

WASTE PLASTICS AS A SOURCE FOR 3D PRINTING FILAMENTS

PLASTIČNI OTPAD KAO SIROVINA ZA 3D FILAMENTE

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SAŽETAK

Zbog stila života proizvodnja različite vrste otpada u kućanstvu i industriji je u konstantnom porastu. Kako bi se smanjila količina otpada, koja svoj životni ciklus završi kao smeće na odlagalištima radeći ekološki problem sadašnjoj i budućim generacijama, važno je pravilno razvrstavanje i odlaganje kako bi se mogao koristiti kao sirovina. Plastika je nezamjenljivi materijal današnjice u područjima pakiranja, transporta, građevinarstva, medicine i elektronike. Zbog svojih svojstava i cijene koristi se u različite svrhe i praktički ne možemo zamisliti život bez nje što rezultira ogromnim količinama producirane plastike. Nesavjesno postupanje s plastičnim otpadom dovodi do onečišćenja okoliša, a posebice mora i oceana. Dugotrajna razgradnja plastičnih materijala je veliki problem, a dodatni problem je što njenom razgradnjom nastaje mikroplastika koja dospijeva u vodu, zemlju i zrak, a time i u prehrambeni lanac. Stoga je recikliranje najbolja opcija gospodarenja plastičnim otpadom. Oznaka otisnuta ili utisnuta na plastičnoj ambalaži daje informaciju o vrsti plastičnog materijala i mogućnosti reciklaže za određene svrhe. Mehaničkim postupcima plastični otpad se pretvara u reciklat koji se može koristiti za nove plastične proizvode. Plastični materijali zbog svojih svojstava i mogućnosti oblikovanja imaju gotovo neograničene mogućnosti te ih je moguće prilagoditi zahtjevima tržišta, a u današnje digitalno doba manualnu i tvorničku proizvodnju sve više zamjenjuje tehnologija 3D tiska usmjerena na izradu raznih proizvoda od polimernih materijala odnosno plastike. Ovaj rad obuhvaća pregled aktualnih mogućnosti u 3D tisku, te razmatra pristup pretvaranja plastičnog otpada u 3D filamente koji bi doprinjeli nužnom smanjenju plastičnog otpada.

Ključne riječi: *plastika, reciklat, 3D tisak, 3D filamenti*

ABSTRACT

Due to the lifestyle, the production of various types of waste in the household and industry is constantly increasing. To reduce the amount of waste, which ends its life cycle as garbage in landfills creating an environmental problem for current and future generations, proper sorting and disposal is important so that it can be used as raw material. Plastic is an irreplaceable material today in the fields of packaging, transport, construction, medicine, and electronics. Due to its properties and price, it is used for various purposes, and we practically cannot imagine life without it, which results in huge amounts of produced plastic. Careless handling of plastic waste leads to pollution of the environment, especially seas and oceans. The long-term decomposition of plastic materials is a big problem, and an additional problem is that its decomposition creates microplastics that end up in water, soil, and air, and thus in the food chain. Therefore, recycling is the best option for managing plastic waste. The symbol printed or embossed on the plastic packaging provides information about the kind of plastic material and the possibility of its recycling for certain purposes. By mechanical processes, plastic waste is turned into recyclate that can be used for new plastic products. Due to their properties and design possibilities, plastic materials have almost unlimited possibilities and can be adapted to market requirements, and in today's digital age, manual and factory production is increasingly being replaced by 3D printing technology aimed at 3D printing various products made of polymer materials, i.e. plastic. This research gives the overview of today's possibilities in 3D printing and considers the approach of converting plastic waste into 3D filaments that would contribute to the necessary reduction of plastic waste.

Keywords: *plastic, recyclate, 3D printing, 3D filaments*

1. INTRODUCTION

Waste recycling is a necessity today. Namely, the production of huge amounts of different types of waste, both in the household and in industry, is constantly increasing. Therefore, the first step in reducing waste is its proper sorting and disposal so that it has the opportunity to become a raw material, and not end its life cycle as garbage in landfills causing problems for current and future generations. Plastic as a relatively new material, that didn't start becoming popular till the 1950's, due to multi-functionality of its long chains or networks of monomer molecules which can be fabricated in desired shape, colour and specifications became an irreplaceable material today. It is inexpensive, has excellent barrier properties and is lightweight, making it a preferred material in various industries like packaging, transport, construction, medicine and electronics. The use of plastic has largely replaced natural products like metal, wood and fibres, which made it an integral part of society. Global plastic production reached 370 million tons in 2019 (Figure 1), and it is expected to increase to 900 million tons by 2050 [1].

