

# POLITECNICO MILANO 1863

# School of Industrial and Information Engineering Master of Science in Automation and Control Engineering

# Sustainability and Mechanical Performance Analysis of Recycled Filaments for 3D Printing Applications

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### NOMENCLATURE

In order of appearance:

Microplastics	MPs
Thermoplastics	TP
Polyurethanes	PU
Thermosets	TS
Polypropylene	PP
Polyethylene Terephthalate	PET
Polyamide	PA
North American Free Trade Agreement	NAFTA
United States of America	USA
Commonwealth of Independent States	CIS
Norway	NO
Switzerland	СН
Overseas Development Institute	ODI
Temperature in Celsius	°C
Plastic Solid Waste	PSW
Carbon Dioxide	CO2
Nitrogen-Oxides	NOx
Sulphur-Dioxide	SO2
Greenhouse Gas	GHG
Million tonnes of Carbon Dioxide equivalent	Mt CO2e
Fourier-transform near-infrared	FT-NIR
Fused Deposition Modelling	FDM
Additive Manufacturing	AM
Stereolithography	SLA
Rapid Prototyping	RP
STereo Lithography interface format	STL
Three-Dimensional	3D
Computer Numerical Control	CNC
Digital Light Processing	DLP

Powder Bed Fusion	PBF
Research and Development	R&D
Large Scale Additive Manufacturing	LSAM
Centimetres Cubic	cm <sup>3</sup>
Circular Economy	CE
Selective Laser Melting	SLM
Ultra-Fine Particles	UFPs
Laser Material Deposition	LMD
General Electric	GE
Direct Metal Laser Sintering	DMLS
Poly-Ethylene Terephthalate Glycol	PETG
Polylactic Acid	PLA
Polymethylmethacrylate	PMMA
Polypropylene	PP
Nylon	PA
Business-to-Business	B2B
Business-to-Costumer	B2C
Millimetres	mm
Seconds	S
Millimetres per second	mm/s
Revolutions per minute	rpm
Life Cycle Analysis	LCA
High-Density Polyethylene	HDPE
Kilowatt hours	kWh
Mega Joule	MJ
Design of the Experiment	DoE
Ultimate Tensile strenght	UTS
Young's Moduls	E
Strain At Break	εMax
Yield Strenght	Rp02

### ABSTRACT

Additive Manufacturing (also referred to "AM" or 3D printing) is claimed to be a disruptive technology that is changing modern industry, with multiple benefits for the development and the manufacturing of a large variety of products. Moreover, during the last decade, AM has proven to be the leading technology in terms of sustainability, especially for the production of plastic-made parts. In fact, since plastic production and usage has drastically increased over the last fifty years, causing multiple irreversible environmental problems, a new way of managing the waste and consumption of synthetic polymers was needed, and AM could lead to the change required in order to keep our environment safe.

This study wants to underline the sustainable characteristics of AM, by analysing the case study of Reflow, a recycled filament producer for Fused Deposition Modelling (FDM) printers. The following research calculated the emission generated by the Reflow production line in Netherlands and compared them with the ones related to the primary plastic production of plastic and first recycling. Results showed how the Reflow production has low impact to the environment, especially due to the energy green sources used by the company.

Moreover, tensile and mechanical tests on recycled and virgin filaments were performed, showing that the first-recycled filaments produced by Reflow do not lead to a loss in mechanical performance, but on the contrary presented better test results overall compared to the virgin ones, proving that a circular economy could be reached without losing in performance.

# 1. PLASTIC RECYCLING AND ADDITIVE MANUFACTURING 1.1 THE PLASTIC POLLUTION PROBLEM

#### **1.1.1 PLASTIC PRODUCTION**

Plastic materials, also referred to as synthetic polymers or plastics, are an industrial product made by the joining of polymeric molecules in long chains in order to obtain the desired characteristics (Scott,1999). This type of materials is characterized by a huge variety of mechanical and chemical proprieties, enabling a wide range of applications in different industries, from the high-performance ones, as automotive and aerospace, to the food packaging production (Cruz Sanchez et al., 2020). Even if the importance of plastic is evident in our everyday life, since almost every product is made with certain amount of plastic or is covered by a plastic packaging, a critical view on the production and waste generation of synthetic polymers has to be made.

According to Al Salem et al. (2009) the first industrial scale manufacturing of plastic materials started in the 1940s, and from those years the production, consumption and waste generation of synthetic polymers has always increased. Moreover, over time, the technological development in all the industries gave as results a high-quality plastic product, with a significant increase of durability and resistance of plastic materials to the external environmental conditions. The low-cost production, combined with the high durability and easy modelling of plastics made this type of materials essential for every type of industry, but on the other hand has caused several environmental problems. To get an idea of how much plastics is present in our everyday life, just think that in 2017 the plastics global market was valued at \$523 billion and was forecast to grow to \$721 billion by 2025 (Grand View Research, 2019). Moreover, considering the whole value chain into account, from raw-material production to the conversion to product and the successive waste management and recycling, revenue in the sector in 2015 was estimated to be equivalent to 3% of the world total economy (Ryberg et al., 2018).

Environmental problems related to plastics are due to three main factors: the nature of synthetic polymers, the energy needed for their production and the low life cycle that usually addressed for products made with this type of materials. Plastics are made from inorganic and organic raw materials as carbon, silicon, hydrogen, nitrogen, oxygen and chloride, also involving a huge consumption of gas and oil used for their extraction (Seymour RB, 1989). In fact, as reported by Hopewell et.al, in 2009 the 4 % of world's oil and gas production was used as feedstock for synthetic polymers and another 3-4 % was used for providing energy for their production. As results, production of plastics

generates huge amounts of CO<sub>2</sub>, contributing to the 4% of global Greenhouse Gas (GHG) emissions in 2015 (Zheng and Suh, 2019). Looking at those numbers, it is important to underline that not only oil and gas are fossil fuels, thus a non-renewable source that contributes to air's pollution, but also that the major portion of plastic made is used to produce short-lived products that have a life cycle of less than one year.

Plastic pollution affects the most of habitats present in our earth, ruled by fragile relations. It has been established that plastic materials are the cause of the 60 to 80% of the entire marine pollution (Gregory and Ryan, 1997) and that the 10% of the total mass of plastics produced every year ends and accumulates in the ocean (Barnes et al., 2009), creating "islands" of plastic, such as the Pacific Trash Vortex showed in Figure 1, that causes huge effects on the marine global ecosystem.



Figure 1- The Pacific Trash Vortex

Moreover, the effects of plastic refuse generation are not limited to the marine environment or air pollution. As pointed by Yooeun et al. (2018) plastic waste can be found in form of microplastics (MPs) also on lands and soils, but almost nothing is known about it. In general, Nizzetto et al. (2016) stated that approximately the 16–38% of the heavier than water MPs hypothetically added to soils are predicted to be stored locally. In a river, the MPs with a dimension lower than 0.2 mm are generally not retained, independently from their density, while larger MPs with densities marginally higher than water can be retained in the soil.

Nowadays, an overview made every year by Plastic Europe (Plastics – The Facts, 2020) reports and analyses all the data on plastic materials production, demand, waste and recycling of the previous year. The following data includes information on thermoplastics (TP), polyurethanes (PU), thermosets (TS), elastomers, adhesives, coatings, sealants and Polypropylene (PP)-fibers, while Polyethylene Terephthalate (PET)-fibers, Polyamide (PA)-fibers and Polyacryl-fibers are not included. As showed in Figure 2, the world's plastic production in 2019 amounted to 368 million tonnes, with an increase of 2,5% with respect to the previous year. However, the distribution of global plastics manufacturing is not uniform across all continents: 51% of synthetic polymers last year were made by Asian countries (31% just in China), 19% by the NAFTA (North American Free Trade Agreement made by USA, Canada and Mexico in the 1992), 16% in Europe, 7% in Middle East and Africa, 4% in Latin America and 3% by the CIS (Commonwealth of Independent States signed between Russia, Belarus and Ukraine in 1991). Fortunately, in 28 European country plus Norway (NO) and Switzerland (CH), thanks to sensibilization campaign made during the latest years against plastic pollution and the Paris Agreement (2015) on climate change, the plastic production decreased of 6,5%, passing from 61.8 (2018) to 57.9 million tonnes (2019).



Figure 2 - Plastic Europe Report (2020)

Another important data to consider is the plastic demand by sector. Hopewell et al. in (2009) established that the 50 per cent of the world's plastics made that year were involved for single-use applications (packaging, disposable items, etc.), between 20 and 25% for long-term infrastructure (pipes, cable covers for construction, etc.) and remainder for durable consumer applications with an intermediate life cycle (electronic items, furniture, vehicles equipment, etc.).



Figure 3 - Plastic Europe Report (2020)

Eleven years later, the situation in Europe has not changed that much. The total demand amount last year was of 50.7 million tonnes, and the Figure 3 clearly shows that almost the 40% is still related to packaging, thus to a production that generates mostly single use products that affects significantly our environment.

This introductory overview showed how, even if during the last year in Europe the plastic production decreased, revealing an increasing interest by governments and consumers to the plastic pollution problem, the world's amount of plastic produced every year is still increasing and too high for reaching the sustainability goals discussed during the Paris Agreement in 2015. In fact, as reported by the Overseas Development Institute (ODI) in an analysis conducted this year, emissions from plastics are going to increase threefold by 2050, dashing any hopes of limiting global warming to 1.5 °C, above pre-industrial levels. However, the study suggests that halving plastic consumption within three decades, recycling 75% of remaining plastic, and decreasing the amount produced without fossil fuels, could cut emissions from plastic from 1,984 million tonnes of carbon dioxide equivalent (Mt  $CO_2e$ ) in 2015 to 790 Mt  $CO_2e$  in 2050 (ODI, 2020). Therefore, the plastic industry should re-invent

itself looking both to the benefits for today and the needs for tomorrow, trying to use renewable sources for the production and creating recycling routes for plastic solid waste (PSW).

#### **1.1.2 PLASTIC RECOVERY**

As explained in Sub-Section 1.1.1, the life cycle of plastics starts with the elaboration of natural resources, like oils and gases, that are transformed into plastic materials. The resulting synthetic polymers are further processed until they reach the shape and properties of the desired product, such as food packaging, cable covers or car's equipment. As showed by Figure 4, regardless their use, every product will reach the end of his first life, opening different routes to the management of the PSW. This Chapter will analyse what are the different possibilities for plastic recovery and which ones are the more suitable for reducing the impact of plastic pollution on our environment.



Figure 4 - Lifecycle of Plastic Products

#### **1.1.2.1 Production Reduction and Re-Use**

The most direct route for recovering plastics, consisting in reducing consumption of plastic products and fossil fuels emission related to them, is essential, but almost impossible without a strong stance of governments towards the plastic industry and a deep sense of conscience of both consumers and producers. In general, over history, the climate agreements for reducing worldwide emissions stipulated by the United Nations (UN) have always constitute one of the most complex multilateral process to be agreed. The first one was the Kyoto Protocol, signed in 1997 but entered into force only in 2005, demonstrating the difficulties in reaching an agreement on common goals and responsibilities. In short, the protocol operationalized the UN Framework Convention on Climate Change by committing industrialized countries to limit and reduce GHG emissions in accordance with agreed individual targets. The Convention itself only asked those countries to adopt policies and measures on mitigation and to report periodically. These targets added up to an average 5% emission reduction compared to 1990 levels over the five-year commitment period 2008-2012 (UN, 2008). Successively, in 2012, the Doha Amendment to the Kyoto Protocol was adopted for a second commitment period, starting in 2013 and lasting until 2020. However, the amendment has never entered into force because a total of 144 instruments of acceptance were required and never reached. Nowadays, the Paris Agreement signed in 2015 and entered into force in 2016, adopted by 196 Parties at the Conference of the Parties (COP), is the first binding agreement that brings all nations into a common cause for the environment. In order to undertake the ambitious efforts to combat climate change and adapt to its effects, the goal is to limit global warming to 1.5 degrees Celsius compared to pre-industrial levels. However, even if according to a recent study of Climate Action Tracker (2020) the Paris Agreement is driving the climate action, since that the temperature spikes predicted for the end of the century (2100) are decreasing from 3.5 C° (2015) to 2.9 C° (2020), the real-world action is still far from meeting long-term temperature goals of limiting warming to 1.5 C°.

In addition, over reducing consumption, re-using the same products should be the parallel sustainable way for consumers in order to prevent pollution, but not always is easy to change the habit of people, that are used to buy products and use them just for one life cycle. Al Salem et al. (2009) pointed that re-using plastic is preferable to recycling as it uses less energy and fewer resources, with multiple advantages as conservation of fossil fuels, reduction of energy and PSW and reduction of carbon-dioxide (CO<sub>2</sub>), nitrogen-oxides (NOx) and sulphur-dioxide (SO<sub>2</sub>) emission.

#### 1.1.2.2 Landfill

Other routes for reducing the impact of plastics on climate change are not related to the consumption or production of synthetic polymers, but on the management of the already existing PSW. Landfill is the most conventional approach for managing the plastic waste from decades, but during the last years the counter indications for this type of treatment have started to emerge. In fact, according to Hopewell et al. (2009) the space for landfills is becoming scarce for all countries, and even a well-managed one causes impacts on the environment in term of energy consumption for the collection and transport. Moreover, landfills can create long-term risks of contamination of the soils and groundwater (Oehlmann et al.,2009) and during the process none of the material resources used to produce the plastic is recovered, creating a material flow that is linear rather than cyclic.

The current uselessness of landfills is moreover justified by Plastic Europe, that warns how zero landfilling is required in order to reach a circular economy of plastic materials and shows the decreasing usage of landfills around the continent during the last decade. In fact, in 2018, 29.1 Mt of plastics were collected in order to be treated at the end of their lifecycle, but only the 24.9% was destinated to landfills, with higher percentage for energy recovery (42.6%) and recycling (32.5%).



Figure 5 - Plastic Europe Report on Recycling (2020)

It is important to underline that during the period between 2006 and 2018 the total amount of plastic recovered in Europe increased by the 19%, while the usage of landfills decreased by the 44%, due to technological improvements in energy recovery (+77%) and recycling (+100%). Another factor that influenced the usage reduction of landfills are the restrictions implemented by governments. As showed in Figure 5 all the countries that have imposed restrictions on landfills creation are able to use that waste in order to recover energy.

