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Influence of fused deposition modeling process parameters on the transformation of 4D printed morphing structures

Lukas Kačergis, Rytis Mitkus  and Michael Sinapius 

Institut for Adaptronic and Function integration, TU Braunschweig, Braunschweig D-38106, Germany

E-mail: r.mitkus@tu-braunschweig.de

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Abstract

4D printing combines additive layer manufacturing processes with smart materials to create structures that are able to change shape or properties over time under the influence of environmental stimuli. The article presents 3D printed multi-material shape-variable structures imitating a hinge. Fused deposition modelling was used because it provides the ability to pre-program structures during the printing process by varying printing parameters. The structures are printed with PLA and TPU and remain flat after printing until they are exposed to a stimulus - heat. The main objective of this article is to present the possibilities of the aforementioned pre-programming step which can be adapted by varying the printing process and design parameters of the printed part. Experimental results are presented investigating the influence of printing speed, temperature of the build plate and number of active layers in the structure. Furthermore, the repeatability of deformations after a small number of cycles is investigated. The obtained results prove that the deformation of the structures can be controlled by printing parameters and a variety of bending degrees can be obtained by manipulating them. Hot water is used as a stimulus in the study to activate the structures but it is believed that other direct and indirect heating sources are also applicable. The research could help predict the behaviour of deformation of shape-morphing structures by selecting certain printing and design parameter values.

Keywords: 4D printing, FDM, PLA TPU, morphing, shape morph

(Some figures may appear in colour only in the online journal)

1. Introduction

A few years ago, 4D printing was introduced as a technology promising improvements to existing benefits of 3D printing. Current 3D printing technology mostly provides static printing, while 4D printing aims to create structures that are able to transform or change their properties over time when exposed to environmental stimuli. Hence, the fourth dimension here is time. The status of 4D printed structures can be achieved by using different smart materials and combining them with

appropriate design options for specific applications. This technology could also potentially lead to improved material savings and enable a number of new applications in different fields. Last few years have seen an increase in publications discussing 4D printing, and while these reports have analysed a lot of challenges, there are still many to solve for this technology to move forward.

Most of the conventional ALM technologies are suitable for use in 4D printing processes, but not all can be used equally well. Initial attempts to print shape-changing structures were made with PolyJet type 3D printers. Early work on 4D printing have been published by Tibbitts *et al* [1] and Raviv *et al* [2], where they analysed structures made of the combination of rigid plastic and hydrophilic UV curable polymer. A simple bi-layer structure was printed, which bent



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when dipped in hot water. Bending was driven by the hydrophilic polymer as it expanded while absorbing water. When the structures were removed from water, structures returned to their original shape. The transformation itself took a long time, and, besides that, mechanical degradation of hydrophilic polymer was observed over time.

In the work of Ge *et al* [3], 4D printed structures were realized by 3D printing Shape Memory Polymer (SMP) fibres within the matrix of an elastomer. Initially flat, after manual thermomechanical programming step, these structures change their shape. Direction and volume fraction of polymer fibres in the elastomeric matrix were used to characterize their shape change. Later this design was improved by Wu *et al* [4] so that the printed structures were able to transform into different shapes with only one thermomechanical programming step. This was achieved by introducing a second type of polymer fibres with different glass transition temperature T_g .

Mao *et al* [5] used PolyJet technology and combined its two standard materials with different proportions to investigate sequential self-folding of 4D printed structures. Mao *et al* [6] further investigated the possibilities of PolyJet technology and successfully printed the structure with reversible shape-changing. The geometry itself consisted of shape memory polymer and elastomer, where the inside of the structure was filled with hydrogel. Although it no longer needed thermomechanical programming step, the entire transformation process was slow due to the nature of hydrogel. It is reported that the entire cycle lasts 10–20 h. While hydrogels have very slow response rates (hours to days) and low modulus [2, 6], SMPs on the other hand have much faster response rates (seconds to minutes) and higher modulus, making them more suitable for various applications [7, 8].

Huge advantage that PolyJet process allows is material combination (up to 3 materials can be combined together to form a digital material) and deposition on the voxel level, where each voxel can have different material combination from adjacent voxels. Ge *et al* [7] addressed the problem of the lack of custom materials with tailorable properties when using Stratasys PolyJet printers. Poor long-term material properties combined with lack of custom materials counter the advantages the technology provides. FDM-type 3D printers, on the other hand, extrude material through a nozzle, where this extrusion process is controlled and perfectly suits special processing required for shape memory effects to appear in polymers [9]. Machines are cheap and easy modifiable, which allows to deposit pastes or inks for future updates. Wide choice of materials is available for FDM and materials are relatively cheap compared with other AM technologies. Moreover, SME exists in a large group of thermoplastics [10, 11] and various common types of filament can be used to print shape-changing structures. Polymers used in FDM suit for the end-use applications better than photocurable polymers, that are used, for example in PolyJet.

Both single [8–10] and multi-material 4D structures [12] were successfully printed using FDM technology. Manen *et al* [8] conducted an extensive research on influence of various FDM printing parameters on structures made of only PLA material. PLA is extruded and fixed on the build plate or

previous layer, cools down and the internal stresses generated inside the material are locked. These stresses can be released while heating the material above its T_g . Mannen *et al* [8] found that lower extrusion temperature together with lower layer height (0.05 mm) produces the highest strain in the structures, while printing speed and flow of the material has very small influence. Contradictory, An *et al* [10] reported that printing speed of PLA has high influence and the structures printed with higher speeds bend more.

The reviewed articles make it clear that the current state of 4D printing still requires answers to a large number of questions. Various printing parameters might have different influences that are dependent from other printing parameters at the same time and complex influence matrix can be constructed and must be understood. Challenges such as reversible deformation and requirement of programming step are yet not fully analysed and vary from structure to structure. The progress and achievements in 4D printing can be found in a series of reviews [13–15].

