3D PRINTING OF SHAPE CHANGING POLYMER STRUCTURES: DESIGN AND CHARACTERIZATION OF MATERIALS

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Abstract

Additive manufacturing (AM) gives engineers unprecedented design and material freedom, providing the ability to 3D print polymer structures that can change shape. Many of these Shape Memory Polymer (SMP) structures require multi-material composites, and different programmed shapes can be achieved by designing and engineering these composites to fold and unfold at different rates. To enable SMP applications involving shape-changing geometries, it is important to have an understanding of the relationships between intermediate shapes and the initial and final designed shapes. To accomplish this, we investigated readily available 3D printable polymer materials and their thermo-mechanical characteristics to create multi-member structures. This paper demonstrates a way to generate different temporary geometric profiles on a single 3D printed shape with the same material. This paper also includes insights from thermo-mechanical analysis of the materials to help create multi-member shape-changing geometries using 3D printing.

1. Introduction

Stimuli-responsive materials can change their shape, mechanical properties, electrical properties, optical properties, etc., upon application of external stimulus such as an electrical field, pH, light, magnetic field, thermal field, etc. [1]. Shape memory polymers (SMP) are such stimuli-responsive materials as they can change from a temporary shape to a permanent shape in response to a stimulus. Heat [2], light [3], and chemical solvents [4] are the few commonly used external stimuli to actuate SMPs [5]. Multi-material 3D printing can be used to create soft polymer composite structures with elastomeric matrix reinforced by such SMP fibers [6,7]. These soft polymer composite structures are also known as Printed Active Composites (PAC) in engineering literature [7] due to the printed composite's ability to respond to the environment and change shape; however, we use the term SMPs for the remainder of this paper to refer to 3D printed shape memory polymer structures.

3D printing is a modern manufacturing method to fabricate 3D objects using layer-by-layer material deposition [8]. CAD data of the 3D object is initially sliced to generate the layer-by-layer contour information. There are seven 3D printing processes, and our focus in this paper is the polyjet process, which has been shown to fabricate SMPs [9]. Polyjet-based 3D printing deposits a liquid polymer material and then cures the deposited material with UV light [10]. This continuous deposition and curing occurs until the entire layer is filled with the material, and then the process repeats for the remaining consecutive layers. The Objet 260 Connex3 system [11], which is based on the

polyjet technique, is used to manufacture the SMPs in this paper. Printing these shape-changing structures is also termed as 4D Printing, where the 4th dimension is time. Recently, 4D printing has been gaining attention after the concept was introduced in [12].

The 4D printing concept is relatively new, and different research groups are trying to establish its foundation with early investigations into design and materials for SMPs. For instance, researchers have demonstrated methods to use 3D printing to print active materials that can be stimulated with water [13] and heat [14]. Different programmable shapes are also demonstrated in the literature [7,14,15]; however, *design guidelines that relate the initial shapes with temporary shapes are still required to enable designers to create complex SMP-based structures*. Although current applications of SMPs are limited due to the difficulty in design and manufacturing of these structures, there are many potential opportunities in future technological applications such automotive seating, customized prosthetics, and gas sensors [20,21]; however, realizing these technological applications is contingent upon being able to design and fabricate different geometries that transform into the desired shapes when activated.

There are other classes of materials that exhibit shape memory effects, for example, shape memory alloys [16], but SMPs are unique due to their characteristics of being light weight, flexible, soft, biodegradable and biocompatible [17–19]. Many of these polymer structures require manufacturing of polymer composites with embedded fibers made of a material different from that of the polymer matrix. Manufacturing thin polymer fibers is a critical aspect of manufacturing these SMPs, and melt-spinning has been used extensively to create functional fibers in different profiles [22]. However, this process requires manufacturing fibers first and then embedding them in usable designs for applications. This limits the application of SMPs from design and manufacturing aspect due to the constraints in realizing complex shapes for many different applications. Thus, it is important to develop modern manufacturing methods to create SMPs with flexibility in design, length scales, and allowing for customization from one application to other. Combining SMPs with 3D printing enables creation of such complex geometries because 3D printing is not only suitable for intricate functional geometries, but also allows customization with negligible effect on the manufacturing cost [23–25].