#### 1.1.2.3 Energy Recovery

The Energy Recovery process, also called incineration, allows to produce energy in form of heat, steam or electricity by burning the PSW. Since plastic materials when burned possess a calorific value comparable to the one of gas oil, heavy oil or petroleum (Al Salem et al., 2009) they are widely used in this type of process. The useful energy recovered can vary considerably depending on whether it

is used for electricity generation, combined heat and power, or as solid refuse fuel for co-fuelling of blast furnaces or cement kilns. Moreover, the incineration results in a 90/95% volume reduction of the PSW, that leads in a reduction of the usage of landfills as a way for the management of plastics waste.

However, different enviroment concerns are related to the incineration of plastics, like the emission of pollutants as CO<sub>2</sub>, NOx and SO<sub>2</sub>. In fact, according to Al Salem et al. (2009), the combustion of PSW is known to generate volatile organic compounds (VOCs), smoke, particulate-bound heavy metals, polycyclic aromatic hydrocarbons (PAHs), polychlorinated dibenzofurans (PCDFs) and dioxins. Also, some carcinogenic substances (as PAHs, nitro-PAHs, dioxins, etc.) have been identified in airborne particles from incineration of plastic materials such as PVC, PET, PS and PE. Despite environmental problems that seems are generated with every process that works with plastics, according to Hopewell et al. (2009) energy-recovery processes may be the most suitable way for dealing with highly mixed plastic such as some electronic wastes and automotive shredder residue. This reason, combined with the reducing use of landfills, is why the number of incinerators in Europe has increased a lot (Figure 1.1.2.2.1) during the last decade, and that is why together with recycling are the most valid way for PSW management.

#### 1.1.2.4 Recycling

By definition, the primary recycling process is the re-introduction of scrap, industrial or singlepolymer plastic edges and parts to the extrusion cycle in order to produce products of the similar material (Al-Salem, 2009). However, the terminology for plastics recycling is not limited to the primary one but depends on the wide range of recovery and recycling activities.

ASTM D5033 definitions	equivalent ISO 15270 (draft) definitions	other equivalent terms
primary recycling	mechanical recycling	closed-loop recycling
secondary recycling	mechanical recycling	downgrading
tertiary recycling	chemical recycling	feedstock recycling
quaternary recycling	energy recovery	valorization

Figure 6 - ASTM D5033 Standards

According to the ASTM D5033 standards and as reported in Figure 6 (Hopewell et al.,2009) the plastics recycling processes can be divided in four main categories:

- *Primary Recycling*: Mechanical processing of PSW into products with properties that are equivalent to the original plastic material. Currently, this process is the most used worldwide. In fact, 2019, according to Plastic Europe, close to 5 million tonnes of primary recycled plastic was produced in European recycling facilities and more than the 80% was re-introduced in the European industries in order to manufacture new products, while the rest was exported outside Europe to re-enter in other regions of the world's economies. Specifically, the 46% of plastic recyclates used in new products in Europe was addressed to the building and construction sector, the 26% for packaging industry and the rest lower percentages for agriculture, automotive and electronical sectors.



Figure 7- Plastic Life Cycle (Plastic Europe, 2020)

Since recycling plastics via mechanical way involves a large number of treatments and preparation steps, with a costly and an energy intense process to be considered, recyclers try to reduce these steps and working hours as much as possible. The flow of this type of process is explained in Figure 7, starting from the collection and separation of the end-of-life products into the different material and polymer categories to the shredding, washing and extrusion processes that allows to gain the final recycled plastic product in form of pellets, ready to use for manufacturing new products. In detail, the mechanical recycling process presented by Plastic Europe is divided in six steps:

**1) Collection**: The first step consists in the collection of the PSW from industries and cities, separating it from other mixed waste streams. Usually, there are two main types of collection process, by "bring- schemes" or through kerbside collection. The brings-schemes method results in a specific

low volume collection and works only if there is a highly committed public behaviour or with an imposition of a direct economic bonus by single countries in order to incentivize people to participate. The kerbside collection works better for a large volume collection and is more used in all the European countries, even if it is important to develop effective 'on-the-go' and 'office recycling' collection schemes, since a lot of PSW addressed to the single consumer is produced away from home.

**2) Sorting**: A first sorting is mandatory in order to separate plastics from others streams of waste (paper, dust, aluminium and other types of impurities) and to divide the different categories of polymers. In general, it is possible to closed loop recycle most thermoplastics, however, some types of PSW as packaging frequently uses a wide variety of different polymers and other materials such that increases the difficulty of the overall process. The majority of recyclers use automatic systems, generally Fourier-transform near-infrared (FT-NIR) spectroscopy for polymer type analysis and optical colour recognition camera systems to sort the streams into clear and coloured fractions (Hopewell et al., 2009).

**3**) **Shredding**: Rigid pieces of plastic of the same polymer are milled together into flakes. This is usually the first step for all recycles around the world, since the PSW arrives to them already separated.

**4) Washing**: Plastic flakes are cleaned in order to remove impurities as food residues or adhesives. Generally, this stage is executed with water, but in some cases (usually for glue removal) also chemical washing is adopted using caustic soda and surfactants (Al-Salem et al, 2009). According to Hopewell et al. (2009), wash plants use 2–3 cubic meters of water per tonne of material, about one-half of that of previous equipment, while more innovative technologies for the removal of small organics and surface contaminants include a dry cleaning, a process that cleans surfaces through friction without using large amounts of water.

**5**) **Control**: Before sending flakes to extrusion, those are controlled and sorted again to avoid contamination of the final product.

**6) Extrusion**: Cleaned plastic flakes of the same polymer are finally extruded and converted into pellets, used in the manufacture of new products. For example, in Reflow recycled pellets are colored and re-extruded in order to produce filament for Fused Deposition Modelling (FDM) 3D printing applications.

- Secondary Recycling: Mechanical processing of PSW into products with properties that are lower with respect to the original plastic material. For this reason, even if the mechanical process is equal

to the primary recycling, the secondary recycling is also called downgrading, and is usually performed on waste that was already recycled with a primary process.

- *Tertiary Recycling*: Chemical conversion of plastic waste into smaller molecules, usually liquids or gases, which are suitable for use as a feedstock for the production of new petrochemicals and plastics (Mastellone, 1999).

- Quaternary Recycling: Energy Recovery process explained in Sub-Section 1.1.2.3.

### 1.2 ADDITIVE MANUFACTURING AS A ROUTE FOR A SUSTAINABLE MANUFACTURING

#### **1.2.1 INRODUCTION TO ADDITIVE MANUFACTURING**

Three-dimensional (3D) printing technologies, also addressed as Additive Manufacturing (AM), enables the production of solid objects from a digital model using an additive process, thus by creating the desired item laying down successive layers of material on top of each other. According to R.Bogue (2013), the first Stereolithography 3D printer was created in 1984 by Charles W. Hull, founder of 3D Systems Corp, but was very expensive, not feasible for the global market and thus used mainly for Rapid Prototyping (RP). With the occurrence of the 21<sup>st</sup> century and the transformation of the mass production industry introduced by Henry Ford into the new Industry 4.0, the cost of using AM decreased and the number of possible applications of this new technology multiplied, together with the range of materials, design freedom and market competitivity. Nowadays, according to the Wohlers Associates Report (2021), this year the additive manufacturing industry grew by 7.5 percent to nearly \$12.8 billion in 2020, despite the ongoing global Covid-19 pandemic. However, compared with an average growth of 27.4 percent over the past decade, the growth was shown to be down considerably due to the worldwide sanitary emergency of the last year.

Is important to underline that besides the most famous and used AM technologies as Stereolithography (SLA) and Fuel Deposition Modelling (FDM), all the process that involve the creation of a solid 3D object are identified as AM technologies. In fact, according to a study of 3D Hubs and as showed in Figure 8, nowadays the AM world includes different types of processes, each one with specific characteristics, materials (plastics, resins, rubbers, ceramics, glass, concretes and metals), applications and companies involved (M.Attaran, 2017). The ASTM F2792-12a standard generically defines seven process classifications for Additive Manufacturing, specifically: Binder Jetting, Directed Energy Deposition, Material Extrusion, Material Jetting, Powder Bed Fusion, Sheet Lamination, and Vat Photopolymerization.



Figure 8 - AM Technologies

As explained by I. Gibson et al. (2010) in the book "Additive Manufacturing Technologies", the main categories of AM processes and the further divisions in the actual technologies are based on three main criteria:

- *The materials that can be used*: Not all the AM machines are able to process all the possible materials addressed to 3D printing technologies. For example, an FDM printer can extrude a large variety of plastic materials and composites but is not able to extrude materials like metals or ceramics, due to the high temperature required to melt alloys and the brittle behaviour of ceramic materials. New technologies like the one patented by Desktop Metals allows to extrude a mixed feedstock of metals and plastic, however sintering and curing post processing steps are still required in order to remove the plastic and to gain a full metal object at the end of the process.

- *How the layers are created*: The single layers can be created using different type of feedstock, in solid or liquid form. Plastic and composites materials are usually feeded to the printers as filaments, pellets or resins (liquid), while metals are processed as powder. In sheet lamination, the feedstock is in form of sheets, thus a thin piece of rectangular material, usually composite or paper.

- *How the layers are bonded to each other*: Depending on the technology, every layer is bonded to the previous one in different ways. For extrusion systems the feedstock is melted trough the hot end and every layer is bonded to each other simply due to gravity, while more complex 3D printing

systems need a more controlled process. SLA bonds every layer due to a laser light that cures the resin, the Digital Light Processing technology (DLP) needs a projector, and the Continuous DPL cures the polymer with LED light and oxygen. Metals, in form of powder, are printed through Powder Bed Fusion (PBF) technologies that cure it using a lasers or electron beams.

Such differences in the processes determine important factors of the final part like the dimensional accuracy and its mechanical properties. Moreover, they will also determine manufacturing factors like how quickly the part can be made, how much postprocessing is required, the size of the machine used, the overall cost of the process, and the convenience of using it instead of a classic machining one (Gibson et al.,2010). Due to the large number of factors involved, comparing AM with respect to the classic Computer Numerical Control (CNC) machining is not that easy. As all the new technologies, AM enables a lot of advantages with respect to the traditional additive manufacturing, but it still carries with itself some problems and possible challenges for the future. In detail, AM technologies can be considered as an added value to the Industry 4.0 for the following characteristics:

- Design Freedom and Topology Optimization: The main advantage of AM is the possibility to directly manufacture an object from the digital model, overcoming the difficulties of traditional manufacturing processes. Different CAD software (SolidWorks, Fusion 360, Rhinoceros, etc.) allow designers and engineers to design complex 3D models without worrying about the possible machining problems that can occur during traditional production (accessibility, fixturing, etc). That is because the model, exported as a STereo Lithography interface format (STL) file and then sent to the 3D printer as G-Code, is directly reproduced layer by layer, drastically reducing the numbers of parts required for obtaining it, and therefore the assembly time. Design freedom also allows to produce more complex part that would be impossible to produce with traditional machining, as objects with



Figure 9 - AM advantages

channels for internal cooling or complicated parametric features, like the coffee cup showed in Figure 9.

Moreover, the Topology Optimization process allows to simulate the stresses that the part will face during its lifecycle, generating an optimized STL with better mechanical characteristics and less weight as demonstrated in Figure 10, where an original full dense aerospace bracket was transformed through topology optimization in a better solution for the manufacturer.



Figure 10 -Topology Optimization

Those characteristics of AM processes implies a high customization of the product, allowing to create particular objects for specific applications. In fact, during the latest years, all the teams participating to the automotive Formula 1 championship stared using 3D Printing for manufacturing high customized pieces for their cars, trying to find the best balance between mechanical performance and weight, obtaining a great advantage on competitors during a Gran Prix.

- *Small Volume Manufacturing*: Traditional manufacturing was introduced by Henry Ford during the industrial revolution in the 20<sup>th</sup> century in order to gain a mass and rapid production for low customized products, as the parts of a car, which have to be all equal and produced as fast as possible. This type of production allows to gain thousands of pieces in a short time, with the cost of manufacturing being indirectly proportional to the number of units. As shown by the graph in Figure 11, AM technologies, due to their characteristics, are significantly changing the unit/cost curve, transforming it in a straight line, since the cost of production is not dependent on the unit produced.



Figure 11- Cost/Volume Curve for AM

That is why AM is usually feasible for a small volume manufacturing industry and high customized products, resulting in a lower cost for unit with respect to conventional manufacturing and a consequentially advantage for the company. Is important to underline that is duty of the company to evaluate all the possible solutions and find the Breakeven point, thus the limit number of units for which is convenient to adopt an AM technology.

- On Demand and Decentralized Manufacturing: Due to the ease of printing directly from a STL file, Additive Manufacturing allows an On Demand production, making possible to print parts in different remote locations by local distributors or service providers that have no knowledge of the specific technology. Therefore, the delivery of goods is no longer a restriction, resulting in the shortening of the supply chain and cost saving for tasks as shipping and inventory management. Moreover, AM could potentially reduce the need for logistics, since every design could be transferred digitally from one production site to the other, leading to a decentralization of manufacturing. By manufacturing items closer to the end destination, it reduces logistical costs, environmental impact, and the overall time from production to sale.

Despite those significantly advantages, AM technologies are not suitable for every type of application, because of different drawbacks, as:

- *Production time*: One of the main drawbacks of AM with respect to conventional manufacturing is the slower production time for the same number of units. As stated by R.King (2012), even if in AM there is no change time in between different production runs, the overall time for manufacturing a batch of products with conventional methods is still a lot lower, due the nature of AM and the relatively short Research and Development (R&D) history for additive processes. For those reasons, AM technologies are more likely to be used in mass customization manufacturing, as it offers the possibility to create highly customized products for a limited inventory.

- *Size Restrictions*: All the 3D printers, independently from the technology, are able to reproduce an object from the correspondent digital model only if its size is equal or less then the printing volume of the printer. This unchangeable characteristic restricts the maximum size of the possible 3D printed products to the size of the printer itself, thus reducing the number of possible applications of AM technologies in different industries. The only possibilities for creating large size products are to divide the model in different parts to be assembled after the printing process or to use Large Scale Additive Manufacturing (LSAM) machines as the MasterPrint showed in Figure 12 by Ingersoll, that was able to produce an entire 3D printed composite boat. Is important to remark that those types of printers are mainly made on demand and thus their cost are significantly high (the manufacturing of the MasterPrint Ingersoll Machine costed around 10 million dollars according to an article of Luca Orlando in 2019).