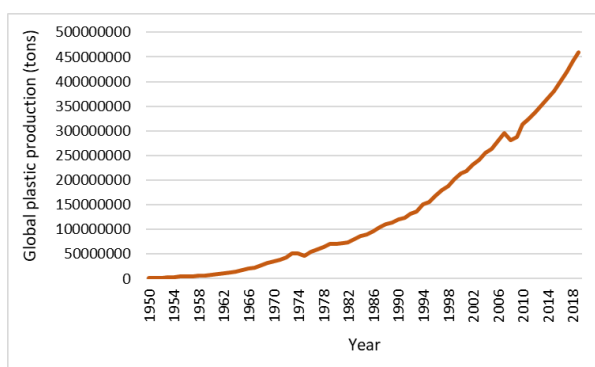


Figure 1 Global plastic production [2]

Given its very short shelf life (typically around 6 months or less), packaging is the dominant generator of plastic waste, responsible for almost half of all global waste [3, 4]. Packaging, as the main plastic waste fraction, in the European Union generate 15.8 million tonnes of plastic waste per year [5]. And in total, close to 26 million tons of plastic waste is generated annually in the countries of the European Union alone [6]. The huge amount of produced plastic waste creates increasing problems because careless handling of plastic waste leads to environmental pollution [7],

especially seas and oceans where plastic makes up 80% of all marine waste [8]. Plastic pollution is globally widespread in all oceans, and it is estimated that surface waters worldwide contain about 269,000 tons of plastic [9].

Plastic thrown into the environment remains in it for hundreds of years, and during decomposition it breaks down into smaller parts (Table 1), which in the form of microplastics end up in water, soil and air, and then through the food chain they reach the human body itself. Additionally, when plastic decompose, it leaks and adds pollutants (bisphenol A and phthalates) to the soil and surrounding environment [4]. Durability makes plastics an increasing problem for the environment, as it cannot easily be broken down by microorganisms [10]. The long-term decomposition of plastic materials is a big problem, and an additional problem is the entry of microplastics into the food chain [11]. Plastic can be degraded via a number of mechanisms (photo-oxidation, biodegradation, chemical and thermal), depending on their molecular structures. As a result of physical, biological and chemical processes, the structural integrity of plastic waste decreases over time and fragmentation occurs, resulting in small particles of microplastics (Figure 2, Table 1).

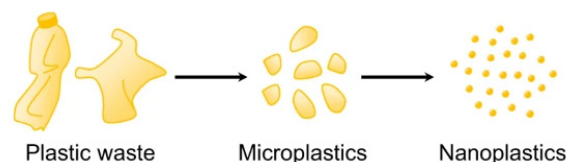


Figure 2 Fragmentation of plastic waste into micro- and nanoplastics [12]

Table 1 Plastic particles size categories

Particle category	Diameter range (mm)
Nanoplastics	< 0.0001
Small microplastics	0.00001 – 1
Large microplastics	1 – 4.75
Mesoplastics	4.76 – 200
Macroplastics	> 200

2. PLASTIC WASTE MANAGEMENT

Recycling is the best option for managing plastic waste, but when recycling is not a viable option, the alternative is energy recovery as a

replacement for fossil fuels. It is important to make aware that plastic products are too valuable to be thrown away. Namely, they can be recycled into a product for the same purpose or another product. The recycling processes require the public participation for the segregation of the waste material at the source, as a proper collection and sorting of plastic waste add more efficacy to recycling methods. All types of plastic are polymers, but all the polymers are not essentially plastic [4]. Polymer materials generally can be divided into three basic groups: plastomers (thermoplastics), duromers and elastomers. The major portion of used plastics belongs to thermoplastic which can be reformed into a fresh new product upon applying pressure and heat.

There are four major plastic waste recycling methods: primary or physical recycling, secondary or mechanical recycling, tertiary or feedstock or chemical recycling and quaternary recycling (incineration with energy recovery/thermal recovery) [4, 13].