In our study, 4D printed structures, made of PLA and TPU are investigated that are printed initially flat with FDM-type 3D printer, bend into U-shape when environmental temperature higher than T_g (in our case higher than 60 °C) is applied. While in U-shape (above T_g), structures can be manually deformed or twisted into any arbitrary shape and can be cooled down to retain that new shape. Repeated heating above T_g will cause the structure to deform back to U-shape. Printing parameters influence on the U-shape (bending angle) of the structure is experimentally investigated as well as the influence of number of active layers.

2. Approach

Printing parameters, influencing the shape-changing of a hinge-type structure are investigated. The structure is made from a combination of two common FDM materials: PLA and TPU. PLA is used in our study for active layers that are programmed (pre-stressed) during printing as the temporary shape is formed by heating the filament above its melting temperature and extruding along the set path. According to van Manen *et al* [9] heating and extruding the filament stretches and aligns the polymer chains along the directions of this path. Stresses generated by this stretching are then stored in the material as it is constrained by depositing it on the build plate or the previous layer. Finally, the residual stresses are fixed when the structure cools down. When the structure is removed from the printer and heated over its glass transition temperature, active material (in this case PLA) shortens along the direction of printing and slightly expands in other two directions. The second material - TPU is an elastomer that is flexible in the room temperature and it is used for the passive layers of the structure. TPU in room temperature is above its T_g and it is assumed that it cannot contract, it can only bend and slightly elongate. When PLA and TPU are combined layerwise into a single structure, the shortening of PLA layers is converted into an out-of-plane bending. This is because the layers of TPU are resistant to the shortening of PLA. TPU

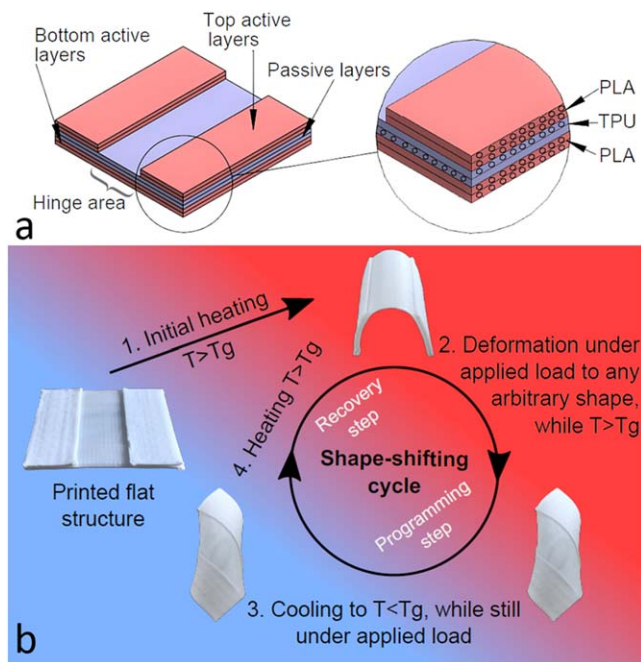


Figure 1. Shape-shifting ‘hinge’ structure: (a) Design of a rectangular multi-ply structure, printed using PLA (active layers) and TPU (passive layers). Printing direction for each layer is marked by additional lines and circles. (b) Shape-shifting cycle of 4D printed structures. Red and blue colours indicate hot (above T_g) and cold environment.

also has a lower Young’s modulus, hence the layers of TPU increase the flexural stiffness of the structure only slightly. Moreover, our early experiments with only single layer PLA structure (without TPU) showed, that usage of only single layer PLA without any additional layers or other materials produces highly varying, unpredictable bending-twisting motion which is not desirable. This problem arises on single layer PLA structure.

When printing 4D structures with single-stimulus responsive material, it is important to produce layers with different directional strains [9]. Layers with unidirectional filling patterns exhibit anisotropic deformation behaviour in contrast to layers where material is deposited in multiple directions, resulting in greater isotropic behaviour. The combination of these layers leads to an out of plane bending, twisting or curling, depending on the direction of layers with unidirectional filling pattern. For this reason all PLA layers in our study were always printed in same direction so that only bending would occur. The speed of printing of PLA layers was varying throughout the study and is described in each experiment.

The structure analysed in our study is hinge-type basic element, which can be used in various, more complex shape-changing designs. Figure 1(a) depicts the design of the multi-material shape-changing structure. The basic structure itself consists of six layers of which four are continuous and two are split. All sides of the structure are of equal length of 30 mm. The shorter sides of the top layers are 10 mm wide. The total height of the structure is 1.2 mm, with each layer height set to 0.2 mm. As shown in figure 1(a), red layers are

printed with PLA and blue layers are printed with TPU. The intended purpose for the two separated top active layers are to control the width of the hinge section and to compensate for bending on two edges, generated by the bottom active layers. Only this print sequence was tested and therefore all structures affected by the stimulus bent downwards due to the shortening of PLA layers. An upwards bend can be achieved by reversing the order of material deposition, but first experiments showed lower deformation and this printing sequence was not investigated further.

Figure 1(b) illustrates the shape-changing cycle for the analysed hinge-type structures. Due to the stresses that build up in the active layers during the rapid deposition of the molten filament, which then cools and solidifies while still stretched [9] these structures are able to transform to the bent shape that can be considered their permanent shape. Whenever the structure is manually programmed to a different shape (figure 1(b), steps 2 and 3) and then subjected to a recovery step (step 4), it deforms back to its permanent shape (U-shape). The main drawback of these structures is that they cannot regain their flat form without straightening them manually. However, by taking a programming step they can be deformed into any shape (both shapes at the bottom of the figure 1(b) represent random deformation) and regain their U-shape after heating them above the glass transition temperature of PLA (step 4).