The goal in this work is to investigate the mechanical performance of 3D printable materials at different temperatures and demonstrate different temporary shapes on a single 3D printed initial shape. The following section describes the design of the SMPs and 3D printable materials used in this study, followed by results from thermo-mechanical tests. The later section discusses preliminary results obtained from experiments to generate different programmed shapes on the same initial shape for two different materials.

2. Design and Materials

The SMP composites in this study are made of two layers: (1) a flexible layer; and (2) a flexible layer with rigid fibers (see initial shape in Figure 1). The top layer does not contain any rigid fibers, and it is made of a flexible material, available in the Objet

Connex3, called Tangoblack+. The bottom layer is a composite structure comprised of Tangoblack+ as the matrix material and a blend of Tangoblack+ and Veroclear (a rigid polymer available in the Object Connex3) as the fiber material. Both the Tangoblack+ (flexible material) and Veroclear (rigid material) are proprietary materials of STRATASYS [26]. Combining rigid (Veroclear) and flexible (Tangoblack+) materials at different proportions within the Objet Connex3 system creates a hybrid material with intermediate properties; therefore, fibers with different strengths can be fabricated. For instance, a mixture of 5% Tangoblack+ and 95% Veroclear is rigid compared to 100% Tangoblack+, and flexible compared to 100% Veroclear.

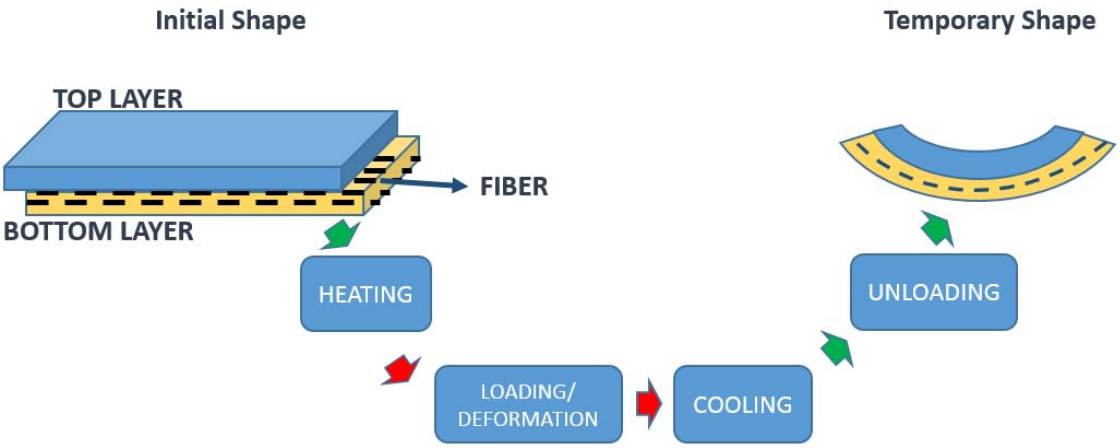

Figure 1: Shows horizontal bilayer composite transforming to a curved shape

The shape memory behavior of the composite is due to the difference in the elastic moduli and glass transition temperatures of Tangoblack+ (T_g = -10 to -5 ^oC) and Veroclear (T_g = 52-54 ^oC). At room temperature, the rigid material is below the glass transition temperature and exhibits a glassy nature; meanwhile, the flexible material is above the glass transition temperature and exhibits a soft rubbery nature. When these polymer composites are heated to elevated temperatures (above the glass-transition temperature in this case), they can be deformed by applying small external forces. When the temperature is decreased below the glass-transition temperature, the deformed shape remains fixed in the temporary shape of the structure. When the polymer is re-heated to above the glass-transition temperature, then it is free to regain the original shape at much lower rigidity [18,19]. The process of heating, deformation, unloading and reheating is illustrated in Figure 2.

