

## MATERIAL RECYCLING AND IMPROVEMENT ISSUES IN ADDITIVE MANUFACTURING

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**Abstract:** *The objective of this study is to find ways of reusing polymeric material left over in Selective Laser Sintering (SLS) in other Additive Manufacturing (AM) processes, i.e. Fused Deposition Modelling (FDM). Furthermore, attempts are made to improve the material's mechanical properties. This paper provides information about the changes of tensile properties taking place in the un-sintered polymeric material after it has been exposed to SLS process and the tensile properties that the recycled material obtains when additives are added into its matrix.*

*Key words:* Additive Manufacturing, Selective Laser Sintering, Fused Deposition Modelling, material recycling, Polyamide-12

### 1. INTRODUCTION

Additive Manufacturing is being falsely advertised as ‘the technology that creates no waste’. Additive and subtractive manufacturing are both widely used for fabricating customized products. Subtractive manufacturing begins with a block of material from which the desired object is cut out by removing material producing waste as a by-product. In contrast, AM adds raw material layer by layer in order to form the desired object; this doesn't mean absolutely no waste is created. There are a number of different types of AM processes; most of them create waste in the form of supporting material used in the manufacturing process.

In Selective Laser Sintering a part's three-dimensional design is created in a

computer aided design program. That 3-D design is virtually sliced into layers with a thickness 0.1 mm [1]. A recoater in the SLS machine applies a layer of polymer powder onto the print bed and a laser beam traces the model's cross section fusing the polymer particles into a solid object layer after layer, leaving the non-traced powder loose [2].

One of the most widely used polymer powders used in SLS is polyamide. All powder present in SLS process is subjected to high heat, which deteriorates polymer particles [3]. The heat and radiation initiated degradation of polyamides may involve the following: change in chemical structure of the polymer molecule, change in crystallinity (molecular orientation), dipole rearrangement, hydrogen bridging and change in the amount of molecularly associated materials that have plasticizing effect, such as water. The polymer molecule breaks at the C-N bond; moreover, breaking of a very small proportion of the C-N bonds can already result in an appreciable decrease in molar mass. Increase in crystallinity is proposed accompanying the decrease in molar mass. All these degradation effects may result in appreciable change in physical characteristics of polyamide [4]. Therefore, only certain amount of processed (presumably degraded) material can be used in mixture with non-degraded, new material to preserve characteristic mechanical properties of the material. This results in excess of polymer powder with changed properties that cannot provide sintered parts with as high quality as would

be achieved by using virgin powder. Therefore a considerable amount of raw material goes to waste; only a certain percentage, 30 - 70% (depending on the material) [1], of leftover can be blended with virgin material and re-introduced into the process assuring production of high quality parts. Since the powder deteriorates differently at different parts of the build chamber the quality of the recycled powder can vary greatly. If too high percentage of heavily deteriorated recycled powder is blended with virgin powder, the produced parts will have surface defects such as 'orange peel'. In order to avoid the risk of producing damaged parts, producers frequently decide to blend just a small percentage of recycled powder into virgin powder [1]. By producing more polymer powder waste than is recycled there will always be leftover powder that will be permanently scrapped.

This research encourages environmentally friendly thinking and acting by studying the waste generated in SLS process and aiming to develop new materials of this waste that would be suitable for using in FDM process. This way all leftover polymer powder which otherwise would be scrapped, could be made into new raw material.

The ultimate goal of this study is to develop a set of polymeric materials that could function as the raw material for FDM printing hand prostheses. Currently a USA based non-profit organization e-NABLE and its global network of volunteers FDM print prosthetic hands at their own expense for people in need across the world [5]. Plastic filament makes the bulk of the cost of such prostheses. If material left over from SLS could be made into filament that would classify as acceptable prostheses material, it would be possible to make printing hands economically more viable. In addition to tackling the economical factor, opportunities to make a version of that recycled material that is stiff and a version that is flexible are explored. Another goal is to make an electrically

conductive polymer that is suitable for using in mechatronic equipment.

In this research, polyamide-12 (PA-12) that is left over from SLS process in Tallinn University of Technology has been studied with a focus on giving the material new mechanical properties. PA's mechanical properties were manipulated by adding thermoplastic polyurethane (TPU) ground pellets and aramid fibres into recycled PA-12's matrix.

