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3D PRINTING TPU COMPOSITES WITH CONTINUOUS CARBON FIBERS FOR IMPACT ABSORPTION APPLICATIONS

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Abstract: In recent years, Additive Manufacturing (AM) has become a pivotal method in the manufacturing industry, especially in producing high-temperature continuous fiber-reinforced thermoplastics via 3D printing. This innovation holds immense potential for sectors like automotive, aerospace, and biotech, offering structural advantages over traditional materials. To overcome limitations in strength and durability, Composite Filament Co-extrusion (cCFC) techniques integrate carbon fibers into thermoplastic matrices. The current study investigates the mechanical properties and energy absorption of 3D-printed composites using Thermoplastic polyurethane (TPU) and Carbon Fibers (CF) through Anisoprint cCFC. Quasi-static tests validate the improved mechanical response and flexural performance of these composites, highlighting their suitability for impact absorption applications such as automotive panels and protective gear. This research paves the way for the broader integration of TPU composites with continuous carbon fibers in various industries.
Key words: Additive manufacturing, Process Optimization, TPU/CF, Composite Filament Co-extrusion, Mechanical Testing.

1. INTRODUCTION

Industry 4.0 has emerged the additive manufacturing (AM) technologies in the spotlight [1]. AM enables the production of components with high geometric complexity and facilitates the fabrication of objects with a high stiffness-to-weight ratio through advanced topologically optimized design and the integration of composites as construction materials [2]. Until the utilization of AM in composite manufacturing, the development of composite products was an expensive and usually unsustainable process due to the special required equipment. The advancements in AM processes led to the development of the continuous composite fiber co-extrusion (cCFC) method. cCFC technique is categorized in material extrusion AM technologies creating composite components with polymeric or elastomeric matrix and reinforced with fibers made of carbon, glass, aramid, etc. cCFC has a wide range of applications in high-tech industries, such as automotive, aerospace and bioengineering [3].
cCFC technique is a subcategory of CFC 3D printing, indicating the continuous fibers of reinforcement material inside the object's volume. Generally, CFC methods can be categorized in two different types, depending on the way that the fiber is impregnated in the matrix material. In the first type, the fiber is already impregnated with the plastic matrix resulting in a ready-to-print composite material. This type limited the research around CFC in commercially produced filaments. On the other hand, in the second type of CFC, the impregnation of the fiber inside the matrix takes place in a heated printhead and is simultaneously extruded through a single nozzle in order to form the desired composite structure. Currently, there is a plethora of scientific studies that reveal a robust mechanical behaviour of 3D prints combining polymer matrix materials, such as polyamide (PA), polyethylene terephthalate glycol (PETG), acrylonitrile butadiene styrene (ABS) or polylactic acid (PLA) with continuous carbon fibers (CF) [4,5,6]. In detail, Becker et al. [4] found from bending tests of CF/PA6 composite, 38.3GPa flexural modulus and 547MPa of flexural strength. Savandaiah et al. [5], conducted bending tests for both PLA and PA composites and observed an increase in both flexural strength and modulus at 497MPa and 50GPa, respectively, for PLA/CF and 488MPa and 45 GPa for PA/CF. Gao et al. [6] concluded in an increase in flexural strength in tensile tests, for CF-reinforced PA at 149MPa. Moreover, Fedulov et al. [7] provide a comprehensive case study employing the cCFC AM technique in a real-life application for an automotive pedal, increasing significantly the component's overall stiffness-to-weight ratio. Reviewing the existing literature, it is obvious that the majority of existing research around cCFC 3D prints focuses on the problem-solving of the highest stiffness-to-weight ratio, revealing a research gap concerning the employment of elastomeric material as matrixes in order to enhance energy absorption capabilities, electrical conductivity, etc. Indicatively, Salo et al. [8] employed cCFC

3D printing in order to produce stretchable electronics, constructed of thermoplastic polyurethane (TPU) and CF reinforcement. Furthermore, Cho et al. [9] developed liquid crystal elastomer-based woven composites to improve the structure's low-velocity energy absorption revealing sufficient amounts of absorbed mechanical energy during loading.
In this context, the current study aims to fulfil the existing research gap around the mechanical behaviour and energy absorption capacity of 3D printed TPU/CF composites fabricated with the cCFC AM technique. The novelty of this research is located in the conducted experimental study of TPU/CF composite structures following international testing standards. The objective of the current study was to explore the potential of cCFC 3D printing for the fabrication of components with demanding energy absorption behaviour. Therefore, in the context of this research, the TPU elastomer was selected as a matrix material and CF as reinforcement material for the developed composite. Figure 1 graphically presents the flowchart of the current study.