For instance, consider the initial shape shown in Figure 1. Upon cooling, the bottom layer has increased in length as the fibers are stretched and fixed, and the top layer is at the initial/original length. As a result, the polymer attains a curved configuration to accommodate the two layers of different lengths (see temporary shape in Figure 1). Thus, a temporary curvature can be programmed in a straight structure using this process. The composite turns soft and flexible upon heating above the T_g of the fiber material, and the composite returns to its original shape. As a result, the composite changes to its initial shape if left unloaded at high-temperatures.

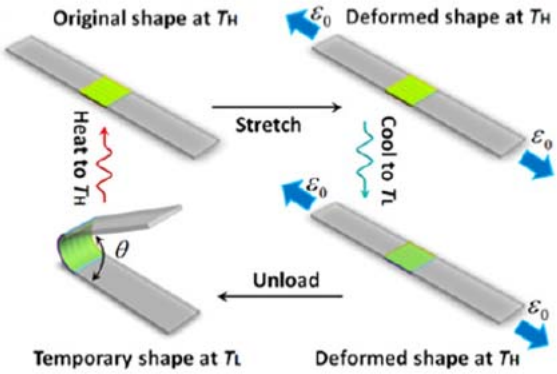

Figure 2: Example of thermo-mechanical programming to induce folding in a hinge [15]

Different temporary shapes can be achieved by changing the internal fiber arrangement within these structures. Three structures with different fiber arrangements are fabricated as illustrated in Figure 3. Blue and green in the figure represent the matrix (Tangoblack+) and fiber (Veroclear) materials, respectively. In Figure 3(a), the fibers are continuous in the top layer; whereas in Figure 3 (b)-(c), fibers of different lengths are present in both the top and bottom layers. These composites are fabricated with a blend of 5% Tangoblack+ (for the matrix) and 95% Veroclear (for the fibers). The 3D printed parts are subjected to the process shown in Figure 2, and the resulting temporary shapes are shown in Figure 4.

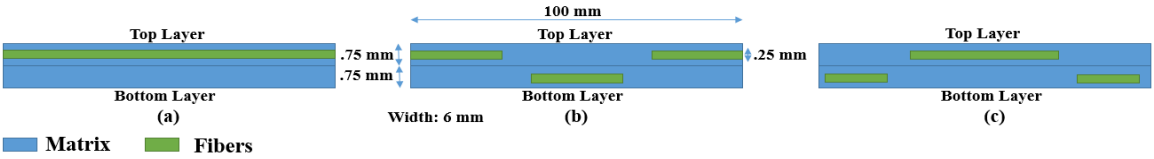

Figure 3: Illustration of different fiber arrangements in composites

As illustrated in Figure 3, if the fibers are placed in the bottom layer, then the temporary shape is curved towards the top layer. Conversely, if the fibers are placed in the top layer, the shape is curved towards the bottom layer. To create a 'sine' shape, the fibers are placed in alternate fashion: in the top layer on the left and right ends, and in the middle in the bottom layer (see highlight box in Figure 4 (c). The lengths of these fibers are varied (fibers at the left and right ends are shortened and fibers at the middle are made longer) to generate the omega (Ω) shape (see Figure 4 (d)). These structures regain their initial shape when placed in hot water (around 80 °C). The roll, sine, and omega shaped structures unfold back into the same initial rectangular shape.

Although the temporary shapes differ substantially, the initial (flat) shape of these structures is identical; therefore, by controlling the lengths and placement of the fibers in the composite, different temporary shapes can be generated with SMPs. These different shapes can consolidate into a large SMP structure with more complex profiles.

