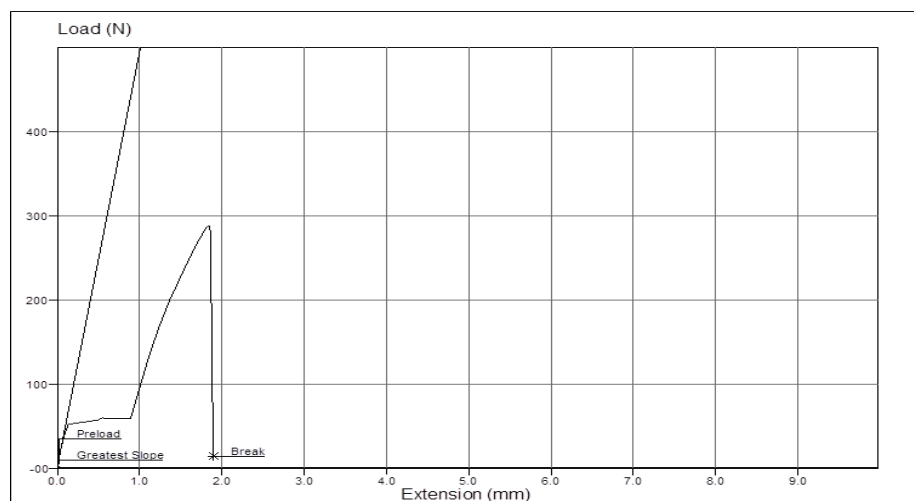


FIGURE 15. Tensile strength test, 6 mm diameter nozzle, 3.6 mm layer height (Horizontal)

Figure 16 introduces the moment when the stress of 1000 Newton load was put on vertical (as labelled XZ) cross-section of 6 mm nozzle 3D printed specimen with 3.6 mm layer height, the deviation on extension axis happened at 5.7 mm while the specimen was tolerating maximum load 737.0 N until it was broken at 9.2 mm. Consequently, the stiffness of this specimen was estimated 573k N/m.

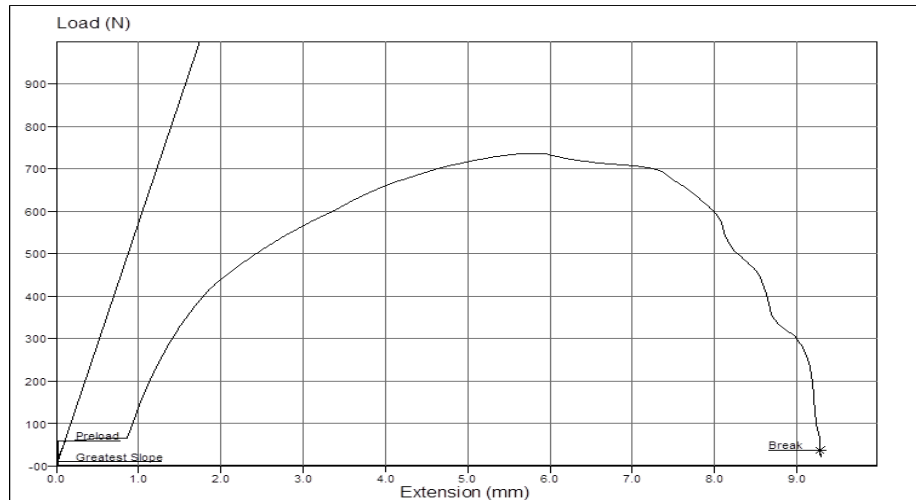


FIGURE 16. Tensile strength test, 6 mm diameter nozzle, 3.6 mm layer height (Vertical)

As (Figure17) illustrates, when the stress of 1000 Newton load was exerted on horizontal (as labelled XY) cross-section of 8 mm nozzle 3D printed specimen with 4.8 mm layer height, the deviation on extension axis occurred at 0.2 mm while the specimen was tolerating maximum load 510.8 N until it was broken at 3.4 mm. As a result, the stiffness of this specimen was equated to 497k N/m.