Figure 12 - 3D Printed Boat

- Costs: The biggest problem that AM technologies had to face, especially during the first years of its life as disruptive technology, was the cost of the printing equipment and overall process. The main costs addressed to AM are usually due to the high pricing of machines and materials. In fact, according to a study conducted by D.Thomas et al. (2015) on the cost effectiveness of AM for metal parts production, the machine costs accounted an average of 62.9% of the total cost estimates. This cost was the largest even when building rate (cm<sup>3</sup>/hours) was more than tripled and other factors were held constant. The second largest cost is the materials, which, on average, accounted for 18.0% of the costs; however, it is important to note that this cost is likely to decrease as more suppliers enter the field. Post processing, preparation, oven heating, and building process fix were approximately 8.4%, 5.4%, 3.3%, and 1.9%, respectively. However nowadays, with increases in technical developments, coupled with more manufacturers entering the industry and more financial helps from investors and governments, the price of AM equipment and process will decrease. In fact, as claimed by an investigation by Siemens in 2017, the predicted cost of metal AM applications during the decade between 2013 and 2023 will decrease of the 50%, while the production speed of the process could increase from 10 cm<sup>3</sup>/hours to 80 cm<sup>3</sup>/hours, thus 400% faster. It is important to underline that the analysis of costs for adopting an AM over to traditional manufacturing is strictly related to the value that a 3D printer can add to the production and the benefits of a possible application. For example, Formula 1 teams are forced to use AM even if the implementation cost is extremely high, because the better performance in a race justifies the expense.

- *Regulations*: As all new technologies, AM has to be regulated properly or can cause social and commercial implications in the imminent future. The regulations must follow innovation at the same pace, or a large number of applications can be considered in a "grey area" between right and wrong. One of the most relevant examples of the lack of regulations was cited by R.Bouge in 2013. In fact, in that year, a free to download STL model of a handgun was available on the internet. This case created a high concerning since the gun, if printed in plastic, it could not be detected by metal detectors and thus if created by someone with malicious intent, could be a threat to public safety. Nowadays, as the limits of AM continue to be tested and as soon as new cases become known, regulations and government intervention could and should restrict who can perform specific prints and what can be printed or not.

Those advantages and drawbacks can be perfectly summarized in a SWAT (Strengths, Weaknesses, Opportunities) analysis conducted by C. McAlister et al. (2016) and showed in Figure 13:



Figure 13 - S.W.A.T Analysis for AM

This critical overview on AM technologies has generally explored the main advantages and drawbacks of 3D printing and its possible applications. Specifically, According to C. McAlister et al. (2014), nowadays 3D printing is used in a wide range of industries as automotive, aerospace, jewellery, plastic packaging and more. In order to investigate in detail the possibilities that can be achieved with AM, the main applications for different types of productions and related industries were analysed by M.Attaran (2017) and summarized in the following Tables 1 and 2:

#### Table 1 - Applications of AM

Industry	Applications	Benefits Gained
Dentistry and Dental Technology	Dental coping Precisely tailored teeth and dental crowns Dental and orthodontic appliances Prototyping	Great potential in the use of new materials Reduced lead-time Prosthetics could be fabricated in only a day, sometimes even in a few hours
Architectural and Construction	Generating an exact scale model of the building Printing housing components	Producing scale models up to 60% lighter Reduce lead times of production by 50–80% The ability to review a model saves valuable time and money caused by rework Reduce construction time and manpower Increase customization Reduce construction cost provide low cost housing to poverty-stricken areas
Retail/ Apparel	Shoes and clothing Fashion and consumer goods Consumer grade eyewear Titanium eyeglass frames Production of durable plastic and metal bicycle accessories	On-demand custom fit and styling Reduce supply chain costs Create and deliver products in small quantities in real time Create overall better products Products get to market quicker
Food	Chocolate and candy Flat foods such as crackers, pasta and pizza	The ability to squeeze out food, layer by layer, into 3-D objects Reduce cost Feasibility of printing food in space

#### Table 2 - Applications of AM

Industry	Applications	Benefits Gained
Aerospace	Prototyping Component manufacturing Reducing aircraft weight Engine components for the Airbus Flight-certified hardware Manufacturing of satellite components	Produce very complex work pieces at low cost Allow product lifecycle leverage Objects manufactured in remote locations, as delivery of goods is no longer a restriction A reduction in lead-time would imply a reduction in inventory and a reduction in costs On-demand manufacturing for astronauts Eliminate excess parts that cause drag and add weight Improve quality
Automotive	Prototyping Component manufacturing Reducing vehicle weight Cooling system for race car	Help eliminate excess parts Speed up time to market Reduce the cost involved in product development Reduce repair costs considerably Reduce inventory Could effectively change the way cars will look and function in the future Improve quality
Machine Tool Production	Prototyping Reducing grip system weight End-of-arm for smarter packaging	Quick production of exact and customized replacement parts on site Allow for designs that are more efficient and lighter
Healthcare and Medical	Fabricating custom implants, such as hearing aids and prosthetics Manufacturing human organs Reconstructing bones, body parts Hip joints and skull implants Robotic hand	Reduced surgery time and cost Reduced the risk of post-operative complications Reduced lead-time

#### **1.2.2 ADDITIVE MANUFACTURING AND SUSTAINABILITY**

During Chapter 1.1 the plastic production industry has been critically explored, and from the data analysis and literature review conducted has been clear that a global change in manufacturing should happen as soon as possible in order to preserve our nature and health for the following generations. Furthermore, different ways of plastic recycling were discussed in detail, showing how even if the majority of governments and industries are committed to do radical changes for aligning with the Paris Agreement's goals, the way to a complete sustainable world is still long. In this subchapter a view on the potential of AM for achieving a sustainable manufacturing will be made, analysing the possibilities that 3D Printing can unlock for achieving a Circular Economy (CE).

In 2014, C.McAlister et al. stated that the balance of lifecycle impacts of 3D printing has been investigated in some initial studies in the area, with the conclusion that electricity in the in-use phase is the dominant environmental factor of AM. However, there were many uncertainties and variations in such analyses, because whether 3D printing has lower or increased environmental impact to alternative manufacture methods strictly depends on which conventional manufacture technique the 3D machine is replacing and its application. For example, for a high production scenario, a UC Berkley study (J.Faludi, 2013) found that 3D inkjet printers had significantly worse ecological lifecycle impact than traditional CNC machining, but also that FDM 3D printer had a significantly lower one. Moreover, another study (S.Wagner, 2010) compared Selective Laser Melting (SLM) with conventional machining and found that even if the energy use at the production stage was comparable, AM leaded to major savings during the material production phase (for the less material needed) and during the in-use phase of the product, since the parts were lighter and thus leading to a lower consumption of fuel during applications in the automotive and aerospace industries. In fact, 3D printed parts can be up to 50% lighter than machined parts and result in carbon savings in the aeronautical parts use stage equalling three to four orders of magnitude more than the amount of CO2 emitted to make them.

In order to understand what factors influence the environmental impact of AM, during the first studies it has been observed that for every AM technology the energy use is the biggest lifecycle problem of 3D printers (J.Faludi, 2013), and that the energy consumptions are mainly influenced by machine design factors (C.McAlister et al.,2014), as:

- *Build Volume*: This factor determines how many pieces can be printed trough a single process in a 3D printer given its capacity. Of course, more objects can be printed simultaneously, less will be the energy required for the single piece.

- *Layer Thickness*: Low layer thicknesses will lead to improved surface finish and higher dimensional accuracy but will also result in a lower process speed and higher energy consumption.

- *Material*: Every material has different specific heat capacities and densities, that have an influence on energy required during the printing process. 3D Printers using materials that can be processed with lower temperatures will lead to lower environmental impacts.

- *Printing Speed*: As already explored in the previous subchapter, the mass production of standard parts using conventional manufacturing methods is faster than 3D printing. Since process speed can vary considerably between printers depending to the factors mentioned (build volume, layer thickness and material selected) ,the lower is the process speed, the higher the energy impact will be for every part made.

Besides the energy needed during the printing process, which as stated previously is with no doubt the main cause of 3D printing consequences on the environment, other factors have been addressed as leading points for the sustainability impact of the AM industry, as:

- *Embodied Energy*: Considering a full life cycle of an AM machine, the processing energy in order to perform the printing operation is not the only one to have an impact. The embodied energy includes all the processes occurring before and after printing, as extraction of raw materials, processing of the raw materials into feedstock, assembly of the printers and heating and cooling operations of the production space. The embodied impacts for printing machines can be high, due to the use of electronics components, metals and plastics. However, those impacts depend on how much the machines are used .According to J.Faludi (2013), the effects of the manufacturing and end of life stages of 3D printers have been found to represent a small proportion of the environmental impacts in high use scenarios, although they become more significant in low use ones.

- *Emissions*: As stated by B.Stephensa et al.(2013), 3D printers that process plastics are high emitters of ultra-fine particles (UFPs) and fumes, that contain toxic by-products as a result of the plastic being heated to high temperatures. Those emissions mainly depend on the materials used and their chemical characteristics. For example, ABS performs worse than PLA, creating fumes while being extruded which can be dangerous for people with chemical sensitivities or breathing difficulties. When measured, the levels of UFPs emitted by 3D printers appear to be the same as cooking indoors, but further work is necessary to determine the type of UFPs and what can be the long terms damages for the environment and human health. In order to absorb the fumes, one or more fans can be used, but they may have an impact on the extruder temperature and thus on the print outcome.

- *Durability*: Due to the additive nature of 3D printing technologies, parts made with conventional manufacturing as injection moulding can have a higher structural integrity than AM manufactured

products. Even if there are great advances in this area, this difference is persisting due to the layerby-layer construction of 3D printed parts, that results in a lower bonding effect of the material in Z direction with respect to the X/Y plane. In fact, part orientation on the build plate is always a primary consideration before the printing process, especially for objects that are known to be more stressed in a particular direction. Moreover, due to post-processing operations usually needed for some 3D printing applications to ensure an adequate strength of the part (e.g. sintering, curing) there may be more energy required for the whole process and less dimensional accuracy in parts. As result, depending upon the application, 3D printed parts may require more frequent replacement, with corresponding additional impacts in material and energy use than their traditionally mass-produced alternatives (C.McAlister et al.,2014).

- *Transportation*: One of the great advantages of AM explained in the previous subchapter is the possibility to create a decentralized production, allowing to significantly reduce the costs and the impacts of transportation. However, there are still impacts in transportation as the delivery of the printer to the user from the factory, the movement of feedstock to the printer location, and the transport of any complex electrical components for use in printer manufacturing to the assembly location. Is important to underline that due to the reduced emissions of transporting vehicles over the years, transportation does not affect the AM impact on the environment as the other factors already explored.

Despite the factors previously analysed, there are multiple sustainability advantages already addressed to 3D printing and different possibilities for reducing the AM impact on the environment. In an article from the "Journal of Cleaner Production" by S.Ford et al.(2016), investigating AM's adoption through a life cycle perspective, four major categories have been identified in which AM is enabling sustainability benefits:

- *Product and Process Redesign*: Thanks to the design freedom advantage, AM enables to design more complex parts with less material, leading to a production with almost no need of assembly and products with new material structures, such as porous mesh arrays and open cellular foams. Those structures can create objects that have better mechanical proprieties with less density, resulting in a more sustainable production with respect to products or components made with traditional manufacturing methods. One of the industries that most benefited from AM applications is the aerospace sector. In fact, besides from the significant environmental impacts of the airline industry, the manufacturing of components has a huge impact on the environment, especially due to the waste arising from the manufacturing process. Typically, the ratio of input material to final component for aerospace application is 4:1 using traditional 5-axis milling processes, with some components

reaching even a ratio as high as 20:1 (S.Ford et al.,2016). Since the most part of materials used for aerospace applications cannot be recycled, in order to overcome to this huge amount of waste the European FP7 MERLIN project sought to address this environmental impact through the application of the Laser Material Deposition (LMD) technology in civil air transportation. As result, the LMD process achieved a 60% waste reduction and 30% of time savings. Another example of the sustainability impact of AM presented by S.Ford et al. is the application of AM technology made by General Electric (GE) in 2016, that redesigned a fuel nozzle of 20 components in a single made one. Successively, GE included nineteen additively manufactured redesigned fuel nozzles in the engine, resulting in a five times stronger component, a better fuel flow geometry for combustion efficiency and a 25% weight reduction with respect to the previous existing nozzle made with conventional manufacturing methods.

Besides the sustainable advantages that AM enables to product design, the improvements are not limited to products but also to some processes. Since AM cannot be applied for every part made in the past with traditional manufacturing, the application of 3D Printing technologies can be useful for redesign the process, thus introducing AM for moulding and tooling. In fact, according to Chen et al.(2015), incorporating AM-produced components as moulds or tools can lead to a more energy and resource efficient production. For example, Salcomp, a company leader in electrical plugs and power supplies production, decided to produce moulds in collaboration with EOS using the Direct Metal Laser Sintering (DMLS) technology. As result of this process redesign, the cooling time was reduced from 14 s to 8 s, enabling 56,000 more injection moulded units produced each month with a better quality.

Over this example, one of the major industries that is exploring AM for redesigning the production process for sustainability improvement is the construction one. In fact, according to Buyle et al. (2013), since the construction sector consumes huge amounts of material, energy and water, applications like the MX3D bridge, the 3D Print Canal House and 3D printed apartment buildings in China demonstrated how AM is capable to reduce the consumption maintaining the aesthetics and the structure resistance. Nowadays, projects like the 3D printed bases for the exploration of Mars made by NASA and the 3D printed houses designed by WASP in Emilia Romagna are confirming the sustainable advantages of AM technologies in the construction industry.

In general, is important to underline that current AM systems are still not suitable in every type of production, especially due to need of post-processing operations to eliminate the stair stepping effect that results from the layer-by-layer type of manufacturing. The best solution could be the use of hybrid manufacturing processes, integrating AM with traditional subtractive, joining and transformative

processes and creating hybrid manufacturing techniques that can lead to advantages like improved finish quality, shorter production time and reduced tool wear.

- *Material input processing*: AM offers a huge range of materials for different applications, most of them allowing opportunities for sustainability improvements. The main advantage is the possibility to produce the same object with less material with respect to traditional manufacturing, using less infill density or even creating 90% hollow parts that can keep the same mechanical characteristics. Secondarily, according to C.Mc Alister et al. (2016), AM offers the possibility to manufacture parts with materials like PLA, a corn-starch based and bio-degradable plastic, that has a low heating requirement both in production and use of the feedstock, thus leading to reduced energy consumption, lower emissions, e and lower embodied energy impacts (27–59 MJ/kg compared to 95 MJ/kg for ABS). Others recycled plastics materials were started to be used in AM , even if there was uncertainty about the potential to recycle waste material from printed parts due to potential changes in the material properties post-printing and pigments that if used may interfere with plastic separation processes (M.Kurman et al., 2013). Nowadays, companies like Reflow are pushing the boundaries of recycling plastic processes in order to reduce the impact of plastic pollution and create feedstock materials for AM applications, maintaining a high quality of the parts being printed with recycled materials.