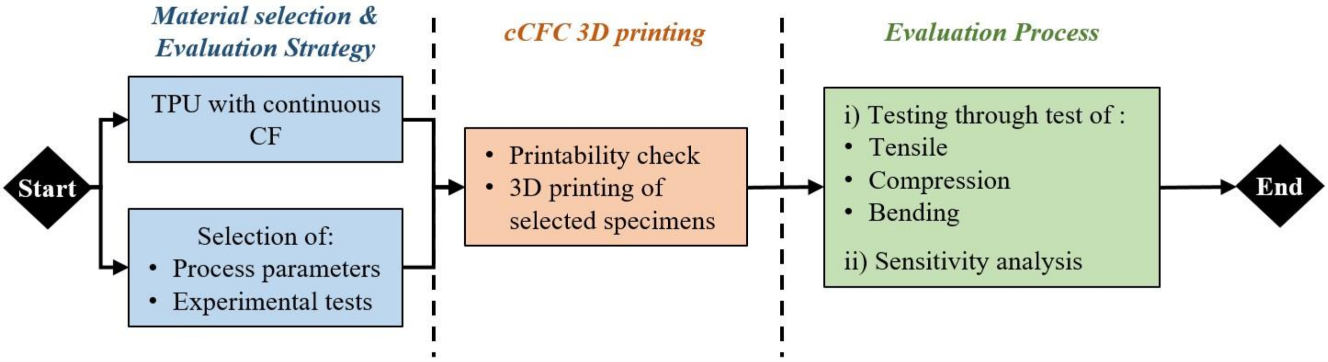

Fig. 1. Flowchart of the current study

In detail, during this research, a sensitivity analysis was performed regarding the influence of the process-related parameters on the printability and mechanical behaviour of TPU/CF composites. Test specimens of TPU/CF composite were additively manufactured with sets of different process-related parameters mainly focusing on the process temperature and speed. Then, a series of mechanical experiments were conducted, namely, tensile, compression and bending tests, in order to extract the main mechanical properties and evaluate the 3D prints' energy absorption characteristics. Finally, the acquired experimental data were compared with the properties of origin materials, TPU and CF, and the most crucial conclusions were stated. Section 2 presents the properties of the construction materials and the corresponding 3D printing parameters along with the employed methodologies for 3D printing and mechanical testing. Moreover, Section 3 lists and analyses the results of the current study and Section 4 summarized its most crucial conclusions.

2. MATERIALS AND METHODS

2.1. Material Specifications

TPU is a commonly used elastomeric with remarkable elasticity and crashworthiness. Thus, it is implemented in numerous energy absorption applications, such as protective equipment, automotive bumpers, etc. On the other hand, CF was chosen as reinforcement in order to enhance the mechanical response of pure TPU. CF material is consisted of primarily tightly bonded carbon atoms in a crystal alignment transfusing exceptional stiffness, strength-to-weight ratio, and corrosion resistance. For this study, PolyFlex^{TM} TPU90 (Polymaker, Shanghai, China) was used along with the Anisoprint CCF (Composite Carbon Fiber). Table 1 lists the main properties of the selected materials based on the provided datasheet from the manufacturers. It is worth noting that during the cCFC 3D printing process the CF material remains in the same phase therefore its diameter should be much thinner than the diameter of the printhead's nozzle.