3. Methods

3.1. Materials and specimens

The structures were made using inexpensive and easily accessible PLA and TPU filaments (Das Filament, Germany). Geometry of the structures was kept the same throughout the whole study, except the last experiment (changes that were made are described further). All structures were composed of six layers, of which, counting from the bottom, first two were made from PLA, third and fourth - TPU, and the last two divided layers - PLA again (layer distribution is shown in figure 1(a)). Each layer had a standard height of 0.2 mm. All structures were printed using rectilinear filling pattern. Infill amount was set to 100% to create layers with no gaps inside. The printer was equipped with a standard nozzle, which has the diameter of 0.4 mm. Extrusion width was set to 0.45 mm for all printer moves. While several parameters were varied during the investigation, most of them remained constant throughout this study: printing temperature for PLA and TPU was 215 °C and 235 °C respectively, printing speed for TPU was always 30 mm s⁻¹.

A single extruder FDM type 3D printer (Anycubic Prusa i3) was used in our study. The extruder itself required small modification in order to print flexible TPU. The filament was prone to buckling during printing and could not be successfully extruded at 30 mm s⁻¹. To solve this challenge, a special adapter was printed and attached to minimize the gap between the drive gear and the filament feed tube.

FDM was selected to produce 4D printed structures because it is a cost-effective, accessible technology that can be configured for multi-material printing. The ability to easily modify these machines can be adapted for further investigation of 4D printing technology, and the results obtained in this investigation can be transferred to more sophisticated designs that could include multiple ALM processes simultaneously.

Although methods are described in several reports [1, 10, 16], there is currently no openly accessible software to design shape-changing structures. Furthermore, the use of two different materials requires material-specific printing parameters for each material. Required parameters for each layer such as printing speed, nozzle temperature, layer orientation, etc were set using the slicing software. In order to print the structures with two different materials on a single extruder printer, the printing process must be interrupted and material must be changed. This was achieved by inserting several command lines into the control code of the printer to initiate pauses during printing and change the material as required.

3.2. Testing and measuring procedure

Most of existing 4D structures respond to actuation either by direct [7, 10, 17–19] or indirect [20, 21] heating. In all experiments done, the transformation of our structures was achieved by placing them in hot water. Hot water was chosen as an activation medium, because it is fast, easy and exact method to apply uniform heat on the structures. The temperature was set to 85 °C and kept constant. The temperature was selected to be around 20 °C higher than the glass transition temperature of PLA. All structures used in the experiments were kept in water until they were no longer exhibiting visual signs of deformation, but no longer than 2 min.

It is well known that polymers tend to absorb water. As literature suggests [22–25] PLA absorption of water is dependent on the temperature of the water and the exposure time to water. However, the weight gain of PLA up to around 2% in 24 h seems to have no significant effect on behaviour of our structures because the longest period our specimens under investigation were in the water is 2 min. Anyway, a few structures were heated in the oven to investigate if swelling of the polymer is happening and if it has significant influence on the deformation. The specimens from the same batch bended to the same angle in both hot water and the oven. It was assumed that water absorption can be neglected and no further investigations on water absorption were done. However, if the structures would be planned to be used in water for prolonged periods of time, further experiments with water absorption and degradation are suggested.

In order to statistically secure the evaluation, a batch of seven structures was printed for each tested value of investigated parameters. To quantitatively evaluate the deformations, an optical 3D digitizer ATOS Triple Scan (GOM, Braunschweig, Germany) was used to measure the geometry of morphing structures after deformation. 3D scanning was chosen because of the simplicity and speed. The structures were placed on the surface prepared for scanning and scanned

from several different angles to capture the entire surface shape and create a detailed ‘point cloud’. The obtained ‘point cloud’ data was merged into one three-dimensional model. GOM Inspect 2019 software (GOM, Braunschweig, Germany) was used for post-processing and evaluation of the acquired 3D models. Cylinders have been added on scanned models in the software as it is shown in figure 2 in order to measure the radius of inner curvature for strain calculation.

As a main measurant the strain on the outer surface (strain surface) of the specimens was chosen as it is illustrated in figure 3. The strain on the outer surface of the specimens were calculated with the following equation:

$$\varepsilon = y/R,$$

where y - the distance from neutral axis to the outside of the structure (strain surface), R - the radius of neutral axis of the structure.

It must be mentioned that the radius measured in the 3D scanning software is the inner radius of the bended structure and for strain calculation the radius of the neutral axis must be used. Since our structures are made of two different materials with different Young’s modules, the neutral axis is shifted towards material with higher Young’s modulus, in our case - PLA. Moreover, the position of neutral axis is also dependent on the number of layers of each material. To calculate the exact position of the neutral axis, equivalent area method was applied. A sketch showing the principle of equivalent area method is shown in figure 4. It is assumed that all the structure is made of single material. The height of the layers cannot be changed. While keeping the height of the layer unchanged, the area of PLA layer which would have the same stiffness as full specimen length made of TPU layer was calculated (see figure 4). The length of respective PLA layer was calculated with the following equation:

$$L_{\text{Equivalent}} = \frac{E_{\text{TPU}}}{E_{\text{PLA}}} * L_{\text{specimen}},$$

where $L_{\text{Equivalent}}$ - length of PLA layer, which under same layer height would represent the same stiffness as full TPU layer (see figure 4(b)), E - Young’s modulus of respective material, L_{specimen} - length of the specimen being investigated. We took into calculations hinge length that is 10 mm in our case as a length of the specimen.

The exact position of the neutral axis from the inner, measured radius is calculated with the following formula:

$$x = \frac{A_{\text{TPU}}y_{\text{TPU}} + A_{\text{PLA}}y_{\text{PLA}}}{A_{\text{TPU}} + A_{\text{PLA}}},$$

where A_{TPU} - area of TPU layers (height of the layer multiplied by $L_{\text{Equivalent}}$), A_{PLA} - area of PLA layers, y_{TPU} and y_{PLA} - the distances from the inner radius to the centroids of respective materials respectively (in simple 2 PLA and 2 TPU layer structure, with single layer thickness of 0,2 mm, y_{TPU} and y_{PLA} values are 0,6 mm and 0,2 mm respectively).