Only homogeneous or well sorted waste plastic can be recycled by primary or physical recycling method, while it is not suited for multi-layered and heterogeneous mixed plastic wastes [4]. Secondary or mechanical recycling is the simplest and the most inexpensive method. Therefore, plastic waste is most often recycled by this method which includes cutting, shredding, contaminants separation, floatation extrusion, and palletisation. Produced secondary raw material (recyclate) can be used to manufacture new products having different characteristics from original plastic waste materials, as it implies thermal processing (mostly operated at the temperature of 200 - 300 °C) of the homogeneous old plastic material by melting with the aim of producing granules of recyclate. Recycling is carried out by extruding, that is, by continuously pushing the heated and softened polymer through the extruder head. This process is employed for a few times till the polymers break down and the quality aspects of the plastic deteriorates. About 80% of thermoplastic can be recovered by mechanical recycling process [6], but the problems in this method are mainly related to the heterogeneity of waste plastic materials.

Tertiary or feedstock or chemical recycling is the process which reduces a polymer chemically to its basic monomeric unit and then it can ultimately be re-polymerized and reformed. Namely, most plastic in use today comes from hydrocarbons derived from organic materials found in nature, such a crude oil, natural gas and coal, minerals and cellulose. Through a polymerisation or polycondensation process in presence of a suitable catalyst low-molecular weight compound (monomer) is produced which are further connected in a chains of a high-molecular weight compound (polymer). These monomeric units gained by chemical recycling can be utilized as virgin material substitutes in manufacturing new plastic polymers (new plastic products). This chemical method comprises manufacturing of some chemicals and energy fuels from plastic waste scrap and it can be applied to all forms of plastic waste mountains containing multilayered polymeric plastics which cannot be recycled using first two recycling methods. There is a wide range of chemical recycling methods (plastic to energy/fuel) as: pyrolysis, hydrocracking, catalytic cracking, solvolysis and gasification. Quaternary recycling or energy recovery is based on the use of heat contained in polymers, which are generally combustible materials. Waste that can no longer be used in any other way is burned in order to reduce its volume in landfills. This is of waste is the oldest, the easiest and the cheapest method to get rid of the discarded materials, but due to the potential health risks from contaminants emitted during combustion it is not favourite any more. The emissions from combustion mainly comprises carbon monoxide, sulphur oxides, nitrogen oxides, hydrochloric acid, volatile organic compounds, trace amounts of poisonous chemical such as styrene, benzene, dioxins, formaldehyde, furans, polychlorinated biphenyls (PCBs), heavy metals like lead, mercury and arsenic etc., all of which can severely impact the entire population of this plane [4].

However, the quality of recycled materials decreases over time with the accumulation of waste, while the quality of the polymer regularly decreases after each step of reprocessing [14], which results in poor mechanical properties and limits the application of recycled products [13].

It is estimated that there are about 900 different types of plastic materials that, due to their properties and design possibilities, have almost unlimited possibilities and can be adapted to the market's requirements. According to the classification developed by the SPI (Society of the Plastics Industry), there are seven types of plastics [11] listed in Table 2.

In many plastic products, the polymer is just one ingredient that is almost always combined with other ingredients or additives that are mixed during processing and manufacturing. Therefore, every plastic product has, or should have, a specific marking of a triangle made up of three clockwise arrows forming a triangle, inside which there is a number. Below such a label comes the abbreviation of the exact chemical name, although this abbreviation can often be absent. The number mark written inside the triangle gives us an information about type of plastic material. Therefore, plastic products that have the number marks help recyclers to sort it after usage into appropriate groups, as the recycling rate of domestic packages is limited by waste quality and the citizens' behaviour and

environmental awareness.

By volume, plastic waste makes up more than 30% of household waste. One of the benefits of plastic is its suitability for different applications. However, the diversity of polymers on the market is also a drawback, since it makes sorting and separation a challenge [15]. Therefore, marking plastic products with defined symbol gives a valuable information for end consumers to make proper waste separation in homes (Table 3). Sorting is essential for a well-functioning recycling system. Sorting to a high degree of purity means polymers are more likely to retain their value and stay in circulation longer. The high variety of polymers in plastic collected causes life cycle issues and only 42% of packaging waste is recycled in Europe [5].

Data from 2020 indicate that in the Republic of Croatia, a total of 54,748.0 t of plastic packaging waste was separated, and the total amount in municipal waste was 329,281.9 t. In sorted plastic waste, PET, LDPE, PS and HDPE were found as the predominant materials, while PVC was the least represented [6].