- Make-to-order component: The on-demand manufacturing characteristic of AM makes it ideal for make-to-order product manufacturing, allowing a fast production of spare parts for replacement, lower costs per part, together with a high customisation of the components. Every product or component's design can be stored in a database and reproduced in every part of the world, reducing the inventory waste and the emissions generated by production and transportation. According to S.Ford et al.(2016), before the born of AM, every part that broke in production was either repaired or reproduced. Repairing the product usually require obtaining replacement components from the manufacturer, but for those organisations maintaining an inventory of replacement parts is costly and there is great uncertainty over future demand for these parts. The alternative, thus producing spare parts on demand, was prohibitively expensive using conventional manufacturing technologies. Nowadays, thanks to AM, the shift from traditional mass production methods and economies of scale to small batch production of personalised products is made possible at a lower cost. Moreover, the possibility to download products or components for free from open-source websites as Thangs3D or Thingverse, has dramatically reduced the need of a centralized production, especially for small businesses and single users. In fact, the "explosion" of consumer 3D printers in homes and offices, such as Ultimaker, Prusa and Craftbot, are re-defining the line between consumers and manufacturers, creating new opportunities for young entrepreneurs and thus generating a passionate network of people interested in AM technologies.

- Closing the loop: The possibility of closing the loop, thus of recycling materials, products or waste into new feedstock in order to obtain a CE, can be achieved in different parts of the AM processes. As first, the feedstock material can be recycled if there is any waste during the production process. As explained by Petrovic et al.(2011), the highest recovery possible is achieved during the manufacturing process when the unused AM material (powder or resin) is reused. Especially, for metal powders it is estimated that the 95/98% can be recycled. Secondarily, for production that use feedstock as filaments or pellets, the implementation of a shredding and extrusion devices enables the creation of recycled feedstock from failed prints, support structures, and/or plastic filament that did not pass the quality test. The embedded energy and additional energy use with such machines will still be more sustainable than using or buying new material, especially for large scale productions (C.Mc Alister et al., 2016). Finally, at the product end-of-life stage, different recycling systems can be suitable for AM, diverting material from classic waste streams into new applications. However, the main problem of this process is related to the materials standardization after recycling, since most of them can be downgraded during the large number of steps required and the difficulties during the separation process and colouring. Is it clear that this is the main challenge for obtaining a complete CE for AM applications, underlining the need for the further development in recycling processes and validation of material properties.

In conclusion, as a proof that the interest on the relation between AM and sustainability has been of interest significantly over the latest years in the academic and manufacturing world, a recent investigation conducted by L.Suárez et al.(2020) analysed all the most recent articles and studies regarding different keywords as sustainability, Eco-Design, Environmental assessment and AM, discovering that the combination of AM publications related with the other categories has increased exponentially over the last decade, as showed in Table 3:

Year	Ecodesign	Environmental assessment	AM	Sustainability	Combination
2009	115	23,770	1245	22,979	200
2010	137	27,129	1622	27,179	267
2011	181	31,150	1533	33,186	216
2012	195	34,641	1819	37,847	278
2013	258	37,797	2312	40,173	359
2014	267	40,888	3791	43,087	575
2015	391	43,260	5039	45,396	758
2016	440	47,545	7361	52,310	1116
2017	525	52,363	9871	58,617	1726
2018	482	57,864	12,698	68,110	2269
2019	378	45,299	10,052	53,513	2086

Table 3 - Studies on AM over the years

#### **1.3 REFLOW'S MISSION AND CHALLENGES**

#### **1.3.1 AN OVERVIEW ON THE FDM TECHNOLOGY**

Nowadays FDM is the most common extrusion-based AM technology (Gibson et al.,2010), due to its wide range of application, the relative cost of the machines (a basic FDM machine can be purchased on Amazon with a few hundred euros) and the different compatible materials with respect to other type of AM processes. First developed by Stratasys Ltd (P.Dudek,2013) in 1992, FDM systems can fabricate prototypes, spare parts or final products in a wide range of materials as thermoplastic or thermosetting polymers, elastomers and composites. The process starts with the loading of the material in the machine, usually in form of pellets or filament, that flows in a heating chamber transforms the material in a molten or semi molten state (P.Dudek,2013) and let flow it into an extruder that guarantees the correct diameter at the exit of a nozzle tip. In this way the molten material is deposited on a drop-down platform (following a route given by the G Code) that should be pre heated in some applications. After that, a layer of material is successfully deposited, the platform moves downward allowing the machine to extrude another layer upon the precedent one.



Figure 14 - FDM Technology

The bonding of the extruded material to the previous layer usually occurs thanks to two primary approaches (Gibson et al., 2010). If the material is a polymer or a composite, usually it is common to use temperature as a controller of the material state. Passed the chamber, the molten material can flow out through the nozzle and bond with the adjacent layer before solidifying. Another approach regards paste materials, that can need a chemical change to cause solidification. This change can happen using curing agents, solvents or simply due to the reaction with air. It is important to notice that, since gravity is the only principle that holds layers together during printing, it is mandatory to consider from the design phase also the inclusion of supports in the part in order to produce complex geometric shapes. Supports are usually made of the same material of the final part or of a different one (Gibson et al.,2010). In the first case the printing process is easily since only one extrusion chamber is needed, but it is more difficult to separate supports from the part without causing fractures. In the second case having a different material allows to distinguish better the support from the part (e.g. using a different colour) and to separate them (e.g. using a weaker material for the supports), but of course a second extruder is needed. At the end of the process supports are carefully removed and the final part is ready to be post processed if needed. The major advantages and disadvantages of the FDM with respect to the others main polymer-based AM processes as SLA and SLS are briefly reported in Figure 15.



Figure 15 - AM Technolgies in detail
As can be seen, FDM is a relative flexible process that allows to have better mechanical proprieties of the resulting parts, at the point that products made with this technology are among the strongest of any polymer-based processes (Gibson et al.,2010). On the other hand, the built speed is limited by the maximum velocity and accelerations of the printing head, that is obviously slower than a laser, since it has to support more weight. In addition, during the process we have a lot of changes in direction, that increases the lead time. We have also to notice that FDM requires a lot of heat in order to melt the material, that will increase the amount of energy (and so the cost) required for the production, even if the machine costs are smaller.

### **1.3.2 RECYCLED REFLOW FILAMENTS FOR FDM APPLICATIONS**

Reflow Filaments is a start-up founded in Amsterdam six years ago with the mission of reducing the global plastic waste trough the production of filament feedstock for FDM 3D printers from industrial scarps. As stated previously in Chapter 1.1, the total amount of plastic material produced over the 2019 was over 350 million tonnes according to Plastic Europe, and less than 20% of this is recycled. As the 3D printing industry rapidly expands each year, the materials market for AM applications is increasingly becoming a contributor to the issue of plastic pollution. As the general population mobilises always more around environmental issues, manufacturers and consumers within AM industry are now prioritising sustainability as a key factor to consider, alongside quality and pricing. When fabricators use Reflow materials, a source traceable sustainable input that delivers high performance, their creations acquire a tangible competitive advantage in the marketplace. Consumers want to know that the products they purchase are not harming the earth, and this consideration is now weighing heavily on their decision making. In fact, according to Reflow's studies of the market, one on three global consumers actively choose brands they believe are doing real environmental good, and the 80% of 3D Printing designer are seeking high performance sustainable 3D printing materials. While just the 14% of 3D printing materials manufacturers offers recycled products, Reflow is a 100% sustainable company, that:

- Uses renewable windmills sources from Netherlands as embodied energy for the production of filaments.

- Produces only recycled products with 97% to 100% of recycled content. The 1 to 3 percent of virgin material is selected as the lowest percentage possible needed for the colouring.

- Does not use plastic packaging apart from the vacuum recipient mandatory for avoiding moisture absorption in the filament, and thus ensuring the highest printing performance as possible while keeping a high sustainable production .

- Selects as partners for transportation the ones with the lowest emissions impact for the environment.



Figure 16 - Reflow Main Steps

As showed in Figure 16, the process of materials collection and production of recycled filaments in Reflow can be summarized in four main steps:

1) *Collection*: The first phase regards the collection and separation of waste at the end of the production. Usually this operation is made by the company that produces the scarps or by recycling centre.

2) *Processing*: After collection, the waste in processed following the primary recycling process explained in Chapter 1.1. This process transforms general waste into pellets of the same polymer, used as input for the Reflow's production.

**3**) *Transformation*: The transformation of recycled plastics from pellets into filament is made in Reflow trough a dedicated extrusion line. In the next chapter the whole Reflow extrusion process for polymers will be explained in detail, from the initial drying until the final packaging of the produced spool.

*4) Creation*: The output of the transformation stage, thus the spool of recycled filament, is used as input for FDM 3D printers. Reflow produces filaments of the two different standard sizes of diameters, 1.75mm and 2.85mm.

The current product range of Reflow is divided in five materials, as reported in Figure 17. At the moment Reflow produces recycled Poly-Ethylene Terephthalate Glycol (PETG), recycled Polylactic Acid (PLA) and recycled Polymethylmethacrylate (PMMA). During this year Reflow is working with different partners in order to extend its material portfolio adding materials as recycled Polypropylene (PP) and recycled Nylon (PA) reinforced with carbon fibres.



In conclusion, the Reflow Business Model (BM), and thus the different sources of revenue, is presented and divided as follow:

- Direct Material Sales (B2B): The business-to-business operations are the one conducted by the sales team in order to sell usually large amount of material to other small companies or large enterprises, in form of recycled pellets or filament.

- *E-Commerce (B2C):* The business-to-costumer operations are made by single consumers trough the Reflow website. Those type of operations usually regards just filament feedstock and are typically made by single costumers, designers or small businesses for purchasing small quantities of material.

- *Resellers (3rd Party Sales)*: Operations made in collaborations with other parties that buy large amount of material at lower price not for manufacturing purpose but for reselling on their websites.

- *Circular Projects*: The circular projects are made in collaboration with other companies that want to re-use their production waste for additive manufacturing applications, allowing to unlock a CE for their product. Reflow helps those companies to find the best recycling route for their material and then transform it into recycled filament for FDM printers. The main difference between Circular Projects and B2B sales is that the produced filament is produced exclusively for the company that has provided the initial waste. A successful example of Reflow's circular projects is the collaboration with Ace & Tate, manufacturing leaders in the eyewear industry. Ace & Tate provides to Reflow industrial scarps from their PMMA lens production in order to produce recycled PMMA filament. Successively, the filament is used for printing trough FDM spectacle frames for Ace & Tate glasses.

- *Material Development Process*: Other companies cannot provide industrial waste in large quantities, but they still want more sustainable materials for their production while keeping high performance. Reflow collaborates with those companies in order to find a suitable waste stream that can be converted into a customized production of recycled material for 3D printing applications.

- *Printer Partnerships*: During this year Reflow will start collaborating with different printer manufacturers in order to sensibilize designer and companies to a more sustainable production. Who buys a printer that collaborates with Reflow will receive with the purchase also some recycled material to test on the printer. In this way Reflow can cover more possible clients and can help creating a network of creators interested in a more sustainable AM production.

# 2.PLASTIC EXTRUSION FOR FDM FILAMENT PRODUCTION

Plastic materials are so widely used across different industries due to their unique characteristics, as easy shaping and durability. In fact, almost every shape can be processed with plastics, and almost all the products made worldwide are obtained with those materials, contain a percentage of microplastics or are covered with a plastic packaging. In the AM industry, plastic is mainly used as feedstock for the production of highly customized products. As already reported in Chapter 1, 3D printers are generally feeded with plastic in form of pellets, resin or filament, depending on the AM technology adopted. Specifically, for FDM printers, the common shape for the feedstock is the filament one, that is rolled around a spool creating a 1 kg plastic feedstock for the printers. However, in order to obtain the desired filament, the original plastic in pellet shape has to be processed, following the extrusion process. This Chapter will analyse the main steps of the Reflow extrusion process for producing recycled filaments spools for FDM printers, from the initial drying to the final spooling and packaging. Is important to remark that the whole process will be explained with the highest detail as possible, however no numerical data or graphs of the main extrusion parameters will be presented, in order to preserve the Reflow intellectual property of the specific production method.



Figure 18 - Extrusion Process

In general, according to John R. et al. (2005), the main steps of a plastic extrusion process can be summarized as showed in Figure 18, starting with the initial quality inspection and drying of the original pellets and ending with the final inspection of the extruded polymer and the following packaging, storing and shipping. Moreover, the study reports that in order to gain a high-quality filament that respects all the desired characteristics, is essential to have a perfectly working equipment (extruder, dies, water tanks, sensors, etc.) that has to achieve five principal goals:

- Correct polymers melt temperature
- Uniform melt temperature
- Correct melt pressure in the die
- Uniform melt pressure in the die
- Homogeneous, well-mixed product

In addition, is important to underline that in order to optimize the overall extrusion process is not sufficient to understand the required equipment and its behaviour, but it is critical also to study the polymers characteristics and the possible reaction that can have with the equipment. In fact, since different polymers behave differently at different melt temperature, pressure and water temperature, the extrusion line is not so easy to control, especially when the material portfolio is large and different changes have to be made on the line every day.



Figure 19 - Reflow Production Line

The Reflow extrusion line, showed in Figure 19, is meticulously checked every day before every production run, and the overall system is live controlled by at least one experienced operator, ensuring a high-quality output and an immediate intervention in case of problems. The Reflow line produces filaments spools of 1kg (for Business-to-Customer clients) or 8kg (for Business-to-Business clients, as reported in the Figure), with a filament diameter of 1.75 or 2.85 mm, depending on the client's needs and on the available stock in the warehouse for that combination of diameter, material and colour.

# 2.1 INITIAL INSPECTION AND DRYING

The first and essential step of the extrusion process in Reflow is the quality control and storage of the recycled material that comes from the recycling centres. Reflow has two different recycling facilities partners (that cannot be mentioned due to the non-disclosure agreement) providing 100% transparent recycled PLA and PETG in pellets form ready to be processed in filament. Is important to storage the raw material at room temperature before starting the extrusion process, in order to keep uniform the polymer melting and softening temperatures across the different seasons during the year. This is a really critical point because a drastic change in the raw material initial temperature can change the

polymer characteristics during the process, leading to a product inconsistency and a lack of performance. After the initial inspection and the storage in a room temperature environment, the raw material needs to be dried before starting the production. The drying process is fundamental for the all the polymers contained in the Reflow portfolio, since they are all very hygroscopic, meaning they absorb a lot of moisture rapidly from the air. The presence of moisture in the raw material drastically affects the production process, since it is converted in steam once the polymer is heated in the extruder, creating bubbles of air inside the produced filament. Bubbles are visible by eye at the end of the process, and the printing results obtained with this product are terrible, since the presence of air leads to a under extrusion effect during the print, creating low performance parts with a lot of surface imperfections.