The distance y from neutral axis to strain surface was found simply subtracting x from the thickness of the specimen at the hinge before deformation.

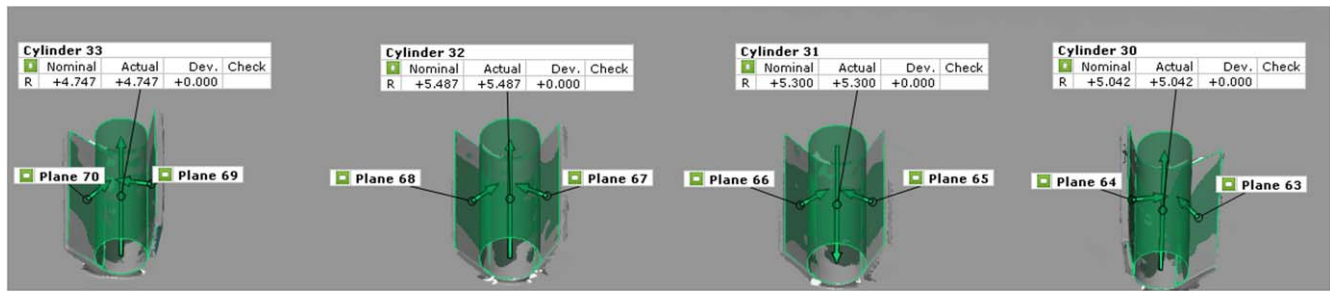


Figure 2. 3D model of shape-shifting structures with constructed planes and measured angles.

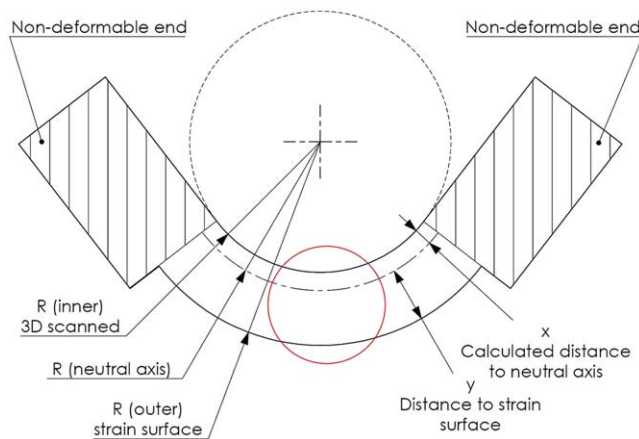


Figure 3. Schematic representation of the specimens. Measured inner radius and calculated strain surface are shown. Circled area is zoomed in figure 4.

4. Experimental tests and results

4.1. Influence of print speed

The print speed for the active layers was the first parameter to be investigated. Statements by Manen *et al* [9] and An *et al* [10] are contradictory where one group of researchers found that printing speed has very low influence and another group found that it has big influence on the deformation of the structure.

Five batches of samples with varying print speeds for the active layers were prepared for testing. The speed was distributed in increments of 15 mm s^{-1} from 35 mm s^{-1} to 95 mm s^{-1} . Build plate temperature was kept 60°C . Figure 5(a) shows the actual deformation of printed structures after morphing and figure 5(b) shows the strain dependence of the print speed of the active layers.

The results obtained proved that the deformation is strongly influenced by the printing speed of the active layers. Increasing the printing speed for the active layers has been shown to possess a significant effect on the deformation, as the results changed more than just in the $\pm\sigma$ range. This difference confirmed that the results are by no means randomly scattered over a larger range, but rather influenced by the parameter change itself. Material deposition on higher speeds produce larger residual stresses because material is stretched more during faster extrusion. Results prove that higher printing speed produces higher residual stresses. Same

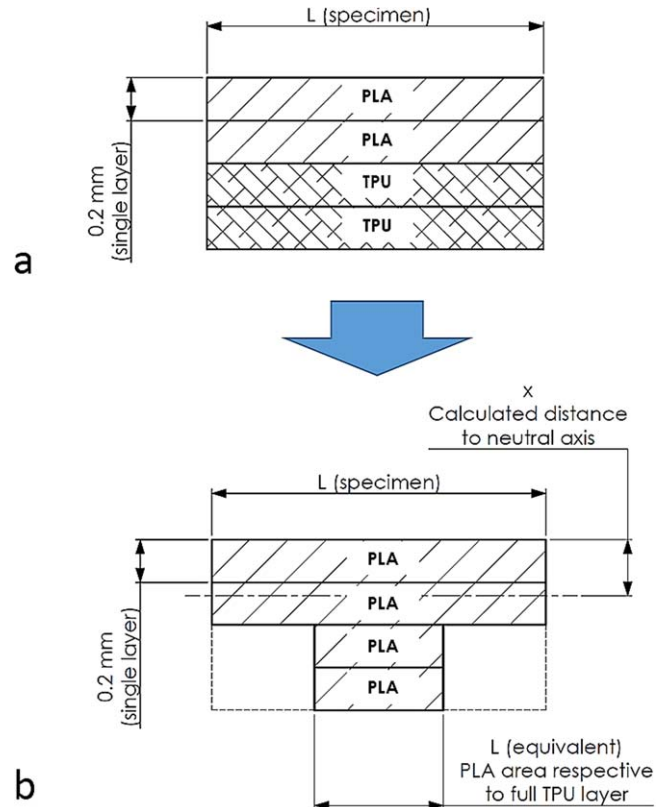


Figure 4. Calculation of equivalent area method: (a) Zoomed area of the specimen (figure 3). (b) Transformed area for calculation of position of neutral axis.

behaviour was reported by An *et al* [10]. However, Mannen *et al* [9] stated that printing speed has very low influence. They used 4 times thinner layers (0.05 mm instead of 0.2 mm) and it is possible that printing speed has much lower influence if thinner layers are used. If the printing speed is constant, and only the layer height varies, the amount of material extruded through the nozzle is lower when thinner layers are used and thus lower stresses are stored in PLA material. However, in our work the influence of layer thickness was not investigated and is left for future experiments.