Table 1. Main properties of the selected matrix and reinforcement material

Properties	TPU PolyFlexTM TPU90	Anisoprint CCF
Density	1.12 g/cm^3	19.65 g/cm^3
Feedstock form	Regular filament	Thin filament
Diameter	1.75 ± 0.03mm	0.36 ± 0.02 mm
Printing Temperature	210 - 230°C	Not applicable

2.2. 3D Printing Process

The 3D printing process of pure TPU via material extrusion technology is widely characterized as a difficult procedure due to its hyper-elastic behaviour that provokes feed and retraction issues. This difficulty is further increased in cCFC 3D printing due to the existence of reinforcement material. Therefore, the first step was to identify the optimal set of 3D printing parameters for TPU which are listed in Table 2. It is important to note that in order to overcome the feed issue, the room temperature was set below a certain temperature providing sufficient stiffness to the TPU material to successfully pass through the feeder. The second step was the employment of cCFC 3D printing process. For this procedure, the Anisoprint Composer A3 3D printer (Anisoprint, Monnerich, Luxenbourgh) was utilized which is equipped with two printheads, one for common plastic extrusion and one for composite extrusion. The majority of process-related parameters were the same with TPU 3D printing due to the fact that the composite TPU/CF showed increased printability without any visible defects. However, there were certain parameters that had to be modified in order to extract high-quality results. These parameters concerned mainly the retraction changing corresponding to these parameters until the fabrication of a flawless 3D printed object. Moreover, the printing speed of TPU/CF composite was a tricky puzzle, as a specific range of speed values ensure acceptable 3D printing results. Hence, in the context of this research, it was decided to perform a sensitive analysis of the influence of printing on the quality of composite 3D prints. Through this analysis, three distinct values of printing were selected, namely 10mm/s, 20mm/s and 30mm/s, and test specimens were fabricated in order to be tested.

Table 2. Main properties of the selected matrix and reinforcement material

Parameters	TPU PolyFlexTM TPU90	cCFC TPU/CF
Room temperature	<18°C	<18°C
Temperature of building platform	50°C	50°C
Printing Temperature	220°C	220°C
Retraction speed	40mm/s	10mm/s
Retraction length	3mm	5mm
Layer height	0.3mm	0.36mm
Nozzle diameter	0.4mm	0.4mm for perimeter / 0.8mm for infill
Printing speed	45mm/s	10 - 30mm/s

Furthermore, the slicing process was performed in Aura™ slicing software. The 3D printing strategy was designed in order to produce specimens with pure TPU perimeter and enhanced infill consisting of 100% composite TPU/CF. In Figure 2, the aforementioned slicing strategy for a tensile specimen is illustrated (left side) along with indicative images of the employed cCFC 3D printer and a tensile 3D printed specimen (right side).

