

# Cactus-inspired design principles for soft robotics based on 3D printed hydrogel-elastomer systems

Anil K. Bastola<sup>a</sup>, Nadia Rodriguez<sup>a</sup>, Marc Behl<sup>a</sup>, Patricia Soffiatti<sup>c</sup>, Nick P. Rowe<sup>d</sup>, Andreas Lendlein<sup>a,b,\*</sup>

<sup>a</sup> Institute of Active Polymers and Berlin-Brandenburg Centre for Regenerative Therapies (BCRT), Helmholtz-Zentrum Geesthacht, Kantstr. 55, 14513 Teltow, Germany

<sup>b</sup> Institute of Chemistry, University of Potsdam, Karl-Liebknecht-Str. 24-25, 14476 Potsdam, Germany

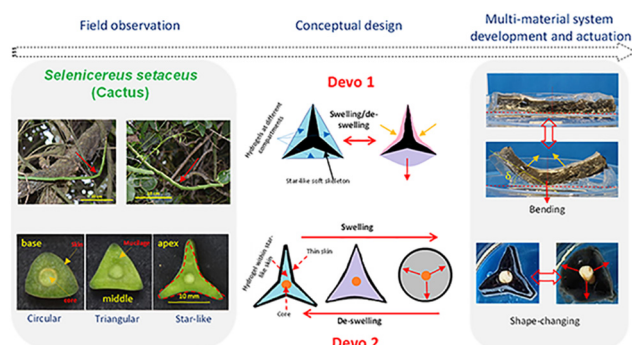
<sup>c</sup> Department of Botany, Federal University of Paraná State, Curitiba, PR, Brazil

<sup>d</sup> AMAP, Univ Montpellier, CIRAD, CNRS, INRAE, IRD, Montpellier, France

## HIGHLIGHTS

- Inspired by functional traits of a climbing cactus, two developmental designs (D1 and D2) are presented
- In D1, hydrogel-elastomer system is presented, which aims at a geometrical configuration similar to the natural cactus
- In D2, multi-material system is presented, which aims at a similar morphology and physical behaviour compared to the natural cactus
- D1 provided a controlled bending whereas D2 was capable of a shape-change from star to circular shape upon swelling/de-swelling of the hydrogel

## GRAPHICAL ABSTRACT



## ARTICLE INFO

### Article history:

Received 30 November 2020

Received in revised form 21 January 2021

Accepted 21 January 2021

Available online 28 January 2021

### Keywords:

Cactus-inspired  
Hydrogel-elastomer  
Multi-material  
3D printing  
Soft-robotics

## ABSTRACT

Plants have evolved many capabilities to anchor, position their stems and leaves favourably, and adapt themselves to different environmental conditions by virtue of growing. *Selenicereus setaceus* is a cactus and is an impressive example of a climbing plant found mostly in the Atlantic forest formations of southern Brazil. This cactus displays striking changes in stem geometry along different stages of growth: older parts are circular while the younger parts are star-like in shape. Such a transformation in shape optimizes its flexural rigidity and allows the cactus to search in three-dimensionally complex environments. Its organisation offers novel schemes for the design of plant-inspired soft robotic systems. In this paper, we have created multi-material systems for soft robotics that display controlled movements as well as mimicking the cactus stem geometries from star-like to circular. The unique star-shaped geometry is 3D printed using a soft elastomer and hydrogel is used as an actuating component. Through anisotropic swelling, the hydrogel-elastomer system adjusts its configuration and shows a controlled movement. Furthermore, the isotropic swelling of the hydrogel of the artificial cactus multi-material system result in the change in shape from star-like to circular as the cactus does naturally in the tropical forest.

© 2021 Helmholtz-Zentrum Geesthacht. Published by Elsevier Ltd. This is an open access article under the CC BY license (<http://creativecommons.org/licenses/by/4.0/>).

\* Corresponding author at: Institute of Active Polymers, Helmholtz-Zentrum Geesthacht, Kantstr. 55, 14513 Teltow, Germany.

E-mail address: [andreas.lendlein@hzg.de](mailto:andreas.lendlein@hzg.de) (A. Lendlein).

## 1. Introduction

Nature has extensively served as a great source of inspiration for humans to design and develop innovative technologies. Plants are increasingly one of the great sources of inspiration. In nature, climbing plants display several different kinds of functional traits by virtue of their growth processes such as anchoring, vining, twining, coiling, attaching, and searching. Such adaptive traits offer a huge potential for novel schemes for the design of new adaptive plant-inspired robotic systems. In recent years the soft-robotics community has been therefore highly interested in the subject and is now focused on innovative plant-inspired robots [1–6]. Plant-inspired robotic systems consider how plants perform and adapt their growth as well as how they vary biomechanical properties (stiffness and rigidity) to anchor, attach, and climb. A few examples of the bio-inspired soft robotic investigations based on plants include artificial growth and movement like plant stems [7], tendrils [8], tendril and searcher like structures [9,10], and roots [11,12]. Moreover, several other studies on the development of plant-inspired actuation systems are available in the literature. For example, a lotus effect-inspired flexible and breathable membrane [13], a soft robotic gripper inspired by the cabbage plant [14], a plant-inspired soft pneumatic eversion robot [15], a soft tendril-inspired gripper [16], plant-inspired soft bistable structures [17] and reversible shape-changes of the cone seed scales of the Bhutan pine (*Pinus wallichiana*) [18].

Cacti are recognized for their amazing adaptations to dry and seasonally-dry conditions [19]. Recently, Soffiatti and Rowe [20] explored the functional behaviour of a climbing cactus, *Selenicereus setaceus*, found in the dry lowland forests of southern Brazil. This cactus displays several interesting functional traits, for example, climbing through unstructured habitats, attaching to highly variable substrates as well as changing the shape of the stem to optimize rigidity. Therefore, with such an incredible variety of functional traits, the cactus offers highly innovative schemes for the design of plant-inspired robotic systems. A number of different mechanisms and potential applications to implement the functional traits of this cactus for the design and development of plant-inspired robots have been exhaustively discussed [20]. One of the interesting functional traits of the cactus is that the stem displays highly variable shape changes along the different stages of growth; the cross-sectional geometry of lower, older stems is circular in cross-section, the later-formed, mostly climbing stems are triangular and the youngest searcher stems that reach to new supports are star-shaped in cross-section, see Fig. 1(a). Such modifications in the cross-sectional geometry of the cactus stem optimizes the flexural rigidity of the searcher stem in the harsh environment conditions (e.g. strong wind and rain) in forest to scrub habitats and is particularly important for young stems that have not yet developed a woody core by secondary growth.

In recent years, 3D/4D printing methods have attracted a huge amount of interest to develop soft robots with sophisticated capabilities, such as jumping, complex 3D movements, gripping, releasing etc. [21]. 3D printing is a digital manufacturing process which uses a layer-by-layer technique to create a 3D object directly from the computer added design (CAD) [14,18,21,22]. Therefore, 3D printing would be the best choice in order to realize intricate geometries and interesting multi-material systems offered by plants for the plant-inspired soft robotics. A few revolutionary achievements in soft robotics using 3D printing are a high-force soft actuator [23], biomimetic hydrogel composites [24], jumping robot powered by combustion [25], highly flexible elastomeric gripper [26], growing robot [27], soft micro-actuators for soft microrobots [28], untethered and fast performing soft-robots [29], millimeter-scale flexible robots [30] and shape transforming magnetic soft materials [31]. Furthermore, in recent years, various computational studies and mathematical modelling have also been devoted to investigate and understand the interesting biological observations of plants such as a plant-inspired growth-driven system [32], growth-induced

deformations [33], biophysical mechanism [34], plant tropism [35–37] and growth control of the plant-inspired soft robot [38]. Such mathematical studies provide an in-depth understanding of various functional behaviours of plants like bending, twisting, coiling, twining etc. In addition, the theoretical studies also provide framework for investigation and understanding the plant-inspired systems for the development of novel plant-inspired robots e.g. growing robots.

To this end, the aim of this study is to develop a multi-material system inspired by the functional traits of *Selenicereus setaceus*. The first aim is to design and create a soft actuator with a controlled movement inspired by the interesting structural configuration and behaviour of the searcher component of the cactus (star-like apex); the second aim is to mimic the changes in the geometrical structure of the cactus at the different stages of the growth (circular, triangular, and star-shaped).

## 2. Scientific question/hypothesis and concept

The highly modified geometrical configuration of the cactus at the searcher apex provides an extremely novel design scheme to create functional artificial systems. The soft tissue of the cactus represents the main structure at this stage and is one of the crucial components; it is like a hydrogel (a water-swollen polymer). Therefore, we can directly learn from this cactus to design and create novel and functional artificial systems involving hydrogel-like tissue combined with other structural elements of the stem. We hypothesize that a synthetic polymer and a hydrogel can be used to not only mimic the changes in geometry but also control the movement of an artificial system. One of the key questions for the development of artificial functional systems is the use of energy. Here, we propose a minimal or near zero external energy supply to develop an actuating system from just swelling and deswelling of the hydrogel within a cactus-inspired robotic system. The complex geometrical configuration offered by the cactus will be realized by taking the advantage of a disruptive digital manufacturing technology, widely known as 3D printing.

Two developmental designs are considered and are directly inspired by the functional traits of the cactus as presented in Fig. 1.