The mean values are characteristic for each set. The dispersion is quite large, which means that deformation results can vary between different specimens. For example, the largest differences from the mean strain value in the sets were achieved where active layers of parts were printed at 35 mm s^{-1} and 95 mm s^{-1} . The variations are around 0,75%

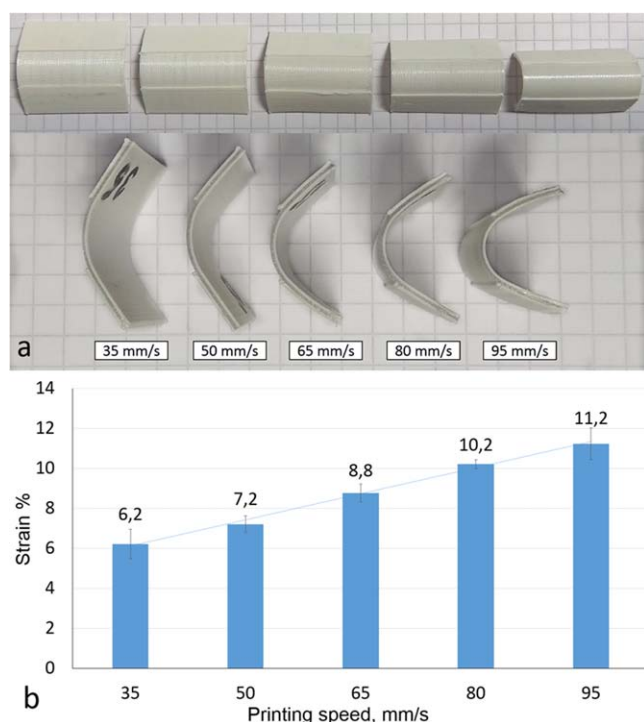


Figure 5. Printing speed influence: (a) Structures after deformation. Printed at constant build plate temperature of 60 °C. (b) Generated strain dependency from printing speed.

of strain. The smallest difference between the results of calculated strains is 0,22% and it occurred in the batch, which had parts with active layers printed at 80 mm s⁻¹. Therefore, it can be stated that deformation of investigated structures can be controlled by varying printing speed.

The maximum strain value of 11,2% was reached when parts were printed at the speed of 95 mm s⁻¹. The compensation line in the diagram indicates a continuous change in the deformation with a simultaneous increase in the printing speed, so that it can be assumed that this dependency is almost linear. Under this assumption, it is possible to predict the approximate deformation angle, i.e. to select the printing speed according to the expected result.

4.2. Influence of build plate temperature

The influence of temperature of the build plate on the performance of the shape-changing is also investigated. It was expected that printing the structures on a cooler build plate would increase their deformation because the deposited layers of PLA can cool faster. Higher residual stresses could be retained in the layers as the rapid temperature decrease would reduce the chain mobility of polymer faster [9]. This would result in macroscopic shape fixation that would prevent any further stress relaxation. The change in temperature of build plate should have a greater effect on thinner structures (i.e. structures composed of fewer layers) as the influence of build plate temperature would mostly influence the first layer. Since the layers are built on top of each other, the second layer would be extruded on the first, which should keep its upper part relatively hot anyway. Therefore, the temperature

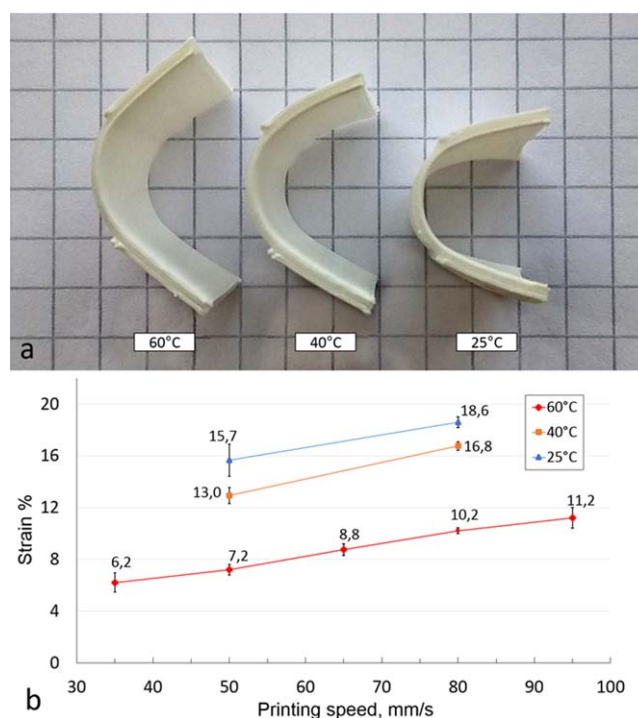


Figure 6. Build plate temperature influence: (a) Structures after deformation. Printed at 80 mm s⁻¹. (b) Generated strain dependency from printing speed and build plate temperature.

between layers should gradually normalize and the deformation in the thicker structures would be more influenced by other parameters such as printing speed and thermal history.