Figure 20 - Drying Equipment

In the Reflow plant in Amsterdam, the drying step is performed by the line operator every evening before production. The recycling centres ship to Reflow large amounts of raw recycled material in pellets contained in big fibre bags. The auxiliary drying equipment showed in Figure 20 is managed by the operator in order to dry the material over night before the production of the polymer that will be extruded the day after. A mechanical pump moves the raw pellets from the fibre bags to the dryer, which dries the polymer for at least 8 hours before the production start. Obviously, every polymer

has different recommended drying temperature that allows to remove all the moisture from it and ensure a high-quality production.

# **2.2 COLOURING**

The colouring step is essential for all the products in the Reflow portfolio that has to be colored and not transparent as the initial raw recycled material. This process is performed right after the drying step and consist in mixing the recycled polymer with a small not recycled percentage of coloured virgin polymer. The virgin polymer is usually the same of the recycled one, so for example for producing a recycled and coloured PLA filament a small percentage of colored PLA virgin pellets will be used.



Figure 21 - Colour Mixing Equipment

The Figure 21 shows how the colouring process is performed. The raw material is charged into the production line through a controlled loader, while the correct quantity of coloured polymer is loaded in the Movacolor equipment input. The Movacolor machine performs the mixing between transparent and colored polymer, creating a homogeneous output that can be successively extruded obtaining the desired coloured filament. Is important to underline that Reflow performs hundreds of tests before launching a new product in order to find the minimum quantity of not recycled polymer that delivers

the wanted final filament colour. Nowadays in the Reflow portfolio are present only filaments with maximum the 3% of not recycled content needed for the colouring, while all the transparent filaments are 100% recycled.

# 2.3 SINGLE SCREW EXTRUSION

The extrusion process is definitely the most important step of the filament production, since it involves different type of equipment and sensors, while transforming the polymer in a molten state and then re-solidifying it in the desired filament form. This process in Reflow can be divided in two different stages that works simultaneously when the production is launched:

### - Feeding and Heating

After the colour mixing process, the homogenous polymer (made by the recycled raw material plus the small percentage of virgin colored one) is feeded directly into the heating line, formed by five different heaters with increasing temperature that transform the pellets from a solid form into a molten state.



Figure 22 - Single Screw Extrusion

As reported in Figure 22, the feeding is made thanks to a single screw and its correspondent motor, that allows the material to be moved horizontally through all the heaters. Every heater has a different temperature, that increases from the first to the last, and is managed by a thermocouple and a controller. The thermocouple reads the actual temperature of the correspondent heater and sends the information to the controller, which tunes the cooling fans and the heating generator in order to keep the temperature as much constant as possible. Every single controller on each heater has both a derivative and integral action (PID), ensuring a controlled temperature both at the start and in saturation. Moreover, a pressure transducer reads the pressure in the die and ensure the correct rotational speed of the screw. The last heater is settled to the melting temperature of the processed polymer and the breaker plate contains just one die for the exit of the melted polymer, as showed in Figure 23.



Figure 23 - Heaters Output

- *Solidification*: Once the material is melted, it has to be re-solidifycated into the desired form, thus the filament one. The melted polymer is pulled from the heater die to the spooling rollers using a pulling system, which will be explained in detail in the next sub-chapter. During the travel, the material flows through two water tanks, in which the polymer is transformed again in a solid state. In order to perform this transformation properly, the polymer cannot be shocked directly into cold water, or it would break instantly. For this reason, the first water tank is settled to the glass transition



Figure 24 - Water Tanks

temperature of the material, which is the temperature needed for passing from the molten to the solid state.

This temperature is usually between the  $70^{\circ}$  and  $90^{\circ}$  degrees Celsius depending on the Reflow material produced. The second tank contains water at room temperature, needed for stabilizing the material with the external environment. At the end of the second tank, a small dryer (Figure 24) eliminates the moisture created by the water during the transformation and a dimensional sensor measures the actual diameter of the filament and its circularity, and represents it graphically on the control screen, in order to ensure that the tension of the spooling system is settled correctly for the diameter dimension required (Figure 25).



Figure 25 - Dimensional Control

### **2.4 SPOOLING**

The spooling system is most important equipment for the extrusion of plastic in order to produce a filament-form product, since it controls the force that pulls the polymer through all the line and giving it the desired filament shape, from the die of the last heater until the final spool. The spooling system is mainly composed by four elements, represented in Figure 26 and 27:

- *Puller:* The pulling machine (Figure 27) and its correspondent motor allows the polymer to be moved through all the line and gives it the characteristically circular filament shape, with a 1.75 mm or 2.85 mm diameter. The dimensional sensor at the end of the line reports information on the X and Y diameters and its circularity (X/Y), so the production operator can regulate the correct tension in order to obtain the wanted diameter. The produced filament is accepted if the diameter has a maximum tolerance of  $\pm 0.05$  mm with respect the nominal dimension of the diameter (1.75 or 2.85 mm) and circularity (1 for instance).

- *Tension controller:* As explained previously, the tension of the puller has to be regulated properly in order to reach the perfect dimension of the filament during the spooling. This is possible with the use of a tension controller, that reports the actual pulling force in Newton and allows to regulate the puller tension for the production. As can be imagined, the logic that regulates this equipment is very easy : in order to create a filament with larger diameter (2.85 mm) less tension is needed, while for a filament with less diameter (1.75 mm) more force is required. Obviously, in absence of particular complications, the tension should be kept as much constant as possible during the production in order to avoid fluctuations around the dimensional accuracy of the product.

- **Buffer:** During the daily production, several spools are made in the production line. Since the production has to be as much continuous as possible in order to be optimal, the puller can not be stopped at every spool change but has to run during all the time the line is active. For making it possible, a large buffer is installed on the top of the line (Figure 27) in order to keep pulling the polymer during the spool changes or in presence of a problem. The buffer allows to never stop the production during the day and keeps constant the tension on the material, ensuring no changes in the dimensions of the product.

- *Spooler*: The spooler is the last equipment element of the spooling system, and it is required for spooling the filament in the FDM spool, thus the final product. The Reflow spooler has two spots for the spools (Figure 26), which are placed in position one by one by the production operator. The spooling machine gives information on the speed of spooling in revolutions per minute (rpm) and on the numbers of rotations of the filament around the spool. Every type of filament (1.75 and 2.85 mm) has a nominal number of rotations for producing a 1kg or 8kg spool.



Figure 26 - Spooling System



Figure 27- Buffer, Puller and Tension Controller

Once the number of rotations reaches the nominal number, the production operator cuts the filament rolled in the first spool and passes it to the second one. During the change the filament is still pulled by the spooling system, but it moves through the buffer without losing tension and dimension. This operation is the only one performed manually during production and is continuously made by the operator during all day. At the end of the shift, several spools of the desired dimension (1.75 mm or 2.85 mm) and weight (1 kg or 8 kg) are made, ready to be inspected, packed and stored for the future sale. It is important to notice that, as for the puller, also in this case the logic that regulates the nominal number of turns is easy to imagine: In order to reach 1kg, less turns are required for a 2.85mm spool, while more are necessary for a 1.75 mm one, due to the less volume occupied in the same circular space.

# 2.5 FINAL INSPECTION, PACKAGING AND STORAGE

In general, during the production shift, multiple 3D printing test for a final inspection on the final product can be performed in order to determine:

- *Correct dimension*: The correct dimension of the filament has to match the correspondent 3D printer input, otherwise the printer won't be able to pull the filament through the extruder continuously, causing clogging problems during the print.

- *Correct Colour:* The correct colour extruded by the printer has to be as specified by the material portfolio or as agreed with the client.

- *Moisture absence:* No moisture has to be present the filament, the presence of air or water can cause several defects on the printed products.

The following tests, which are usually simple prints that require low time, are performed by the 3D Printer Engineer using a small sample from the line while the production is running in order to give rapid feedback to the manufacturing team.



Figure 28 - Reflow Final Product

At the end of the production line, if no problems emerged during the extrusion process and the consecutive spooling, and if the requested printing test were accepted, every single spool (regardless its weight, dimension or colour) has to be properly sealed in a vacuum bag. The vacuum sealing is performed by the production operator during the production shift, using a vacuum machine with different bags sizes for 1kg (Figure 28) or 8kg products and using the proper pressure for each one.

The final finished product is then packed in the Reflow cardboard packaging and properly stored in the dedicated warehouse as showed in Figure 2.1. The warehouse storage is kept at room temperature, in order to preserve the characteristic of the polymer and guarantee the quality of the product over time.



Figure 29 - Reflow warehouse

Moreover, at the end of the production, every packed spool is labelled with recyclable stickers containing the following information:

- *Serial Number:* The Serial Production number is essentially for tracking the product in the Reflow Data System and thus keeping storage of its information, clients, related compliances and essential production data as sensors reports and conditions.

- Material : PETG, PLA or PMMA.
- *Diameter* : 1.75 mm or 2.85mm
- Weight : 1kg or 8kg.

- *Colour* : Reflow has a wide material colour portfolio for its materials, different colour collections are usually added every year or requested on-demand by a specific client.

- *QR Code:* Enables the client to directly access to the Reflow website and social channels and allows users to share their experiences with Reflow.

# **3.EMISSION ANALYSIS AND COMPARISON**

# **3.1 METHOD**

During the last Chapter the Reflow plastic extrusion process for producing high quality recycled filament for 3D Printing applications has been explored, analysing the main equipment and actions required in order to guarantee a flawless production flow and obtain a recycled product that can fulfil the client requirement in terms of both performance and sustainability. Is important to remember that the main mission of Reflow is to reduce the most as possible the production of virgin plastic used in the AM industry, maintaining all the material advantages that it can offer. At this point of the study is it clear that AM is being part of the Industry 4.0 revolution, helping to create a circular economy type of manufacturing and reducing the waste generated by traditional methods.

For this reason, the main goals of this Chapter are two:

- *Analyse:* Analyse the emissions generated by the Reflow production for 1 kg of rPETG and rPLA, using real measured data from the last year production.

- *Compare:* Compare the Reflow data with respect to the data available on the Granta EduPack database related to the virgin plastic production and recycling.

Is important to remark that this study will analyse the production emissions in order to manufacture recycled filament for FDM printers, but this analysis cannot be considered as a Life Cycle Analysis (LCA) of the final product. This is due to various reasons:

- A full LCA should provide all the emission data related to the complete plastic stream, from its first production to the different stages of recycling, and not only about the filament production. Unfortunately, those data are not available for the Reflow product in analysis or are too generic and not precise as they should.

- The LCA should contain precise data on all the emission related to the operations needed in order to provide and move the material, as transportation and storage, which are difficult to calculate or can provided only with a lot of uncertainty.

- The LCA can be considered "full" if it tracks all the Life Cycle of the product. Since the Reflow filament could be recycled more than one time, this analysis cannot be considered as a full LCA, but just as an emission data collection for the first filament production. Moreover, emissions data generated during the 3D Printing process can vary significantly based on the time of the print, material selected and printer.



Figure 30 - Reflow Impact Comparison

In conclusion, as showed in Figure 30 (that represents first research of Reflow impact comparison of virgin and recycled filament production conducted last year), this study will be limited in analysing the second production GHG emissions (thus the ones emitted by Reflow for producing the filament after the first recycling) and compare them with the available data on the primary production emissions (from oils to polymers). Both processes taken in exam are highlighted in red in the Figure. Despite this research, different studies during the last years have performed a LCA or emission analysis of plastic materials used in the manufacturing industry, demonstrating as a circular economy system can be applied to the Industry 4.0 for different materials and can help reducing the emission related to the polymers production. For example, In the 2012 Li Shen et al. reviewed the environmental profiles of petrochemical PET, partially bio-based PET, recycled PET, and partially recycled bio-based PET, and compare them with other bio-based materials, like PLA (polylactic acid, a bio-based polyester) and man-made cellulose fibres (i.e. Viscose, Modal and Tencel), showing that both recycled and bio-based materials offer important environmental benefits over single-use petrochemical PET. In detail, the paper demonstrates that Among the four PET product systems studied, the partially recycled bio-based PET has the lowest impacts, followed by recycled PET, partially bio- based PET, and petrochemical PET, while the PLA and man-made cellulose have lower impacts than both petrochemical virgin PET and the bio-based PET. Another study, more focused on the AM application and conducted by M.A. Kreiger et al. (2016), demonstrated how a distributed recycling LCA of High-Density Polyethylene (HDPE) for 3D printing filament results in less embodied energy emitted than centralized recycling. Moreover, the paper states how with the opensource 3D printing network expanding rapidly the potential for an adoption of distributed recycling of HDPE represents a novel path to a future of circular manufacturing, addressing it lower environmental impacts than the current centralized system.

### **3.2 ANALYSIS**

The analysis of GHG emission of the Reflow production is made distinctly for the two materials taken in exam, recycled PLA and recycled PETG. Is important to remark that the two materials have different production rates, depending on the efficiency of the line and the characteristics of the polymer. The production rate was calculated over the last year of production and is defined as the percentage of acceptable filament produced, thus represent the difference between the recycled pellet inserted at the start of the production line and the waste generated during the production (Figure 31). In fact, since the rPETG is more difficult to process and has a wider range of colours, the production rate calculated, of 83,3%, is lower than the PLA one (95%).



Figure 31- Production Rate

Obviously, the production rate influences the number of spools produced in one hour, and consequently the correspondent amount of kg manufactured and impact of the material on the emissions data. As result of this difference, even if both materials follow the same production line and use the same equipment and the amount of energy (in MJ or kWh), the impact on the emissions (kWH/kg) will be different for the two materials. In general, this study analysed the energy required for the line of all the necessary equipment presented in Chapter 2, thus:

- Dryer
- Loader
- Dosing unit (Color Mixer)
- Extrusion Line
- Air Compressor
- Vaacum Sealing

The calculation of GHG emissions is performed following those four steps:

**1**) Reporting the energy consumption for every single production item provided by the equipment manufacturer (both in kWh and MJ).

**2**) Measuring the number of the maximum kg produced in one hour based on the production rate and equipment capacity (kg/h).