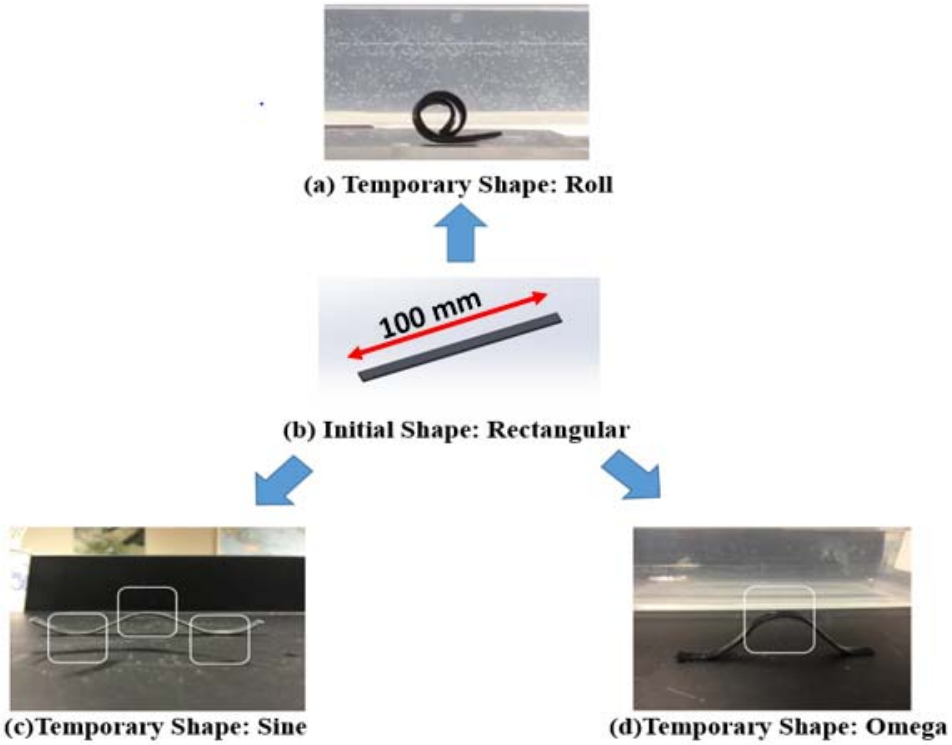

Figure 4: Few programmed shapes on an initial rectangular shape

In the remaining sections of this paper, we investigate the temperature-dependent mechanical performance, and a method to generate different shapes using the same initial structure. Section 3 presents the results of Dynamic Mechanical Analysis (DMA) and tensile tests on different compositions of rigid and flexible materials. Section 4 contains experiments with different stretching lengths to demonstrate various temporary shapes on a single 3D printed shape. Section 4 also presents the recovery times for various materials. Table 1 summarizes the details of these material characterization tests and shape generating experiments.

Table 1: List of samples and related characterization

Type of characterization	Initial shape	Materials	Repetitions	Other parameters
DMA	-	Sample 1(5% T) to Sample 4 (20%T)	-	Temperature sweep from -50 ^{o}C to 100 ^{o}C
Tensile tests	-	5%, 10%, 15%, 20%, 25%, and 60% of T	-	Upto 10% strain
Generating different shapes	See Figure 3(a)	5% T	-	Stretch lengths: 5.5, 6.5 and 7.5 mm
	See Figure 3 (c)	5% T and 20% T	-	Stretch lengths: 5,6,7, and 8
Recovery times	See Figure 11	Polymer 1 (RGD 05), Polymer 2 (RGD 20), Polymer 3 (RGD 30), and Polymer 4(FLX 85)	2	

3. Material Characterization of Different Blends of Rigid and Flexible Materials

We use DMA to evaluate the gap between the transitions temperatures in the different material compositions of Tangoblack+ and Veroclear. Polymer materials are viscoelastic in nature, and the individual contribution of viscous and elastic properties varies with the temperature of the polymer; therefore, moduli in polymers can be denoted by two components: (1) storage modulus, representing the elastic nature of the polymer, and (2) loss modulus, representing the viscous nature of the polymer. As the polymer is heated above the glass transition temperature, the storage modulus (or elastic nature) drops while the loss modulus (or viscous nature) increases. At the transition-temperature of the polymer, where the material starts to exhibit a rubbery nature, the storage modulus decreases significantly, and the loss modulus reaches a maximum. This transition temperature is also indicated by the peak in the tan (δ) curve that represents the measure of viscous to elastic natures [27].