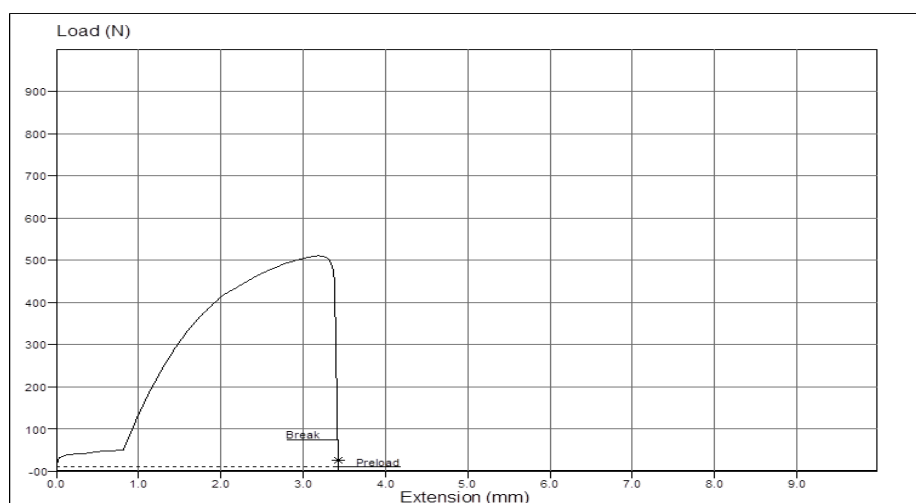


FIGURE17. Tensile strength test, 8 mm diameter nozzle, 4.8 mm layer height (Horizontal)

Figure 18 represents the moment when the stress of 1200 Newton load was applied on vertical (as labelled XZ) cross-section of 8 mm nozzle 3D printed specimen with 4.8 mm layer height, the deviation on extension axis took place at 7.0 mm while the specimen was tolerating maximum load 1042.6 N until it was broken at 9.6 mm. By doing so, the stiffness of this specimen was estimated 995k N/m.

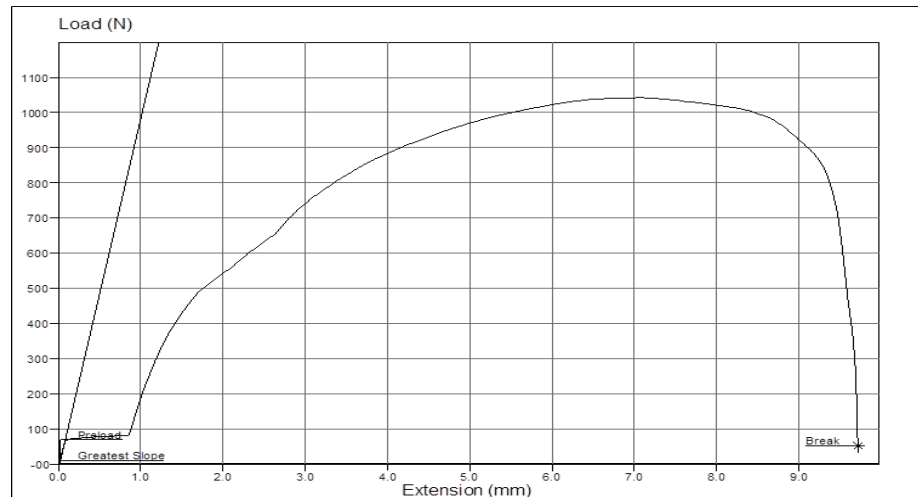


FIGURE 18. Tensile strength test, 8 mm diameter nozzle, 4.8 mm layer height (Vertical)

In summary, regardless of nozzle diameter and layer height as defined parameters, when tensile strength and strain of the two-printing orientation (e.g., XY and XZ) were compared, the vertical (as labelled XZ) specimens were almost twofold as much as horizontal (as labelled XY) specimens. This explained lower mechanical properties had obtained in XY orientation. In general, the mechanical properties of the XZ specimens showed higher values than those of the XY specimens. Comparing these two-layer heights of 4 mm nozzle with 2.4 mm and 3.2 mm, layer height didn't have a meaningful impact on increasing tensile strength. However, it had a positive impact on the tensile strain.

