

Investigation of 3D printing conditions for composite material based recycled HDPE and hay

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ABSTRACT: The main aim of this paper is to present the research findings which come out from the experimental investigation of the 3D printing conditions for composite material based waste materials (recycled HDPE and meadow hay). During the hay-plastic composites (HPCs) production, important raw material parameters such as hay sawdust particle size, hay/plastic concentration ratio or type of plastic matrix can be recognized. In this research study, the aim was to produce filament for 3D printer from hay-plastic composite material of an acceptable and competitive level of quality which could be used for 3D printing. Particle size of hay sawdust used for production of HPC has significant influence on mechanical properties of composites and also on other important properties (water absorption, hardness, frost resistance, etc.). The paper deals with the investigation of the impact and the relationship between the input hay-plastic composite material properties and production processes – filament producing and 3D printing. As the input raw material, meadow hay, HDPE plastic matrix and recycled HDPE, represented by lids from PET bottles, was used.

KEYWORDS: 3D printing, hay sawdust, hay-plastic composites, filament, particle size, recycling.

I. INTRODUCTION

In recent years, the global demand for plastic-based products has risen considerably. According to PEMRG [1], global plastic production reached 359 million tons in 2019, with Asian countries accounting for 51% and Europe accounting for 17%. The supply of virgin plastics consumes 4% of global demand (1.3 billion barrels a year) [2]. The polymers used in manufacturing are often non-degradable and can last for several years in the environment [3]. As a result, environmental

contamination caused by this type of waste may be a big issue. According to statistics, up to 90% of plastics can be reused. Currently, 80 percent of plastic waste is disposed of in landfills, with just a small percentage recycled. The most serious issue is HDPE, LDPE, PP, and PVC plastics, which are widely used by factories and pollute the environment with gas emissions [4]. PLA-related pollution, which has a much smaller global effect due to its natural roots, is a much smaller global concern. Unfortunately, products made of this material are less physically stable, discouraging future producers from using it more often. The most significant drawback associated with fabric reuse is the issue of properties being lost following several recycling cycles. Furthermore, there is a lack of equilibrium, which can harm public wellbeing [5].

One of the possibilities for the production companies to ensure the EU strategic plan is implementation of the circular economy. The circular economy, also called “green economy”, is a new economic model that is the opposite of the current model - the linear economy [6]. The essence of the profit of the current system of “take-produce-throw away” is above all the high consumption of renewable and also non-renewable raw materials, which logically cannot work in the long run.

There are few reports on WPC (Wood Plastics Composites) that help recycled thermoplastics. Most of the studies have focused on using only one kind of waste plastic or a combination of recycled and virgin plastics to provide WPCs, though some have also looked at recycled plastic blends. The effect of recycled or waste thermoplastics in WPCs, on the other hand, is still unknown, leaving research possibilities for improving product properties available.

When we add other negative factors to this, such as the use of cheap manpower from developing

countries, the population explosion, increasing consumption and the devastating human impact on the environment, we can rightly consider the current system to be unsustainable [6]. Economically, ecologically and socially. On the contrary, the circular model is intended to ensure the competitiveness of countries, their stable economic growth and a healthy environment. The yield in the circular economy is based on the efficient use of natural resources achieved by the efficient recovery of the materials, products and components used. Their constant return to the technical and biological cycle represents the closure of material flows. This radically minimizes waste and the costs of material inputs and energy required for the production of new products. The main features of this concept are the use of renewable energy sources, eco-innovation, rental, sharing or support of local trade. The rising concern towards environmental issues and, on the other hand, the need for more polymer-based materials has led to increasing interest about polymer composites based waste materials [7].

Humans have been making composites out of biomass sawdust and waste plastics for many years. Waste-based composites are becoming increasingly popular in both commercial and non-commercial applications. Due to global demand for waste-based composite materials, a global shortage of trees in many areas, and environmental awareness, research on the manufacture of composites manufactured from various waste materials is being pursued vigorously. The global industry is currently interested in the development of waste-based composites made from waste straw, glass fibre, waste plastics, wood sawdust, agricultural wastes and other products. The alternative of using waste-based composites materials is not only one possible solution to the shortage of accumulated capital in some areas, but also a means of helping to conserve the atmosphere, as the wood industry already produces a vast amount of waste that is not reused or recycled. 3D printing is a developing additive manufacturing (AM) technology that enables the printing of lightweight and complex structures, which are hardly achievable by other manufacturing methods. The distribution of AM throughout 2015 to 2019 in prototype manufacturing, production, the research sector, and mechanical part manufacturing is graphically shown. It is seen that 3D printing has been mainly used in prototype manufacturing

throughout these years, and the growth of this technology in actual production is still at a lower level[8].

