Bio-inspired and computer-supported design of modulated shape changes in polymer materials

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Abstract

The Venus flytrap is a fascinating plant with a finely tuned mechanical bi-stable system, which can switch between mono- and bi-stability. Here, we combine geometrical design of compliant mechanics and the function of shape-memory polymers to enable switching between bi- and mono-stable states. Digital design and modelling using the Chained Beam Constraint Model forecasted two geometries, which were experimentally realized as structured films of cross-linked poly[ethylene-co-(vinyl acetate)] supported by digital manufacturing. Mechanical evaluation confirmed our predicted features. We demonstrated that a shape-memory effect could switch between bi- and mono-stability for the same construct, effectively imitating the Venus flytrap.

Introduction

In nature, evolution has had millions of years to develop some of the most ingenious solutions for survival. The carnivorous Venus flytrap (*Dionaea muscipula*) comprises of two trap lobes, which upon stimulation of its inner spikes slams shut to trap prey inside. It is with this snap-through behavior, which is occurring within 300 ms,^[1] that is of particular interest for biomaterial applications. Previously, the mechanism has inspired the invention of pressure-dependent multi-stable origami cells^[2] and was implemented as a bi-stable reversible hydrogel construct.^[3] Many researchers have speculated what processes are behind the quick actuation of closing the trap lobes. Water displacement from the inner surface of the lobes to the external surface plays a critical role,^[4] but pre-stress is important for the speed with which the lobes are closed.^[5] In its open position, the lobes of the Venus flytrap are concave [Fig. 1(a)] and when activated and stimulated the displacement of water acts as an actuation of the lobes from a concave to a convex configuration. The switching between an open and closed lobe position, can be seen from a mechanical perspective as a bi-stable system—such a system has per definition at least two stable states that remain unchanged until further activation. These positions are points of local energy minima, or in other words where the static forces are balanced. Bi- or meta-stable systems are also present in the field of compliant mechanisms. They are defined as flexible entities that transmit motion through elastic body deformation.^[6] In comparison to classical mechanisms, e.g. barrel hinges, which require a minimum of three parts; two leaves and a pin connector, a compliant mechanism should enable the same motion in one single part. The benefit is minimized abrasion, since no parts

are grinding against each other, and no need for assembly.^[7] With the advance of digital manufacturing such as Fused Deposition Modelling (FDM), Multi-Jet Printing (MJP), and Stereolithography (STL), the ability to design and manufacture complex single-part objects have become easier and require less processing time. Mechanisms with bi-stable behavior can be used in a range of applications, including the design of Microelectromechanical Systems (MEMS),^[8] animatronics,^[9] or grippers for soft robotics.^[10] The forecasting of the behavior for such a design is non-trivial and requires analysis and computation by non-linear models. The two major models are the Pseudo Rigid Body Model (PRB)^[11] by Larry L. Howell and the Beam Constraint Model (BCM)^[12] from Shorya Awartar, both of which are developed with the intention of incorporating the effects of load stiffening, kinematic and elastokinematic effects while still accessible enough to be used as design tools. This paper incorporates the Chained Beam Constraint Model (CBCM) which is a further improved BCM model proposed by Chen et al.^[13] To add another degree of functionality, we investigate how one can modulate bi-stable behavior, turning it on or off. The conception of switching between bi-stable and mono-stable states was achieved by active materials, which are well known in the fields of robotics, sensors and autonomous processes.^[14–16] Here, a thermally responsive shape-memory polymer (SMP) was chosen, as they can shift between preprogrammed and original geometries under certain temperatures governed by melting transition.^[17, 18] It is the combination of geometrical features, the shape of the lobes, and material properties, the displacement of water that have enabled the Venus flytrap to develop its “snap-through” mechanism. Similarly, by combining compliant mechanisms and the shape-memory effect