Moreover, it's crystal clear that by increasing the nozzle diameter, tensile strength had also been amplified accordingly. The reason behind high tensile strength might be attributed to presence of is Aluminum particles in the polymer matrix as a whole. By default, Aluminum particles have been distributed uniformly. Through experimental observation, Tensile strength and strain test indicated that mechanical properties of material had not reduced after two heating/extrusion processes despite of 100% use of recycled materials.

## 6 CONCLUSION

This study focused on using large-scale 3D printing by Fused Granular Fabrication method as part of material extrusion with Poly-Al material (recycled non-fiber fraction of beverage carton). The acquired result from this experimental study pointed out that 3D printing (Distributed recycling) could be strongly considered as a supportive practice for the recycling partner in order to increase recycling rate of Poly-Al as a wasted material. To diminish Poly-Al waste, it additionally contributes recycling partners to design, manufacture rapid prototyping and on-demand complex products by 3D printing process with the recycled materials for their clients in the market. Fused Granular Fabrication or direct extrusion was selected as a well-suited method for Poly-Al material because of numerous benefits e.g., lower cost, lower carbon emission, higher production speed, less heating cycles and higher availability of materials and larger quantity of recycled feedstock, resulting in time-money consumption. The challenge was that it was relatively difficult to anticipate a thermo-mechanical behavior of Poly-Al material. Given that it was composed of heterogeneous material, various polymers. By and large, Poly-Al 3D printed parts exhibited a remarkable layer interfacial adhesion. Furthermore, it was mechanically characterized by appreciable mechanical properties e.g., high specific resistance against corrosion, might be attributed to Aluminum particles in the polymer matrix. Arguably, the drawback was high “Coefficient Thermal Expansion” (CTE), high viscosity which had lowered the material flow (MFI), thereby causing lower printing speed. If Poly-Al will be improved for better material process-ability, consequential attention will be received by recycling industries for 3D printing purpose. Regarding added value proposition to the Poly-Al granule as a secondary material, the transforming granule to 3D printed parts could intrinsically bring added value to the recycling activity. Because, the recycled material in the new shape could be kept in use for a longer time and be used again and again. The value propositions for 3D printed parts as a decoration furniture included a high functionality for outdoor application, aesthetic features e.g., eye-catching surface texture due to the Aluminum particles which was offering a shining gray color to the end-product or complex organic shapes, fine-tune design for the designers.

## RECOMMENDATIONS

A possible circular solution in response to mass production of waste would be harnessing full capacity of both chemical and mechanical recycling. Distributed recycling in this case 3D printing and its techniques increases additional capacity beyond centralized recycling. The reason behind this is that distributed recycling is so versatile and functional in the sustainability field.

So, without a shadow of doubt, Fused Granular Fabrication as a direct extrusion is a well-suited method for Poly-Al material because of numerous benefits e.g., lower cost, lower carbon emission, higher production speed, less heating cycles and higher availability of materials and larger quantity of recycled feedstock, resulting in time-money consumption. For this purpose, the material should meet the requirements such a low “co-efficient of thermal expansion” (CTE), a satisfactory index of melt flow and fine mechanical properties, which the Poly-Al material didn't comply with this.

As obtained results validate the drawback of Poly-Al material was high “Co-efficient Thermal Expansion” (CTE), high viscosity which lowered the material flow (MFI), thereby causing lower printing speed. By and large, lower printing speed immensely affects both the quality and efficiency.