Our DMA tests were conducted on TA RSA-G2 [28], and the experiments were performed over the temperature range of -50 °C to 100 °C at a heating rate of 3 °C /min. The oscillation frequency in the experiment was 6.28 rad/s. Figure 5 shows the peaks of tan (δ) curves that represent the corresponding transition temperatures. The temperature gap (see Figure 5) between the transition temperatures of these materials is narrow (10 °C); therefore, precise thermal stimulation is required to exploit this difference in transition temperatures to actuate folding structures comprised of hinges with different materials. This precise temperature management is difficult if the hinges are not sufficiently far apart, because thermal actuation of one hinge can indirectly actuate a nearby hinge due to heat transfer.

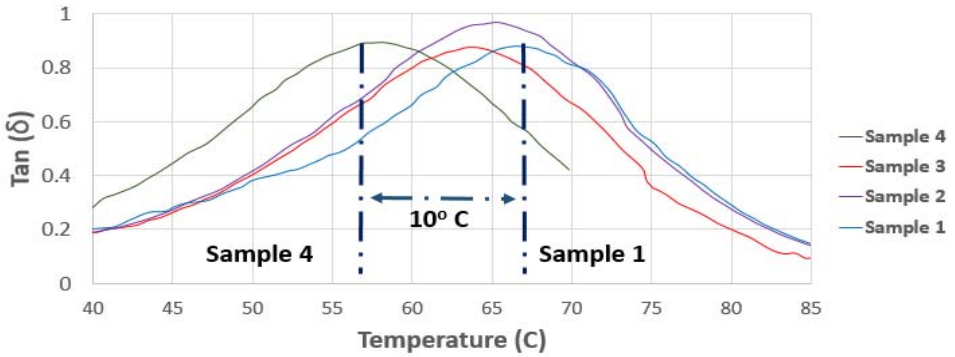

Figure 5: Results of the temperature sweep test on four different material compositions of Tangoblack+ (T) and Veroclear (V); sample 1(5%T) to sample 4 (20%T) are in the increasing order of flexible nature

For many polymer materials, the tensile modulus of the material is also dependent on the strain in the material; therefore, tensile testing is performed to capture the effect of strain on tensile modulus for different material compositions. The tensile test results shown in Figure 6 for the material samples show a general trend that materials that are more flexible have lower modulus for the same strain percent. Material sample with 10% flexible material deviates from the observed trend at higher strains; however, the trend in tensile modulus at lower strains (less than 1.5%) is still followed. This deviation in

pattern may be due to manufacturing defects such as lack of proper curing, material non-uniformity, contamination with support material, induced-physical defects, etc.

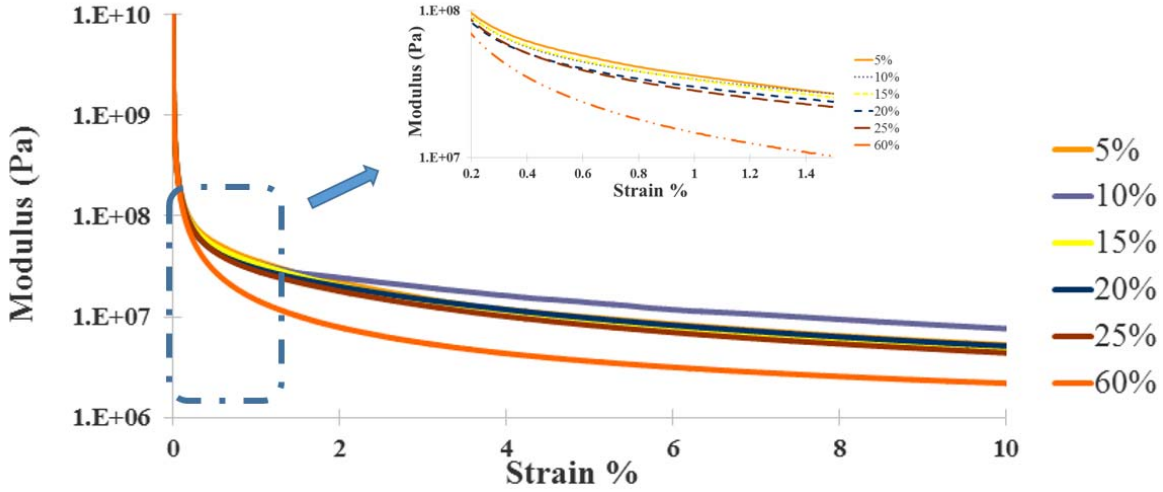

Figure 6: Tensile modulus versus strain curve for different compositions of Tangoblack+ and Veroclear: 5%, 10%, 15%, 20%, 25%, and 60% of flexible material (Tangoblack+)