The general purpose of this paper is to present the research findings regarding the investigated conditions for 3D printing when using a waste based materials. Effect of particle size and percentage amount of hay sawdust in composites is very significant and this paper this effect on production processes were determined. Authors would like to present this effect also in case of HPCs based virgin thermoplastic and also in case of HPCs based waste raw materials. Such of results are very important and interesting from the production possibilities and applications of HPCs based waste raw materials point of view. Obtained future research findings can be very helpful at HPCs production using 3D printers and shown the possibility of using also waste raw materials for HPC products, and thus increase the environmental responsibility with the environment protection.

II. MATERIALS AND METHODS

The main aim of our experiment is to determine the effect of raw material properties (particle size of hay sawdust) on the filament production and on the 3D printing of material based waste hay and HDPE. For purposes of determination this effect, the basic raw materials had to be chosen and prepared. HDPE (high-density polyethylene named TIPELIN 1108J from Slovak company Slovnafta.s. Bratislava, with the melt index 8.0 g / 10 min), HDPE rec. (recycled high-density polyethylene originating from lids of PET bottles) were used as plastic matrices. Meadow hay sawdust originating from South Slovakia was obtained from agricultural company. This kind of agricultural crop is a typical crop in Slovakia and are widely and usually cultivated in our country. Samples of meadow hay in the untreated form were obtained from an agricultural company. For processing the hay to the form of hay sawdust the hammer mill STOZA ŠV5 equipped with screen 8.0 mm and 4.0 mm in diameter was used. On Fig. 1 can be seen the achieved samples of hay sawdust after shredding. Hay is used as the biomass fibre in this experiment. The hay's bulk density was 64.76 kg/dm³, the hemicellulose content is 45.2 %, and the cellulose content is 33.82 %.



Figure 1 Shredding of hay to the samples of hay sawdust – with particles up to 0.5 mm (left), with particles up to 1.0 mm (right)

Shredding on two levels was used, for obtaining proper amount of samples with suitable level of fineness. Initially, Retsch Vibrating Sieve Equipment AS 200, according to the EN ISO 17827-

1 [7], for analysing of the particle size distribution (Fig. 2) was used. Fig. 2 shows the raw material particle size distribution

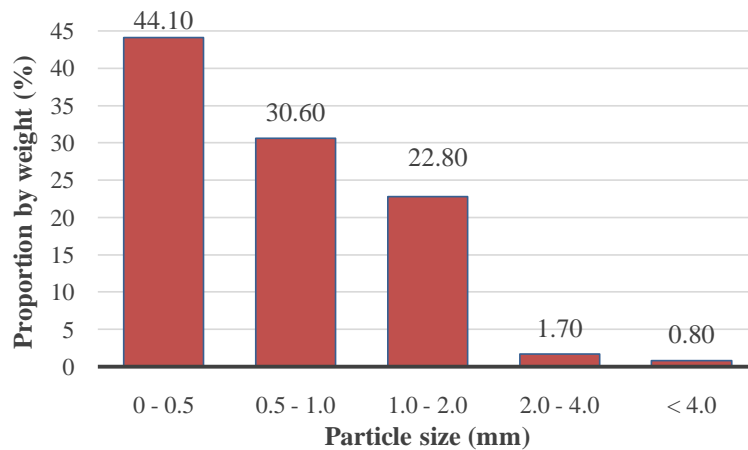


Figure 2 Raw material particle size distribution of the samples studied

In our case, determination of the mutual interaction between production processes (filament producing, 3D printing) of HPC's samples, type of the plastic matrix used in HPCs, hay/plastic concentration ratio and particle size of hay sawdust used in HPCs were chosen [9]. Experimental research was done according to the designed experimental plan where the full factorial

experiment was used [10]. Experimental research consisted of 3 influencing parameters, the specific type of polymer matrix - on 2 levels, particle size - on 3 levels and concentration ratio of plastic/hay where the 90:10, 80:20, 70:30 ratios were used (see Table 1). That was our plan for experimental investigation.