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Figure 1. (a) The closing “snap-through” mechanism of the Venus flytrap. (i) In its dehydrated state the Venus flytrap is unlikely to snap when stimulated. (ii) When hydrated it is in its activated state, “ready-to-snap”, and upon stimulating its inner spikes (iii) a rapid displacement of water leads to a configuration change of the lobes from concave to convex and thus shuts close. (Photos with permission of Izabella Bedő, Pexels GmbH.) (b) 3D-printed master molds for PDMS mold creation. (i) and (ii) are combined and casted with PDMS to create the top PDMS mold in c(i). Subsequently a casting step is performed with c(i) to create the negative mold c(ii) by PDMS. By combining the PDMS molds c(i) and c(ii) with a PEVA film through hot embossing the final structured cPEVA structure is manufactured (d). The mechanical testing setup (e) shows how the cPEVA films are tested. Here, 3D-printed heat-resistant parts were printed which are able to sustain temperatures up to 200°C shown in orange. The red-dashed line in e(i) shows the plane cut for the cross-section in e(ii). The cPEVA film is clamped down by two opposing plates with a hole in the shape of the truncated tetrahedron. Two magnets on the top and bottom of the cPEVA film respectively couples the mechanical probe and the sample, as can be seen in cross-section in e(ii) indicated by the black arrows.

(SME) we hypothesized that we could create the same effect controlled by temperature instead of humidity.

We explore the design, realization, and characterization of switching between bi-stable states on and off in 3D-structured films prepared from cross-linked poly[ethylene-*co*-(vinyl acetate)] (cPEVA).^[19] A truncated tetrahedron structure was designed and simulated to predict bi-stable or mono-stable states by varying geometrical parameters. The predicted structures were created through a two-step manufacturing method: (i) master molds were 3D-printed to cast the PDMS molds and (ii) the cPEVA films were created through hot embossing in the PDMS molds. The programming of the cPEVA film was achieved by first compressing the film between the PDMS molds of the targeted shape at the melting temperature and then fixing the temporary shape by cooling to a low temperature.

Experimental details

Modelling

Modelling was carried out using the equations for the Chained Beam Constraint Model (CBCM) as outlined in the previously reported publication by Chen et al.^[20] The relevant equations are outlined in the supporting info as a supporting method. Each beam was subdivided into 15 elements. The solution of the non-linear models were carried out through Python 3.8 using the packages NumPy, SciPy, Pandas, and Matplotlib as part of the Anaconda Distribution (Anaconda, Inc., Austin, USA). The Young's modulus, E , used for the modelling was set to 40 MPa (Storage modulus $E' = 44$ MPa, and loss modulus $E'' = 6.4$ MPa at 25°C as reported by Liu et al.^[21]).

Materials

Poly[ethylene-*co*-(vinyl acetate)] with a vinyl acetate content of 18 wt% (PEVA18) was obtained from DuPont de Nemours (Neu-Isenburg, Germany). Crosslinking agent dicumyl peroxide (DCP) was purchased from Sigma-Aldrich Chemie GmbH (Taufkirchen, Germany) and Sylgard™ 184 silicone elastomer kit from Dow Corning Corp. (Midland, USA). All chemicals were used as received. The 3D printing materials used are VeroWhite™ and HighTemp™ (Stratasys, Rehovot, Israel) and printed with the Stratasys Connex 3 Objet 260 Polyjet printer (Stratasys, Rehovot, Israel).

Preparation of 3D films

The master molds to create the PDMS molds were first designed in Autodesk Inventor (Autodesk, San Rafael, USA) and printed with VeroWhite™ [See Fig. 1(b)] using the Stratasys Connex 3 Objet 260 Polyjet printer (Stratasys, Rehovot, Israel). To ensure good demolding Ease Release™ 200 (Smooth-On, Inc., Macungie, USA) was applied before PDMS casting. The PDMS soft molds were then replicated from the 3D-printed master molds [see Fig. 1(c)] and synthesized from a precursor mixture of 90 wt% prepolymer sylgard 184 and 10 wt% curing

agent by polymerizing at 40°C for 24 h. The PDMS molds were further cured at 80°C for 2 h after removed from the 3D-printed master molds. The cPEVA precursors were prepared by mixing 196 g PEVA18 and 4 g DCP in a twin-screw extruder (Euro-PrismLab, Thermo Fisher Scientific) at 110°C and 50 rpm. The blends were compression molded into films with a 0.5 mm thickness and ready for further molding and crosslinking into defined 3D structures. Finally, for synthesizing the cPEVA 3D films, the PEVA blended films were sandwiched between the pair of PDMS molds and clamped between two metal plates, which was heated in the oven at 200°C for one hour. Then, the resulted 3D film [Fig. 1(d)] was separated from the molds after cooling.