4. Experiments to Program Different Temporary Shapes in Same Initial Shape

One way of programming different temporary shapes into SMPs is by leveraging the difference in thermo-mechanical performance of different material compositions; unfortunately, it is difficult to achieve the precise thermal stimulation required to exploit the narrow temperature gap. Another method of achieving different temporary shapes is by controlled deformation of the material during the stretching/deformation phase (see Figure 2). By inducing different deformation lengths, different temporary geometric profiles can be obtained. The experimental setup used in this work to stretch the test specimen by pre-determined values is shown in Figure 7. The setup consists of an assembly of fixed and movable linear stages. The gauges attached to these stages can measure the deformation lengths at any moment.

A rectangular test specimen is tested by stretching (i.e., deforming) it to different lengths: 5.5 mm, 6.5 mm, and 7.5 mm. The test specimens are subjected to the programming and actuation process illustrated in Figure 2. Once the temperature of each specimen is above the glass transition temperature for the material, then the material can be stretched by turning the knob of the gauge on the movable stage in Figure 7. The deformed/stretched specimen is cooled to room temperature to fix the temporary shape. Three different profiles are programmed on the same initial shape as shown in Figure 8. This method of inducing measured-deformation at slow rate also prevents the mechanical failure of the material.

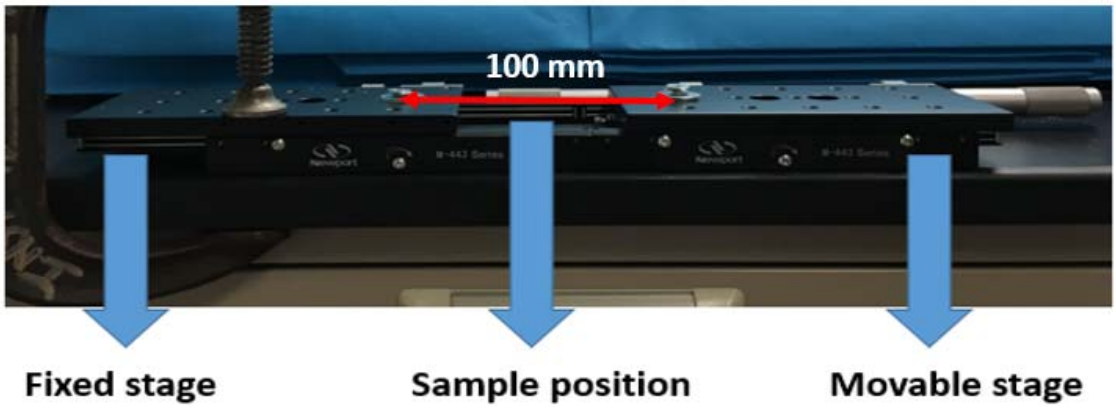

Figure 7: Experimental setup to induce measured deformation in the material

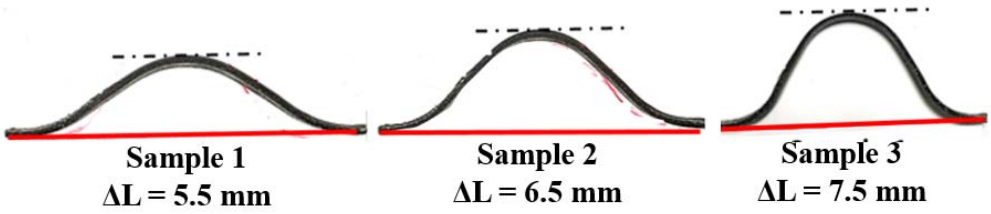

Figure 8: Different programmed shapes obtained by different induced deformation lengths (ΔL): 5.5 mm, 6.5 mm, and 7.5 mm