Table 1 Input controllable variables of the experiment [12]

| VARIABLES | | | |
|-----------|-----------------------|--------------------|-------------------------|
| LEVELS | Hay/Plastic ratio (%) | Particle size (mm) | Polymer matrix type (-) |
| 1 | 0 / 100 | 0 - 0.5 | HDPE |
| 2 | 10 / 90 | 0.5 - 1.0 | HDPE rec. |
| 3 | 20 / 80 | - | - |
| 4 | 30 / 70 | - | - |

Because the experimental research with raw waste materials was dealt and according to given experimental plan hay for the experiment had been treated. For obtaining the given hay particle sizes the disintegration and separation processes were used. According to our knowledge as a particle size only particles 0-0.5 mm and 0.5-1.0 mm were chosen [9]. Bigger particles are not usually used during the production of SPCs. For this experiment a meadow hay sawdust with up to 0.5 mm and up to 1.0 mm particle size was used (Fig. 1), with

properties shown on Fig. 2. The moisture content of chosen straw before mixing, extrusion and injection were measured with the aid of a Kern MRS 120-3 balance. This measurement consisted of heating the raw feedstock (gravimetric method of moisture content measuring) [11] at $105 \pm 2^\circ\text{C}$ until a constant weight was achieved. The moisture content of hay sawdust on the level of 2.2 % was prepared. For disintegration of PET bottle lids (Fig. 3) cutting mill Retsch SM 300 was used.



Figure 3 The disintegration of PET bottle lids

The whole treatment and samples preparation consisted of following steps:

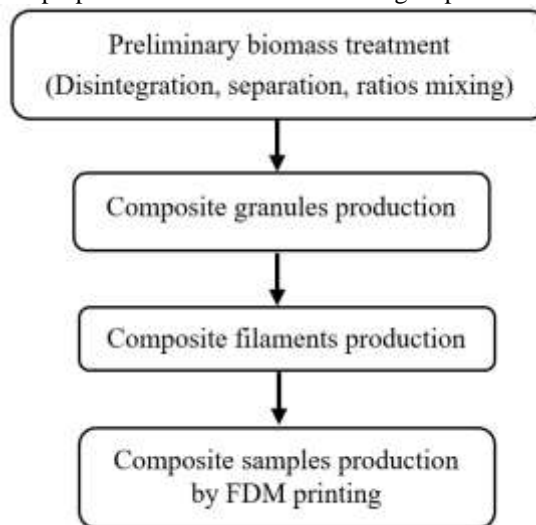


Figure 4 Material treatment and samples production process

Composites granules for filament production by twin-screw extruder LTE26 will be produced [10]. Final composite filaments for each setting by Filament Maker Composer will be produced (Figure 5).



Figure 5 Extrusion and production of composite granules by twin-screw extruder

For plastic parts production by FDM technology are used as standard plastic materials ABS (Acrylonitrile Butadiene Styrene), PC (Polycarbonate) or PLA (Polylactic acid). In our case was used composite mixture based HDPE and biomass (meadow hay). They have different processing requirements as for example required temperature but also very important material properties. Selection of printing device type for produced specimen depends mostly from used input material to be printed because not all FDM devices are able to process all available materials[12]. Temperature is necessary for plastic material melting and layer thickness. Each producer of

filaments states just range of suitable temperatures, not the exact temperature [13]. This is because different 3D printers work with different software and have different technical design, so there could be necessary different temperature setting, but also settings of many other parameters. The temperature affects the consistency or the flow of semi-melted thermoplastic what will affect the diffidence of deposited fibres [13, 14]. Layer thickness is affecting the dimensional accuracy, so we try to figure out if there is also influence to shape and positional tolerances [14]. Each 3D printer allows to set layer thickness and it is easy to change and affect the final quality of produced parts.



Figure 6 Composite filament production

For the filament production the filament maker 3devo was used. In our experiment, we are planning to print specimens with standardized

dimensions. A set number of specimens for testing will be produced by FDM 3D printer Prusa i3 (Fig.7)



Figure 7 FDM 3D printer used for the experiment

III. RESULTS AND DISCUSSIONS

According to the properties of filament maker but mainly according to the properties of 3D printer, the experiment had to be reduced on the base of output parameters of 3D printer. Because the output diameter of printing nozzle is 1.0 mm and

optimal diameter of filament is 1.76 mm, we have used only such compositions where 0 - 0.5 mm hay particle size was planned to avoid troubles during printing. So the full-factorial experimental plan listed in Table 1 was reduced to plan shown in Table 2.