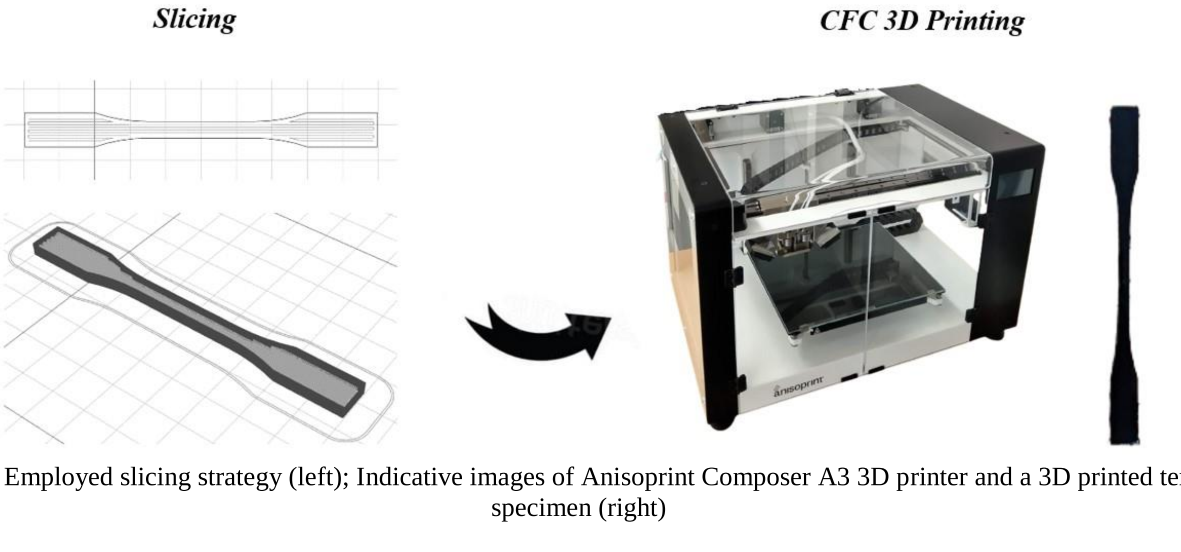

Fig. 2. Employed slicing strategy (left); Indicative images of Anisoprint Composer A3 3D printer and a 3D printed tensile specimen (right)

2.3. Mechanical Testing

After the cCFC 3D printing process, the next step is mechanical testing of the TPU/CF composite specimens. In order to obtain a complete image of the developed composite's mechanical response, a series of mechanical testing experiments were conducted including tensile, compression and bending tests, through which the basic mechanical properties were evaluated. All these experiments were performed at room temperature utilizing the universal testing machine Testometric M500-50AT (Testometric Company, Rochdale, UK) was employed and integrated with a 50kN load cell. Moreover, in order to ensure reliability and high-quality results all tests were conducted at least five times. In addition, the strain rate for all mechanical tests was set at 5mm/min [10]. All developed test specimens were designed based on international standards, and more specifically for the tensile compression and bending tests the ISOs 527, 604 and 178 were employed, respectively [11, 12, 13]. Figure 3 presents the developed test specimens along with the most crucial dimensions. It is essential to state that the spam length of the bending specimen was set at 50mm.

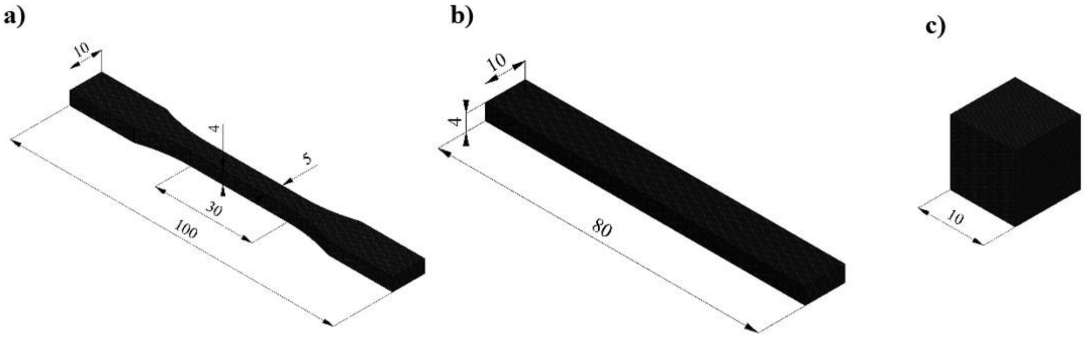

Fig. 3. 3D CAD models for: a) Tensile, b) Bending and c) Compression tests specimens

One of the most significant aspects of TPU/CF composite that the current addresses, is the material energy absorption performance. Therefore, it is essential to observe and measure the absorbed energy of the developed composite in terms of energy absorption per volume, energy absorption efficiency and energy absorption per mass, also known as Specific Energy Absorption (SEA). According to the existing international standards, the absorbed mechanical energy of a structure can be derived from the evaluation of the surface area below the experimental curve of compressive stress to strain diagram until 60% of strain, i.e. before the densification phase begins. Furthermore, the corresponding mathematical formulas for energy absorption per volume (W), SEA and energy absorption efficiency (W_e) are listed in Eq. 1, Eq. 2 and Eq. 3, respectively. It is worth mentioning that the energy absorption efficiency is a valuable index of the energy absorption performance of a structure due to the fact that compares the structure performance with the performance of an ideal absorber [14].

