Progress in Additive Manufacturing (2022) 7:445–452
https://doi.org/10.1007/s40964-022-00300-y

FULL RESEARCH ARTICLE

Under-extrusion challenges for elastic filaments: the influence of moisture on additive manufacturing

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Received: 11 October 2021 / Accepted: 5 April 2022 / Published online: 29 April 2022

Abstract

The applicability of Additive Manufacturing for operational parts expands with the availability of new materials with specific properties. For elastomeric components produced with Fused Filament Fabrication, challenges associated with the printing process due to the nature of the material are faced. This paper investigates the effect of under-extrusion in this process regarding the feeding system and, predominantly, the moisture for thermoplastic polyurethanes with 3D printing experiments and thermomechanical testing. In particular, the filament flow control with a Bowden extruder provides a challenge. A microscopic analysis reveals the signs of under-extrusion, along with the influence of material drying to reduce the moisture content. The drying may depend not only on time and temperature, but also on mass and surface effects. Water uptake measurements exhibit absorptions up to 1.89% in weight, most of which take place during the first 24 h of the experiments. Tensile tests performed on samples with different moisture contents show their influence in the ultimate stresses. The moisture in the material causes under-extrusion induced failures. Those failures are less likely to happen at lower moisture levels, resulting in occasional higher tensile strengths. Overall, the importance of proper storage of the material throughout printing is verified, even under moderate humidity conditions due to its hygroscopic nature.

Keywords Fused filament fabrication · Moisture · Thermoplastic polyurethanes · Microscopic analysis · Tensile tests

1 Introduction

Elastomers constitute a significant portion of applications in various industrial segments, which may benefit from the advent of Additive Manufacturing (AM) processes in the late twentieth century. The layer-by-layer process of joining materials to make objects from a CAD file [1], has allowed a higher design flexibility, with the exemption of individual tooling and reduction of waste by-products [2–4]. Initially for prototyping purposes, the rather young AM method is under continuous development and research, becoming a feasible alternative to conventional manufacturing for some applications. Consequently, elastomeric components may not only be produced on demand with reduced warehousing costs [5], but also considered for a quicker replacement of damaged parts discontinued by original manufacturers.

Much progress has been achieved in, e.g., medical applications and metallic parts [5, 6]. The same trend has been reflected for flexible parts, and today there are several materials and technologies available with good mechanical properties compared to conventional manufacturing. These flexible materials display an ease of deformation, but not necessarily the recovery to the original state. Overall, AM itself has a great potential for the elastomeric field. However, along with the benefits there are also downsides. Thus, research becomes a fundamental stage to better understand the behaviour of the associated materials and, consequently, help in improving the printing process for the manufacturing of functional parts.

One of the main drawbacks nowadays is the inferior mechanical properties of the 3D printed elastomers concerning operational performance and service life. The combination of high stretchability with a complete recovery to the initial state after unloading and long-term stability, characteristic of traditional elastomers, are not usually achieved. One reason is the usual vulcanisation process not being

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easily transferred to AM. Moreover, each AM technology requires a specific semi-finished product, e.g. thermoplastics as filaments for Fused Filament Fabrication (FFF) and as powder for Selective Laser Sintering (SLS), UV-cured resins for Vat Photopolymerization and Material Jetting. Since none of those make use of traditional rubber vulcanization, the achievable material properties are not similar. Usually, 3D printed elastomers possess poorer performance [7]. The question at the moment lies in improving such materials to better reach these properties.

Thermoplastic elastomers (TPEs) are the rubber-like option for AM processes operating with thermoplastics, being thermoplastic polyurethanes (TPUs) typically employed. TPU is known to result from physical crosslinks between soft segments (based on polyester/polyether chains, forming the elastic matrix) and hard segments (based on isocyanates, acting as multifunctional tie points). The combination of these segments gives different TPU grades, and, when balanced, can lead to unique and high-performance properties [8–10]. The availability of those materials enabled their use in FFF printers and, therefore, the fabrication of parts with rubber-like behaviour on one of the most popular AM processes.