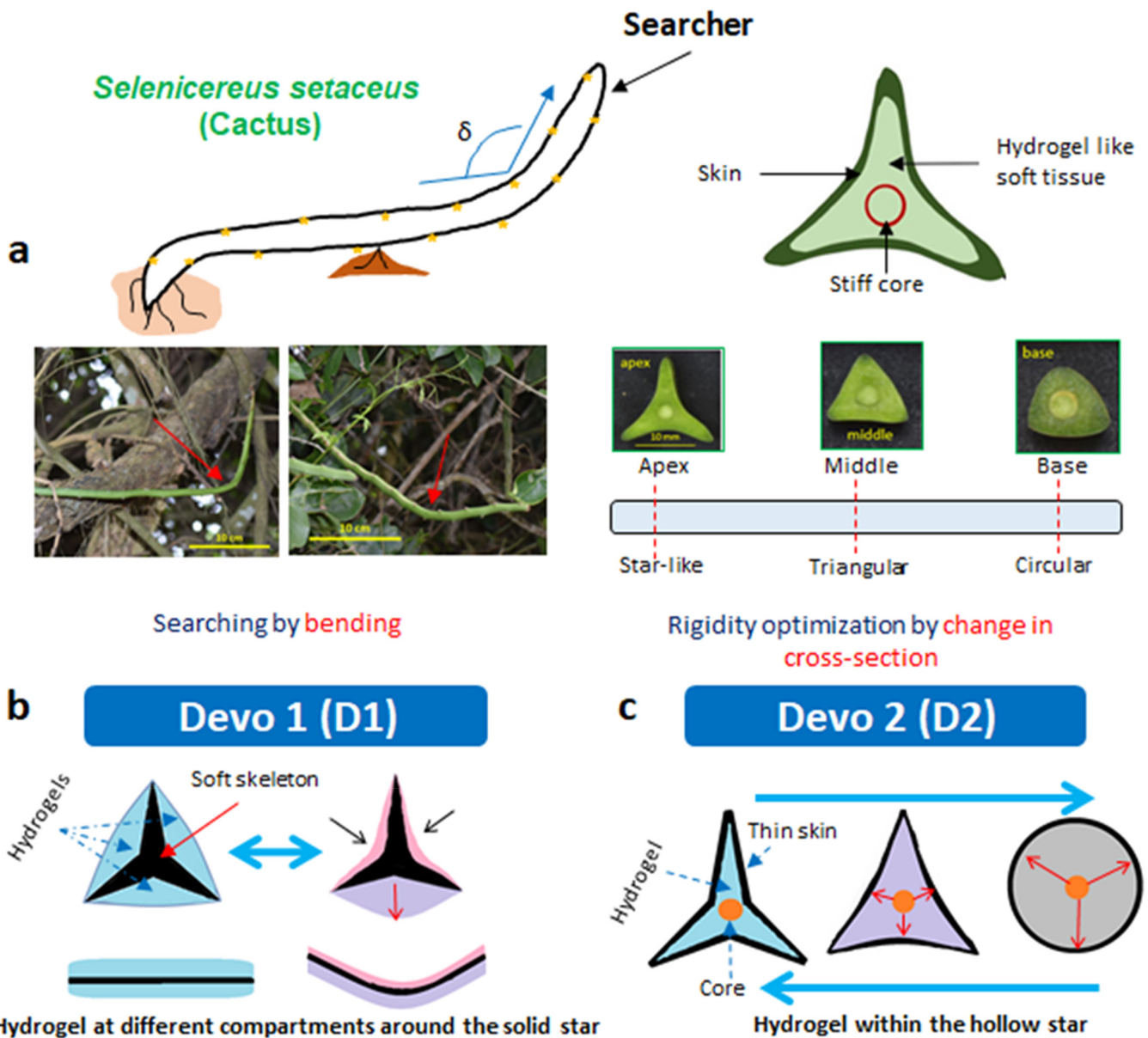
Development 1 (D1) targets a controlled movement of a star-shaped searcher of the natural cactus in the presence of external stimuli available in nature, such as sunlight or humidity. Significant changes in the angle of young star-shaped stems is commonly seen in natural conditions and effectively allow the plant to steer towards optimal conditions of light and/or supports to climb on (Fig. 1(a)).

Development 2 (D2) targets a change in stem cross-sectional geometry as the natural cactus does by means of growing from circular at the base of the plant to star-shaped near the searching apical branches (Fig. 1(a)).

### 2.1. Development 1

The core concept of D1 is to develop a soft robotic system inspired by the shape and structure of the young “searcher” component of the cactus. A multi-material system can be developed with a star-shaped cross-section made up of a soft polymeric elastomer representing the “skeleton” surrounded by hydrogels (Fig. 1(b)). In this model, the elastomer skeleton, therefore, represents the soft star-shaped body of the young cactus stem and the outer hydrogel layer an additional one necessary for actuation. This organisation differs from the true biological organisation where the hydrogel-like tissue is actually incorporated within the main body and covered by the skin, rather than on the outside. The skeleton is surrounded by hydrogels in the three different compartments of the unique geometry, see Fig. 1(b). Anisotropic swelling and de-swelling of the hydrogels in the three different compartments are expected to provide controlled movement.

Development of the star-shaped skeleton geometry is the first challenge. 3D printing is a fabrication method where the structures are constructed in a layer by layer fashion. It is a fast and precise manufacturing



**Fig. 1.** Design scheme of the cactus-inspired multi-material system. Structural configuration of the natural cactus and two kinds of developmental design: Development 1 (Devo 1) and Development 2 (Devo 2). (a) Structural configuration and graphics depicting the functional traits of the cactus such as bending and change in cross-sectional geometry of the stem. (b) Design concept of Devo 1: anisotropic swelling of the hydrogel at different compartments of the star-like soft skeleton to induce the controlled movement of the hydrogel-elastomer system. (c) Design concept of Devo 2: isotropic swelling of the hydrogel within the elastomeric skin to mimic the different growth stages of the cactus.

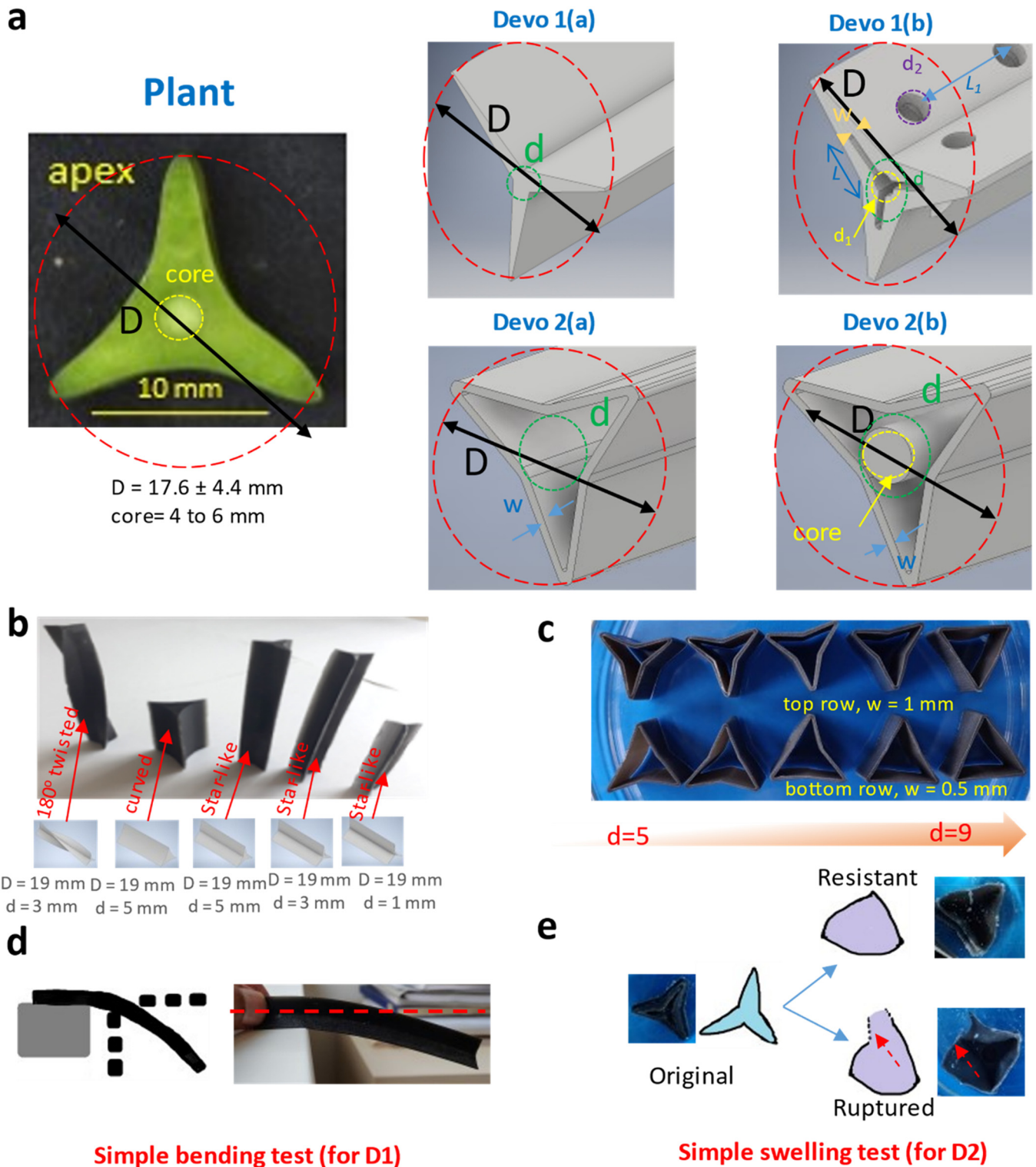
process, suitable for producing highly customizable products using a wide range of materials [22,39–41]. We, therefore, employed a 3D printing technique to develop the star-shaped “skeleton” inspired by this climbing cactus. The next challenge was to successfully synthesize the hydrogel on the skeleton. Peeling tests (mechanical observations) followed by FTIR spectrum analysis (chemical observation) were used to investigate the attachment of the hydrogel to the elastomer surface. Here, the hydrogels will be allowed to swell and de-swell outside of the elastomeric skeleton (star-shaped) in an anisotropic manner to realize the controlled movement similar to the searcher of natural cactus.