3D printed part made up of recycled material could contain lower “aesthetic quality” (surface finish) at the same time that it is functional. Demand for high quality could lead to perception problems associated to recycled materials. To gain much competitiveness, forming profitable circular business model, and be assured that material properties make it suitable for a range of applications, upgrading mechanical performance of material to functionally-graded material should be ascertained by researchers.

### **Adding reinforcement**

According to Katalagarianakis et al., “The combination of recycled fiber reinforcement with recycled polymer as a feedstock material for extrusion-based AM

creates an opportunity for a more sustainable material use” (Katalagarianakis et al. 2022). So, one solution would be making innovative materials by deploying material engineering knowledge.

Sanchez et al. (2022) asserted that loading polymers with fibers decreased the Co-efficient Thermal Expansion (CET<sup>6</sup>) and increases their stiffness, resulting in better mechanical behavior in response to internal tensions of the deposited material as it slightly cools down, as well as improved “heat dissipation” and a reduction in distorted parts like warping. Basalt Fibers (BF), carbon fibers (CF) and glass fibers (GF) are the most researched fibers for this function.

Also, previous works have shown that alignment of fibers with the loading direction or printing direction due to the fiber’s flow-induced orientation massively impact both tensile strength and modulus (Katalagarianakis et al. 2022). As described earlier, Poly-Al material was recognized as a high “co-efficient Thermal Expansion” (CTE) material, might be attributed to due to presence of uniform Aluminum particles in the polymer matrix, causing warping and defects limitedly. In this regard, Katalagarianakis et al. (2022) asserted that additionally, carbon fibers decrease distortion of the printed part through reducing the coefficient of thermal expansion. Shrinkage and warping occur to semi-crystalline thermoplastics as a result of their crystallization when cooling. With the cooling down of the deposited thermoplastic, the free volume drops, which is more noticeable for semi-crystalline materials as opposed to amorphous materials because of the higher density of the formed crystals. Since this can be troublesome for parts which require a high dimensional precision, it is recommended that shrinkage and warpage be prevented by including fillers into the feedstock polymer.

Coupled with this, Sanchez et al. (2022) discovered that due to the deficient adhesion between layers, “printing direction” has a strong influence on the “CTE”, which leads to unpleasant “reinforcement properties” when the material is vertically printed. For instance, increasing the Basalt Fiber content (5-10 wt%) results in a significant decline in  $\Delta L$  of the specimens tested for both XY and XZ specimens. This phenomenon is caused by the nature of Basalt Fiber, having an

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<sup>6</sup> CTE is usually measured by Thermomechanical analysis (TMA).

intrinsic “CTE” close to zero. When the two orientations are compared with each other, a noticeable anisotropy is consequently observed, with 60% lower  $\Delta L$  values for XY specimens.

There is history of adding up to 40% wt% short carbon fiber to the recycled PET or other polymers as a feedstock for FFF in the previous studies. According to what we have realized so far, no research has taken place on uniting the effect of CF on heterogeneous materials such as Poly-Al until now, neither for traditional manufacturing nor for AM technologies. Therefore, there is scant information on effect of fiber content's upper limit, percentage of CF weight and impressive length of CF on the mechanical performance of Poly-Al (Recycled material). Despite this, there is an assumption that adding carbon fiber to Poly-Al material can enhance viscosity, causing increased flow rate, thereby resulting high speed of 3D printing accordingly.

### **Gantry-based solution**

As evidenced earlier, large-scale 3DP with Poly-Al material was carried out by CEAD SLAM (AM Flexbot) which couldn't enable the researcher to control the room temperature (uncontrolled ambient temperature). Due to the presence of Aluminum (metal) particles in the polymer matrix and dynamic melt-solidification cycle of metals, making them hard to 3DP process.