However, in FFF several issues arise; some associated with the printing technology itself, some with the material. Most FFF printers, with nozzle diameters from 0.25 to 0.8 mm, restrict the part size because of the limited build area. FFF is relatively time-consuming due to the single nozzle material deposition. Interlayer adhesion also plays an important role, being achieved by a good balance of airflow and printing temperatures to allow sufficient line attachment without overheating the material to a too low viscosity or to the polymer thermal degradation [11].

Moreover, the adhesion between the build plate and the first layer of the object, the printing of complex geometries and extrusion failure mechanisms are common problems faced on FFF [12]. Bed adhesion can be improved by coating it with an adhesive and increasing the contact area and/or bed temperature. Support material allows the printing of overhangs and bridges in complex geometries. Extrusion failure may result from [13]:

- improper filament diameters, being avoided by stricter manufacturing tolerances;
- annular backflow, i.e. polymer flow in the opposite direction of the extrusion inside the die annular region and consequent solidification and clogging of the nozzle;
- filament buckling, causing resistance and hindering a constant material flow to the hot end.

Concerning elastic filaments as source material for FFF, extrusion failure is predominant and should be addressed. Particularly, filament buckling is typical due to the low elastic modulus of TPEs [13]. The extrusion, therefore, is not as trivial as for most common, commercially available filaments, which are rigid enough to reduce buckling effects. Typical solutions include: reducing extrusion speeds, applying minimal (at times, deactivated) retraction, and printing with a constant flow [12]. Faster print head movements in the *X*–*Y* plane also compensates the low retraction without inducing too much stringing into the process.

When printing with TPEs, the extruder type comes to evidence, as the distance between the motor and the nozzle influences directly on buckling. The remote Bowden extruder (Fig. 1a) reduces the print head weight and associated vibrations but imposes a great length of compressed filament towards the heated nozzle. Buckling can be reduced by using a Direct Drive (Fig. 1b), mounted directly on the print head, offering more extrusion control. The latter feeding system is typically preferred when dealing with elastic filaments. It is important to mention, however, that it is possible to use the Bowden extruder, but more challenging. In contrast, a Direct extruder does not fully eliminate buckling issues. A progressively apparent alternative to overcome these issues is the use of pellet-based systems, where the chances of buckling or bending are eliminated [14].

Extrusion failures may also have physical sources. Generally, TPEs are hygroscopic. Hence, moisture absorption prior to printing will condition the extrusion to a streaming with voids due to steam formation on the hot end [12]. A common solution is drying the filament. Proper storage before, after and, depending on the printing environment and hygroscopic levels, during printing must be implemented to preserve the material and ensure good printing quality.

In the light of the discussed issues, this work comprises experimental investigations for a better understanding of the 3D printing of elastic materials and is a contribution to the material database in the matter for prospective studies and subsequent industrial implementations. The aim of this paper is to investigate and discuss the under-extrusion of commercially available TPUs caused by buckling and

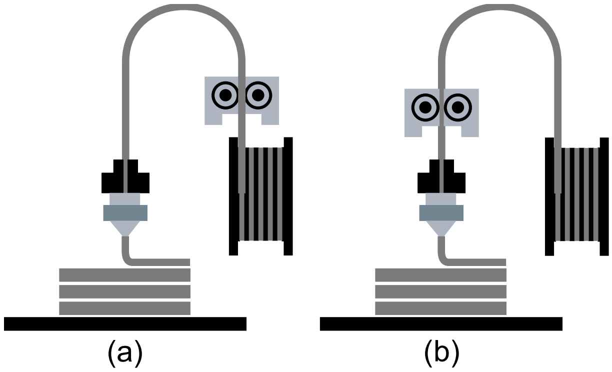

Fig. 1 FFF printer with **a** Bowden and **b** Direct extruder

moisture. Buckling is discussed in a qualitative analysis of the feeding system. Moisture, a recurrently faced situation, has a special attention and is verified with a microscopic analysis, water uptake and tensile tests. The samples were fabricated using conventional FFF printers. Different material conditions were considered to study the influence of moisture on the printed parts, its impacts on visual aspects and the mechanical behaviour.