For the Devo 1 design, two types of the skeleton were considered for representing different developmental organisations and potential behaviours of the cactus, see Fig. 2(a). Devo 1(a) represents a relatively simple or (biologically younger) development organisation, where vascular conduits are not yet highly developed and fast water transport would be limited across the plant body. Devo 1(b) represents a more

complex (biologically older) development where vascular conduits would be more developed allowing faster more general water conduction throughout the plant body. The 3D printed skeleton of Devo 1(b) therefore contains a central axial canal and lateral perforations permit movement of water through the skeleton and adjoining compartments of hydrogel (Fig. 2(a)). Consequently, the multi-material soft robotic objects with asymmetrical (Devo 1(a)) and symmetrical structure (Devo 1(b)) can be realized and are analogous to the different biological phases of development in the cactus.

## 2.2. Development 2

The primary notion of Developmental design 2 (Devo 2) is to learn directly from the structural configurations at different stages of growth of the cactus and to develop a multi-material system that potentially displays similar performances. Devo 2 possesses a more complex design



**Fig. 2.** Design and development of the two developmental cactus inspired artificial systems. (a) CAD design and control parameters for the devo 1 and devo 2 compared with natural system. (b) Prototypes of the devo 1. (c) Prototypes of the devo 2. (d) Facile method to optimize the design parameters for devo 1. (e) Facile method to optimize the design parameters for devo 2. Variation of the design control parameters: (1) for devo 1(a)  $D = 19$  mm,  $d = 1-5$  mm, (2) for devo 1(b)  $D = 19$  mm,  $d = 1-5$  mm,  $d_1 = 2-4$  mm,  $d_2 = 0.5-2.5$  mm,  $L = 0.5-2.5$ ,  $L_1 = 0.5-2.5$  mm (3) devo 2(a)  $D = 19$  mm,  $d = 5-9$  mm,  $w = 0.5-1$  mm and (4) for devo 2(b)  $D = 19$  mm,  $d = 5-9$  mm,  $w = 0.5-1$  mm and core = 4 mm.

compared to Devo 1 as the target is to develop a similar artificial cactus at an older stage of growth compared to the star-shaped stage, which includes a stiff central core representing the woody cylinder of the cactus surrounded by a layer of hydrogel, see Fig. 1(c). As illustrated in Fig. 2(a), again, Devo 2 also consists of two designs, Devo 2(a) is a

simpler system which only depicts the star-like shape and Devo 2 (b) is a complex system which has a similar physical arrangement like the natural cactus. The artificial cactus stem consists of a 3D printed thin elastomeric layer in a star-like shape (apex of the natural cactus) which forms the outer part of a sandwich structure enclosing the soft

hydrogel (for Devo 2(a)) and the hydrogel and PU foam inner core at the centre (for Devo 2(b)). The thin elastomer layer is the representation of the outer cactus skin, the hydrogel is a representation of soft cortical tissue of the cactus, and PU foam core is a representation of the cylinder of wood surrounding a soft pith of the natural cactus. Both the elastomer and PU foam are passive components of the structure whereas the bulky hydrogel layer is the active component of the artificial cactus. Here, the sandwiched hydrogel will be allowed to swell in a uniform manner, and it is expected that the shape of the artificial cactus will be changed from star-shape to circular by swelling and vice-versa by de-swelling. One of the aims of this device is that no external energy will be used to modulate the change in shape.

### 3. Experimental

#### 3.1. Materials

Acrylic acid (AA) (purity 99%), *N, N'*-methylene bisacrylamide (MBA) (purity 99%), and Ammonium Persulfate (purity >98%) were obtained from Sigma–Aldrich (Taufkirchen, Germany) and used as received. TangoPlus was used as ink material and SUP706 was used as support material for the 3D printer (Object260, Stratasys, Eden Prairie, MN, USA). Polyurethane (PU) foams were prepared by using the mix and pour expanding foam (FOAM-iT™ 3, Smooth-On, Inc. Macungie, PA, USA).

A soft elastomeric skeleton is desired to realize the movement of the multi-material system. We employed Objet 260 Connex 3™ (Stratasys, USA) 3D printer to develop the skeleton. This 3D printer provides the capability to develop a wide range of materials from soft (TangoPlus) to rigid (VeroClear). Soft elastomer TangoPlus is selected due to its softness (28–28 Shore A hardness) and elasticity (elongation at break 170–220%) [42]. TangoPlus did not readily swell in water, however, swelling behaviour in other solvents was not studied. The water jet can remove the support materials. The elastic properties of hydrogels can be controlled by regulating the cross-linker amount. It was found that the higher cross-linker resulted in the stiff hydrogel, but such hydrogels broke into pieces after swelling. Therefore, to realize the actuation, a soft and elastic hydrogel is desired, before and even after the swelling. The details about the chemical structure and swelling of hydrogels with different cross-linker amounts are provided in the Supplement data, Fig. S1.

#### 3.2. Method

First, we investigated the biological variation and components of the natural cactus at different stages of growth. *Selenicereus setaceus* displays an amazing range of cross-sectional shapes from circular at the base, triangular in the middle, to star-shaped at the apex, depending on stages of growth. The overall diameter of the stem increases from round to triangle to star-shaped from 12.8 mm ± 3.4 mm to 11.5 mm ± 2.7 mm to 17.6 mm ± 4.4 mm, respectively. In addition, circular and triangular shapes are found in the forest while star-shaped is found at the end tip when the plant points out of the vegetation towards the light. The searcher of the cactus optimizes its flexural rigidity by adjusting its shape from triangular to star-like. The different compartments can be seen in the searcher of the cactus. The lobed cross-section of a star-like searcher, rather than a more regular cylindrical or triangular, offers a novel biological model for the development of the plant-inspired artificial system.

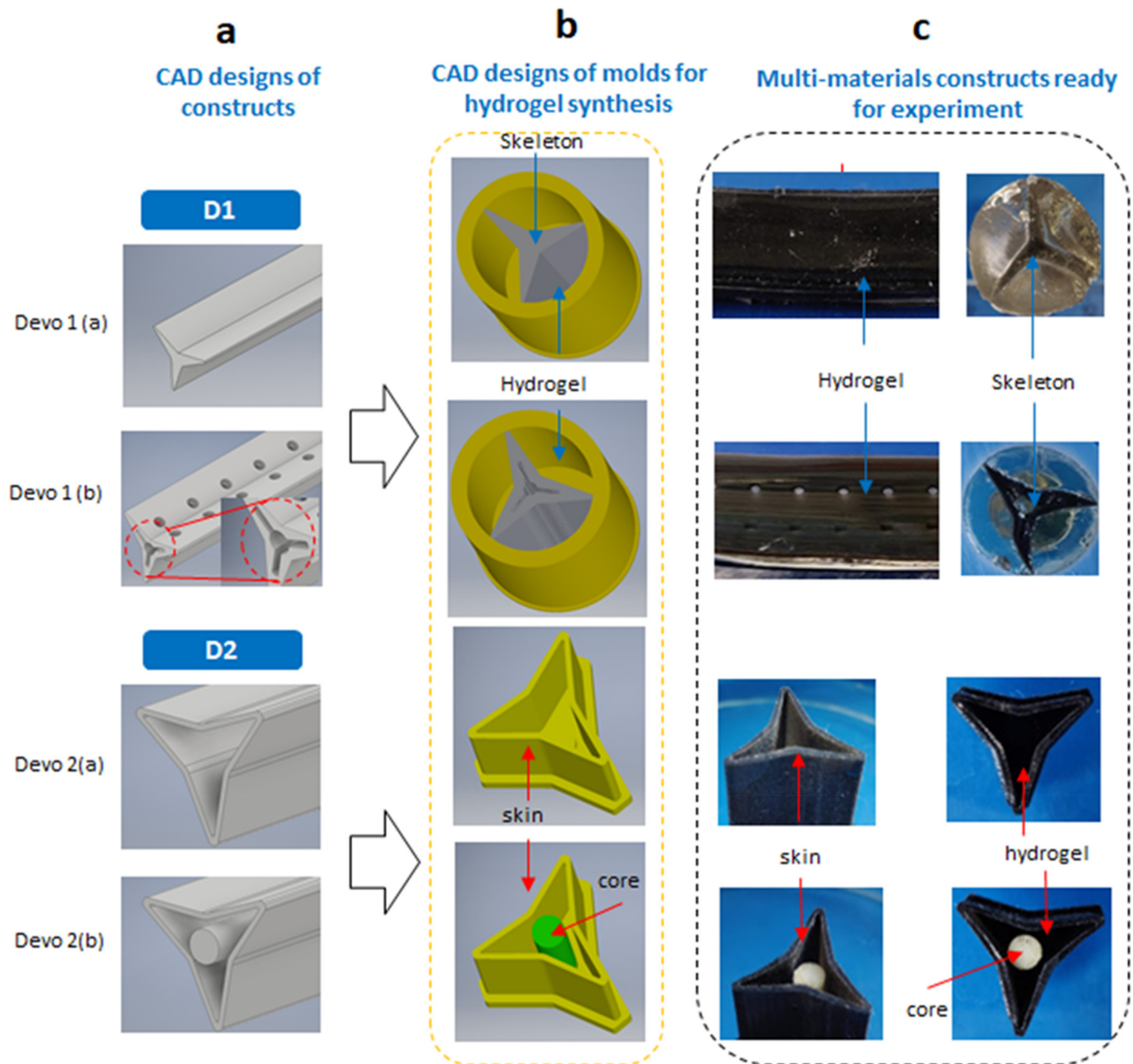
The old circular stem is composed of the three major components outer skin, cortex, and wooden core. The outer skin is a mix of lignified dead cells, thin-walled living cells, and irregularly thick-walled living cells. The cortex is a mix of large soft thin-walled cells (for photosynthesis and water storage functions) and scattered mucilage-producing cells. The wooden core consists of an inner layer: lignified cells (mechanical tissue) and the pith: soft thin-walled cells (water storage)