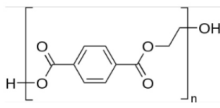

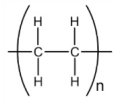

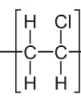

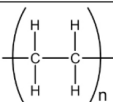

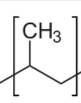

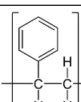


Number mark	Plastic type	Structure	Acronym	Symbol
1	polyethylene terephthalate		PETE/PET	
2	high-density polyethylene		HDPE	
3	polyvinyl chloride		PVC	
4	low-density polyethylene		LDPE	
5	polypropylene		PP	
6	polystyrene		PS	
7	other		O	

Table 2 Plastic markings and their meaning

Table 3 Sources of plastic waste for recovery depending on the numerical designation

Number mark	Acronym	Sources of plastic waste
1	PETE / PET	beverage bottles such as water, juice and milk; containers for food products; bottle caps; jars for various spreads, etc.
2	HDPE	bottles from detergents, shampoos, pesticides and other chemicals; pots; shopping bags, etc.
3	PVC	chemical packaging; packaging films; labels on bottles; pipes; Windows; door; various building materials, etc.
4	LDPE	bread bags; freezer bags; foils; nylons, etc.
5	PP	containers of dairy products; bottle caps; medical bottles; straws; disposable plates, cups and cutlery; hangers, etc.
6	PS	containers for catering; glasses for hot drinks; trays; for stronger packaging, etc.
7	O	multi-layer (laminated) materials: acrylic, polycarbonate, nylon, fiberglass, polyactide

3. 3D PRINTING AND PLASTIC MATERIALS

The goal of humanity is to prevent any generation of plastic waste through regenerative and cost-effective production cycles, and 3D printing could be a potential solution for the implementation of waste polymers to the greatest extent possible.

Fused filament fabrication (FFF) is one of the additive manufacturing (AM) technologies commonly known as 3D printing. It gained its popularity in the early 2000’s with the open-source project RepRap [16] and the 3D printing market is one of the fastest growing sectors (Figure 3) [17].

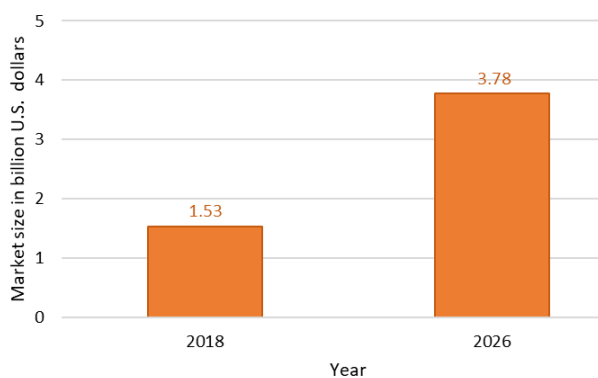


Figure 3 3D printing material market in 2018 and estimation for the 2026 in billion US dollars [17]

Namely, the digital age has brought drastic and rapid changes to the entire way the world functions. In the past, objects were produced either manually or by factory production in thousands of copies, but today this has started to change with the advent of the 3D printer, which enabled fast prototyping and production of unique products at home. There are currently about 130 types of 3D printing materials, mainly classified from polymers, metal, ceramics, and concrete. Among these materials, polymer materials, i.e., plastic are mainly utilized in 3D printing [18]. 3D printing is similar to inkjet printing, in which the ink is propelled from the nozzle of the printer in form of jet. The raw material in 3D printing is in the form of filament that is slightly heated at the time of ejection and the extruded on the bed, where the component is manufactured. On the exit, the outer core of the filament is in the semi molten form, that is fused with the earlier formed layer and solidifies to form the product [19]. So, we can say that 3D printing is a process of making three dimensional solid objects from a digital file. The creation of a 3D printed object is achieved using additive processes where an object is created by laying down successive layers of material until the entire object is created. Each of these layers can be seen as a thinly sliced horizontal cross-section of the eventual object [20].

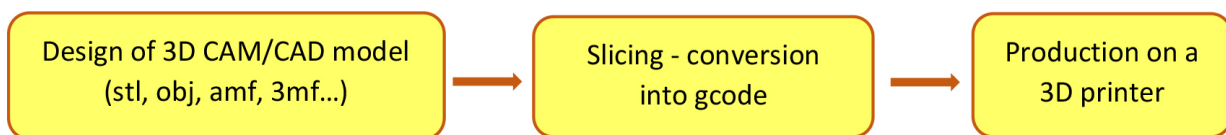


Figure 4 Common steps of the 3D printing process

Generally, 3D printing process is categorized on the basis of raw material form used and is always consisted of several steps listed in Figure 4.