3) Normalizing the energy consumption on the quantity produced (kWh/kg).

**4**) Calculating the GHG emission (CO2/kg) by using the conversion rates for Dutch Energy sources (P.S Ortiz et al.,2020) for both filament produced and waste.

It is important to underline that the following steps will be performed for the acceptable filament production but also for the related waste, through the use of the production rate data, that as already explained divides the consumption between filament and waste.

# 3.2.1 rPETG

The Table 4 shows in the detail the first three over cited steps and the related calculations in order to obtain the energy consumption required for every kg of filament produced. As can be seen, it is clear to notice that the consumptions are divided between filament and its related waste thanks to the production rate factor, that for rPETG is 83,3%. Moreover, is important to remark that, once the raw material is inspected correctly, the possible waste can be generated only through the extrusion line and the compressor.

#### Table 4 - rPETG Energy Consumption

REFLOW rPETG PRODUCTI	EFLOW rPETG PRODUCTION ENERGY CONSUMPTION					
				тот	FILAMENT	WASTE
	kWh	MJ	kg/h	kWh/kg	kWh/kg	kW/kg
Dryer	10	36	16,67	0,599880024	0,599880024	
Loader	1,1	3,96	11,64	0,094501718	0,094501718	
Dosing	0,08	0,288	12	0,006666667	0,006666667	
Extrusion Line	25	90	12	2,083333333	1,736110417	0,347222917
Compressor	2,25	8,1	12	0,1875	0,156249938	0,031250063
Vaacum Sealing	1,5	5,4	120	0,0125	0,15	
тот				2,984381742	2,743408763	0,378472979

Successively, in order to perform the last step, thus the calculation of the GHG emissions (kg of CO2 emitted on every kg of filament produced) a conversion rate table (Table 5) is required. The following table is based on a study of P.S Ortiz et al.(2020) and shows the different conversion rates addressed for different energy sources in the Dutch territory, where Reflow operates. This is the most recent study of the energy impact in Netherlands and the following data are the most updated one for the country.

Table 5 - Dutch Energy Conversion Rates in GHG

<b>DUTCH ENERGY CONVERSIO</b>			
Source	Conversion F	Rate (kgCO2	/kwh)
Coal-fired		0,579	
Oil-fired		0,737	
Natural Gas-fired		0,478	
Biomass-Coal		0,143	
Nuclear Power		0,039	
Windfarms (Reflow source)		0,003	

The Reflow production plant is powered by an energy supplier that produces green energy thanks to windfarms, which are an energy source widely used in Netherlands due to the country's position in Europe and its characteristic weather. Since windfarms are a very low emission generator type of energy source, Reflow has always chosen it as its energy provider in order to impact less as possible

#### Table 6 - rPETG Emissions

REFLOW rPETG GHG EMISSIO	ON CALCULATION		GHG (kg CO2/ kg product)			
	Coal-fired	Oil-fired	Natural Gas-fired	Biomass-Coal	Nuclear Power	Windfarms
FILAMENT	1,5884337	2,021892258	1,311349389	0,392307453	0,106992942	0,00823023
WASTE	0,2191359	0,278934586	0,180910084	0,054121636	0,014760446	0,00113542
тот	1,727957	2,199489344	1,426534473	0,426766589	0,116390888	0,00895315

the environment. Thanks to those data, the last analysis step is performed simply multiplying the energy consumption and the energy conversion rate, and the results are displayed in Table 6.

As can be seen, windfarms energy sources (that represents the Reflow production) have the lowest emission impact with respect all the others available sources in Netherlands. In general, the production impact of the Reflow plant can be assumed relatively low (0,00895315 kg CO2/kg produced), demonstrating the importance of the use of green energy sources (such as Biomass coals, nuclear and windfarms) that generates less than 300 times the emission produced by traditional energy sources as Oil or Coal.

# 3.2.2 rPLA

The steps performed in the last Sub-Section for rPETG are repeated in the same order and method also for rPLA. As already explained, the main difference between the two materials is represented by the production rate, which for rPLA is at 95% and thus higher with respect the rPETG one. This difference is due to two main reasons. First, the rPLA has entered in the Reflow production portfolio during the last year, when the production process was already efficient and less test were required in order to find the optimal parameters, thus less waste has been generated. Moreover, rPLA is easier to process and the coulor portfolio is limited with respect the rPETG one, resulting in less possible problems in production, thus in a higher production rate. In addition, apart from the production rate, also the quantity (kg/h) is different, due to the different chemical characteristic of the polymer that limits the traction speed of the puller during production and consequentially the maximum quantity produced in one hour.

As for rPETG, the Table 7 represents the first three steps required for the calculation of the rPLA energy consumption:

REFLOW rPLA PRODUCTION ENERGY CONSUMPTION						
				тот	FILAMENT	WASTE
	kWh	MJ	kg/h	kWh/kg	kWh/kg	kW/kg
Dryer	10	36	14,28571429	0,7	0,7	
Loader	1,1	3,96	11,64	0,094501718	0,094501718	
Dosing	0,08	0,288	12	0,006666667	0,006666667	
Extrusion Line	25	90	12	2,083333333	1,979166667	0,104166667
Compressor	2,25	8,1	12	0,1875	0,178125	0,009375
Vaacum Sealing	1,5	5,4	120	0,0125	0,15	
тот				3,084501718	3,108460052	0,113541667

Table 7 - rPLA Energy Consumption

Successivly, referring to the Dutch conversion data Table 3.2 (already showed in the previous subsection related to rPETG) is possible to calculate the GHG emissions for rPLA using windmills (Reflow provider) and other different energy sources, both green (Biomass Coal or Nuclear) or traditional (Oil and Coal). Results are displayed in the following Table 8:

REFLOW rPLA GHG EMISSION CALCULATION		GHG (kg CO2/ kg product)				
	Coal-fired	Oil-fired	Natural Gas-fired	Biomass-Coal	Nuclear Power	Windfarms
FILAMENT	1,799798	2,290935058	1,485843905	0,444509787	0,121229942	0,0093254
WASTE	0,065741	0,083680208	0,054272917	0,016236458	0,004428125	0,0003406
ТОТ	1,785926	2,273277766	1,474391821	0,441083746	0,120295567	0,0092535

Table 8 - rPLA Emissions

As for rPETG, the Reflow rPLA production impact (in blue) on the environment is relative (0,0092535 kg CO2/kg produced), and drastically lower with respect the traditional energy sources.

# **3.3 COMPARISON**

During the Analysis step, the data collected of the Reflow production during the last year (2020) were processed in order to obtain the GHG emissions related to the manufacturing of recycled filament in Netherlands. Those data are very specific, related to a particular company that operates in a particular country (with related energy conversion rates). Moreover, they represent emission for just a single line of production, and not a mass one as it can be for virgin plastic. However, a comparison between the GHG emission of recycled filament production and the emission related to the primary polymer production has to be made, in order to understand the impact of plastic production and the importance of the adoption of circular economy during the following years. The data related to primary production and recycling were obtained using the Granta EduPack Database, that contains several and updated data on all the materials characteristics and production methods. Energy required to make 1 kg of the material from its ores or feedstocks. The quoted energy and CO2 footprint data are based on those published in literature and life cycle inventory databases. The sources and values used for each data point are provided. In cases where data is unavailable, the energy and CO2 footprint are estimated using a statistical model.

Table 9 -	Virgin	PETG	Production	Emissions

VIRGIN PETG PRODUCTION AND RECYCLING EMISSION DATA (GrantaEdupack Database)					
kwh/kg MJ/kg GH					GHG (kg/kg)
PETG Primary Production			26,013889	93,65	4,375
Reclyled Pellets Production			8,833333	31,8	1,485

VIRGIN PLA PRODUCTION AND RECYCLING EMISSION DATA (GrantaEdupack Database)						
	kwh/kg MJ/kg GHG (kg/kg)					
PLA Primary Production		15,38889	55,4	2,84		
Reclyled Pellets Production		4,875	17,55	0,9475		

In Table 9 and 10 the main emission data related to the virgin PETG and PLA production and first recycling are showed, demonstrating how the production of virgin plastic from zero require much energy and is drastically more pollutant than the recycling phase.



Figure 33 - Emission Comparison for PETG



Moreover, as reported by Figure 32 and 33 the Reflow production line of recycled filament consumes less energy and produce a lot more less pollutant emissions compared to the virgin one, as calculated previously. These results are explicable by different reasons:

- Low energy is required for transforming plastic from recycled pellets to filament, while a lot more energy is needed in order to produce virgin plastic from raw materials.

- The extraction of raw materials (carbon, silicon, hydrogen, nitrogen, oxygen and chloride) into plastics generates much more pollutant gas emissions with respect the extrusion for recycling or filament production.

- The energy source adopted by Reflow, thus windmills, has the lowest impact possible on the generation of GHG, while the traditional plastic production generally adopts traditional methods for powering the huge production plants and extraction sites.

By looking at those data, is clear that something has to be done in order to reduce the impact of the plastic production on our enviroment. In fact, as explored in Chapter 1, the embodied energy and its emissions are not the only factor that affects our planet, but also waste, bad recycling and consumers' carelessness. However, the change that we need as people is not complicated as it appears to be. For example, imagine if all the plastic waste present in our planet could be recycled and re-used for 3D Printing : this process could generate less emission than producing the same amount of plastic from zero, and would create a circular economy that can benefit consumers, recyclers centres and our planet. The reasons why this is not happening are multiple. Nowadays, in plastic industry and in several other industry sectors, the traditional production methods are so rooted and efficient that is too expensive for industries and governments to change and be adapted to the sustainability goals. Moreover, even if different sensibilization campaigns were made by different counties and associations, most of the people are indifferent to this problem. In conclusion, is desirable that the new generation of student, entrepreneurs and mangers investigate furthers to this problem and the governments apply more restricted rules to big enterprises and consumers, or the Paris Agreement goals will never be reachable.

# 4. 3D PRINITING PROFILE OPTIMIZATION AND MECHANICAL TESTING

During this Chapter, the recycled Reflow materials will be compered, in terms of 3D printing and mechanical performance, with respect to virgin filaments made by a Reflow competitor in Europe, which will remain nameless. The selected competitor was chosen between different candidates due to its similar mechanical characteristics and price with the Reflow filament, in order to perform a reasonable comparison between virgin and recycled filament. All the filaments used (rPETG, rPLA, virgin PETG and virgin PLA) are of the same dimensions (2.85mm) and will be tested in the same printer (Ultimaker 2+) in order to find the optimal printing parameters for each one of them. Successively, following ISO standards, different mechanical specimens will be 3D printed with different infill directions and optimal settings. In conclusion, the final tensile and bending tests will be performed at the material testing laboratory of the Politecnico di Milano, in order to evaluate and compare the mechanical performance between the two couples of materials taken in considerations (rPETG with respect to virgin PETG, rPLA with respect to virgin PLA) and the different infill directions. Therefore, the research process is mainly divided in the following steps:

1) Design of the Experiment (DoE) in order to find the optimal 3D printing parameters.

- 2) 3D Printing of mechanical specimens following the ISO standards for testing plastic materials.
- 3) Tensile and bending ISO testing of the 3D Printed specimens and analysis of the results.

Over this research, is important to underline different studies in the latest have already analysed and compared recycled and virgin plastic material, 3D Printing and traditional manufacturing processes or multiple-times recycled polymers for FDM applications. In fact, already in 2016, Sanchez et al. have investigated on the feasibility of recycling PLA multiple times and compered the mechanical tests of the 3D printed specimens, providing evidence of the feasibility of using recycled PLA for open-source additive manufacturing. However, as a main result, is important to highlight that the 3D printing process and further recycling reduces the mechanical properties. Successively, also Mazher Iqbal Mohammed et al.(2017) have studied the feasibility of using 100% first recycled ABS for 3D printing applications using and FDM printer. Results showed that ABS can be recycled and reformed into filaments without the addition of virgin material. However, some changes in polymer characteristics were observed, leading a limited lowering of mechanical properties during tensile tests and a decrease in the polymer melt flow. Despite these limits, it was demonstrated that ABS first recycling and reprinting is possible (with acceptable loss of material characteristics), and thus could

provide different opportunities for a sustainable use of waste ABS using FDM technology. Another considerable study made the same year was performed by Isabelle Anderson (2017), who compered the mechanical performance of virgin and recycled PLA. The virgin test specimens were first made using a desktop FDM printer, tested, then ground up and recycled into 3D printing filament. This recycled filament was used to 3D print identical specimens that were tested following the same procedures used for the original filament, resulting to have similar properties with respect to the virgin one(slightly decreased in most areas with a moderate increase in property variation). Furthermore, a 2018 study conducted by Nicole E. et al. showed how recycled PET bottles transformed into filament can be a suitable material for FDM printing, provided the material is properly cleaned and dried. The research demonstrated that even if printed specimens achieved only half of the tensile strength of their injection moulded counterpart, the same data was equivalent to printed parts made from virgin PET pellets and commercial ABS filament, meaning that the recycled PET from plastic bottles could be a valid alternative as feedstock for 3D printers. To conclude, is important to mention the research made by F.De Fazio in 2018, who was the first to test the recycled Reflow rPETG filament and compare it with other different filaments, both virgin and recycled: recycled PET from industrial waste, recycled PET from post-consumer waste, virgin PLA and virgin PET. The results showed how the best recycled material tested is the Reflow rPETG, since its tensile stress at yield is very close to the one of a virgin PLA (-4,50%), the elongation at yield and at break is higher (+48,01%, +50,27%) and its flexural properties are comparable with PLA, with a flexural strength 12,65% lower. In fact, according to De Fazio, the good quality of this filament can be due to the glycol added to the chemical composition of PET, which also enhanced the printability of the material.

### **4.1 DESIGN OF THE EXPERIMENT**

The Design of the Experiment (DoE) step is fundamental in order to determine the optimal printing parameters of all the material taken in exam, thus rPETG, rPLA, virgin PETG and virgin PLA (all without any colours added). The DoE process is required because every material can behave differently depending on the printer used and the relative settings adopted in the slicer, thus the goal is to find the best settings related to every single material for the printer used (Ultimaker 2+). Hence, for example, using the same printing setting (e.g. the best for Reflow's filaments) for all the materials could lead to faked results. In order to prevent so, three main tests will be performed: temperature tower, warping and shrinkage. Those tests, reported on Figure 34, have the objective of finding the best printing values in order to overcome the main problems that every FDM user faces during the first use of a new material, which are usually:

- The selection of the best extruder temperature range for correct the flow of the material during the print.

- The selection of the optimal first layer speed and bed temperature in order to limit the warping effect on the base of the print.