CEAD company, is a technology supplier of 3DP equipment on the frontier of large scale of composite additive manufacturing. This company offers gantry-based solutions in which pellet/granule extrusion is carried out in a fully enclosed environment, resulting in controlled ambient temperature. Also, some gantry-based technologies e.g., Continuous Fiber Additive Manufacturing (CFAM) enables addition of continuous fiber to the extruder polymer. In addition, comparatively, Gantry system has a higher speed and enables more accurate printing than robot-based AM Flexbox (CEAD 2022). Because, in robot-based system, the printing speed is limited by the speed of robot arm whereas, in gantry-based system, higher speed of robot arm provides optimal layer adhesion time better for composite materials particularly for metal materials.

## **Viscosity-modification chemistry**

Recently, Milliken and Company, a worldwide manufacturing leader with a focus on materials science has offered latest viscosity modifier based on latest viscosity-modification chemistry. This modifier was designed to specifically assist rPP by doubling the melt flow rate for specific purpose. This serves to reduce energy consumption, enhances circularity and advances the goal of creating a healthier future (Milliken and Company 2022).

As we discussed earlier, some materials in the recycled Poly-Al matrix exhibited high viscosity and low flow rate during 3DP process. As a future scenario, viscosity modification most probably will go beyond one recycled material, and modify the whole recycled polymer matrix.

In the future, to unlock full potential of fiber-reinforced Poly-Al material, Fused Granular Fabrication by LSAM especially gantry-based solution will procure a lot of new possibilities in those where large 3D printed parts are demanded for outdoors application.

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## APPENDICES

### Appendix 1. Results of 3D printing test on Poly-Al material

TABLE 5. Tensile strength test results of specimens

Batch Reference	pre-defined layer height (mm)	cross-section	Breadth (mm)	Area (mm <sup>2</sup> )	Speed (mm/min)	Maximum Load (N)
4 mm	2.4	H <sup>7</sup>	14.7	64.6	10	243.2
4 mm	2.4	V <sup>8</sup>	14.7	64.6	10	789.0
4 mm	3.2	H	14.7	48.5	10	168.8
4 mm	3.2	V	14.7	48.5	10	691.7
6 mm	3.6-	H	14.7	54.3	10	288.4
6 mm	3.6	V	14.7	54.3	10	737.0
8 mm	4.8	H	14.7	80.8	10	510.8
8 mm	4.8	V	14.7	70.5	10	1042.6

TABLE 5.1 Tensile strength test results of specimens

Batch Reference	pre-defined layer height (mm)	cross-section	Deflection at Maximum Load (mm)	Stress at Maximum Load (MPa)	Work to Maximum Load (J)	Stiffness (N/m)
4 mm	2.4	H	1.4	3.7	0.1	428644.2
4 mm	2.4	V	4.7	12.1	2.3	646369
4mm	3.2	H	1.4	3.4	0.1	342058.3
4mm	3.2	V	5.1	14.2	2.2	550831.4
6mm	3.6	H	1.8	5.3	0.2	492456.1
6mm	3.6	V	5.7	13.5	2.7	573876
8mm	4.8	H	-0.2	6.3	-0.1	487241.3
8mm	4.8	V	7.0	14.7	4.9	995501.5

<sup>7</sup> "H" is abbreviation for Horizontal.

<sup>8</sup> "V" is abbreviation for Vertical.

TABLE 5.2 Tensile strength test results of specimens

Batch Reference	pre-defined layer height (mm)	cross-section	Load at Break (N)	Deflection at Break (mm)	Stress at Break (MPa)	Work to Break (J)
4 mm	2.4	H	26.1	4.6	0.3	2.5
4 mm	2.4	V	39.4	7.0	0.6	3.8
4mm	3.2	H	8.4	2.4	0.1	0.1
4mm	3.2	V	34.5	8.8	0.7	4.1
6mm	3.6	H	14.4	1.8	0.2	0.2
6mm	3.6	V	36.8	9.2	0.67	4.80
8mm	4.8	H	25.5	-0.002	0.31	-4.3E-05
8mm	4.8	V	52.1	9.6	0.7	7.4