$$W = \int_0^{\varepsilon_0} \sigma(\varepsilon) d\varepsilon \tag{1}$$

$$\text{SEA}= \frac{1}{\rho_m} \int_0^{\varepsilon_0} \sigma(\varepsilon) d\varepsilon \tag{2}$$

$$W_e = \frac{W}{\sigma_{peak} \times \varepsilon_0} 100 \tag{3}$$

Where ε is the strain, $\sigma(\varepsilon)$ is the stress for a given strain (ε), σ_{peak} is the maximum observed stress, ε_o is the maximum strain (60%) and ρ_m is the density of the 3D printed composite structure. The integrals of the above-mentioned equations were numerically calculated utilizing the acquired experimental and the approximation mathematical formula of the trapezoidal rule. Therefore, integrating the trapezoidal in Eq.1 and Eq.2 the following equations are derived. Then, the experimental data for compression tests were utilized in order to evaluate the energy absorption indexes employing the approximation Eq.4 and Eq.5.

$$\int_{x_0}^{x_n} f(x)dx = \frac{\Delta x}{2}[f(x_0) + 2f(x_1) + \ldots + f(x_N)] \begin{cases} W = \dfrac{1}{2}[\sigma(0) + 2\sigma(\varepsilon_1) + \ldots + \sigma(\varepsilon_{60\%})]\Delta\varepsilon & (4) \\ SEA = \dfrac{1}{2\rho_m}[\sigma(0) + 2\sigma(\varepsilon_1) + \ldots + \sigma(\varepsilon_{60\%})]\Delta\varepsilon & (5) \end{cases}$$

3. RESULTS AND DISCUSSION

Having set the methodology of the current study, the next step was the conduct of mechanical experiments and the presentation of the results. Due to the fact that the different printing speeds extracted sufficient quality for the TPU/CF composite, it was decided to perform a sensitivity analysis in order to reveal the optimum printing speed for this cCFC 3D printing process. Three different printing strategies were selected with three different speeds within the speed range that provided printable results. Hence, printing strategies P1 (cyan) was set at 10mm/s, P2 (black) at 20mm/s and P3 (red) at 30mm/s. Figure 4 shows the experimental stress-strain and load-deflection diagrams of tensile tests and bending tests, respectively, for all employed 3D printing strategies. Through Figure 4a, it is obvious that the TPU/CF composite follows a typical stress-strain curve of rigid polymer without the existence of necking phenomena, brittle fracture and extensive plateau. Moreover, for these experimental data, it is revealed that the P2 3D printing strategy showed the best tensile performance in terms of strength and stiffness, followed by 3D printing strategies P1 and P3. The same trend is also observed during the bending tests. However, during the bending experiment, all specimens showed extensive deflection highlighting the remarkable plasticity of the developed TPU/CF composite.

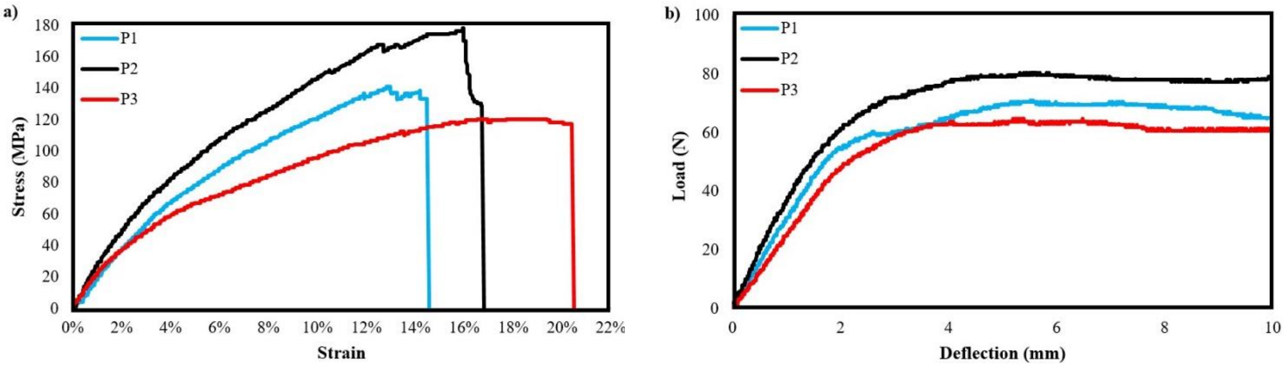

Fig. 4. a) Experimental stress-strain diagram of tensile tests and b) Experimental load-deflection diagram of bending tests for the three different 3D printing strategies