Mechanical testing

To characterize the bi-stable behavior of the cPEVA 3D film, a Zwick Z1.0 tensile test machine for mechanical testing (Zwick, Ulm, Germany) equipped with a thermo-chamber and temperature controller (Eurotherm Regler, Limburg, Germany) was used. For testing the structure a custom-made test jig was produced [Fig. 1(e)]. The components of the jig were printed with high temperature resistant material (HighTemp™, Stratasys, Rehovot, Israel) using Stratasys Connex 3 Objet 260 Polyjet printer (Stratasys, Rehovot, Israel). These parts are heat resistant up to 230°C (Heat deflection of 0.45 MPa at 230°C), which was sufficient for our testing. Here, two flat plates having triangle holes with side length of 30 mm were designed to clamp and fix the film, which only allows freedom at the location of the truncated tetrahedron structures. Ample space was made below the film to allow the truncated tetrahedron to deform. To apply force on top of the truncated tetrahedron, a cylindrical probe with a diameter of 5 mm was designed and clamped to the top clamp. A pair of magnets, one fixed at the end of probe and the other touching the underside of the truncated tetrahedron, was used to keep the probe in contact with the sample to track the force during on- and off-loading. The force curves of compression on the truncated tetrahedrons of original, programmed and recovered samples were measured at room temperature. The recovery of the programmed sample was achieved by heating to 100°C in the chamber and cooled down to room temperature before measurements.

Optical microscopy

The optical microscopy of the cPEVA 3D film was measured by DVM6 microscope (Leica Microsystems, Wetzlar, Germany) with the objective PLANAPO FOV 43.75. Each sample was measured by $\times 50$ magnification with 20 to 48 pictures. The Z dimension was stacked with 14 to 20 steps. The 3D image was integrated and the critical angles of the pyramid structures at three sides were analyzed by software Leica Applications Suite X (LASX, Version 3.0.14). Each side was measured at three positions.

Programming and fixing

The programming was achieved by compression between PDMS molds with the targeted angle. The cPEVA 3D films were aligned with the pyramid molds and sandwiched between two metal plates and compressed with four clamps. The whole device was put in the oven at 70°C for 1 hour during the programming. Finally, the programmed sample was accomplished by cooling in the fridge at 5°C for 1 hour before being removed from the molds.

Results and discussion

For a typical bi-stable structure, the definition as hinted in the name is that it has two stable positions, i.e., two stable equilibrium points. At these stable points, the structure will remain unchanged until further acted upon. Figure 2(a) shows the typical behavior of a mono-stable and a bi-stable structure. The requirement for a bi-stable structure is that at some point in the force–displacement curve the force reaches zero at some displacement $d > 0$. In a mono-stable structure this does not occur. The critical forces acting on the structure switching between the two stable points in a bi-stable structure are