TPU is a linear block copolymer that is made up of hard and soft segments, which are separated during extrusion or injection. This separation of microphase segments gives the material unique properties including high tensile strength, high elongation at break, good wear and tear resistance and low temperature elasticity. The shortcoming of TPU is expressed in low temperature resistance, which may become a problem during processing the material. Blending TPU with other (heat resistant) materials, such as polyamide, helps to overcome this issue [6]. Aramid is considered to be a high performance fibre [7]. High levels of molecular alignment result in high levels of stiffness and strength of aramid fibres [8].

Polymers are reinforced with short fibres to increase their strength and load carrying capacity of the components. Among the fibre reinforcements as glass and carbon, aramid fibres are widely used. Polymer composites reinforced with these fibres are usually one to four times stronger and stiffer than their unfilled equivalents. The reinforcement reduces also coefficient of friction and hence allows the material to be used for higher duties without exceeding the softening point of the matrix [9]. Good adhesion (which is necessary for reinforcing effect) between PA matrix and aramid fibres is expected due to similar chemical nature.

TPU and polyamide mixture provide material with toughness, low temperature flexibility, tensile strength, tear strength, abrasion resistance, transparency, resistance to oils, resistance

to hydrolysis, resistance to kinking and hardness without brittleness [10].

In addition to perfecting current materials' composition and structure, further studies will tackle PA's electrical conductivity and visual properties. Depending on the achieved properties of the material some possible applications are considered, e.g. limb prostheses, mechatronic equipment, etc.

## 2. IMPROVEMENT OF MATERIALS

This study investigated the change in mechanical properties of un-sintered PA 2200, which is a modified version of polyamide 12 supplied by EOS GmbH [11], after the polyamide powder was processed in Formiga P100 (EOS GmbH) and after that reused powder was added TPU and aramid.

All raw materials were dried in a convection oven at 105°C for four hours; after drying the raw materials were stored in airproof containers until processing them in the injection moulding machine. Four different sets of test specimens were prepared at 215°C using micro-injection moulding machine Babyplast 6/10P [12]. Specimens were prepared of the following materials 1) 100% virgin PA-12, 2) 100% reused PA-12, 3) 90% reused PA-12 and 10% mechanically milled TPU pellets, 4) 95% reused PA-12 and 5% aramid fibres.

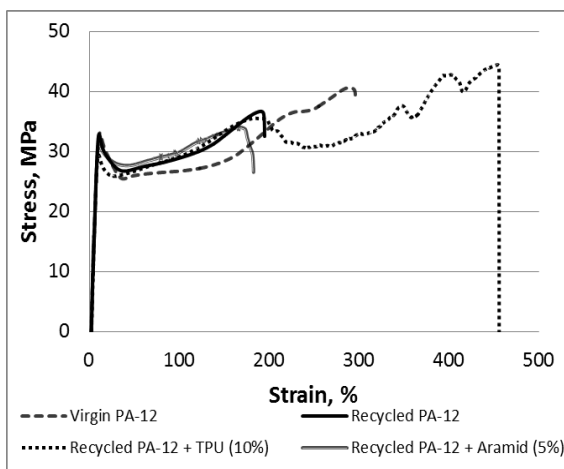


Fig. 1. Comparison of tensile stress against tensile strain of specimens made of different materials.

The brand name of TPU that was used is Elastollan® which is made by BASF. This material has excellent low temperature flexibility [13]. Short fibre reinforced PA-12 was prepared using aramid fibres with a length of 2 mm.

## 3. TESTING OF MATERIALS

All specimens were prepared according to the standards of ISO 527-2 type 5B. A small specimen size was chosen in order to preserve unnecessary waste of materials and to shorten the testing time.

Tensile testing was carried out on all four different sets of prepared specimens using Instron 5866 machine. The crosshead speed was set at 50 mm/min. Each specimen was individually measured. The results of 10 successful tensile tests of each set of specimens were recorded.

A sample filament was produced of recycled PA-12 using a laboratory compounder manufactured by Brabender.

Following properties were measured: maximum tensile stress, tensile strain at break, modulus and energy at break. Recycled PA-12 filament was used as raw material in Vellman K8200 FDM printer [14] to 3-D print an object in order to be able to analyse FDM-printability of produced material.