2 Methodology

Print jobs were carried out on an Ultimaker S5 (Bowden extruder, 2.85-mm diameter) and a German RepRap x500 (Direct extruder, 1.75-mm diameter) printer with a 0.4-mm-diameter nozzle. TPU filaments from companies Recreus (Spain) and Volaprint (Germany) were selected due to their soft nature. Bed heating for improved bed adhesion was not necessary. Moreover, due to the absence of shrinkage, skirts were preferably used instead of brims to ensure good flow before printing. Flat parts were printed and complex geometries were avoided, as support structures cannot be broken away and easily removed due to the material rubber-like nature.

The feeding systems were comparatively analysed for the two extruder types, exploring the related under-extrusion causes and solutions. A qualitative microscopic analysis was performed with material Recreus Filaflex 70A, printed on the S5 printer. The filament was stored in vacuum bags but exposed to air (humidity 40%; 23 °C) during printing. Dryings in a Nabertherm TR60 convection oven ($450 \times 380 \times 350$ mm) were performed at 50 °C for 4–5 h, considering common parameters for TPUs in the 3D printing community (at least 2–4 h at 50°–60 °C [15]). The extrusion for different moisture conditions was analysed, noting the influence of drying time associated with material amount.

Water uptake tests according to Standard DIN EN ISO 62 with materials Recreus Filaflex 70A and 82A and Volaprint TPU 70A contribute to the filaments hygroscopicity analysis. Three 25-mm-long specimens per filament were previously dried in the convection oven (72 h; 50 °C), immersed in distilled water (23 °C), dried with a paper tissue and weighed after specific time intervals on a Sartorius ME235P electronic analytical scale (range 230 g; accuracy 0.01 mg). The water absorption was fitted to an ideal model in accordance with Fick's Law. Constant water absorption properties over the thickness were considered. The absorbed water content $c(t)$, i.e., the percentile rate of the difference between actual and initial weights over the initial weight of the sample, is described as follows. Parameters c_s, k, D and s are, respectively: water content at saturation, index of summation, diffusion coefficient in the surface normal direction and filament diameter.

Table 1 Printing parameters from Mechanical Analysis

Temperature	Infill	Perimeter outlines	Retraction
235 °C	100% ǀ 90°–0°	2	3.5 mm ǀ 40 mm/s

Table 2 Variations from Mechanical Analysis

Variations	Layer height [mm]	Printing Speed [mm/s]
A	0.1	15
B	0.1	7.5
C	0.2	15
D	0.2	7.5

$$c(t) = c_s \left\{ 1 - \frac{8}{\pi^2} \sum_{k=1}^{20} \frac{1}{(2k-1)^2} \exp\left[-\frac{(2k-1)^2 D \pi^2}{s^2} t \right] \right\}$$

Dogbone samples with several degrees of moisture were printed with Recreus Filaflex 82A on the x500 printer, since a stiffer filament and a Direct extruder reduce buckling and allow a focus only on under-extrusion by moisture. Printing parameters were selected from a combination of manufacturer suggestions and personal experiences (Table 1). Printing speed and layer height were varied (Table 2). The first affects directly the extruded material amount, and feasible values for good but not too long printings were chosen. The latter influences the total number of layers and, hence, the number of possible failure points in under-extruded flows. Tensile tests according to Standard DIN 53504-S3A evaluate the samples strength. The goal was to verify a possible relationship between the moisture content and the ultimate stress, and investigate the effect of under-extruded processes on printed parts.

The material was initially dried in the oven for three hours at 60 °C and then exposed to room air (23 °C; 40% humidity) for four durations: 0 h (printed right after drying), 24 h, 48 h and 168 h. From experience, seven days of moisture absorption already visibly affected the prints. Thermogravimetric (TGA) measurements were performed on a TGA Q500 V20 to determine the filament moisture content for each exposure time, with heating from room temperature to 130 °C (10 K/min-rate), followed by an isotherm at 130 °C for at least 70 min. Two samples were also analysed prior to drying to check the moisture contents of spool strands at the surface (direct contact with the environment) and in the centre (no direct contact). Tensile tests were performed for 5 samples per exposure time and per variation on a ZwickRoell universal testing machine (force sensor 500 N) at ambient temperature and a displacement rate of 15 mm/min.