called the *snap-through force* ($F_{\max}$) and *snap-back force* ($F_{\min}$), respectively.^[20] As the force F acting upon the structure reaches the critical value $F_{\max}$ the structure will “snap” to the second stable position, as this is more energetically favorable. Similarly, this occurs when switching back to the original position with $F_{\min}$. These structures are mainly fabricated as 2D-planar structures.^[8, 22–24] Here, we create a 3D structure with modulated bi-stability, i.e. a structure that could switch from being mono- to bi-stable. We devised a stable structure in CAD, which could later be evaluated with a mechanical tester. The design was settled on a truncated tetrahedron, a “pyramid-like” structure [Fig. 1(d)]. The truncation allows for an area load, which is more stable than a point load. Here, we left it open to see what slope angle θ of the sides of the tetrahedron would lead to a mono-stable and bi-stable structure. The side of the tetrahedron has a “sigmoid” like shape and could be modelled as a beam with a fixed and a free end. This allows the structure to be modelled with non-linear beam models. The previously reported Chained Beam Constraint Model (CBCM) was implemented.^[13] The computational model predicted that a slope angle θ of 30° and 45° would show a mono- and bi-stable behavior, respectively [Fig. 2(b) and Supplementary videos S1

Figure 2. (a) A typical mechanical bi-stability diagram showing the static equilibrium curve (black) and dynamic equilibrium curve (purple). Here, three equilibria can be identified (black points) but only where the static and dynamic equilibrium passes through are stable; i.e. only two. The areas in which the structure would return to an equilibrium point is indicated in the areas A (blue) and B (green). The third equilibrium in the middle is unstable and will fall back into a stable equilibrium if only a slight disturbance occurs. (b) Modelled behavior using the CBCM model for slope angles θ of 30° (blue) and 45° (red). The structure for a truncated tetrahedron with a slope angle θ of 45° shows bi-stable behavior. (c–f) Photos of prepared cPEVA 3D films with pyramid slope angles of 30° (c, d) and 45° (e, f) from top view (c, e) and side view (d, f). The scale bar in each picture is 10 mm.

and S2]. In order to, in the same structure, switch between a mono-stable and a bi-stable structure we needed an active material. We show here how the use of an SMP can achieve this. To fabricate these structures we decided to cast cPEVA using PDMS molds instead of directly using 3D-printed molds due to three considerations. First, the PDMS mold is stable at this high curing temperature of cPEVA (200°C), compared to the 3D-printed mold made from VeroWhite™, which has a low heat deflection temperature (HDT) of 60°C. Second, the PDMS has low surface potential and is chemically inert to the crosslinking reaction of PEVA, while the 3D-printing material is more reactive to the polymerization of PEVA. Third, the PDMS mold is more elastic than the rigid 3D-printed mold and the elasticity is helpful to peel the mold off from the structured cPEVA films. Therefore, a pair of 3D-printed master molds was created with the design of truncated tetrahedron structures, as shown in Figure S1a, b and a pair of PDMS molds

(Fig. S1c,d) were replicated from the 3D-printed counterparts. Then, the cPEVA films with truncated tetrahedron structures [Fig. 2(c)–(f)] were successfully fabricated by soft lithography approach using PDMS molds (Fig. S1e,f). The slope angle of the truncated tetrahedrons were measured by an optical microscope and calculated to be $29.2^\circ \pm 3.3^\circ$ for the 30° tetrahedron and $39.3^\circ \pm 1.3^\circ$ for the 45° tetrahedron. The results prove a success of the molding approach and reveal a slightly higher difference for the 45° tetrahedron to the realized sample.

To mechanically test these films, a custom-made test jig was created from high-temperature-resistant material for insertion into a mechanical tester [Figs. 1(ei), 3(a)]. To measure the force throughout the displacement of the structure two magnets were used to couple the mechanical probe to the cPEVA film; one affixed onto the probe and one underneath the cPEVA film at the truncation of the tetrahedron [Figs. 1(eii), 3(b)–(d)]. From the mechanical testing it was

Figure 3. (a) Photos of 3D-printed parts used for mechanical test. (b) Photos of assembled self-build device for bi-stable test (c-d). Photos of self-build device working in the tensile test machine. Force-displacement curve of (e) 30° and (f) 45° truncated tetrahedron samples. CBCM model prediction (blue dashed line); mechanical test curve (orange). The black arrows indicate on- and off-loading directions. Areas A (blue) and B (green) indicate stable domains for each construct.