Samples made of two different material compositions (i.e., 95 % rigid and 80% rigid compositions) are subjected to four different elongation lengths to generate different temporary shapes. The resulting temporary shapes along with geometric dimensions are shown in Figure 9. R is the radius of curvature calculated at the center of the temporary shape. For this sample (initial length of 100 mm), decreasing the elongation lengths from 8 mm to 5 mm significantly affected the temporary shape. This procedure demonstrates the ability to induce different geometric profiles by inducing different deformation lengths; however, the repeatability of the programmed profiles is still unknown. To investigate the repeatability, multiple specimens with same material can be tested with the same deformation lengths. Understanding this relationship between material, deformation length, and temporary shapes can help create a mapping between different programmable shapes and initial shapes.

The plot in Figure 10 shows the radius of curvature versus the elongation length. We see that a 35% change in radius of curvature can be achieved with a change in elongation length of 20%. Although the difference in the radius of curvature for two different compositions at same elongation is less, the composition with the higher percentage (95%) of rigid material always yields a lower radius of curvature. This pattern demonstrates that higher radius of curvature can be achieved by using more rigid materials while the stretching length remains same. These preliminary results also demonstrate the ability to achieve different temporary geometric profiles by varying material composition.

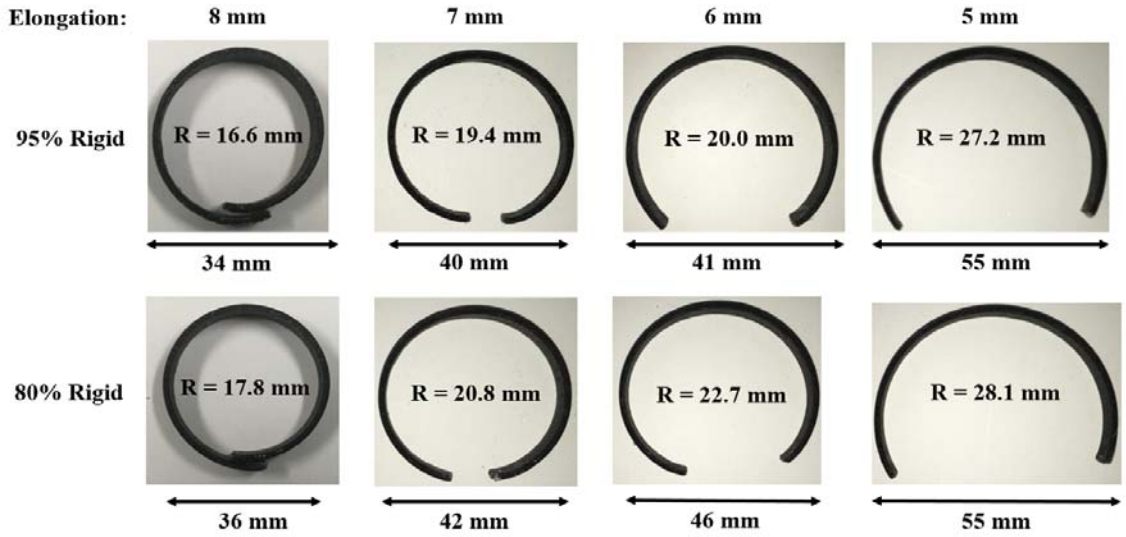

Figure 9: Different temporary shapes programmed on two different materials by stretching at various lengths (8 mm, 7 mm, 6 mm and 5 mm); R is the radius of curvature at center of the shape