and scattered mucilage producing cells. The outer skin of the triangular middle part is composed of a thick waxy layer covering irregularly thick-walled living cells. The outer skin of the apex has the same composition as the skin of the triangular stem, but the skin of the star tip is reinforced with more layers of irregularly thick-walled cells. Cortex, and pith of the base, middle, and apex have the same composition. Such an incredible change in shape from circular to star-like shape is mostly governed by the positioning of the soft-tissues of the cactus stem. The soft tissue provides another simple biological model where soft and porous polymers can be used to realize the shape morphing structures directly learning from the cactus biological arrangement.

In order to create demonstrators for two developmental designs, the first Computer-Aided Design (CAD) models were generated and dimensions were optimized by rapid prototyping. For Devo 1, the solid star-like skeletons have been developed, there are two different designs in D1; Devo 1(a) and Devo 1(b). Devo 1 (a) is a solid and star-like skeleton and Devo 1(b) is a porous and also star-like skeleton. The rationale for Devo 1(b) was to mimic the actual plant in the upright condition where hydrogels could take up water through internal channels of the porous structures. The design control parameters for Devo 1(a) were outside the overall diameter (*D*) and internal diameter (*d*) of the star section. *D* was fixed as 19 mm, which is similar to the natural cactus, while, *d* was varied from 1 mm to 5 mm. Some prototypes of the solid skeleton are given in Fig. 2(b). A simple bending test was conducted to study the flexibility and length of the skeleton. It was observed that the larger *d* made a much stiffer skeleton while the smaller *d* made a weak skeleton, which easily collapsed and even could not hold the star-like shape, see Fig. 2(d). In this way, we found that the *d* equal to 3 mm and 4 mm were acceptable and the overall length of the skeleton should be at least 10 cm. Similarly, the major design control parameters for Devo 1(b) also remains the same as Devo 1(a). In Devo 1(b), channel configuration was important. The channel was configured in such a way that the water could be transported through the star-like channel in the middle of the skeleton and each compartment of the skeleton has openings connected to the central channel, see Fig. 2(a). The details of the design control parameters variation are given in Fig. 2 (caption). In D1, the hydrogel will have semi-confined swelling and drying at the different compartments of the star-shaped skeleton to realize the movement.

Before the development of the actual demonstrators, one of the key requirements is that the hydrogel must be firmly bonded to the elastomer. In order to observe the attachment of the hydrogel to the elastomer surface, a peeling test was conducted. For the peeling test, the hydrogel was bonded onto a rigid surface on one side and the 3D printed elastomer surface on the other side with the size of 100 × 15 × 3 mm (length × width × thickness). The samples were tested with the standard 180°-peeling test [43]. After choosing the compatible hydrogel, in order to develop the demonstrators for D1, the successful synthesis of the hydrogel at different compartments of the skeleton is the key step. The hydrogel was polymerized using a moulding technique, see Fig. 3. First, the 3D printed star-shaped skeleton was inserted into the mould. After that, hydrogel ink was injected into the mould. Finally, the mould was sealed and placed into a water bath or at an oven at 50 °C for the hydrogel to polymerize. After 24 h, the mould was removed, and the hydrogel-elastomer construct was obtained. The same way of hydrogel synthesis can be performed for both Devo 1(a) and Devo 1(b).

For D2, hollow star-like skins have been developed, again there are two designs in D2, Devo 2(a) and Devo 2(b). Devo 2(a) is hollow skin only while Devo 2(b) is hollow skin with a core. The rationale for Devo 2(b) is to provide the strength for the artificial system. Rigid PU foam could provide axial stiffness. The design control parameters for Devo 2(a) were outside the overall diameter (*D*) and internal diameter (*d*) of the thin star section and the wall thickness (*w*) of the skin. Again, *D* was fixed as 19 mm, which is similar to the natural cactus, while *d* was varied from 5 mm to 9 mm and *w* was varied from 0.5 mm to 1 mm. Some prototypes of thin star-like hollow skin are given in Fig. 2(d). It



**Fig. 3.** CAD designs and fabrication method for two developmental designs. (a) CAD designs of solid (Devo 1(a)), porous (Devo 1(b)) star-shaped skeletons and hollow thin star-shaped skin (Devo 2(a) and Devo 2(b)). (b) Hydrogel synthesis process at the different compartment of the solid skeleton (for D1) and within the hollow skin (for D2). (c) Multi-material hydrogel-elastomer constructs for D1 (upper two rows) and artificial cactus for D2 (lower two rows). Panel a is the collection of CAD models of the actuator constructs. Panel b is the collection of CAD models of the mould, where 3D printed constructs are inserted into the mould for hydrogel synthesis for Devo 1 and 3D printed constructs act as mould themselves for Devo 2. Panel c is the collection of the developed multi-material constructs ready for the actuation experiment.

was observed that if  $d$  was bigger than 8 mm the thin skin lost its star shape. Similarly, if the wall thickness was smaller than 0.5 mm the thin skin collapsed and could not hold the star-shape. In addition, a simple hydrogel swelling test was conducted to optimize wall thickness. In swelling, our aim was that the force of the swollen hydrogel should not break/tear the elastomer skin, see Fig. 2(e). After these observations,  $d = 7$  mm and  $w = 1$  mm hollow stars were found to be suitable. Similarly, the major design control parameters for Devo 2(b) also remains the same as Devo 2(a). In Devo 2(b), an extra component, the core was needed, the diameter of the core was 4 mm, which is similar to the plant.

After the optimization of the dimensions of the hollow star-shaped skin for D2, the hydrogel needed to be synthesized within the hollow

star. Again, the technique is moulding. Here, the 3D printed thin star-shape skin itself acted as a mould. First, hydrogel ink was injected into the star-like mould. It should be noted that for Devo 2(b), the PU foam cylindrical rod was inserted into the centre of the star-shaped hollow thin skin before pouring the hydrogel ink. After that, the mould is covered, and the hydrogel is allowed to polymerize at 50 °C. After 24 h, the hydrogel was fully polymerized, and the construct was ready for further use.

For the actuation experiments, for D1, the hydrogel elastomer construct was placed on the flat petri-disc (one compartment of the star-section on the bottom) and DI water was poured until the bottom half of the construct was covered. In this way, hydrogels on the lower section could swell and hydrogels on the upper section could dry.

Thereafter, a photo was taken every 15 min for at least 36 h. Finally, the photos were imported into a CAD software (Autodesk), converted into 2 D sketches from which the bending angles were determined (Supporting Data, Fig. S0). In addition time-lapse videos were prepared from the single pictures.

For the actuation for D2, the artificial cactus was fully submerged in a water bath and the hydrogel was allowed to swell at least for 48 h. Again, a photo was taken every 15 min and finally, photos were compiled to make a time-lapse video. For reverse effect, the same process was repeated by allowing hydrogel to dry, the swollen system was just allowed to dry at room temperature at 23 °C and again photos were taken every 15 min to generate a time-lapse video.

## 4. Results and discussion

### 4.1. Devo 1 properties

First, a soft elastomeric skeleton is required to enable movement of the multi-material system. TangoPlus is selected due to its softness (28–28 Shore A hardness), elasticity (elongation at break 170–220%) [42] and 3D printability for the development of skeletons. Afterward, the synthesis of the hydrogel on the 3D printed elastomeric skeleton is a crucial step. The fundamental requirement is that the hydrogel must be strongly bonded to the elastomer. The widely used method to examine the bonding characteristics of hydrogels to the elastomer is a peeling test [43]. Thus, to detect the attachment between hydrogel and elastomer, a peeling test is conducted. The results of the peeling test are given in Supplementary data Fig. S2 (a & b). Acrylic acid (AA) based hydrogel is found to be compatible with TangoPlus 3D printed elastomer as both of them (hydrogel and elastomer) are acrylic-based materials as marked by C=O bond at FTIR peaks near  $1700\text{ cm}^{-1}$  ( $1699\text{ cm}^{-1}$  for AA hydrogel and  $1716\text{ cm}^{-1}$  for TangoPlus, see Supplementary data Fig. S2 (c)). The steady-state peeling force per unit width ( $\text{N}\cdot\text{m}^{-1}$ ) of the hydrogel sheet provides the interfacial toughness ( $\text{J}\cdot\text{m}^{-2}$ ) between the hydrogel and elastomer complex. The interfacial toughness for the AA hydrogel (in this prepared state) bonded onto the 3D printed TangoPlus elastomer is about  $170\text{ J}\cdot\text{m}^{-2}$ . Moreover, a cohesive failure was observed meaning that failure was not observed at the interface between hydrogel and elastomer; rather it was observed within the hydrogel. During the polymerization of the hydrogel, the acrylic monomer of the hydrogel link can also crosslink with free acrylic-based monomers of the 3D printed elastomer to provide a strong bonding between hydrogel and elastomer. Thus, it can be confirmed that the hydrogel is firmly bonded to the elastomer as shown by both mechanical (peeling test) and chemical (FTIR) observations.