## APPENDIX 2. LIST OF FIGURES

FIGURE1. Waste management stages and the impact of Industry 4.0 (Solid Waste Association 2021)

FIGURE2. Sustainable additive manufacturing ecosystem (Javaid et al. 2021)

FIGURE 3. Life cycle analysis in Additive Manufacturing and Conventional Manufacturing (Langeled et al. 2022)

FIGURE 4. Benefits of 3D printing technology (Thomas & Mishra. 2022)

FIGURE 4. The four flows to achieve circular objectives: Narrow, Slow, Regenerate and Cycle (Fraser et al. 2023)

FIGURE 5. Typical material structure of aseptic beverage cartons (Alliance for Beverage Cartons 2022)

FIGURE6. Generic routes for reprocessing Poly-Al fraction (Alliance for Beverage Cartons 2022)

FIGURE7. Key properties for quality assessment of recycled polymer (Sanchez et al. 2017)

FIGURE8. Multiple processing approach to evaluate thermo-mechanical degradation

FIGURE9. Content of  $Al_2O_3$  in various Aluminum waste, AD, AC, AF. AF is considered as a feedstock with the lowest yield of  $Al_2O_3$  with traces of impurities (Mohamad et al. 2022)

FIGURE10. Typical mechanical recycling approach used in material extrusion (MEX) (Gomes et al. 2022)

FIGURE 11. Tensile strength test, 4 mm diameter nozzle, 3.2 mm layer height (Horizontal)

FIGURE 12. Tensile strength test, 4 mm diameter nozzle, 3.2 mm layer height (Vertical)

FIGURE 13. Tensile strength test, 4 mm diameter nozzle, 2.4 mm layer height (Horizontal)

FIGURE 14. Tensile strength test, 4 mm diameter nozzle, 2.4 mm layer height (Vertical)

FIGURE 15. Tensile strength test, 6 mm diameter nozzle, 3.6 mm layer height (Horizontal)

FIGURE 16. Tensile strength test, 6 mm diameter nozzle, 3.6 mm layer height (Vertical)

FIGURE17. Tensile strength test, 8 mm diameter nozzle, 4.8 mm layer height  
(Horizontal)

FIGURE18. Tensile strength test, 8 mm diameter nozzle, 4.8 mm layer height  
(Vertical)

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### Appendix 3. LIST OF PICTURES

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PICTURE 7. Input material into extruder machine (left) and extruder temperature profile (right)

PICTURE 8. Cooling round-shape granule by cold water (left) and sieving, separating granules from each other (right)

PICTURE 9. Granule bagging station (left) granule as an end-product (right)

PICTURE 10. Dehydrating of Poly-Al granule in a drier's tank prior to 3DP processing

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PICTURE 14. Different kinds of adhesive tapes and fixing sprays applied in 3DP

PICTURE 15. Simulation and 3D printed cube with nozzle 4 mm layer height of 2.4 mm

PICTURE 16. Simulation and 3D printed cube with 4 mm nozzle, layer height of 3.2 mm

PICTURE 17. Simulation and 3D printed cube with 8 mm nozzle with layer height of 4.8 mm

PICTURE 18. Simulation and 3D printed vase-shaped with 6 mm nozzle, layer height of 3.2 mm

PICTURE 19. Tensile strength test equipment at TAMK lab

PICTURE 20. Cross-sections of specimen, 4mm diameter, 2.4 mm layer height

PICTURE 21. Cross-sections of specimen, 4mm diameter, 3.2 mm layer height

PICTURE 22. Cross-sections of specimen, 6mm diameter, 3.6 mm layer height

PICTURE 23. Cross-sections of specimen, 8mm diameter, 4.8 mm layer height

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TABLE 2. Initial characterization of Poly-Al granule material

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TABLE 5. Tensile strength test result of specimens

TABLE 5.1. Tensile strength test result of specimens

TABLE 5.2. Tensile strength test result of specimens