First step is preparation of a model, whose design is first developed in a CAM/CAD software or an already produced component is converted into a CAM/CAD model using a coordinate measuring machine (CMM) or 3D scanner. The 3D model is exported from the CAM/CAD software in a format that describes the geometry of a 3D object (stl, obj, amf, 3mf...). Second step is slicing i.e., conversion of the 3D model file into a machine-readable format (gcode) which contains instructions for the 3D printer production parameters (axis movement, extrusion, temperature control etc.). And the last step is production where the received data consisting of production parameters is executed to produce the desired model.

There are two major variations in 3D printing techniques – fused filament fabrication (FFF), whereby plastic filaments are deposited on top of the same material to produce objects via adhesion or heat, and stereolithography (SLA)/selective laser sintering (SLS), whereby layers of powders or liquids are deposited with the use of photopolymer and UV laser [21].

FFF is by far the most widespread 3D printing technology. It works on the principle of melting the material that is most often brought to the printer in the form of a plastic thread, the so-called filament. This is placed on a spool in the tank of the 3D printer, from which the printer gradually unrolls the material and pulls it, into the print head, where it melts. The print head of the 3D printer then stacks layer after layer into a physical 3D model that it downloads from a computer, memory card, application, etc. Just as an inkjet print head injects a very small amount of ink onto paper, a 3D printer's print head deposits molten material onto a substrate layer by layer, creating a physical model. It is the cheapest 3D printing technology, which is the main reason for its widespread use. The disadvantage of this technology is a long printing time, a large variation in print quality, which depends on the printer model and the material used, and various design limitations (e.g., the molten material must harden first, for more complex models it

is therefore necessary to count on the printing of supports and the like). SLA 3D printing technology is stereolithography, which means curing polymer resin into a specific shape using LEDs or UV rays. The main domain of this 3D printing technology is its exceptional accuracy, which significantly exceeds the printing resolution of FFF printers. However, this technology is very expensive, and another serious problem is that during the hardening and curing of the resin, toxic gases are released so it should be used in controlled, ventilated environment to reduce the health concerns for the user.

Most 3D printing technologies are focused on 3D printing of various products made of polymer materials, i.e., plastic. Among the commercially available materials, polylactic acid (PLA) and acrylonitrile butadiene styrene (ABS) are the two most commonly used (Figure 5).

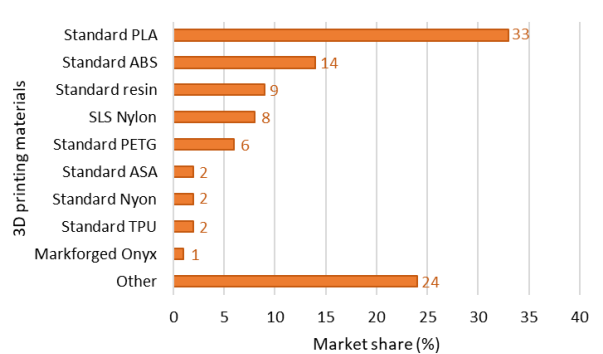


Figure 5 Most commonly used 3D printing materials in 2018 [22]

Table 4 compares the properties of commonly used thermoplastic materials for 3D printing. Thermoplastics (thermos - warm, hot and plast from the ancient Greek plássein - to form, shape), also called plastomers, are plastics that can be deformed by heating in a certain temperature range. The common properties of materials for 3D printing are low melting point and reduced viscosity [23].

The use of recyclates in the production of new plastic goods is an important contribution to a circular economy, as it enables closing material cycles and reduction of dependencies on fossil raw materials and greenhouse gas emission minimization. After the plastic waste is sorted, it is compounded into recyclates. Any impurity and contamination will lead to potential problems in

Table 4 Properties of the common 3D printing filament [13,14]