3 Results and discussion

3.1 Experimental analysis on feeding system

Under-extrusion was the greatest challenge of the Bowden extruder. Common suggestions to help overcoming it include [16]:

- increasing the printing temperature and/or the flow, which increases the amount of extruded material;
- decreasing the printing speed: the higher the speed, the more difficult the filament is sufficiently extruded in a shorter amount of time;
- using the correct filament diameter;
- ensuring a clean nozzle (associated with non-uniform under-extrusion caused by jamming).

Among those, the printing speed was the most impactful parameter, assuming values below 15 mm/s for reduced under-extrusion. Increase in temperature also helped, but it is limited to avoid material degradation. Unfortunately, for elastic filaments those solutions were not effective enough to rectify under-extrusion. The predominant factor observed with the Bowden feeding system was filament buckling.

For buckling reduction, the filament retraction parameters were gradually lowered (showing improvements for a 1.5-mm retraction distance and a 20-mm/s retraction speed). Retraction points can be reduced by keeping a constant flow. Therefore, printed flat plates as semi-finished part for the mechanical testing specimens led to better outcomes compared to the direct printing of dogbone samples. However, the S5 printer Bowden tube has a length of ca. 1 m in which the filament is compressed towards the hot end, promoting the exceeding of the material's critical buckling load. Moreover, the pushing force delivered by the filament to the viscous melt decreases while the required force to extrude the material through the nozzle increases due to the elastomer melt high viscosity [14], undermining the piston movement by the filament for a proper extrusion.

Figure 2 shows cumulative printings T1 (no contour lines), T2 and T3 (2 outlines), with infill at 0° relative to the tensile direction. It can be noted that T1 is very translucent, and the opacity increases with the printing order. This denotes the extrusion inconsistency for the Bowden system, especially for T2 and T3, revealing the compromised reproducibility in sequence printing. However, it was observed that unloading and reloading the filament into the machine relieved the material compression inside the Bowden tube, leading to better results. This evidences the impact of this extruder when dealing with elastic filaments.

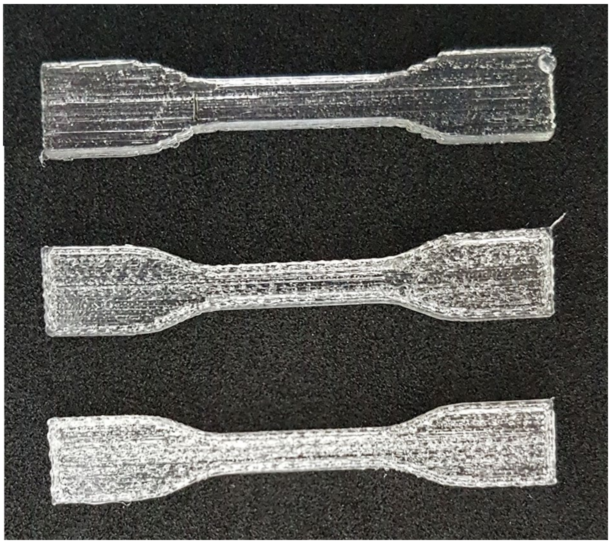

Fig. 2 Volaprint TPU 70A cumulative prints T1, T2 and T3 (from top to bottom) with Bowden extruder

Printings performed with the Direct extruder with similar parameters had reduced under-extrusion, were less prone to buckling and showed a better printing control. However, under-extrusion was noticed to result from filament friction inside the tubes leading to the extruder, as the TPUs exhibited more surface friction than rigid filaments. Therefore, the extruder power needed to pull the filament through the tubes while unrolling the spool, especially due to the material elasticity, indicated to be insufficient to keep the correct flow out of the nozzle. This was particularly observed in printings with the correct flow and, at one point, under-extrusion started and the filament was tensioned in the spool. The unrolling of the spool in advance contributed in avoiding this effort being done by the printer motor itself. Keeping the filament as loose as possible helped reduce friction in the tubes caused beforehand by filament stretching while pulling it towards the hot end. Using a smaller tube length may also provide improvements by reducing the friction area.