Table 3. Tensile and flexural properties of the employed 3D printing strategies compared with pure TPU and pure CF

Properties	P1	P2	P3	TPU	CF
Density (g/cm^3)		1.22 ± 0.2		1.12	1.75
Elastic Modulus (MPa)	1050 ± 30	1130 ± 30	930 ± 20	8 ± 1.5	135,000
Yield Strength (MPa)	113 ± 5	142 ± 7	97 ± 5	9.6 ± 1.5	1704
Ultimate Tensile Strength (MPa)	141 ± 7	178 ± 8	122 ± 7	12.2 ± 1	2130
Elongation at Break (%)	14.6 ± 1.5	16.8 ± 1.5	20.5 ± 2	709 ± 30	1.75
Max. Bending Force (N)	71 ± 3	83 ± 3	64 ± 3	6.4 ± 0.5	-
Deflection at Max. Force (mm)	5.5 ± 0.5	5.6 ± 0.5	5.5 ± 0.5	5.4 ± 0.5	-
Flexural Modulus (MPa)	625 ± 15	728 ± 15	569 ± 15	57 ± 3	-
Flexural Strength (MPa)	33 ± 1.5	39 ±1.5	30 ± 1.5	3 ± 0.5	-

Furthermore, Table 3 lists the tensile and flexural properties of the employed 3D printing strategies, as they were derived from the above-presented diagrams. In addition, in Table 3 the corresponding values for pure TPU and pure CF are also listed in order to provide an order of magnitude for the research. Based on the density data, it can be concluded that the produced TPU/CF composite contains almost 15% CF and 85% TPU. Moreover, the TPU/CF composite revealed remarkable strength (>100MPa) with increased elasticity and plasticity surpassing the 15% strain until the specimens' failure. The percentage differences in 3D printing strategies were observed around 10%-15% for both tensile and bending experiments. These differences can be justified by poor adhesion of consecutive layers at high printing speeds (≥30mm/s) or pore creation due to the thermal strain of the TPU matrix for low printing speeds (≤10mm/s). These results indicate that the TPU/CF composite is a very promising material which combines the extensive elasticity of TPU and the outstanding strength of CF.
Figure 5 presents the stress-strain diagram for compression testing and the evaluated indicators of energy absorption, in the form of a bar chart, as they were derived from the analyses of the stress-strain diagram employing the mathematical formulas of Eq. 3, Eq.4 and Eq.5. Verifying the aforementioned results, the mechanical behavior of three employed 3D printing strategies in the compression experiment follows the same trend with the other test, i.e. P2 withstands the higher load followed by P1 and P3. For Figure 5a, it is worth mentioning that despite the fact that the TPU/CF composite revealed increased loads, the compassion curves show a typical elastomeric pattern with exponentially increasing withstanding loads. This observation led to the conclusion that the developed composite has increased plasticity under compressive loads and up to a certain experience extended stress and strain recovery indicating the enhanced energy absorption behavior of this composite.