To investigate how the deformation is influenced by the temperature of the build plate, specimens were printed on cooler build plate. All structures had the same shape, size and number of layers (2 layers of PLA and 2 layers of TPU) as in previous study. Two different printing speeds of active layers were investigated: 50 and 80 mm s⁻¹. The build plate temperatures were set to 25 °C, 40 °C and 60 °C. The results from first study are also included. Specimens after deformation are shown in figure 6(a) and strain dependency from printing speed and build plate temperature is shown in figure 6(b).

The results show that the temperature of the build plate has a significant influence on the deformation of the structures. Setting the printing surface temperature to 25 °C and printing speed to 80 mm s⁻¹ resulted to significantly higher deformation compared to structures whose active layers were printed at hotter surface (60 °C).

From figure 5(b) it is clear that printing speed has almost linear influence to strain of the structures. By changing the temperature of the build plate (printing on cooler build plate), linear relationship is still kept, because the steepness of the lines almost did not change (see figure 6(b)). Build plate temperature decrease from 60 °C to 40 °C produces higher influence to bending of the structures than temperature decrease from 40 °C to 25 °C. It is believed that after printing any single layer, and just before starting the new one, former layer cools to temperature close to 40 °C. When the build plate temperature is kept at 25 °C, the first layer on the build plate after printing cools to around 40 °C. This assumption

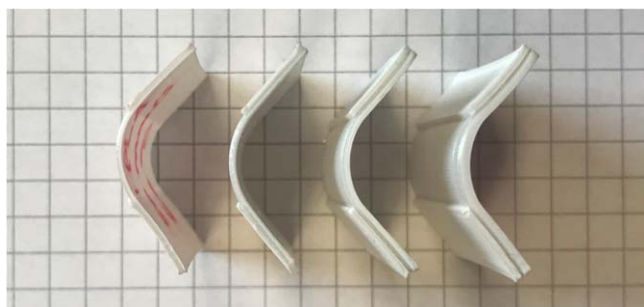


Figure 7. Structures after deformation. Printed at 50 mm s^{-1} and build plate temperature of 60°C . From left to right: 1 active PLA layer, 2 active PLA layers, 3 active PLA layers, 4 active PLA layers.

explains, why the difference of build plate temperature from 25°C to 40°C produces smaller difference of bending angle of the structures.

Assuming that the temperature of the build plate largely influences the first layer and later gradually normalizes over subsequent layers, this dependence could possibly be linearized by cooling or heating all layers together to achieve a more uniform distribution of residual stresses over the whole structure. The enclosure for the printer with regulated temperature control might solve this problem.

The measurements of the structural deformations show as a visible tendency that the deformation of structures was indeed dependent on the temperature of the build plate. The cooler the build plate, the higher deformation structures achieve at any printing speed. Under the assumption that the change in printing speed for the active layers almost always linearly influences the resulting deformation, the entire deformation range can be compensated accordingly by changing the build plate temperature. The depositing of material on a cool build plate helps to maintain higher residual stress by quickly locking polymer chain in the stretched state, which results in the structures bending more. Figure 6(a) contains a picture with side-by-side comparison of all three structures printed at 80 mm s^{-1} .

4.3. Influence of number of active layers

Structures with one, two, three and four active layers printed on both 50 and 80 mm s^{-1} were used to investigate the influence of number of active PLA layers on deformation of the structures. Build plate temperature was kept 60°C for all specimens. Figure 7 shows all specimens of different thicknesses (all printed at 50 mm s^{-1}) after completed shape recovery step. The number of passive TPU layers was kept the same (2 TPU layers) for all structures in this study. As can be seen, reduction or addition of active layers does not change bending angle of the structures. All structures bent to very similar angles.

Figure 8(a) shows the strain dependency from the amount of active layers and their respective printing speeds while figure 8(b) shows the bending angle dependency. Blue, longest line with squares, representing structures with 2 PLA and 2 TPU layers can be taken as a reference point. Figure 8(a) indicates that strain is dependent from amount of

active layers and increases with the amount of active layers while figure 8(b) shows that bending angle of the structures almost does not change, especially for structures containing 2, 3 or 4 active PLA layers.

Because the strain is calculated on the outer surface (strain surface, see figure 3), the strain increases when amount of active layers in the structures increase. It is because the neutral axis of the structure moves away from the strain surface when more active layers are added, since they have much higher stiffness than soft TPU material. It can be concluded that strain in this case is not proper value to define the deformation of the structures, when geometry of the structures varies. For this reason figure 8(b) is added where bending angle dependency is shown.

Specimens with only single PLA layer produce smallest strain and bend to lowest degree. Only single layer of PLA in the structure does not produce enough contraction (has less residual stresses stored in the material) to bend structure to high degree. The structures with 2, 3 and 4 layers on the other hand bent to higher degree but they all bent to very similar angles and the results are slightly overlapping. It is believed that this phenomena arrives from the shift of neutral axis. In structures with 2, 3 and 4 active layers, the neutral axis is almost in the middle of all active layers. It can be assumed that half active layers are compensating other half of the active layers. However, then the structure should not bend at all. It is believed that first layer of active material which is printed directly on the build plate has highest amount of stored residual stresses. This theory can be explained with figure 8(b), where it can be noticed that the more active layers are in the structure, the more structures are bending because the distance from first layer with highest stored residual stresses increased from neutral axis. However, the difference between bending of specimens is very small and the measured data is overlapping.

Very interesting behaviour is observed for structures that are printed at 80 mm s^{-1} and have 2, 3 or 4 active PLA layers. They all deform almost to the same angle. This phenomena currently cannot be explained and it can be measurement error. It was also observed that structures with more active material layers require more time to achieve full deformation because the heat requires more time to penetrate the structure. This problem can be solved or adjusted by incorporating porosity in the structures as Manen proposed [8], however in this case the performance of the structures can be lowered.