As illustrated in Fig. 4(a), if the hydrogel elastomer construct is allowed to swell uniformly, a volume expansion occurs in all directions. Uniform drying results in a uniform shrinkage throughout the sample.

### 4.2. Devo 1 behaviour

We have employed a simple, yet effective, technique of hydrogel swelling and de-swelling phenomena that are based on what we have observed with a species of climbing cactus [20]. As a result, the movement of an artificial hydrogel-elastomer system has been achieved using some of the morphological, physio-chemical and developmental characteristics of the cactus.

In nature, development processes are central to how many plants grow and move in response to the environment. In the seasonally dry/humid tropical climate of the Brazilian Atlantic forest, this climbing cactus can creep along the ground or along and up tree trunks, stems often change direction to move upwards from the ground or sideways to search for supports.

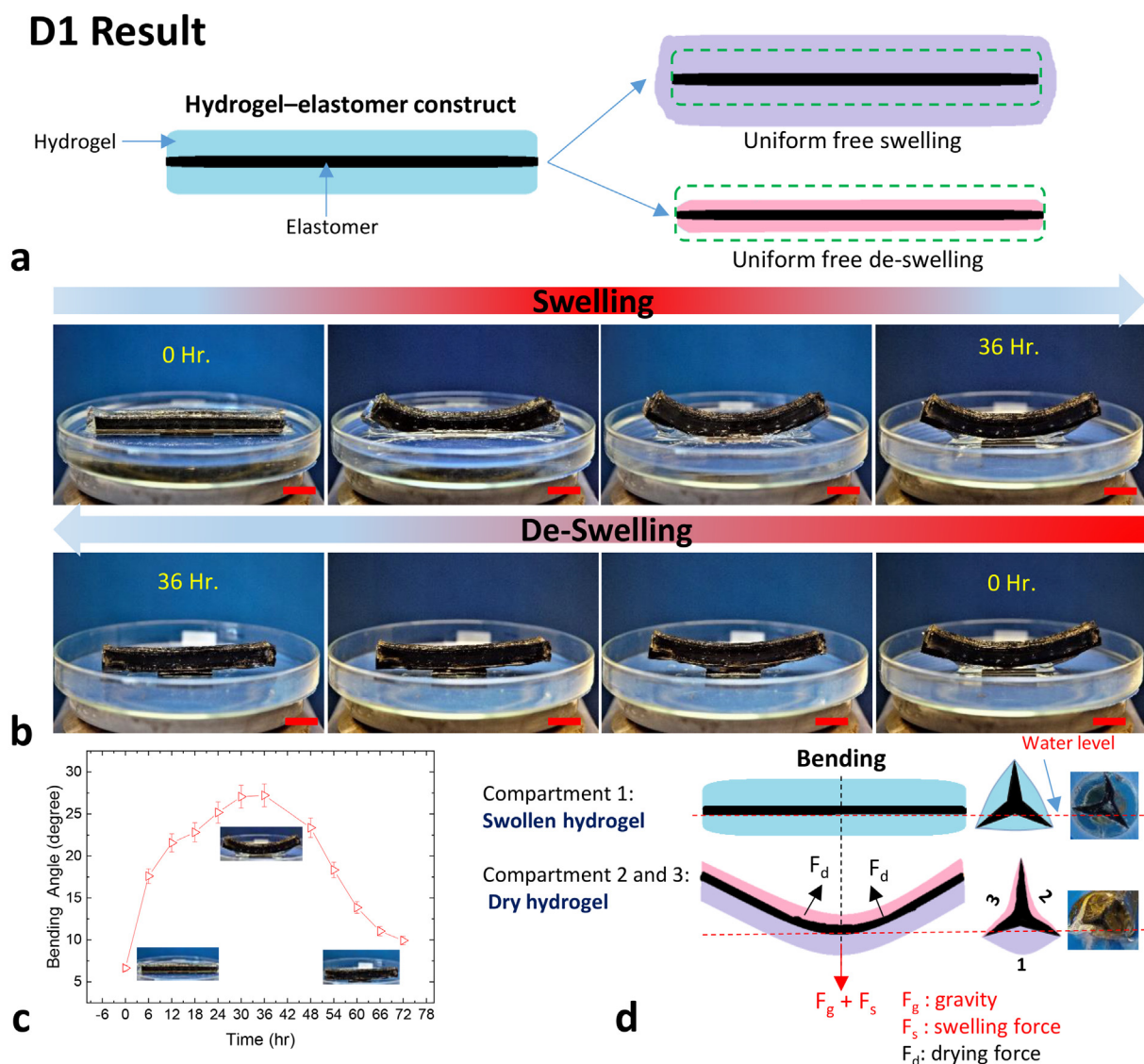
In a simple, non-energy requiring way, and furthermore using the kinds of gradients of humidity and light that might be expected under

natural conditions for a stem that is creeping along the ground or up a tree trunk, the developmental process seen in the Devo-1 concept is capable of a kind of adaptive movement of moving up from the “ground” or moving away from a “trunk”.

Plant stems creeping flat along the ground and flat against tree trunks are likely to be under more humid conditions on the contact side of the stem but under drier conditions on the side facing away. So, in these experiments, the movement effect is analogous to an object being in contact with a damper substrate and from which it turns away in order to explore and grow into new areas towards the light. The behavioural attributes of Devo-1 therefore mimic the behaviour of the cactus stem in relation to the difference between damp or dry properties of the substrate and the air surrounding it.

In the experiments, the hydrogel-elastomer system is exposed to a similar environment as the plants lying on the ground or tree trunks in nature. As can be seen in Fig. 4 (b), the hydrogel-elastomer system is lying in a damp environment - the lower half of it is exposed to a water bath, which mimics damp to wet ground environment in nature. The upper half is exposed to room temperature and low humidity, which mimics sunlight and dry aerial environment in nature. This humid/dry environment makes the hydrogel on the lower side (lower compartment of the star-shape stem) to remain hydrated while the hydrogel in the compartment on the upper part starts to dry. As the hydrogel starts to dry on the top half of the hydrogel-elastomer system, volume shrinkage can be observed. The stress generated due to shrinkage of hydrogel makes the hydrogel-elastomer system bend towards the low humidity environment as can be seen in Fig. 4(b top panel). Moreover, hydrogels in the two upper compartments of the skeleton are under drying and hydrogel in the lower compartment is under swelling. The stress generated due to the drying force must be higher than the stress generated due to swelling force. The reason for the hydrogel-elastomer construct to bend pivoting at the centre is because it is a free system and the location of the centre of gravity (CG), the hydrogel-elastomer construct is untethered and has its CG at the centre. Measurement of the bending angle showed that the hydrogel-elastomer system can bend towards a low humidity environment up to an angle of 30°. A time-lapse video showing the controlled movement of the hydrogel-elastomer system can be found in supplementary data (Video S1.1 (via swelling) and S1.2 (via de-swelling)). This process is reversible. When the bottom half of the hydrogel-elastomer system is also allowed to de-swell, it returns to the original state (Fig. 4 (b) lower panel). A similar kind of bending was realized using both designs Devo 1(a) and Devo 1(b). In terms of the bending angle, it was found that there was no significant difference between the two design demonstrators. In principle, the swelling of the hydrogel can be achieved via the internal channels for Devo 1(b), however, the increment in the bending angle was not significant between two designs. The Devo 1(b) design shows the potential to represent the plant organisation in an upright position where hydrogel “tissue” could take up water through internal channels that could be in contact with moisture at some distance from the targeted point of movement. However, in the current study, a noteworthy observation was not detected, with the test specimen in a horizontal position and further experimentations and optimizations of this more “advanced” (biologically older vascularized) developmental stage are predicted to open new doors for further developmental models of this cactus-inspired soft-robotic system.

The two developmental variations of the Devo 1 design demonstrated new and unique soft-robotic design schemes inspired by functional traits of a climbing cactus at different developmental stages. They provide an innovative framework to develop plant-inspired soft robotic systems by studying and mimicking how plant growth and development can adapt and take advantage of natural environmental conditions e.g. sunlight and changes in humidity. Furthermore, the fabricated structures are able to respond to environmental cues without



**Fig. 4.** Results for Devo 1. (a) Illustration of uniform free swelling and de-swelling of the hydrogel elastomer construct. (b) Result of the controlled movement of the hydrogel-elastomer system: controlled movement shown by the hydrogel elastomer construct as a result of the anisotropic swelling and de-swelling phenomenon. (c) Bending angle plotted against time. (d) Illustration of anisotropic swelling and de-swelling mechanism of the hydrogel-elastomer construct and interpretation of the governing forces.

using external energy for actuation in excess of that available in the surrounding environment; this is a very desirable feature and a close similarity with the living biological models.