Overall, the Direct extruder is more suitable for printing with elastic filaments. Buckling significantly affects the printing control and leads to extrusion failures, and this feeding system assists in mitigating this effect. The use of a Direct Drive, however, is not excluded from under-extrusion, as well as the Bowden Drive does not eliminate the possibility of working with TPUs, although it is more challenging.

3.2 Microscopic analysis of moisture

Regardless of the feeding system, moisture affects the part printing quality. The non-uniformity in the flow was observed after ca. 3 cumulative hours of printing under exposure to room air, with increased opacity on the prints with the transparent filament, crackling sounds, voids on

Table 3 Drying information and printing parameters for Microscopic Analysis

Fig	Drying at 50 °C	Temperature [°C]	100% Infill	Layer height	Speed
3	–	230	45°	0.1 mm	15 mm/s
4a	–	230	0°		
4b	Filament 4 h	230	0°		
4c	Spool 5 h	230	0°		
4d	Spool 5 h	240	0°		
4e	Spool 5 h + Filament 4 h	230	0°		
4f	Spool 5 h + Filament 4 h	240	0°		

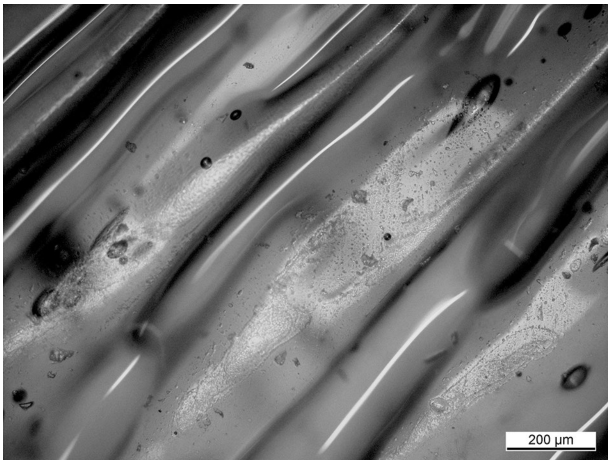

Fig. 3 Top surface of printed sample with low moisture levels

the strands and compromised raster adhesion. This shows that only proper storage between the printings is not enough to preserve the filaments from moisture. Material drying was imperative for the printing quality. The conditions for the analysed samples are summarised in Table 3, where “Filament” stands for drying a piece of filament (ca. 25 g) and “Spool”, for the drying of the whole spool (ca. 500 g).

The Fig. 3 specimen was exposed to air less than 2 h since opening the package, producing a transparent sample, with uniform extrusion and few bubbles trapped inside. This demonstrates the possibility of printing elastic filaments with a Bowden extruder, but also reveals an already present moisture in new spools. In fact, Fig. 3 is in accordance to expectations, as the manufacturer guarantees a maximum of 0.15% humidity [16] (which was not verified here). Two weeks of exposure to air led to a greater moisture absorption by the material, evidenced by non-uniform flow and crackling sounds. Figure 4a proves this by showing several bubbles in the print. Degradation effects are discarded, since the printing temperatures are outside this range. Printing a piece of filament dried for 4 h (Fig. 4b) generated a better extrusion flow and fewer flaws. Prints with a spool dried for 5 h (Fig. 4c) had similar but less intense improvements from Fig. 4b, with impacts on line adhesion in some points. The temperature increase in Fig. 4d reveals the greater extruded material amount compensating the under-extrusion by moisture. Additional