Table 2Reduced experimental plan for production filaments and 3D printings

| SAMPLE # | TYPE OF PLASTIC MATRIX | AMOUNT OF HAY (%) | PARTICLE SIZE (mm) |
|----------|------------------------|-------------------|--------------------|
| 1 | HDPE vir. | 0 | - |
| 2 | HDPE rec. | 0 | - |
| 3 | HDPE vir. | 10 | 0.5 |
| 4 | HDPE rec. | 10 | 0.5 |
| 5 | HDPE vir. | 20 | 0.5 |
| 6 | HDPE rec. | 20 | 0.5 |
| 7 | HDPE vir. | 30 | 0.5 |
| 8 | HDPE rec. | 30 | 0.5 |

On the following Figures 8 - 12 you can see the produced filaments according to the reduced plan (Table 2).



Figure 8Produced filament from virgin HDPE



Figure 9Produced filament from recycled HDPE



Figure 10Produced filament from virgin HDPE with hay sawdust (ratio 90:10)



Figure 11 Produced filament from recycled HDPE with hay sawdust (ratio 90:10)



Figure 12 Produced filament from virgin HDPE with hay sawdust (ratio 80:20)

However we tried do our best, we were able to produce the filament only from 5 samples. Even when we adjusted the filament maker extrusion parameters. Virgin HDPE filament without addition of hay, recycled HDPE filament without addition of the hay and virgin HDPE filament with 10% of hay addition had proper output parameters for printing and stable shape and surface of filament. These seem to be used for 3D printing. Recycled HDPE filament with 10% of hay addition and virgin HDPE filament with 20% of hay addition where unusable for 3D printing, because their shape was not equal throughout the filament and the filament was thin. The settings of filament maker kept during extrusion the filament are shown in Table 3. As a plastic matrixes were used virgin HDPE and recycled HDPE with the addition of meadow hay sawdust. There were some limitations because of the 3D printer. It has its own limitations where the extruder speed was not able to increase as

required. Its maximum speed was 15 Rpm. We have adjusted the extruder speed according to the requirements of the filament. So that the filament can be produced without any damage and in the required quality. Also, the fan output power of the filament maker was adjusted according to the requirement of the filament. During production of the filaments the temperature at the beginning of the screw (T1) was constant for all the filaments. But at the end of the screw (T2), it was changing as shown in the below Table 3. Where we have compared the different temperature at different point. The puller speed influences the output shape and quality of filament, and thus also the filament maker productivity. This parameter of filament maker was also necessary to adjust according to the final filament quality. In some cases, the filament maker was able to automatically control the puller speed, and no operator intervention was required.

Table 3 Filament production parameters according to the material composition

| SAMPLE # | TYPE OF PLASTIC MATRIX | AMOUNT OF HAY (%) | EXTRUDER SPEED (RPM) | FILAMENT FAN POWER (%) | T1 (°C) | T2 (°C) | T3 (°C) | T4 (°C) | PULLER SPEED (RPM) |
|----------|------------------------|-------------------|----------------------|------------------------|---------|---------|---------|---------|--------------------|
| 1 | HDPE v. | 0 | 4.60 | 55 | 240 | 230 | 230 | 220 | automatic |
| 2 | HDPE r. | 0 | 7.10 | 40 | 240 | 230 | 230 | 220 | 14.37 |
| 3 | HDPE v. | 10 | 10.50 | 45 | 240 | 235 | 235 | 230 | 15.04 |
| 4 | HDPE r. | 10 | 10.10 | 30 | 240 | 235 | 235 | 230 | 16.00 |
| 5 | HDPE v. | 20 | 15.00 | 45-50 | 240 | 235 | 235 | 230 | automatic |
| 6 | HDPE r. | 20 | - | - | - | - | - | - | - |
| 7 | HDPE v. | 30 | - | - | - | - | - | - | - |
| 8 | HDPE r. | 30 | - | - | - | - | - | - | - |

After preparing the filaments we tried to print the samples within the 3D printer. But during printing we faced another troubles. Because the input material composition was not proper the basic questions was which 3D printing parameters should be adjusted? This significantly influences the printing process and a shrinkage of printings. Shrinkage of printings can cause due to many reasons, but the main 2 reasons were bed temperature and bed layer. Our 3D printer have limitations regarding the printing temperature, because during the printing can be adjusted only up to 120 °C. Other problem was the adhesion of printings on the bed layer. So we decide to investigate the optimal solution as bed layer for such an investigated material compositions. The general aim was to

increase the adhesion forces between printing surface and bed layer. This can be done with right combination of bed temperature, extruder temperature and type of bed. Material composition of bed layer should be as close as possible to the printed material. Due to the many options and for obtained clear results dealing with influence of bed type on adhesion, only one setting was chosen. The 3D printer was adjusted on following technological parameters, extruder temperature 190 °C, bed temperature 120 °C and printing speed 18 mm/s. As a bed layer the different types were used, which basic composition you can see in Table 4. Bed layers which were used during investigation can be seen on following Figure 13.