Thermoplastics	Temperature (°C)		Properties	
	Printing	Degradation	+	-
PLA	175-200	300-400	eco-friendly, biodegradable, good adhesion at high printing speeds, especially good at producing sharp corners, less prone to warping	fan required at the extruder, fairly brittle, susceptible to heat thus not ideal for long-term outdoor use, limited gluing
ABS	220-240	380-430	resistance to high-temperature environments like sunlight or hot water, good adhesion at high printing speeds, slightly flexible, smooth extrusion, easy sanding, easy gluing	mild chemical odour during extrusion, expands and shrinks in heating and cooling, thus heated plate required. Imperfect bonding between layers, will bubble when exposed to moisture
Nylon	230-270	390-450	excellent layer adhesion great bridging capabilities and durability, tear resistance, dyeing filaments before print provides a tie-die effect, odourless	more prone to curling thus a heated build platform required, potentially emits cyanide at high temperatures (fine at printing temperatures)
PC	≥ 260	400-500	high resistance to scratches and impact, high strength and durability	hygroscopic, can undergo a change in state when exposed to UV, more opaque and brittle overtime, can release toxic fumes
PET	212-235	350-480	fairly stiff and very light weight, strong and impact-resistant, Taulman T-Glase is FDA approved polymer for food contact, 100 % reclaimable	hygroscopic, more brittle than PLA
HIPS	210-230		easy to paint and glue, great for printing lightweight parts	still at experimental stage to be used as a soluble support
PVA	160-205		biodegradable, recyclable, eco-friendly, high bonding power, good barrier properties	very hygroscopic, not suitable in slight humid conditions

the compounding process and finally affect the recycle quality. Insufficient sorting purity of the fractions due to missorting leads to problems in the compounding process. Namely, when recycling mixed thermoplastic fractions, different melting points and processing temperatures (Figure 6) lead to a loss of quality in the recycle. Processing two melting components with similar processing temperatures can change the properties of the final product, such as colour. If, on the other hand, components with a higher melting point are present, they can be separated and removed by the melting filter, but at the same time the melting filter is clogged, requiring a greater cleaning effort. Low-melting components in the recycle will overheat and degrade, which affects the properties of the final product, like reduced mechanical and optical

properties. This problem particularly often affects the polymers polyethylene (PE), polypropylene (PP), polystyrene (PS), polyamide (PA) and polyethylene terephthalate (PET), which are often used for food packaging, because they are not compatible at the molecular level. Processing compatibility issues can also occur within these polymer groups, for example when polymer types PET-A and PET-G or PP-H and PP-C are processed together [24].

Due to their properties and design possibilities, plastic materials have almost unlimited possibilities and can be adapted to market requirements, and in today's digital age, manual and factory production is increasingly being replaced by 3D printing technology aimed at 3D printing various products made of polymer

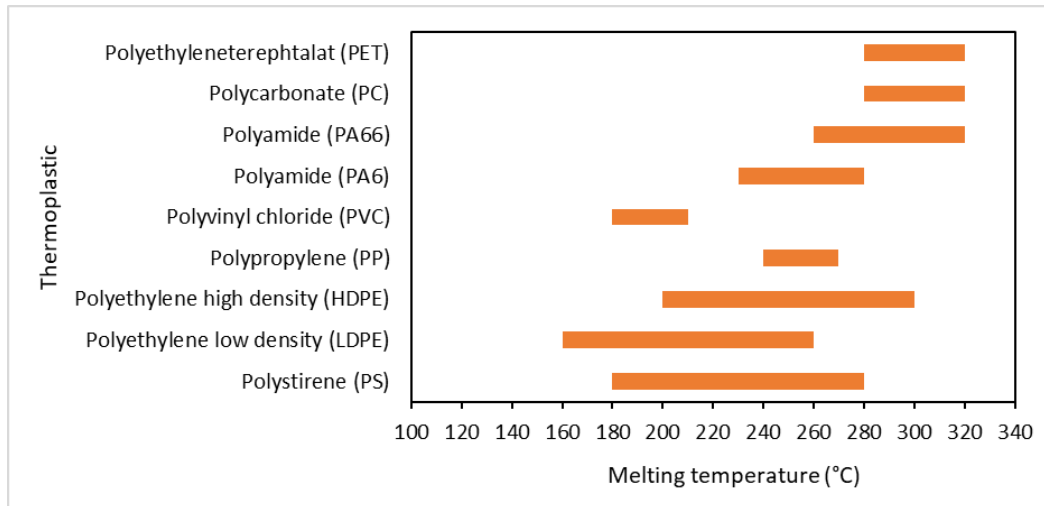


Figure 6
Processing temperatures of common thermoplastics [24]

materials, i.e., plastic. 3D filaments can be made from different types of thermoplastic materials, so the approach of converting plastic waste into 3D filaments would contribute to the necessary reduction of plastic waste. Therefore, turning plastic waste into new filament also contributes to the necessary reduction of plastic waste, which is necessary to preserve our planet as we know today. However, it should be noted that not all plastics can be recycled for these purposes because not all plastics behave in the same way. In order to know whether the plastic material can be turned into new filament or not, it is necessary to correctly identify the plastic waste, which is helped by the recognizable number (from 1 to 7) in the typical "recycle" icon.