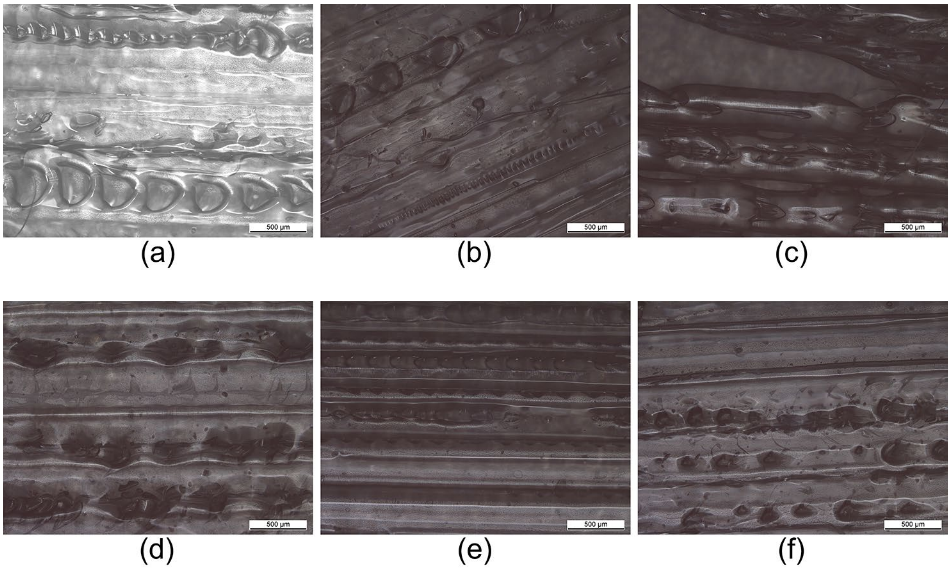

Fig. 4 Brims of printed part with **a** moist filament, **b** 4-h dried filament, **c** 5-h dried spool, **d** 5-h dried spool and 10 °C temperature increment, **e** 4-h dried filament from 5-h dried spool, **f** 4-h dried filament from 5-h dried spool and 10 °C temperature increment

drying led to Fig. 4e with reduced bubbles and lack of adhesion compared to Fig. 4c. Likewise, Fig. 4f with higher temperature decreases the failures further.

The observations illustrate the impact of the drying time related to the dried material amount. Single "Filament" drying showed better improvements than "Spool" drying. This can be explained regarding mass and surface effects. "Filament" material corresponds to only 5% in mass of "Spool", and allows more surface area exposed to the oven air for better moisture removal compared to the compacted filament strands in the spool. Both dryings were beneficial, although insufficient given the remaining presence of bubbles, requiring further studies for optimized drying. Moreover, the flow and the adhesion were improved and flaws were reduced, the more the filament was dried and the higher the printing temperature. Similar results were achieved for the Volaprint filament, which were not here presented.

Hence, this qualitative analysis attests the influence of moisture on the visual quality of TPUs printing performance and the importance of recurrent drying, even for moderate humidity conditions. Moisture induces flaws in the flow, leading to lower quality prints and creating critical cracks points. The drying process duration varies according to the evidence of moisture and cannot be arbitrarily set to the printing community practice of 2–4 h. Additionally, the material amount should be considered, especially if compacted in the spool. Finally, filaments should be preferably protected from the environment in all stages: before, during and after printing.

3.3 Water absorption

Figure 5 shows the water uptake results over the square root of time and Table 4 gives the parameters identified using Fick's Law. The uptake for all materials is very intense during the first 24 h. It also displays that the lower the hardness, the faster the uptake, as both filaments of hardness 70A had the highest diffusion coefficients, although all materials achieve similar maximum water contents.

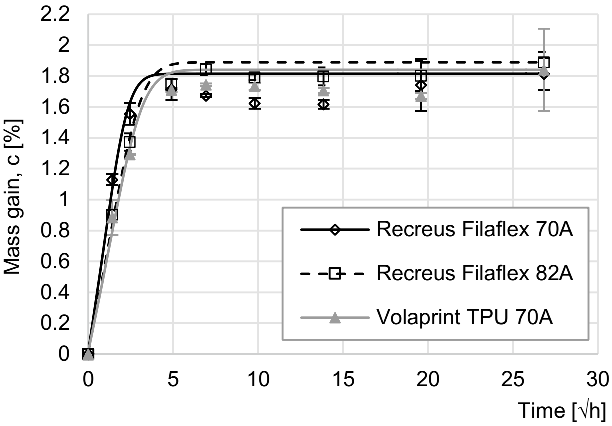

Fig. 5 Measured water absorption (markers) and corresponding fitted curves based on Fick's Law (solid lines)