#### 4.3. Devo 2 properties

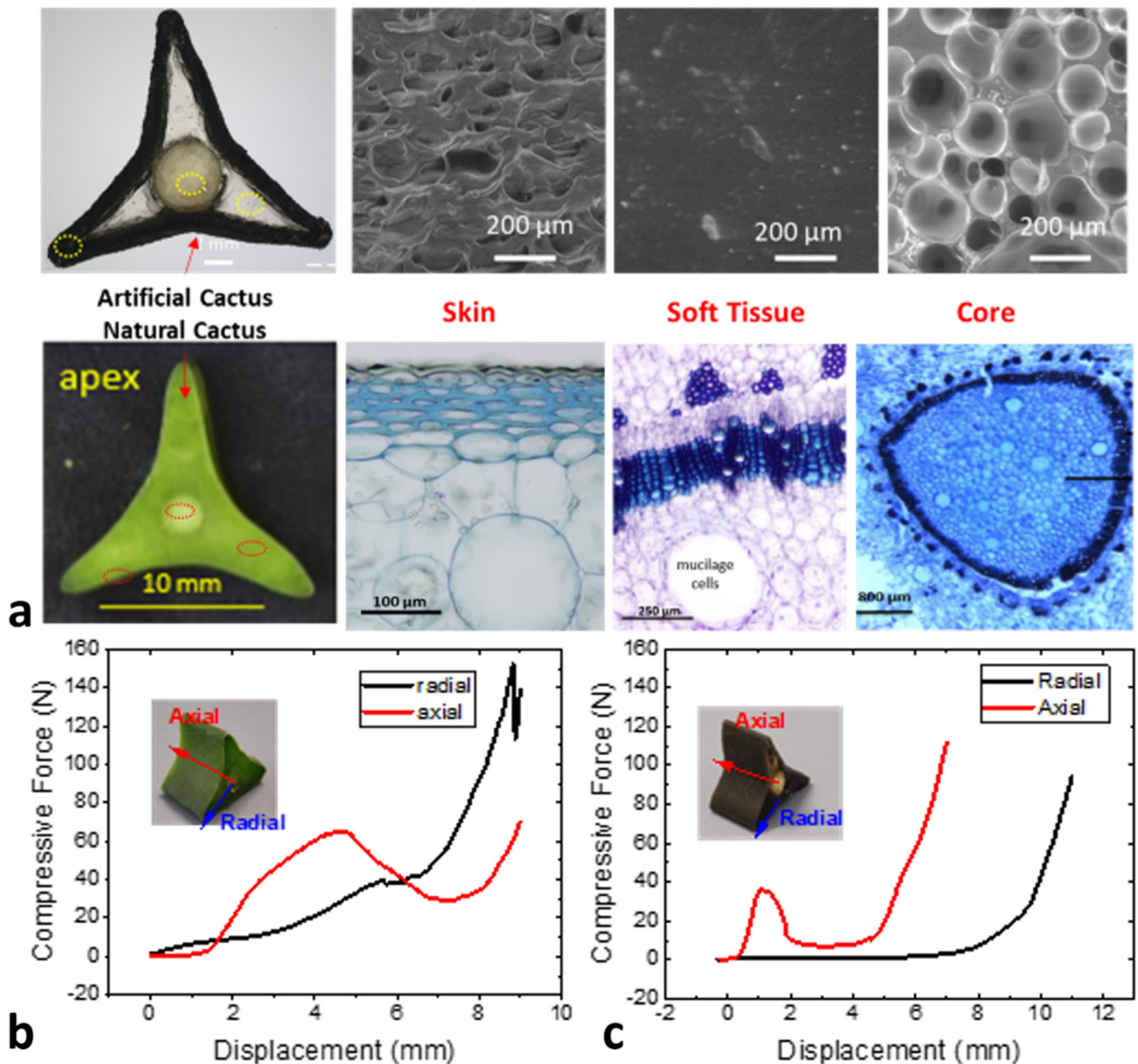
In the Devo 2 design, the main aim was to mimic adaptive, developmental changes in cross-sectional shape, from the young star-like to older circular shapes of the natural cactus during its stem development. The artificial cactus stem was constructed with a closely comparable organisation as the natural structure (Fig. 5). We started with a simple design (Devo 2(a)), however, to closely mimic the structure and mechanical properties of the natural system, a stiff core was required. Therefore, the basic design of Devo 2(a) was not further considered, however, Devo 2(a) provided an initial and progressive guideline to transform to Devo 2(b). The natural cactus possesses a varying geometry at different stages of growth (circular, triangular, and star-like) and the biological organisation of the stem comprises three main different physical components (pith and wood cylinder core, soft cortical tissue, and outer skin, see Fig. 5(a, bottom row)). Like that of the natural

organisation, the artificial cactus also consists of three major components. A 3D printed elastomeric thin layer represents the skin, a soft hydrogel represents the mucilaginous cortical tissue and a rigid PU foam represents the wood-pith core. The organisation, therefore, differs somewhat from the organisation used for the Devo 1 design in which the hydrogel actuating system was positioned outside the elastomer skeleton.

In the Devo 2 design, the outer elastomeric skin delimits and maintains the overall shape. The inner PU foam core provides a centrally placed internal strengthening element as well as porosity for the hydrogel to take up water. The hydrogel is the actuating component of the artificial cactus and is situated in the space between the outer skin and the core and is thus very similar to the biological organisation.

The physical properties of the components of the artificial cactus are given in supplementary data Fig. S3. The drying rate of the hydrogel is found to be higher than the swelling rate. The hydrogel can take up water at up to 80 times its weight in 48 h. Similarly, the hydrogel can be completely dry within 48 h when just left at room temperature. In tensile loading, it was found that the mechanical properties of the 3D printed TangoPlus skin are dependent on the printing directions of the





**Fig. 5.** Properties of components of artificial cactus. (a) Morphology of the artificial cactus (top row) compared to natural cactus (bottom row). Compressive properties in axial and radial loading: (b) natural cactus and (c) artificial cactus.

3D printing process. The tensile modulus of the skin is  $100 \pm 60$  kPa (normal to the printed layer, z-direction) and  $500 \pm 60$  kPa (along the printed direction) however, the elongation at break is always maintained above 100%. The PU foam displays a clear yielding in compressive loading with yield points typically occurring at about 15% strain level. The compression modulus is about  $2 \pm 0.2$  MPa and the compressive strength is about  $130 \pm 20$  kPa.

For comparison, we also include here the response of the natural cactus stem in compressive loading. As seen in Fig. 5(b), the natural cactus shows a marked difference in its response to axial and compressive loading. Axial stiffness is always higher, which is due to the presence of the wooden core in the centre. Axial loading is marked with a clear yield point however, the natural cactus stem does not show a clear yield point in the radial loading. The clear yield point in axial loading indicates the failure of the wooden core of the natural cactus. A similar type of response is shown by the artificial cactus as can be seen in Fig. 5(c). The

artificial cactus shows higher axial stiffness than radial stiffness in compression and the rigidity of the PU foam core is responsible for the axial stiffness. Moreover, the clear yield point in the axial loading is also marked by the failure of the PU foam core. Considering all these features together, the artificial cactus shows both a similar morphological arrangement and physical behaviour compared to the natural cactus.