- The selection of the optimal printing speed in order to avoid an over extrusion effect or shrinkage in the print.



Figure 34 - Optimal Control Variables Selection

Every test, independently from the material in exam, will be characterized by two main variables categories :

- *Control Variables* : The control variables are the ones that will be settled into the slicer software before printing and will vary during every printing test in order to determine which one gives the best result. Each control variable will be tested in a reasonable range determined by the correspondent test and the one that will lead to the best response variable value will be chosen for the successive 3D printing of the specimens for mechanical tests.

- *Response Variables*: The response variables are the outputs values of the 3D printing tests and will determine which control variable is the most suitable for the specific material. The control variable that will lead to the best response variable will be the one used as setting for the 3D printing of mechanical tests samples.

In Table 11 it is possible to have a preview of the control and response variables chosen for every test:

	CONTROL VARIABLES	RESPONSE VARIABLES	
TEMPERTURE TOWER TEST	Extruder Temperature	Visual Inspection	
WARPING TEST	Bed Temperature First Layer Speed	Dimensional Accuracy (%)	
SHRINKAGE TEST	Speed	Dimensional Accuracy (%)	

Table 11 –	Control	and	Response	Variables
------------	---------	-----	----------	-----------

It is important to remark that during the first test only the correspondent control variable will vary (extruder temperature), while the others will be kept constant to a default value. Of course, after the selection of the optimal control variable of the first test (e.g best extruder temperature), the selected value will be used as default for the successive tests (warping and shrinkage). Consequentially, the best control variables founded for the warping test will be used as default value in the final shrinkage test. Moreover, each spool of every material was dried as specified by the manufacturer before every print in order to perform the tests with the best filament conditions as possible. The other setting values, independently from the test performed and material used, are the following:

Table 12	- Default	Printing	Settings
----------	-----------	----------	----------

SETTINGS	VALUES
Layer Height	0,2 mm
Layer Width	0,48 mm
<b>Perimeter Lines</b>	2
Outline	35%
Infill	100%
Top Layers	5
<b>Bottom Layers</b>	5

### **4.1.1 TEMPERATURE TOWER TEST**

The temperature tower test in essential in order to find the optimal range of temperatures that the extruder should keep in order to obtain a flawless extrusion flow. In order to perform this test, the STL model file of a temperature tower was downloaded on Thingiverse (www.thingiverse.com) and sliced with the Simplify3D software using the following range of decreasing temperatures (change happens every 50 layers) for each material as control variable:

PETG	PLA				
265°C	225°C				
260°C	220°C				
255°C	215°C				
250°C	210°C				
255°C	205°C				
250°C	200°C				
245°C	195°C				
240°C	190°C				
235°C					
230°C					
225°C					
220°C					

Table 13	_	Temperature	Test	Control	V	ariables
1001015		remperante	1001	Comitor	v	ununu

Is important to remark that only during this test the control variables differs between PETG and PLA, since the two materials have different melting points and maximum acceptable temperature, thus cannot be tested with the same range of values. After slicing, the prints were performed on a Ultimaker 2+ printer and then visually inspected in order to observe the best temperature for each material. Since a too high printing speed can cause under extrusion problems at low temperatures, the speed for this test was settled to 20 mm/s for every material in order to cover more control variables as possible, while the first layer speed and bed temperature were settled to the values advised by the filament manufacturers.



Figure 36 - Temperature Tower for rPETG



Figure 35 - Temperature Tower for virgin PETG

Figures 35 and 36 represent the results of the temperature tower test on the two PETG spools, thus for the Reflow recycled PETG (on the left) and competitor's virgin one (on the right). As can be seen, in both cases the PETG has a wide range of printable temperatures without any notable defects, covering all the control variable range. For this reason, the control variable selected is the same for both materials, and equal to 235°C, which re-enters also in the temperature range advised by the two different filament manufacturers.



Figure 38 - Temperature Tower for rPLA



Figure 37 - Temperature Tower for virgin PLA

For PLA, results are not optimal as for PETG. In fact, as can be seen from Figures 38 and 37, the temperature towers (on the left rPLA, on the right virgin PLA) cannot cover all the possible control variables selected for the test. This can be due to a too high printing speed for lower temperatures, that produces a under extrusion problem during the print. However, printing at speeds lower than 20 mm/s is not recommended, because a too low speed drastically increases the printing time. For this reason, the discussed test can be considered valid even if not all the control variables were explored, and the selected one is of 220°C for both materials, since from the figures is clear that this value leads to a flawless flow in both towers. Moreover, 220°C re-enters in the temperature range suggested by the two different filament manufacturers.

### 4.1.2 WARPAGE TEST

The second test required for the DoE phase is represented by the warping one. In 3D printing, and especially for FDM, warpage is one of the biggest problems that can occur. The warping effect is caused on plastic materials by the change of temperature between the extruder and the printing bed.



In fact, as showed in Figure 4.4 (M.Alsoufi et al.,2017), due to the polymer's characteristics, the material retracts when is subjected to a drastic temperature change in a short time (high temperature gradient), creating warped layers on the base and several problems during the print of the successive layers. In order to overcome to this problem, three different solutions can be adopted:

- Using a heated printer bed near the glass transition temperature of the polymer in order to limit the retraction. Usually every filament manufacturer tests the material multiple times before adding it to its material portfolio and shares with clients the best bed temperature that allows the molten polymer to stick to the bed for all the duration of the print.

- Use a limited first layer speed allows the first layer to stick perfectly on the bed and leads to better printing of successive layers, and thus of the overall print.

- Adding external adhesives agents, as glue or adhesive spray, can be the optimal solution if the material presents a visible warping effect even with low first layer speed and advised bed temperature.

It is important to underline that during this research the two control variables are represented by the bed temperature and first layer speed. No external adhesive agent will be used for any material, in order to analyse the response of the polymers in exam without the help of external factors. For this test, the 3D object file was designed on SolidWorks and created in order to measure the warping effect as better as possible.



Figure 40 - Warping Test Sample

As showed in Figure 40 (the represented dimensions are in mm), the specimen designed for this test is a low height parallelepiped with a wide base area. This design allows to measure the height at every corner after printing and compare it with the nominal one (5 mm). Since every measurement will be repeated 5 times, the mean value of the height will be:

$$Height (mm) = \frac{\sum_{i=1}^{5} Single \ measurement \ i}{5}$$

All the Height measured were > 5mm due to warping.

Successivly, the warping effect and the response variable, thus the dimensional accuracy at every corner, are calculated as follows:

$$Warping(mm) = Height - 5 mm$$

Accuracy (%) = 
$$\left(\frac{5mm - Warping}{5mm}\right) 100$$

While the error is:

# *Error* (%) = 100 - Accuracy

In conclusion, the control variable that will lead to the warping specimen with the highest Accuracy for every corner will be selected as optimal value for the successive test and printing of mechanical specimens.

The first control variable to be determined for this test is the bed temperature, since it has more influence on the print with respect the first layer speed. During the printing, multiple warping specimens will be printed for every type of filament, everyone at different bed temperature, and then measured with a Digital INSIZE Height Gauge, while the first layer speed will be settled to a default value of 10 mm/s and the nozzle temperature will be the one determined in the previous test. When the optimal control variable for bed temperature is selected, the same test is repeated with the first layer speed as control variable, while a bed temperature will be settled to the best value founded during the first warping test. In detail, for PLA and PETG (recycled and virgin), the bed temperature control variable will vary in the following range:

PETG	PLA
60°C	50°C
70°C	60°C
80°C	70°C

Table 12 - Bed Temperature Control Variables for Warping

While the second control variable (first layer speed), will vary equally for PETG and PLA in the following range:

PETG	PLA
5 mm/s	5 mm/s
10 mm/s	10 mm/s
15 mm/s	15 mm/s

Table 13 - First Layer Speed Control Variables

The results of the test are represented by the following Table, which shows which control variables for every material leaded to the best results in terms of dimensional accuracy (media between accuracy calculated at every corner), thus are selected for the following test and for the printing of the ISO mechanical specimens.

Table 14 - Warping Test Results

	Bed Temperature	Accuracy	First Layer Speed	Accuracy
rPETG	80°C	≥ 96%	10 mm/s	≥98%
rPLA	60°C	≥ 97%	10 mm/s	≥97%
Virgin PETG	70°C	≥97%	15mm/s	≥96%
Virgin PLA	60°C	≥95%	10 mm/s	≥96%

As can be seen, both bed temperature and first layer speed do not differ a lot between recycled and virgin materials. Moreover, all the values founded re-entre in the setting range advised by the filament manufacturers.

### 4.1.3 SHRINKAGE TEST

The last test required is the one related to the shrinkage problem, that can occur on every printed part due to the plastic materials' behaviour. As happens during the warping for the first layers, thus the retraction of plastic, also during the other layers the material retracts leading to a shrinking of the final part with respect the nominal dimensions defined in the 3D model file. This test aims to explore the influence of the printing speed (control variable for this test) on the final shrinkage and to select the one that leads to a final dimension close to the nominal one. In order to perform this test, a 3D cube was designed on Solidworks, with nominal dimensions as indicated in Figure 4.6 (dimensions are in mm).



Figure 41 - Shrinkage Test Sample

As for the warpage test, after every print at different speed for every material in exam, the cube's thickness will be measured 5 times for every direction with an INSIZE Vernier Gauge (on X,Y and Z). Then the mean value on that champion of five measurements of the shrinkage will be calculated as follows:

Thickness (mm) = 
$$\frac{\sum_{i=1}^{5} Single \ measurement \ i}{5}$$

It is important to remark that all the thickness measured were < 5 mm due to shrinkage.

Consequentially, the response variable (dimensional accuracy)with respect the nominal value will be:

Accuracy (%) = 
$$\left(\frac{Thickness}{5 mm}\right) 100$$

And the error will be:

$$Error(\%) = 100 - Accuracy$$
The control variable will vary for every material in a range descripted by Table 4.7, while the other settings will be the ones determined by the previous tests and the default one presented previously (Table 4.2). The control variable that will lead to the best medium accuracy on all directions will be the one selected for the following 3D printing of the mechanical specimens.

PETG	PLA
20 mm/s	20 mm/s
30 mm/s	30 mm/s
40 mm/s	40 mm/s
50 mm/s	50 mm/s

The results of the test, thus the choice of the best control variable and the relative medium accuracy, are presented in the following Table:

Table 16 - Shrinkage Test Results						
	Speed	Accuracy				
rPETG	30 mm/s	≥97%				
rPLA	20 mm/s	≥98%				
Virgin PETG	40 mm/s	≥96%				
Virgin PLA	20 mm/s	≥98%				

Looking at the result, it is clear the PETG (both virgin and recycled), at least for the filaments in exams, presents less shrinkage at higher speed with respect to PLA. It is important to underline that usually the printing speeds reported on the filament manufacturers' datasheets are higher because they represent the highest printing speed that guarantees an acceptable print minimizing the printing time, while for this study the aim is to find the one the minimize the shrinkage.

In conclusion, at the end of the DoE process, all the optimal control variables for every filament were selected and ready to be used as settings for the successive printing of the ISO samples.

# **4.2 3D PRINTING OF ISO SPECIMENS**

In order to determine the tensile and bending proprieties of the 4 filaments in exam (rPETG, rPLA, virgin PETG and virgin PLA) the following ISO standards for plastic material testing will be taken in consideration :

Table 17 -	Tensile	and	Bending	Tests	Standards
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TENSILE TEST	BENDING TEST		
ISO 527	ISO 178		

The specimens were designed on SolidWorks using the dimensions reported in the ISO standard documents, as reported in Figure 42:



Figure 42 - Tensile and Bending Samples

The overall printing settings for every material, determined by the DoE process plus the default ones, are represented in the following Table:

	rPETG	rPLA	Virgin PETG	Virgin PLA
Layer Height	0,2 mm	0,2 mm	0,2 mm	0,2 mm
Layer Width	0,35 mm	0,35 mm	0,35 mm	0,35 mm
Perimeter Lines	2	2	2	2
Outline	35%	35%	35%	35%
Infill	100%	100%	100%	100%
Infill Direction	Х,Ү,ХҮ	Х,Ү,ХҮ	Х,Ү,ХҮ	X,Y,XY
Top Layers	5	5	5	5
Bottom Layers	5	5	5	5
Extruder Temperature	235°C	220°C	235°C	220°C
Bed Temperature	80°C	60°C	70°C	60°C
First Layer Speed	10 mm/s	10 mm/s	15 mm/s	10 mm/s
Speed	30 mm/s	20 mm/s	40 mm/s	20 mm/s
<b>Extruision Flow Multiplier</b>	1	1	1	1

Table 18 - Overall Printing Settings

As can be seen from the Table, the DoE process has selected different control variables for virgin and recycled PETG, apart from the extruder temperature, which is settled equally to 235 °C. On the other hand, the two spools of PLA share the same settings, since the DoE has led to the same control variables selection for both recycled and virgin spools.

Moreover, as represented in the Figure 4.8, it is important to remark that each specimen will be printed laid on the printer bed (thus on the XY plane) five times for each infill direction (X represented by Figure 4.8a, Y by Figure 4.8b, and XY by 4.8c), in order to analyse the influence of the printing direction on the mechanical proprieties. This is applied also to the bending test specimens.



The different infill directions are characterized by the raster angle, thus the angle between the X direction and the printing direction. For example, a "X" direction of the infill means the raster angle is equal to  $0^{\circ}$ , while for the "Y" direction it is correspondent to  $90^{\circ}$ . Consequentially, the "XY" direction corresponds to a diagonal path of the printing head, thus to a raster angle of  $45^{\circ}$ .

After the DoE process and the creation of the G-CODEs using the Simplify3D slicing software, all the ISO mechanical specimens, both recycled and virgin, are printed using a Ultimaker 2+ 3D printer at room temperature. Moreover, as showed in Figure 4.9, the printer is covered on all the sides in order to keep constant the extruder and the bed temperature guaranteeing constant environment conditions during printing. Since every specimen has to be printed 5 times for each infill direction and material as defined by the ISO standards, every mechanical test (tensile and bending) will require 60 samples. In conclusion, a total number of 120 specimens were printed in the Reflow headquarter in Amsterdam and then shipped to Milan in vacuum bags for the final testing.



Figure 44 - 3D Printing Step

# 4.3 MECHANICAL TESTING AND RESULT ANALYSIS

## **4.3.1 TENSILE TEST**

The tensile test is performed at the material testing laboratory of the Politecnico di Milano university, following the procedures described by the ISO 527 standard document for testing plastic materials.