Table 4 Parameters calculated by Fick's Law

Parameters	Recreus 70A	Recreus 82A	Volaprint TPU 70A
Maximum water content [%]	1.81	1.89	1.84
Diffusion coefficient [10^{-7} cm^2/s]	2.96	1.65	4.15
Correlation	0.9918	0.9970	0.9947
Diameter [mm]	1.75	1.75	2.85

The initial linear section of the curve fittings agrees with the experimental data, indicating Fickian diffusion. The curves show some distancing to the oscillating experimental values after the initial steep increase, which is lower for Recreus 82A. This can be attributed to imprecisions on the weighing due to the substantially small sample diameters, difficulties in drying the rough sample surfaces and the scale sensitivity. An increased measurement frequency during the first hours for larger material amounts could help improve the accuracy. The non-negligible water absorption is noteworthy, reaching almost 2% levels in weight, and its majority occurs during the first hours. In conclusion, the hygroscopicity of the TPUs is confirmed, as already observed in the Microscopic Analysis.

3.4 Effect of moisture on mechanical behaviour

Table 5 presents the filaments moisture contents prior to drying at the surface and in the centre of the spool and after drying with exposure times between 0 and 168 h. The "centre" was protected by outer filament strands and exhibited a lower moisture content, close to the manufacturer's delivery guarantee of 0.15% [16]. The "surface", indeed, presented higher moisture levels. The 0 h condition was similar to "surface" (without drying), indicating the inefficiency of drying and need for a longer duration for better moisture removal. The hygroscopicity is again noticeable, as from exposures 0–24 h the moisture increases by 67%, being less pronounced thereafter. This reinforces the necessity of protecting the material during the first printing hours.

Figure 6 presents the measured engineering stresses (regarding the outer cross-sectional area) as function of strain. The ultimate stresses don't necessarily decrease with increased moisture content as a result of more flaws from bubbles and inconsistent extrusion. In general, the stresses decrease with the moisture level, although not in a regular form. Except for variation B, the 0 h exposure had the highest results. Moreover, samples from 0

Table 5 Moisture content from TGA measurements without (centre; surface) and with prior drying (0–168 h)

Condition	Centre	Surface	0 h	24 h	48 h	168 h
Moisture content [%]	0.18	0.37	0.36	0.60	0.65	0.77

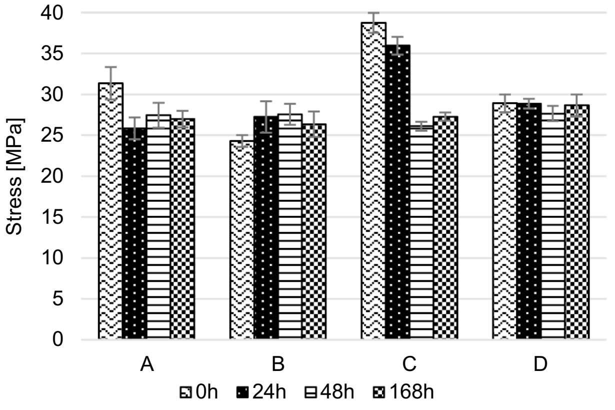

Fig. 6 Ultimate tensile stress for printing variations A-D and exposure times 0–168 h

and 24 h exposures had more irregular results (errors of 19–37% from the highest value within the same exposure) and greater fluctuations for the individual samples. Additionally, 24 h and 48 h exposures had similar moisture contents but dissimilar tensile strengths. An explanation for the lower stress of B (0 h) and the peculiarities of the 0 h, 24 h and 48 h results is the random distribution of flaws from the non-uniform flow caused by the present moisture. The flaws may have concentrated in some areas and induced larger cracks, leading to weaker points for the samples with lower tensile stresses. Besides, calculations were made based on a uniform cross-sectional area, while defective samples have lower effective areas, which would lead to higher stresses. Those effective areas, however, are laborious to obtain.