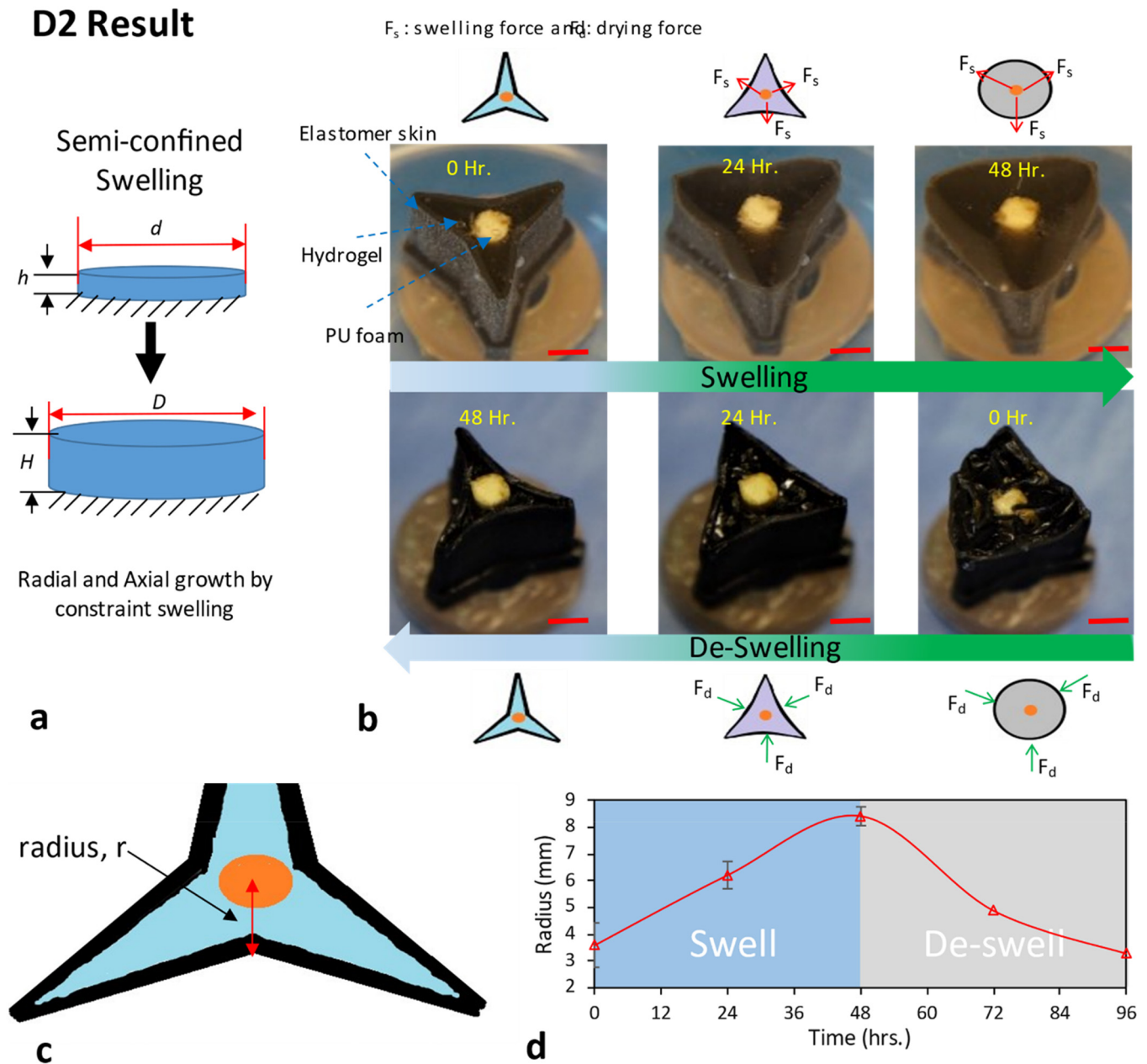
#### 4.4. Devo 2 behaviour

In nature, many kinds of plant display axial and radial growth simultaneously. Plant stems can reach heights from mm to meters by means of axial growth and at the same time increase stem thickness as a result of both (a) primary radial growth of the cortex and (b) secondary radial growth of the wood and outer bark tissue.

In many plants, changes in cross-sectional size and shape involve at least three kinds of meristematic activity: cell division, differentiation,

and maturation. The cactus studied here shows these complex changes during additive growth via meristems (cell divisions). However, swelling and de-swelling tests of the living cactus stem at different osmotic potentials as well as observations in the field of both shrunken and turgescient stems, indicate that water availability and water adsorption and turgescence by mucilaginous tissue, clearly play an important role in assuring stem geometry and likely mechanical properties (A. K. Bastola, P. Soffiatti, M. Behl, A. Lendlein, N. Rowe, Structural performance of a climbing cactus: making the most of softness, unpublished results). So, in the Devo 2 model, we were interested to show to what extent we could mimic developmental changes in stem shape and behaviour using just *one part* of the cactus's developmental system -the mucilaginous tissue swelling, but not more complex levels of additive or equivalent meristematic growth.

When a hydrogel swells the overall volume within a structure can change and both radial and axial growth can be achieved [44], see Fig. 6(a). In the Devo 2 development, the hydrogel is confined by the outer elastomeric skin, so when the hydrogel takes up water the axial growth of the hydrogel is expected to be dominant. The reason for axial growth expected to be dominant is because the hydrogel is compliant at the outer edges by the thin elastomer skin. However, radial/lateral growth is also expected due to the high flexibility and elasticity of the thin skin. Radial growth of the hydrogel will be responsible for changes in the cross-sectional shape of the artificial cactus. In our experiments, to enable changes in geometry, the star-shaped artificial cactus was submerged in a water bath, see Fig. 6(b). As the hydrogel starts to take up water, the change in shape from star-like to circular resembles very much that of the developmental sequence of the natural cactus.



**Fig. 6.** Results for Devo 2. (a) Illustration of radial and axial growth of hydrogel during the constraint swelling. (b) The change in the shape from star-like to circular and vice versa by swelling and de-swelling demonstrated by the artificial cactus similar to that of natural cactus. The scale bar is 4 mm. (c) illustration of the radius of the artificial cactus. (d) Result of the change in radius of the artificial cactus by swelling and de-swelling plotted against time (red curve is a visual guide).

The pressure generated due to the water intake by the hydrogel pushes the elastomeric skin outward from the original star-shaped cross-section. As can be seen in Fig. 6(b), the star-like shape is changed to a triangular and then to a circular outline within 48 h. This process is reversible; when the hydrogel is allowed to dry, the elastomeric skin returns to the original developmental stage of the star-like shape via a triangular section. In order to quantify the results, a change in radius of the artificial cactus (see Fig. 6(c) for definition of the radius) as a result of swelling and deswelling was obtained and plotted against time (Fig. 6(d)). It was found that the change in radius can be as high as thrice that of the original radius. At the same time, it was also observed the rate of change in radius is higher during the de-swelling compared to swelling as it was also detected in Devo 1. The experiment demonstrates that a simple multi-material system can be programmed with an artificial developmental process and perform changes in cross-sectional shape that develop during the natural developmental steps of the cactus. A time-lapse video showing the change in geometry can be found in supplement data (Video S2.1 (via swelling) and S2.2 (via de-swelling)).

## 5. Conclusions

The two developmental models explored here demonstrate that there is much to learn from the ecology, development, and adaptive behaviour of plants. These biological systems provide a ready source of information to design developmentally smart and adaptive artificial systems. The controlled movement (bending) and the changes in shape could be used to monitor, for example, variations in humidity as well as to develop a humidity-based actuator for a plant-inspired ecological robotic system.

We present a variety of soft robotic schemes based on the growth and developmental of a climbing cactus, *Selenicereus setaceus*, from the seasonally dry Atlantic forest of Brazil. In its natural habitat, the cactus stem develops striking modifications in cross-sectional geometry, which adaptively perform varying functional roles in response to external cues and constraints from the environment.

Firstly, we demonstrated a multi-material, hydrogel-elastomer, and biphasic soft robotic system inspired by and based on the structural configuration of the cactus. This first soft robotic system Devo 1 was based on a design mimicking two developmental stages of the young searcher, star-shaped configuration. We demonstrated that it is possible to make a controlled bending movement of the artificial stem via anisotropic swelling and de-swelling of the hydrogel. We demonstrated these movements in a young stage without water transport conduits inbuilt in the stem "skeleton" and a second theoretically "older model" in which water transport is facilitated by interconnecting conduits as seen in older stages of development of the cactus where water transporting tissue is further developed. The movements require no external energy input besides that of the local environment. The plant movements are also reversible and thus mimic an important kind of movement which does not depend on additive growth and which can be used for optimally positioning leaves and stems and in climbing plants points of attachment.

Secondly, in the Devo 2 design, we created an artificial cactus stem based on the morphological arrangements as well as the mechanical properties that are comparatively similar to those of the natural cactus. In this design, the artificial cactus is made up of 3D printed elastomeric skin which sandwiches the soft hydrogel and PU foam inner core. Elastomer and PU foam are passive components whereas the hydrogel is the active component of the artificial cactus. The star-shaped artificial cactus demonstrated the changes in shape from star-like to circular and vice-versa as a result of uniform hydrogel swelling and de-swelling, respectively. The experiment demonstrates that it is possible to recreate similar adaptive changes in stem cross-sectional geometry that are used by the cactus to optimize mechanical properties such as flexural rigidity to explore and colonize its local environment. Furthermore,

whereas the natural development of the cactus includes both meristematic, additive growth and addition of biomass (e.g. wood and bark tissue) as well as swelling and de-swelling of hydrogel-like mucilaginous tissue, the experiments here demonstrate that a similar range of developmental adaptive cross-sectional geometries can be achieved by changes in hydrogel swelling only.

A further advantage is that the cactus-inspired soft robotic system does not require additional sources of energy to actuate the robotic such as heat or magnetic/electric field. The developed cactus-inspired soft robotic systems can be used for sensing (e.g. humidity), energy harvesting, and innovative soft robotics applications without using any external energy. We hope that this study stimulates the design of novel plant-inspired ecological and growing robotic systems. Lastly, it must be noted that, in principle, the proposed design and development of the artificial cactus do not entirely depend on 3D printing for the developmental changes observed, the pronounced flexibility of the additive manufacturing or 3D printing technology is however extremely useful for the successful realization and development of the intricate geometry of the cactus.

Looking closely at nature can provide exciting solutions for new innovative artefacts such as the developing cactus designs here. Interestingly, the results shown by the artificial system may actually shed light on the biological system; such as the roles of cellular growth versus hydrogel function of the soft tissue in maintaining or switching to different shapes. Our study suggests that the desire to learn from nature may also help us to understand nature.

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.matdes.2021.109515>.

## Data availability statement

The raw and processed data required to reproduce these findings will be available upon publication of the manuscript to download from <https://doi.org/10.6084/m9.figshare.13302830>

## Declaration of Competing Interest

The authors declare no competing interests.

## Acknowledgements

Mrs. Yvonne Pieper is acknowledged for cross-sectional images of the artificial cactus. This work was financially supported by the Helmholtz Association through programme-oriented funding and received funding from the European Union's Horizon 2020 research and innovation program under Grant Agreement No. 824074 (GrowBot).