The results lead to conclusion that increased number of active layers increases the deformation of the structures only slightly. It is mainly because of position of neutral axis which is in the middle of active layers and is only slightly moved towards TPU (TPU has much lower flexural stiffness). Interesting phenomena was observed that the first layer of active material that is printed directly on the build plate might have higher residual stresses stored inside. However, further investigation is envisioned in the future because the results are overlapping and it might be simply measurement error.

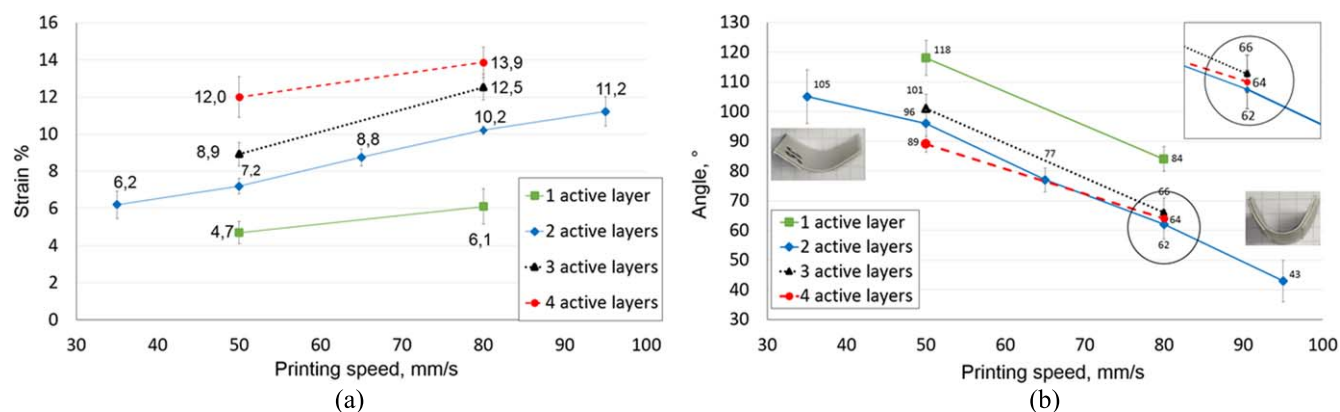


Figure 8. Number of active layer influence to the performance of the structures. (a) Strain dependency from printing speed and number of active layers. (b) Angle dependency from printing speed and number of active layers.

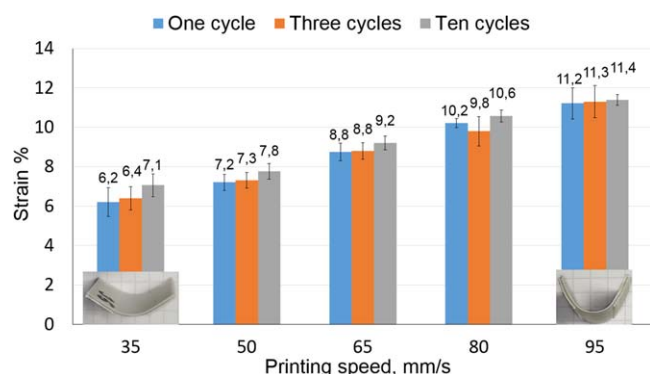


Figure 9. Comparison of deformations after one, three and ten cycles.

5. Shape recovery rate and endurance

The same five sets of samples (build plate temperature 60 °C and different printing speeds) from the first test were re-used to investigate the repeatability of deformations. Firstly, the structures were subjected to two additional shape-changing cycles, i.e. they were heated and placed in a flat position, cooled and then immersed in hot water again to allow shape-changing. This process was repeated twice to increase the total number of deformation cycles to three. Structures were rescanned and the measurement data was collected. To obtain more data in order to compare the repeatability of transformations, structures were subjected to additional seven shape-changing cycles. The structures were rescanned and deformation data were collected for comparison after a total of ten cycles. The data is presented in figure 9.

Results show that the deformation of the structures retain quite similar values even after ten shape-changing cycles. Largest difference between the mean values of strain were noticed between the first and the tenth shape-changing cycle of the structures containing active layers printed at 35 mm s⁻¹. The variance of the results can be a result of manufacturing process, as the non-ideal printing conditions can influence the deformations after several cycles. The parameter values may vary as the 3D printer used to print the specimens is quite inexpensive and not one of high-end 3D

printers. In addition, each part of these batches was printed individually. A combination of these circumstances could have led to a difference in deformation. In other cases, similar mean values of the measured quantities were obtained and the ranges of standard deviation overlap mostly for each number of cycles. In most cases, the standard deviation seemed to slightly increase with increasing amount of shape-changing cycles. This can result in structures requiring several actuation cycles before final deformation values can be obtained.

It was observed that when structures were placed in hot water for a second, third and further shape recovery steps, the recovery speed increased compared to the first step. While the initial transformation took around 15–20 s to reach full range, consecutive shape recovery steps took only 3–4 s to reach full range of morphing.

After straightening the structures during the shape-changing cycle, one specimen out of thirty-five exhibited wear results. The upper of the two passive layers partially delaminated, but this did not seem to have a decisive impact on its structural integrity, as the structure could still bend as before. After all structures had gone through ten cycles of shape-changing, none of the other specimens showed any signs of wear or further delamination. Since only one part reached this condition, there is a possibility that this may have been a result of an error during printing process.

6. Conclusions and future work

The deformation angle can be tuned by printing structures on a cooler surface and by selecting higher printing speeds. These two printing parameters can lead to a higher residual stresses stored in the material and thus be used for structures with more active layers, so that these structures can bend even more after the recovery step while maintaining the increased overall stiffness. Furthermore, it was found that the structures consisting of more layers required more time to reach their final shape. This happens because the inner layers do not reach their rubbery state and try to return to their high entropy state as quickly as those located on the outer regions. Although it may look like a drawback at a first glance, it can

be treated as a feature. The number of active layers can be utilised as a mechanism for a sequential folding of a complex layout of a model.