Exposure times 48 h and 168 h appear uniform for all variants, indicating that the mechanical strength is intensely affected by the respective moisture levels, regardless of the printing parameters. It can be assumed that lower moisture levels have more chances of getting better prints, leading to more result variability. In contrast, filaments with higher moisture contents do not leave much room for less affected extrusion. Furthermore, all exposure times exhibited higher tensile strengths for the 0.2-mm layer height. A likely assumption is the lesser chance of failure points with half the number of printed layers. Since the flow out of the nozzle is automatically increased for a higher layer height, this may compensate the under-extrusion by moisture. Also, the smaller the number of layers, the smaller the interlayer contact area with voids. Nothing can be stated with certainty regarding the influence of printing speed.

All samples registered high elongations at break beyond 1000% of strain and plasticity, unlike an ideal elastomer, withstanding at least 23 MPa. Their behaviour up to 500% strain (Fig. 7) was nearly similar, with stresses of about 8 MPa and, except for the 48 h exposure, decreasing with increasing moisture content. Thus, in lower strain ranges, moisture does not substantially influence the material's behaviour.

Unfortunately, flaws induced by moisture are not easily predicted. TGA measurements are made on a small piece of material, while printings take a large filament length. Moisture absorption is also non-uniform throughout the material. Samples with longer exposure have more time for absorption uniformity, which is a reason for more consistent and regular results. The pre-drying was equally ineffective in uniformly reducing moisture content. Nevertheless, the greatly affected ultimate stress is noticeable for higher strains, even at moisture concentrations less than half the water saturation levels.

4 Conclusions

This paper reports and discusses issues on the AM of elastomeric parts using TPUs. The feeding system has a major impact on buckling and printing control, and using a Direct extruder is recommended for reducing extrusion failures. Nevertheless, for both systems, moisture in the filament is an additional cause of under-extrusion and should be prevented. A qualitative microscopic analysis shows the effect of moisture and drying on sample quality, highlighting the reduction of bubbles for longer drying times and higher printing temperatures. Water uptake tests confirm the filaments hygroscopicity, where most of the uptake occurs in the first 24 h.

Tensile tests verified the moisture influence on the mechanical behaviour, being more prominent for strains above 500%. Longer exposure times of filaments to air provided more uniform moisture absorption and less result variability. Moreover, moisture contents below half of saturation already impact the ultimate stresses. Considering that under-extrusion by moisture occurs similarly in FFF, the results can be generalised to other TPU materials. However,

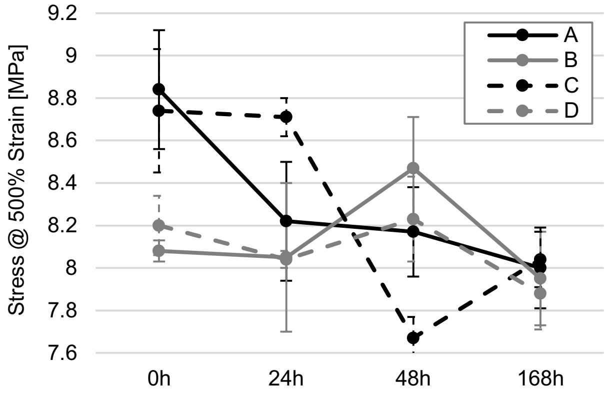

Fig. 7 Tensile stress at 500% for printing variations A–D and exposure times 0–168 h

generalisation to other filaments depends on how the material absorbs moisture.

In conclusion, this work consists of a first step towards further investigations of the effect of moisture on TPUs and drying optimization for better printing control and mechanical behaviour of filaments in such conditions. This work also aimed to gather knowledge on the topic of 3D printing with elastomers and study its feasibility, targeting at the AM of functional parts with solid and stable mechanical properties. Future perspectives include the analysis of long-term properties, the improvement of the 3D printing process and the investigation of definite geometries for specific applications.

Funding Open Access funding enabled and organized by Projekt DEAL.

Declarations

Conflict of interest On behalf of all authors, the corresponding author states that there is no conflict of interest.

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