confirmed that the 30° truncated tetrahedron sample showed a mono-stable behavior and that the 45° truncated tetrahedron sample showed a bi-stable behavior, and was in good agreement with the predicted models [Fig. 3(e), (f)]. Additionally, there was some hysteresis present comparing the on- and off-loading curves, due to the viscoelastic nature of cPEVA. Therefore, the *snap-through force* was measured from the on-loading curve, and the *snap-back force* was measured from the off-loading curve. From the evaluation, it is also apparent that the 45° sample required a greater *snap-through force* (0.90 ± 0.13 N, $n=3$) than the 30° sample (0.41 ± 0.19 N, $n=3$), and that the *snap-back force* for the 30° was positive (0.06 ± 0.08 N, $n=3$) and negative for the 45° sample (-0.29 ± 0.08 N, $n=3$). This granted us to move on to the

next step to create a structure that could switch between mono- and bi-stable modes.

Here, a structured cPEVA film with 30° truncated tetrahedron was programmed by compression between PDMS mold with 45° truncated tetrahedron structure at 70°C and fixation at 5°C, as shown in Fig. 4(a). At room temperature the programmed sample showed a slope angle of $42.6 \pm 0.9^\circ$ and when the temperature was increased to 100°C the slope angle recovered back to $27.4 \pm 0.8^\circ$, as confirmed by the optical microscopy [Fig. 4(b)–(f)]. To confirm the switching between a mono- and bi-stable structure the programmed sample was tested at room temperature, then heated up to 100°C and cooled down to room temperature to be tested again [Fig. 4(g)]. From the results it is apparent that we were able to switch between these two modes, and that the sample

Figure 4. (a) Schematic showing of programming of the structured cPEVA film using PDMS molds. (b–e) 3D stacking optical images of cPEVA 3D films in the programmed state (b, d) and recovered state (c, e). The scale bar in each picture is 10 mm. The blue, red and green lines are the markers of positions where the cross-sections were measured; (f) The recovery curve and slope angles with increasing temperatures from 30 to 100°C; (g) Axial force vs. Displacement curve of programmed and recovered truncated tetrahedron sample.

in its programmed state showed a larger force than in its recovered state, i.e. amplification. Finally, we further test the reconfiguration behavior of the programmed sample starting from the second stable state by first compressing on the truncated tetrahedron structure to the other side. Then, when the temperature increased to 80°C, the programmed sample recovered from bi-stable to mono-stable. The losing of bi-stability introduced a snap movement on the structure, as shown in supplementary video S3. By combining the SME and bi-stable property, a shape recovery with higher magnification was achieved.

This mechanism could be thought of as being implemented as a safety switch, which turns off the moment a critical temperature has been reached—and will remain off until reset. It could also be thought of as a dynamic energy storage in which a structure is activated by a mechanical force and the stored energy is released by an increase in temperature. cPEVA is a relatively soft polymer with a Young's modulus of $E \approx 40$ MPa at room temperature, which shows some viscoelastic behavior.^[21] This explains the hysteresis in the on- and off-loading curves during mechanical testing. As an outlook, it may be an interesting development to make use of the viscoelastic behavior, which would add another dimension of functionality of the structures in terms of time. Here, one could devise a structure with a highly viscoelastic behavior that is bi-stable only if held at a certain force for a given amount of time, and for transient forces it is mono-stable.

Conclusion

In this paper, digital methods for design and manufacturing played a key role in the technical realization. We report on the implementation of modelling of beams using the CBCM model as an important tool in the computational design process for compliant mechanisms. It was shown that a truncated tetrahedron structure with a slope angle of 30° showed no mono-stable behavior, and such a structure with a slope angle of 45° did show bi-stable behavior. Furthermore, it successfully realized to switch between these two structures using cPEVA as an SMP. The modulation of compliant mechanisms by the SME of a polymer is an example for a bio-inspired function, which is derived from the switch-like mechanism of the Venus flytrap.

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Data availability

The datasets generated during and/or analysed during the current study are available from the corresponding author on reasonable request.

Declarations

Conflict of interest

The authors have no relevant financial or non-financial interests to disclose.

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Supplementary Information

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