Plastic mass processing is called extrusion and the machine in which the plastic processing process takes place is called an extruder (Figure 7). Plastic extrusion is a process in which a thermoplastic material, in the form of powder, pellets or granulates, is homogeneously melted and then forced out of the shaping die by means of pressure. The simplest extruder consists of a funnel for dosing the plastic mass, a cylinder in which the molten mass moves and in which one or more screws are placed, which by their rotation transport the plastic mass along the cylinder to the opening where the tool for obtaining desired product. Electric heaters are used to heat the cylinder to melt the plastic mass, while a water circulation system or air is used for tempering and cooling. In this manufacturing process it is very important to keep right operating control inputs and

temperature measurements for required polymer and resin [25].

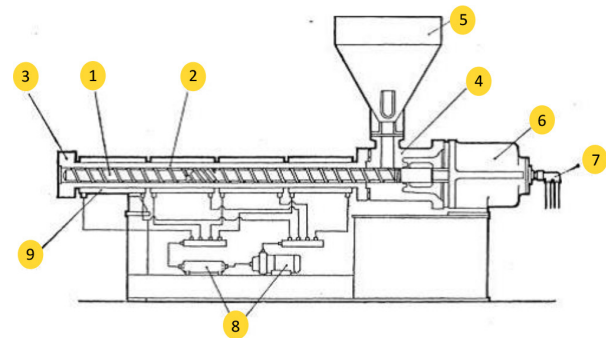


Figure 7 Extruder with basic elements: 1) screw, 2) cylinder, 3) coupling for connection with the tool, 4) water-cooled inlet zone, 5) funnel for dosing the raw material, 6) extruder drive, 7) tempering of the screw, 8) system for cooling and tempering of the cylinder, 9) heating elements for heating the cylinder

Plastic extruders are also used extensively for the reprocessing of recycled plastic waste where the material is typically extruded into filaments. 3D filaments can be made from different types of thermoplastic materials. These are linear or branched polymers in which the molecules are held together by weak secondary bonds, and they are made of polymer resins that become a homogenized liquid when heated and cooled, and are also fusible and soluble. When the plastomer is softened by heat, the macromolecules are free to move because the secondary forces are relaxed and can then be molded by extrusion. By cooling the plastomer, the opposite process occurs, the secondary bonds are re-established and the plastomer acts like glass, is subject to breakage, and offers versatility and a wide

Table 5 Recyclability of plastic waste for 3D filaments

Number mark	Acronym	Recyclability	Difficulties
1	PETE / PET	+	can absorb the smell of food and liquids for which it is used as a container and above all it is designed for blow moulding
2	HDPE	+	to ease the process cooling system recommended
3	PVC	-	its potentially dangerous chemical composition
4	LDPE	+	chopping it is not easy due to the shape of the elements in which we find it
5	PP	+	-
6	PS	+	it is possible that particular types of polystyrene cannot be extruded

array of applications. Due to the character of the secondary bonds, the cycle of softening and hardening can be constantly repeated, therefore it is possible to materially and mechanically recover the plastomer, i.e., recycle it. Although extrusion is possible for many of the materials listed in Table 5, each plastic, even of the same type, can show a different behaviour (due to the injection moulding of the original parts). Furthermore, the quality of the extrusion can depend on various factors, such as the external temperature, air humidity or the size of the starting granules. The research conducted by Sonjaya et al [26] showed that the best temperature to mould the plastic waste is 190°C, not only for polypropylene plastic cups, but also for combined polyethylene terephthalate plastic bottles and polypropylene plastic cups and that these filaments produced by extrusion machines were almost in line with the market PLA 3D printing filament.

4. CONCLUSION

Plastic waste, which mainly comes from households and industry, is a very problematic polluter of the ecosystem, and it is extremely important to involve citizens in the circular economy approach. Citizens can deliver dry and clean recycled materials to recycling containers which will be the raw material for numerous products. In the era of digitization and the increasingly rapid development of 3D printing technology for various types of industries, the production of environmentally friendly 3D filaments based on recycled polymers can help reduce plastic waste pollution.

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