## References

- [1] B. Mazzolai, Plant-inspired growing robots, in: C. Laschi, J. Rossiter, F. Iida, M. Cianchetti, L. Margheri (Eds.), *Soft Robotics: Trends, Applications and Challenges*, Springer, Cham 2017, pp. 57–63.
- [2] B. Mazzolai, L. Beccai, V. Mattoli, Plants as model in biomimetics and biorobotics: new perspectives, *Frontiers in Bioengineering and Biotechnology* 2 (2014) 2.
- [3] I.D. Walker, Biologically inspired vine-like and tendril-like robots, 2015 science and information conference (SAI), IEEE (2015) 714–720.
- [4] E. Del Dottore, A. Sadeghi, A. Mondini, V. Mattoli, B. Mazzolai, Toward growing robots: a historical evolution from cellular to plant-inspired robotics, *Frontiers in Robotics and AI* 5 (2018) 16.
- [5] B. Mazzolai, F. Tramaccere, I. Fiorello, L. Margheri, The bio-engineering approach for plant investigations and growing robots. A mini-review, *Frontiers in Robotics and AI* 7 (2020) 130.
- [6] B. Mazzolai, *Plant-Inspired Growing Robots*, Springer International Publishing, Cham, 2017 57–63.
- [7] E.W. Hawkes, L.H. Blumenschein, J.D. Greer, A.M. Okamura, A soft robot that navigates its environment through growth, *Science Robotics* 2 (8) (2017) eaan3028.
- [8] I. Must, E. Sinibaldi, B. Mazzolai, A variable-stiffness tendril-like soft robot based on reversible osmotic actuation, *Nat. Commun.* 10 (1) (2019) 1–8.
- [9] M.B. Wooten, I.D. Walker, Circumnutation: From plants to robots, in: E. Tuci, A. Giagkos, M. Wilson, J. Hallam (Eds.), *From Animals to Animats 14*, Springer International Publishing 2016, pp. 1–11.

- [10] M.B. Wooten, I.D. Walker, Vine-inspired continuum tendril robots and circumnutations, *Robotics* 7 (3) (2018) 58.
- [11] A. Sadeghi, A. Tonazzini, L. Popova, B. Mazzolai, A novel growing device inspired by plant root soil penetration behaviors, *PLoS One* 9 (2) (2014) e90139.
- [12] E. Del Dottore, A. Mondini, A. Sadeghi, B. Mazzolai, Characterization of the growing from the tip as robot locomotion strategy, *Frontiers in Robotics and AI* 6 (2019) 45.
- [13] R. Chen, Y. Wan, W. Wu, C. Yang, J.-H. He, J. Cheng, R. Jetter, F.K. Ko, Y. Chen, A lotus effect-inspired flexible and breathable membrane with hierarchical electrospinning micro/nanofibers and ZnO nanowires, *Mater. Des.* 162 (2019) 246–248.
- [14] F. Hu, L. Lyu, Y. He, A 3D printed paper-based thermally driven soft robotic gripper inspired by cabbage, *Int. J. Precis. Eng. Manuf.* 20 (11) (2019) 1915–1928.
- [15] F. Putzu, T. Abrar, K. Althoefer, Plant-inspired soft pneumatic eversion robot, 2018 7th IEEE international conference on biomedical robotics and biomechanics (biorob), IEEE (2018) 1327–1332.
- [16] W. Wang, C. Li, M. Cho, S.-H. Ahn, Soft tendril-inspired grippers: shape morphing of programmable polymer–paper bilayer composites, *ACS Appl. Mater. Interfaces* 10 (12) (2018) 10419–10427.
- [17] D. Lunni, M. Cianchetti, C. Filippeschi, E. Sinibaldi, B. Mazzolai, Plant-inspired soft bistable structures based on hygroscopic electrospun nanofibers, *Advanced Materials Interfaces* 7 (4) (2020) 1901310.
- [18] D. Correa, S. Poppinga, M.D. Mylo, A.S. Westermeier, B. Bruchmann, A. Menges, T. Speck, 4D pine scale: biomimetic 4D printed autonomous scale and flap structures capable of multi-phase movement, *Philosophical Transactions of the Royal Society A* 378 (2167) (2020) 20190445.
- [19] A.C. Gibson, P.S. Nobel, *The Cactus Primer*, Harvard University Press, 1986.
- [20] P. Soffiatti, N.P. Rowe, Mechanical innovations of a climbing cactus: functional insights for a new generation of growing robots, *Frontiers in Robotics and AI* (2020) 7.
- [21] T. Wallin, J. Pikul, R. Shepherd, 3D printing of soft robotic systems, *Nature Reviews Materials* 3 (6) (2018) 84–100.
- [22] X. Wang, M. Jiang, Z. Zhou, J. Gou, D. Hui, 3D printing of polymer matrix composites: a review and prospective, *Compos. Part B* 110 (2017) 442–458.
- [23] H.K. Yap, H.Y. Ng, C.-H. Yeow, High-force soft printable pneumatics for soft robotic applications, *Soft Robotics* 3 (3) (2016) 144–158.
- [24] A. Sydney Gladman, E.A. Matsumoto, R.G. Nuzzo, L. Mahadevan, J.A. Lewis, Biomimetic 4D printing, *Nat. Mater.* 15 (4) (2016) 413–418.
- [25] N.W. Bartlett, M.T. Tolley, J.T.B. Overvelde, J.C. Weaver, B. Mosadegh, K. Bertoldi, G.M. Whitesides, R.J. Wood, A 3D-printed, functionally graded soft robot powered by combustion, *Science* 349 (6244) (2015) 161–165.
- [26] D.K. Patel, A.H. Sakhaei, M. Layani, B. Zhang, Q. Ge, S. Magdassi, Highly stretchable and UV curable elastomers for Digital light processing based 3D printing, *Advanced Materials* 29 (15) (2017) 1606000.
- [27] A. Sadeghi, A. Mondini, B. Mazzolai, Toward self-growing soft robots inspired by plant roots and based on additive manufacturing technologies, *Soft Robotics* 4 (3) (2017) 211–223.
- [28] M. Tyagi, G.M. Spinks, E.W. Jager, 3D printing microactuators for soft microrobots, *Soft Robotics* (2020) <https://doi.org/10.1089/soro.2019.0129>.
- [29] Y. Kim, H. Yuk, R. Zhao, S.A. Chester, X. Zhao, Printing ferromagnetic domains for untethered fast-transforming soft materials, *Nature* 558 (7709) (2018) 274–279.
- [30] T. Xu, J. Zhang, M. Salehizadeh, O. Onaizah, E. Diller, Millimeter-scale flexible robots with programmable three-dimensional magnetization and motions, *Science Robotics* 4 (29) (2019) eaav4494.
- [31] C. Ma, S. Wu, Q. Ze, X. Kuang, R. Zhang, H.J. Qi, R. Zhao, Magnetic multimaterial printing for multimodal shape transformation with tunable properties and Shiftable mechanical behaviors, *ACS Applied Materials & Interfaces* (2020) <https://doi.org/10.1021/acsami.0c13863>.
- [32] R. Bastien, A. Porat, Y. Meroz, Towards a framework for collective behavior in growth-driven systems, based on plant-inspired allotropic pairwise interactions, *Bioinspiration & Biomimetics* 14 (5) (2019) 055004.
- [33] C. Kadapa, Z. Li, M. Hossain, J. Wang, On the advantages of mixed formulation and higher-order elements for computational morphoelasticity, *Journal of the Mechanics and Physics of Solids* 148 (2021) 104289.
- [34] F. Xu, C. Fu, Y. Yang, Water affects morphogenesis of growing aquatic plant leaves, *Physical Review Letters* 124 (3) (2020) 038003.
- [35] D.E. Moulton, H. Oliveri, A. Goriely, Multiscale integration of environmental stimuli in plant tropism produces complex behaviors, *Proc. Natl. Acad. Sci.* 117 (51) (2020) 32226–32237.
- [36] Y. Meroz, Plant tropisms as a window on plant computational processes, *New Phytologist* 229 (4) (2020) 1911–1916.
- [37] Y. Meroz, R. Bastien, L. Mahadevan, Spatio-temporal integration in plant tropisms, *Journal of The Royal Society Interface* 16 (154) (2019) 20190038.
- [38] H. El-Hussieny, I.A. Hameed, J.H. Ryu, Nonlinear model predictive growth control of a class of plant-inspired soft growing robots, *IEEE Access* 8 (2020) 214495–214503.
- [39] S.C. Ligon, R. Liska, Jr. Stampfl, M. Gurr, R. Mühlhaupt, Polymers for 3D printing and customized additive manufacturing, *Chem. Rev.* 117 (15) (2017) 10212–10290.
- [40] T.D. Ngo, A. Kashani, G. Imbalzano, K.T. Nguyen, D. Hui, Additive manufacturing (3D printing): a review of materials, methods, applications and challenges, *Composites Part B: Engineering* 143 (2018) 172–196.
- [41] A.K. Bastola, M. Paudel, L. Li, Development of hybrid magnetorheological elastomers by 3D printing, *Polymer* 149 (2018) 213–228.
- [42] STRATASYS: the 3D Printing Solutions Company: Tango: Polyjet Simulated Rubber Material, <https://www.stratasys.com/materials/search/tango> 2020 (accessed 29/10/2020).
- [43] H. Yuk, T. Zhang, G.A. Parada, X. Liu, X. Zhao, Skin-inspired hydrogel–elastomer hybrids with robust interfaces and functional microstructures, *Nat. Commun.* 7 (1) (2016) 1–11.
- [44] W.R. Illeperuma, J.-Y. Sun, Z. Suo, J.J. Vlassak, Force and stroke of a hydrogel actuator, *Soft Matter* 9 (35) (2013) 8504–8511.