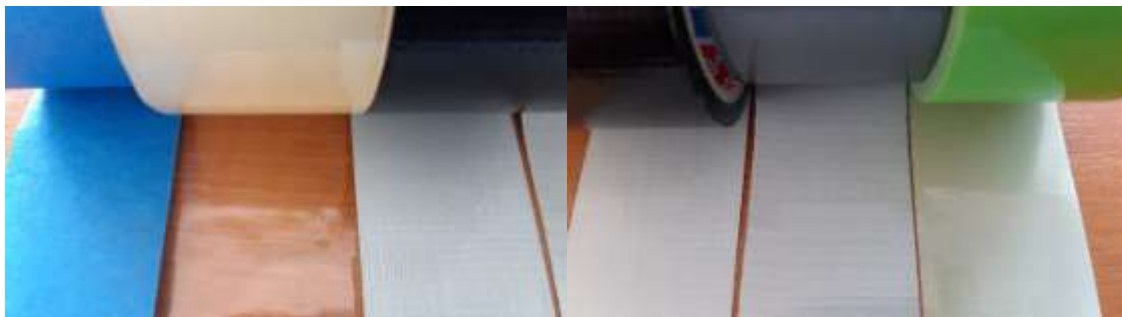


Figure 13 Bed layers used for testing of adhesion during 3D printing (description in Table 4)

Table 4 Description and composition of bed layers

| TAPE COLOUR | BASE MATERIAL OF TAPE | MATERIAL OF 2ND LAYER | STRUCTURE OF 2ND LAYER |
|--------------|-----------------------|-----------------------|------------------------|
| blue | paper | no layer other | no structure |
| transparent | PP | no layer other | no structure |
| black | PVC | no layer other | no structure |
| black | PVC | textile | rectangle |
| grey | LDPE | textile | line |
| green - neon | LDPE | textile | square |

In this phase of investigation only displayed types of bed layers were used due to availability. During this investigation only with virgin HDPE filament without additional hay and recycled HDPE filament without addition of hay were successful. Filament with addition of hay

wasn't able to create enough adhesion for continuous printing. As we can see on following Figures 14 – 17, most of bed layers were not suitable for printing. Except of LDPE tape with second squared textile structure was suitable for printing during both types of printing materials.



Figure 14 Polypropylene (PP) transparent bed layer

On the above Figure 14 we see that during printing in PP bed layer, even if we used recycled HDPE or virgin HDPE as filament, problem was in

the corners of printings, were getting shrink and the sample was not able to stick to the bed.



Figure 15 Polyvinyl chloride (PVC) black bed layer

The same results were achieved also during printing using PVC bed layer (Figure 15) and also during printing using LDPE (grey) bed layer (Figure 16). The corners of printings were getting shrink and the sample was not able to stick

to the bed due to the small adhesion. However it looked like the LDPE tape was better than both previous tapes. The improvement can be attributed to the presence of second (textile) layer of this tape. This textile layer has lined structure.



Figure 16 Low-density polyethylene (LDPE) gray bed layer

Finally, we decided to use another tape based LDPE but with different structure of second

layer. Second (textile) structure of this tape (green neon) was squared. Using this type of bed layer we were able to print stable samples (Figure 17).



Figure 17 Low-density polyethylene (LDPE) green-neon bed layer

IV. CONCLUSION

Investigation of filament production and 3D printing conditions using wasted plastic and hay raw materials was presented in this research. Presented results of preliminary phase relates to the effect of waste based materials and effect of hay sawdust addition into the printing material on printing conditions.

The main conclusions that can be withdrawn from this study are as follows:

- HDPE recycled originating from PET bottle lids can be used for filament production,
- Waste based material (meadow hay) can be used for filament production,
- Composites based HDPE recycled with addition of hay are insufficient for 3D printing within investigated conditions (technological parameters, type of 3D printer),

- Tape based LDPE with second textile squared structure can be used for printing using recycled HDPE based lids from PET bottles.

Additional phase of this experimental research will concern to research of printings shrinkage and optimization of 3D printing technology from the technological parameters point of view. Research of basic material composition suitable for 3D printing and development of HPC's composition based on waste materials which can be used for 3D printing is very ambitious and interesting issue.

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