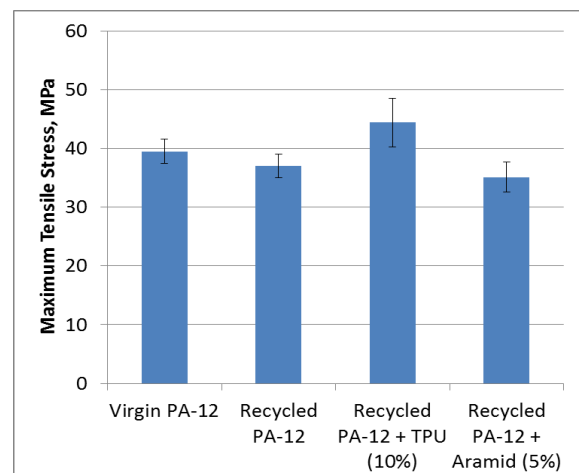


Fig. 2. Tensile strength of different compositions.

Material	Maximum Tensile Stress, MPa		Tensile Strain at Break, %		Modulus, GPa		Energy at Break, J	
	Value	Standard Deviation	Value	Standard Deviation	Value	Standard Deviation	Value	Standard Deviation
Virgin PA-12	39,5	2,1	296,8	21,3	0,60	0,10	5,90	0,60
Recycled PA-12	37,0	2,0	198,5	34,3	0,60	0,10	3,90	0,70
Recycled PA-12 + TPU (10%)	44,4	4,1	421,4	132,1*	0,56	0,05	9,21	3,18
Recycled PA-12 + Aramid (5%)	35,1	2,6	172,6	23,7	0,64	0,07	3,32	0,55

**Table 1.** Mechanical properties of different PA -based materials (\* high variability is caused probably by two defected specimens).

#### 4. RESULTS

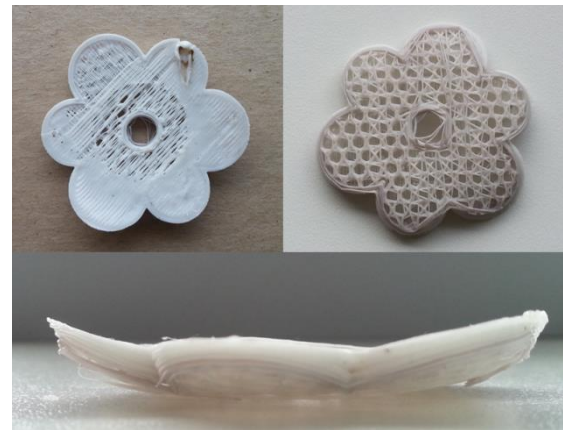
Fig. 1 shows typical tensile test diagrams of the four different material sets of specimens studied. Fig. 2 represents the comparison of average tensile strength of each material type. The standard deviation is marked for each set of specimens.

The mechanical properties of all four tested PA -based materials are shown in Table 1. Some preliminary tests were made to analyse the suitability of recycled PA-12 in an FDM machine. Fig. 3 shows a flower printed with recycled PA-12 in an FDM printer.

#### 5. DISCUSSION

The results show that the biggest difference between virgin and recycled PA reflects at tensile strain at break and also in energy at break (see Table 1). Other results are more similar. Perhaps there is not that much difference between these materials and recycled PA could be successfully used for most applications that virgin PA is used for. However, the quality of virgin material is much better controlled; it is more homogenous and predictable.

Adding 10% of TPU to PA may have been too small amount for making it softer since the modulus did not change



**Fig. 3.** FDM printed Recycled PA-12 flower

significantly compared to pure PA. However, the maximum strain increased considerably, and also the tensile strength increased. More studies for identifying the influence of different levels of TPU in PA are needed.

Adding 5% of short aramid fibres made the material only slightly stiffer. Small difference may have been caused by either too low reinforcement content or non-uniform fibre distribution inside the test specimens. Therefore, some further studies are needed.

Studies show that the recycled PA-12 is FDM-printable, but at this point the quality is far from acceptable level (see Fig. 3). The main issues to deal with are achieving control over the thermal

expansion of the material and assuring good adhesion of the material to the building platform of the FDM machine.

## 6. CONCLUSION

The study focused on recycling issues of polyamide powder left over in Selective Laser Sintering process. The attempts were made to improve the material and re-use it in Fused Deposition Modelling system. In the study four polyamide based materials were tested: a) unprocessed virgin PA; b) recycled PA; c) recycled PA + TPU (10%); d) Recycled PA + short Aramid fibres (5%). Mechanical properties of the materials were measured. The best results showed recycled PA + TPU. The addition of TPU increased tensile strength and maximum strain.

Future studies for identifying optimal content on additives should be made. A study for focusing on FDM processing using the created materials is planned.

## 7. ACKNOWLEDGEMENTS

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