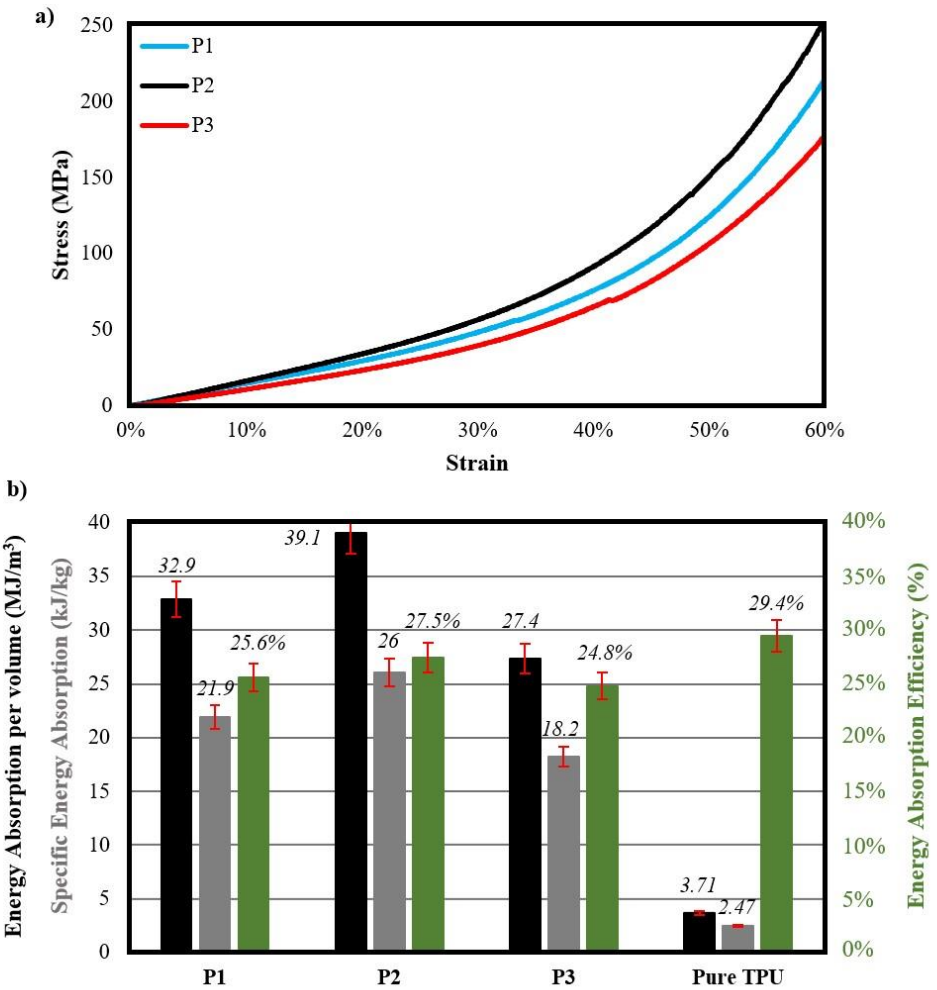

Fig. 5. a) Experimental stress-strain diagram of compression tests and b) Evaluation of energy absorption indicators for all examined specimens

Indeed, as it is shown in Figure 5b, the impermanent of CF in the TPU matrix leads to an almost tenfold energy absorption per volume and per mass compared to pure TPU energy absorption. Moreover, as it was expected the P2 3D printing strategy showed to have also the optimal energy absorption performance. Finally, the energy absorption efficiency of the produced composite was slightly decreased in an order of magnitude of 5% to 10% compared to the pure TPU material. This reduction in plasticity seems to occur due to the embedment of a significantly stiff material, such as the CF, into the TPU matrix. Overall, the TPU/CF composite revealed outstanding energy absorption performance and combining it with the remarkable tensile and flexural behavior, it could revolutionize the field of impact and energy absorption-demanding applications.

4. CONCLUSIONS

In the current paper, the cCFC 3D printing technique was studied and the TPU/CF composites were developed and examined under a series of mechanical tests to present a comprehensive mechanical performance analysis. In detail, TPU/CF composites were additively manufactured and it was observed that the quality of the 3D prints is heavily influenced by the applied printing speed. Therefore, three different 3D printing strategies (P1, P2 and P3) were employed with printing speed 10mm/s, 20mm/s and 30mm/s, respectively. Then, the produced test specimens underwent tensile, bending and compression experimental testing to extract the basic mechanical properties. The 3D printing strategy P2 was revealed as the optimum strategy for cCFC 3D printing of TPU/CF composite. Moreover, despite the fact that the produced composite consisted of only 15% CF, their mechanical strength was remarkable reaching values close to 150MPa. In addition, the elasticity and plasticity of the composites were significantly increased, as the elongation at break in tensile testing surpassed the 15% strain and the strain recovery during compression testing possessed the main role. Furthermore, their energy absorption was ten times higher than the pure TPU, reaching up to 40MJ/m^3 with only a small drop in energy absorption efficiency which ranged between 23-28%. To conclude, the outcomes of this study are expected to pave the way for further testing and eventually the implementation of TPU/CF composite in impact and crashworthiness applications due to their mechanical and energy absorption behaviour.

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