Figure 45 - Tensile Test

The test was performed at 5 mm/s on an MTS Alliance RT/100 tensile test machine using the 60 tensile dog bones shaped specimens (divided in groups of five depending on the life, material and infill direction of the sample) printed in Amsterdam, as showed in Figure 45. The machine was also equipped with a MTS strain gauge in order to measure the strain as indicated by the ISO 527 document. The results were then analysed using the MiniTab statistical software licensed by the Poltienico di Milano.Is important to remark that this Sub-Section will analyze the different output results for this test, while all the stress-strain graphs and specimens'pictures are available in the appendix at the end of the document. The following tensile test will analyze and compare the results for the following outputs over three main parameters (life, material and infill direction):

- Ultimate Tensile strenght (UTS)
- Young's Moduls (E)
- Strain At Break (ɛ Max)
- Yield Strenght (Rp02)



Figure 46 - Stress/Strain Curve

### - Ultimate Tensile Strenght (UTS)

The UTS is the maximum stress that a material can withstand while being stretched or pulled before breaking, and it is measured in megapascal (MPa). The UTS can be calculated as:

$$UTS = \frac{F max}{Ai}$$

Where F max is the maximul load applied on the speciemen before necking, expressed in Newton, while Ai is the original cross sectional area of the sample in exam. The following graph show the results of the test.



Figure 47 - Main Effect Plots for UTS

As can be seen, the recyled filaments showed and impressive results compared to the virgin ones, with almost the double UTS. Moreover, the PLA has better tensile perfromace with respect the PETG, and the infill direction that led to the best results was the X one, probably because for those champions the layers were printed in the same direction of the tensile force applied by the machine, while for the Y infill (worst results) the force is perpendicular to the printing direction.

#### **Analysis of Variance**

Source	DF	Adj SS	Adj MS	F-Value	P-Value
removal point (mm)	1	2,2	2,20	0,08	0,784
life	1	2678,3	2678,29	92,60	0,000
mat	1	285,8	285,80	9,88	0,003
dir	2	5000,6	2500,28	86,45	0,000
life*mat	1	133,6	133,64	4,62	0,037
life*dir	2	439,2	219,61	7,59	0,001
mat*dir	2	636,1	318,04	11,00	0,000
Error	49	1417,2	28,92		
Lack-of-Fit	7	234,5	33,50	1,19	0,330
Pure Error	42	1182,8	28,16		
Total	59	10616,8			

Figure 48 - Analysis of Variance for UTS

In addition, since the aim of the study is to comprare the data more specifically, a more detailed analysis based on the three main study variables (life, material and infill direction) was made. From the analysis of variance (Figure 48) is it clear from the low p-value that there is interaction between life (virgin or recyled), material (PETG and PLA) and infill direction (X,Y an Z) variables, thus an interaction study was peformed, with results showed in the following plots:



Figure 49 - Interaction Plot for UTS

The results show the interaction between the three main categories of variables in exam (life, material and infill direction). As can be seen, both rPLA and rPETG shows a higher UTS compered to the

correspondent virgin PLA and PETG. Moreover, the Y direction has led to the worst results indipendtly from the life and material, while the PETG in general has a lower UTS compered to the PLA, a part from the Y direction.

#### - Young's modulus (E)

The Young's Modulus is the slope of the linear part of the stress-strain curve for a material under tension, and it is measured in MPa. E is caculated by the following formula:

$$E = \frac{\sigma}{\varepsilon}$$

that represent the ratio beetween the force per unit area and proportional deformation in the liner elastic region on the tensile test's curve.



Figure 50 - Main Effects Plot for E

The results showed are similar to the ones of UTS. Recyled filaments showed a better behavior with respect the virgin ones, and the PLA has a medium higher slope with respect the PETG. Moreover, the X direction perfroms better than the other two, due to its parellesim with the applied tension force. As for UTS, also in this case an anlysis of variance and a study on the interaction was made.

Source	DF	Adj SS	Adj MS	F-Value	P-Value
removal point (mm)	1	4315	4315	0,06	0,808
life	1	1359230	1359230	18,87	0,000
mat	1	18177377	18177377	252,38	0,000
dir	2	2220871	1110435	15,42	0,000
life*mat	1	16	16	0,00	0,988
life*dir	2	1061914	530957	7,37	0,002
mat*dir	2	817195	408598	5,67	0,006
Error	49	3529159	72024		
Lack-of-Fit	7	600370	85767	1,23	0,308
Pure Error	42	2928789	69733		
Total	59	28135209			

#### **Analysis of Variance**

Figure 51 - Analysis of Variance for E

In this case, results slightly different with respect the UTS ones. In fact, as for UTS, the rPLA and rPETG presents an higher mean value of E compered to the ones of the virgin filaments, but not on the X direction, which is the one that breaks hardly. Moreover, the PETG presents an higher value of E on the Y direction with respect the XY one. Is important to note that the value of E does not rapresent the strenght of the material, but the slope of the stress-strain curve during the elastic zone.



Figure 52 - Interaction Plot for E

## - Strain At Break (ɛ Max)

The strain at break, also known as fracture strain, is the ratio between changed length and initial length after breakage of the test specimen. It expresses the capability of the material to resist changes of shape without cracking, and it is calculated as:

$$l(\%) = \frac{lf - lo}{lo} \quad 100$$



Where If and lo are respectively the final and initial length of the specimen.

Figure 53 - Main Effect Plot for l

In this case results shows that, even if in general the recyled materials presents higher values of the elongation at break with respect the corrispondent virgin polymers (as for the prevuois outputs in exam), the PETG shows a better elongational behavior with respect PLA, and the XY infill direction allows a incredible higher maximum strain with respect the X and Y ones.

#### Analysis of Variance

Source	DF	Adj SS	Adj MS	F-Value	P-Value
removal point (mn	n) 1	7,999	7,9994	11,15	0,002
life	1	33,272	33,2720	46,39	0,000
mat	1	39,343	39,3433	54,86	0,000
dir	2	131,743	65,8714	91,84	0,000
life*mat	1	1,774	1,7743	2,47	0,122
life*dir	2	16,689	8,3446	11,63	0,000
mat*dir	2	10,120	5,0601	7,06	0,002
Error	48	34,426	0,7172		
Lack-of-Fit	6	10,269	1,7115	2,98	0,016
Pure Error	42	24,157	0,5752		
Total	58	247,761			



Moreover, as can we see from the following interaction graphs, both the recyled PETG and PLA elongates more than the virgin filaments, and the XY direction confirmes to be the one with the higher possible elongation, indipently form life or material in consideration.



Figure 55 - Interaction Plot for l

### - Yield Strenght (Rp02)

The 0.2% offset yield strength is defined as the amount of stress that will result in a plastic strain of 0.2%. Results shows that the Rp02 values registered by the sensors during this test are very similar to the ones of the UTS. In fact, as for UTS, the recyled materials presents higher values with respect the virgins. Moreover, the PLA results to have an higher Rp02 as for the X direction, as it was for the UTS output.



Figure 56 - Main Effects Plot for Rp02

### Analysis of Variance

Source	DF	Adj SS	Adj MS	F-Value	P-Value
removal point (mm)	1	0,61	0,61	0,03	0,874
life	1	1405,73	1405,73	58,07	0,000
mat	1	826,89	826,89	34,16	0,000
dir	2	3618,83	1809,41	74,75	0,000
life*mat	1	6,53	6,53	0,27	0,606
life*dir	2	387,28	193,64	8,00	0,001
mat*dir	2	914,88	457,44	18,90	0,000
Error	48	1161,90	24,21		
Lack-of-Fit	7	97,98	14,00	0,54	0,799
Pure Error	41	1063,92	25,95		
Total	58	8362,75			

Figure 57 - Analysis of Variance for Rp02

Analzing the interactions plot, is it clear that the recyled materials perfrom better than the virgin ones in exam. Moreover, the X direction guarantees an higher Rp02, while the Y direction is not optimal, expct for the PETG, where the results are similar to the XY one.



Figure 58 - Interaction Plot for Rp02

In conclusion, the recycled materials have better charateristics than the virgin ones across all the test outputs, demostrating that recycling could be an effetive solution in order to create a circular manufacturing in th AM industry witouth losing mechanical tensile performance. Moreover, results have shown that the PLA allows to reach higher UTS and Rp02, thus and higher maxium reachable strenght, but the PETG presented an higher strain at break. In general, the X infill direction seems the preferable one for bearing tensile stresses, but the XY direction can reach a higher elongation.

## **4.3.2 BENDING TEST**

The 60 bending specimens printed at the Reflow headquarter in Amsterdam were divided in groups of five depending on life, material and infill direction of the sample. Successivly, the champions were vacuum packed and shipped to the Politecnico di Milano, where the three-point bending test was performed at 5 mm/s on the MTS Bending Testing machine showed in Figure.



Figure 59 - Bending Test

Differently from the tensile test, the three-point bending test output for this machine is represented by only the Flexural Strength, that addresses the highest stress (in MPa) experienced within the material at its moment of yield, and which for a rectangular cross-section is calculated as showed by the following bending curve graph:



Figure 60 - Force/Deflection Curve

As for the tensile, also for this test the flexural stress results were analysed using the MiniTab statistical software licensed by the Politecnico di Milano. Is important to remark that this Sub-Section will analyse the statistical output result for this test based on the different variables (life, material and infill direction), while all the stress-deflection graphs and specimens' pictures are available in the appendix at the end of the document.



Figure 61 - Main Effects Plot for Flexural Stress

Results showed in general a higher flexural stress for recycled materials, similar to what was discovered during the tensile test. Moreover, also in this case the PLA presents a higher output, and the X infill direction is the preferred one with respect the Y and the XY one.

## **Analysis of Variance**

Source	DF	Adj SS	Adj MS	F-Value	P-Value
life	1	797,4	797,41	13,83	0,001
mat	1	2427,2	2427,20	42,11	0,000
dir	2	5374,0	2687,02	46,61	0,000
life*mat	1	1497,1	1497,06	25,97	0,000
life*dir	2	1277,2	638,59	11,08	0,000
mat*dir	2	875,7	437,86	7,60	0,001
Error	50	2882,2	57,64		
Lack-of-Fit	2	421,6	210,78	4,11	0,022
Pure Error	48	2460,7	51,26		
Total	59	15130,8			

Figure 62 - Analysis of Variance for Flexural Stress

Since the analysis of variance showed low p-values for the variables in exam, the interaction plot analysis has to be taken in considerations. The following interaction plots shows how, even if previously the general results showed a better performance for recycled materials, in reality it is valid only for rPLA, while the rPETG presents lower flexural strength with respect the virgin counterpart. Moreover, the virgin filament performs better than the recycled in the X direction, while the Y direction clearly is the worst possible direction for this test, as it was for the tensile.



Figure 63 - Interaction Plot for Flexural Stress

# 5. FINAL CONSIDERATIONS AND CONCLUSION

This research has analysed and studied the environmental plastic impact over the last two decades, highlighting the major problems related to the massive production of synthetic polymers made during the last fifty years. Moreover, the plastic use in the AM industry has been explored, demonstrating how this technology could help to reduce plastic pollution and create a more sustainable circular economy worldwide. It is clear that this transformation cannot happen just researching and studying the impact of polymers, but an effective new way of production should be implemented. In addition, a strong commitment of world leaders, companies and individual should be triggered in order to prevent an ecological point of no return for our climate. After an accurate literature review, the sustainability analysis of the Reflow production was performed, together with a comparison with respect the emissions generated by the primary plastic production and recycling. The results of the analysis and comparison showed that:

- The Reflow filament production line, both for rPETG and rPLA, has a minimal impact on the environment. This is mainly due to the use of green energy sources as windmills for powering the plant, that leads to a low conversion rate from energy consumptions to GHG emissions and allows a sustainable production over time.

- The Reflow GHG emissions are impressively lower with respect the ones related to primary plastic production and recycling, due to two main reasons. First, the heat and power required for the extraction and transformation of raw materials into plastic are higher than the ones required for the extrusion process, leading to a higher amount of energy needed for the primary plastic production. Secondly, the traditional plastic manufacturing is performed in huge production plants powered by no-green energy sources, such oil or gas, that are characterized by high conversion rates from energy consumptions to GHG emissions.

In general, it is clear that the only way to for reducing emissions in the imminent future is to invest in green energy sources, even if the cost would be extremely high. Moreover, the different ways of recycling could help in reducing the production volume of synthetic polymers and create a circular economy system that uses waste as a source and not as scrap.

Later to the sustainability analysis, the 3D printing and mechanical performance comparison between recycled and virgin filaments was performed over three different infill directions. The results showed that:

- The 3D printing optimal settings founded during the DoE process for PLA doesn't differs from virgin and recycled filaments, while a slight difference on speed and temperature emerged between virgin and recycled PETG.

- The tensile test over three main variables (life, material and infill direction) showed in general better results for recycled materials over the virgin counterpart. Moreover, the PLA has a higher strength resistance with respect the PETG, but the maximum elongation is lower. In conclusion, due to the parallelism between the X infill and the tensile force applied, this direction is the preferred in order to obtain a higher strength of the printed part, while the XY direction allows the highest elongation.

- The bending test over three main variables (life, material and infill direction) led to slightly better results for the virgin PETG over the recycled one, while for PLA the recycled nature performed better. Moreover, virgin filaments presented in general a higher flexural strength on the X direction with respect the recycled ones, but not for the XY and Y infills. In addition, as for the tensile test, the X direction is the one that achieved the best results.

In conclusion, as it was meant to prove, the first-recycled filaments produced by Reflow do not lead to a loss in mechanical performance, but on the contrary presented better test results overall compared to the virgin ones. This study has showed how the plastic waste generated could be re-transformed in a feedstock material for FDM application without losing the characteristics desired, allowing to reach sustainable goals for companies and consumers and create a circular economy for the AM industry.

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ISO 527, International Standard for the tensile test of plastic materials

ISO 178, International Standard for the bending test of plastic materials

# APPENDIX

This appendix contains all the specific curves of the tensile and bending specimens tested at the material testing laboratory of the Politecnico di Milano. Each page will contain five specimens of same life (recycled or virgin), material (PETG or PLA) and infill direction (X,XY,Y), for a total of 120 samples.

## TENSILE TEST





























































0,035

0

-0,005

-5 0

0,005

0,01

0,015

0,02

0,025

0

0

0,005

0,01

0,015

0,02

0,025

0,03

-0,005






## **BENDING TEST**





50 40 Load (N)

Crosshead (mm)

Load (N)

Crosshead (mm)









Crosshead (mm)



Crosshead (mm)



