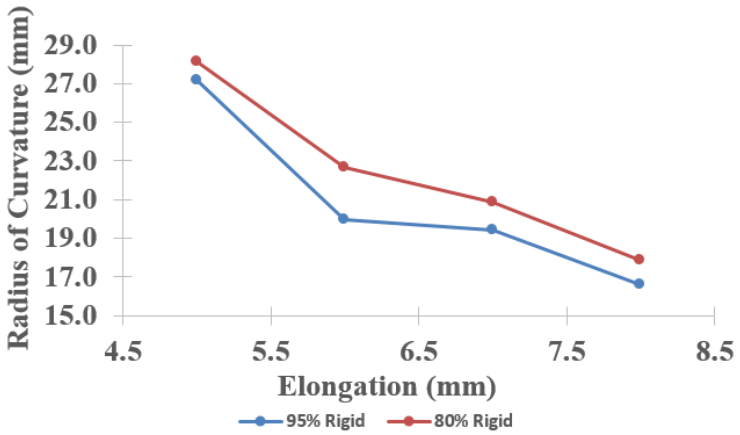

Figure 10: Effect of elongation length on the radius of curvature for two different materials

Recovery times of the hinge structures (see Figure 11) made of different compositions are also calculated. Recovery time is the time taken by the sample to shift from a programmed shape to its initial shape upon application of sufficient external heat. For this geometry, the folding angle between the programmed and initial configurations is around 73.5°. Table 2 presents the recovery times for four different polymer samples. Note that the recovery time is almost doubled when the percentage of flexible material in the composition is increased from 5% to 15%. This difference in recovery time can be useful to create shape-changing structures consisting of members actuating at different rates, but the recovery times are significantly affected by the operating temperatures.

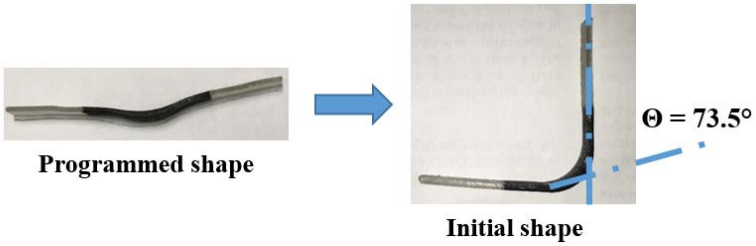

Figure 11: Shows initial and programmed shape of a two-member hinge; Θ is the angle of folding

Table 2: Recovery times for hinges made of four different materials (blends of Tangoblack+ and Veroclear): Polymer 1 (RGD 05), Polymer 2 (RGD 20), Polymer 3 (RGD 30), and Polymer 4(FLX 85)

Material	Recovery Time (s)
Polymer 1	3.5
Polymer 2	5
Polymer 3	7
Polymer 4	-

5. Closing Remarks & Future Work

The ability of 3D printing to fabricate shape memory polymers (SMPs) has already been demonstrated, but it remains unclear how to design and actuate initial shapes to obtain different temporary shapes. Different factors such as fiber arrangement, material, deformation length, operation temperature, etc., are critical to achieve the desired temporary shapes. Investigating the thermo-mechanical response of different materials can improve our understanding of how to design and choose materials for initial shapes to achieve a desired temporary shape. First, the difference in transition temperatures of different blends of materials used in making fibers in a polymer matrix structure is investigated. The results show that this gap is narrow and requires precise stimulation of the material to leverage the difference in transition temperature. Tensile test results of the material samples show a general trend of changing material properties with change in material composition at higher strains. This difference in material properties can be explored to create range of radius of curvatures by changing material composition of the structure.

A method to generate different temporary shapes on a single 3D printed structure by varying the length of stretching (5, 6, 7 and 8 mm) is also investigated. A setup consisting of two linear stages is used to generate temporary shapes of different curvatures. Results demonstrate a potential to generate different shapes by varying material composition and the amount of induced-deformation (stretch length). Higher deformation rates are observed to damage/break the sample; therefore, this method of inducing measured-deformation prevents the obvious mechanical failure of the sample. It is also observed that recovery time of temporary shapes significantly depends on the operating temperature.

Preliminary results in this work demonstrate the ability to achieve different temporary geometric-profiles by deforming the same material at different lengths, illustrating the impact of design decisions on SMPs and their actuation. In continuation of this work, repeatability of these geometric-profiles will be investigated. Additionally, the relationship between materials, deformation length, and temporary shapes will be thoroughly explored to provide engineers with design guidelines that are required to create the complex SMP-based structures.

Acknowledgments

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