All printing parameters investigated have a significant influence on the extent to which structures can deform. By varying these parameters, multi-material morphing structures can be designed. A combination of the parameters would make it possible to create larger structures with differently programmed hinges.

The study with different number of active layers showed only very slight difference between performance of the structures. It is believed that this almost same performance arrives from shift of neutral axis, which is almost in the center of the active materials in all investigated specimens. It is believed that by moving neutral axis away, higher performance of the structures could be achieved.

The actuation stimuli currently used (hot water) may not be the best choice for applications and alternative actuating methods should be investigated. A few early experiments show that actuation of our investigated structures in oven is not a problem and produces the same behaviour of the structures.

While these structures are pre-programmed during printing and the transformation rate can be manipulated by the number of active layers, reversible deformation still remains a challenge. Structures also become quite soft, when heated above glass transition temperature of active layers. The blocking force of our investigated structures is questionable and is left for further investigations.

In order to fully understand and describe our investigated structures, further research is required. It is believed that influence of passive TPU layers to the deformation of the structures should be also investigated. Moreover, the influence of other basic printing parameters such as layer height and width of the raster must be investigated.

ORCID iDs

Rytis Mitkus  <https://orcid.org/0000-0002-6715-1044>

Michael Sinapius  <https://orcid.org/0000-0002-1873-9140>

References

- [1] Tibbits S, McKnelly C, Olguin C, Dikovsky D and Hirsch S 2014 4D printing and universal transformation *ACADIA 14: design agency, Proc. 34th Annual Conf. of the Association for Computer Aided Design in Architecture* pp 539–48
- [2] Raviv D *et al* 2014 Active printed materials for complex self-evolving deformations *Sci. Rep.* **4** 7422
- [3] Ge Q, Qi H J and Dunn M L 2013 Active materials by four-dimension printing *Appl. Phys. Lett.* **103** 131901
- [4] Wu J *et al* 2016 Multi-shape active composites by 3D printing of digital shape memory polymers *Sci. Rep.* **6** 24224
- [5] Mao Y *et al* 2015 Sequential self-folding structures by 3d printed digital shape memory polymers *Sci. Rep.* **5** 13616
- [6] Mao Y *et al* 2016 3D Printed reversible shape changing components with stimuli responsive materials *Sci. Rep.* **6** 24761
- [7] Ge Q *et al* 2016 Multimaterial 4d printing with tailorable shape memory polymers *Sci. Rep.* **6** 31110
- [8] van Manen T, Janbaz S and Zadpoor A A 2017 Programming 2D/3D shape-shifting with hobbyist 3D printers *Mater. Horiz.* **4** 1064–9
- [9] van Manen T, Janbaz S and Zadpoor A A 2018 Programming the shape-shifting of flat soft matter *Mater. Today* **21** 144–63
- [10] An B *et al* 2018 Thermorph: Democratizing 4D Printing of Self-Folding Materials and Interfaces *The 2018 CHI Conference* (<https://doi.org/10.1145/3173574.3173834>)
- [11] Huang W M *et al* 2012 Shape Memory Polymers (SMPs)—Current Research and Future Applications <https://www.azom.com/article.aspx?ArticleID=6038>
- [12] Kim Y, Yuk H, Zhao R, Chester S A and Zhao X 2018 Printing ferromagnetic domains for untethered fast-transforming soft materials *Nature* **558** 274–9
- [13] Momeni F *et al* 2017 A review of 4D printing *Mater. Des.* **122** 42–79
- [14] Li X, Shang J and Wang Z 2017 Intelligent materials: a review of applications in 4D printing *Assem. Autom.* **37** 170–85
- [15] Wu J-J *et al* 2018 4D printing: history and recent progress *Chin. J. Polym. Sci.* **36** 563–75
- [16] Chung S, Song S E and Cho Y T 2017 Effective software solutions for 4D printing: a review and proposal *Int. J. Precis. Eng. Manuf.-Green Technol.* **4** 359–71
- [17] Hu G F *et al* 2017 Increasing dimension of structures by 4D printing shape memory polymers via fused deposition modeling *Smart Mater. Struct.* **26** 125023
- [18] Ding Zhen, Weeger Oliver, Qi H. Jerry and Dunn Martin L. 2018 4D rods: 3D structures via programmable 1D composite rods *Materials & Design* **137** 256–265
- [19] Yang H *et al* 2017 3D printed photoresponsive devices based on shape memory composites *Adv. Mater.* **29**
- [20] Akbari S *et al* 2018 Enhanced multimaterial 4D printing with active hinges *Smart Mater. Struct.* **27** 65027
- [21] Yang C *et al* 2017 Modelling and characterisation for the responsive performance of CF/PLA and CF/PEEK smart materials fabricated by 4D printing *Virtual Phys. Prototyping* **12** 69–76
- [22] Fernandes J, Augusto M D, Reis L, Vaz M F and Leite M 2018 Study of the influence of 3D printing parameters on the mechanical properties of PLA *3rd International Conference on Progress in Additive Manufacturing (Pro-AM 2018)* (Singapore, 4–17 May 2018)
- [23] Wahit M U, Hassan A, Ibrahim A N, Zawawi N A and Kunasegeran K 2015 Mechanical, thermal and chemical resistance of epoxidized natural rubber toughened polylactic acid blends *Sains Malaysiana* **44** 1615–23
- [24] Tham W L, Poh B T, Ishak Z A M and Chow W S 2016 Characterisation of water absorption of biodegradable poly (lactic acid)/halloysite nanotube nanocomposites at different temperatures *J. Eng. Sci.* **12** 13–25
- [25] Ecker J V *et al* 2019 Mechanical properties and water absorption behaviour of PLA and PLA/wood composites prepared by 3D printing and injection moulding *Rapid Prototyping J.* **31** 1213