

Thermo-responsive Self-Folding Strings for Programmable Matter



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Mater artium necessitas

Declaration

I, Quentin Lahondès, hereby declare that, except where specific reference is made to the work of others, the contents of this dissertation are original and have not been submitted in whole or in part for consideration for any other degree or qualification in this, or any other university. This dissertation is my own work and contains nothing which is the outcome of work done in collaboration with others, except as specified in the text and Acknowledgements.

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Abstract

Whether metal, polymer or ceramic, crystalline or amorphous, both the nature and arrangement of the elements constituting matter dictate its physical properties. Traditional processes can be used to fabricate structures relying on processes such as additive manufacturing, self-folding, and modular assembly. Compared to other approaches, self-folding structures present the advantage of shape morphing to 3D using various actuators including smart materials. Nevertheless, self-folding structures face challenges, including universality for shape morphing, scale and folding torque, addressability and collisions, reversibility and reprogrammability, remote actuation, sensors, communication, and computation. In this thesis, three different approaches are presented, each related to distinct challenges. First, I propose an environment with reduced gravity to lower the required folding torque, along with a new design for a modular chain that allows the hinges of the string to be manually reset to their initial state and the string to be reprogrammed in order to self-fold into a different configuration. This was demonstrated by the self-folding of three different voxel configurations used for crystalline structures. Second, I propose a manufacturing process enabling the reversibility of self-folding to be turned off permanently. This was implemented on semi-crystalline structures, including a pyramid, a rectangle and a cube, and the results were extended to small robots whose locomotion could be tuned. Third, I propose a design methodology to render self-folding hinges via magnetic induction to be addressed sequentially by adapting the design of the magnetic receivers and their configurations. In addition, a method was proposed to translate knots based on continuous deformations into folding patterns with discontinuous deformations. These methodologies were tested on a sequential self-folding box, a bio-inspired mimosa pudica leaf and an overhand knot to replicate amorphous structures.

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Chapter 1

Introduction

From the plethora of possible different atomic arrangements occurring in nature, a string-like arrangement is essential for life, to carry and transfer information as in DNA, and to make functional building blocks and nanomachines as in proteins. On the opposite, inorganic matter constructs itself in a 3D structured manner. Inorganic matter comes in two main categories. Highly ordered structures with repeating units denominated as crystalline, whereas disordered structures lack repeating patterns, are designated as amorphous. In amorphous structures, a complete lack of order may appear. In polymers, accordion patterns of various lengths may form and are referred as semi-crystalline, thus making amorphous structures locally ordered or disordered.

More recently, a new type of material has been discovered: metamaterials. Initially thought to be only be born of human ingenuity, these materials are actually present in nature. The prefix meta means "beyond" in Greek and signify here, that the physical properties result from the design of the material rather than its atomic nature, and as a result, exhibit novel optical, thermal, acoustic, mechanical, and other properties.

In the early days of mankind's development, making structures was vital whether by piling and bonding small units to build sturdy shelters or knotting together fibers to make flexible clothes. In essence, human realisations are highly ordered with a clear construction methodology and design. Artificial materials can be fabricated with traditional means but recent progresses demonstrated their fabrication from using origami for deploying structures or solar panels in space, to using strings for engineering nano robots made of DNA strands to be applied for biomedical applications.

To bring "low dimensions" planes, strings, and points to 3D and beyond, these structures are typically self-folded using bending, buckling, or rotary mechanisms. Bending and buckling can rely on miniature actuators, called smart materials, embedded directly in the structures and responding to variations in their environments (temperature, pH, light, etc.) by

changing their volume. This makes smart materials, both actuators and sensors, capable of actuating on a small scale.

Without smart materials, bending and buckling are achieved by inducing mechanical stress within the structure, which allows bending or folding out-of-plane upon stress release. Rotary mechanisms at the micro scale function identically to macro scale joints, but are challenging to fabricate, even with high-precision micro-fabrication techniques.

1.0.1 Challenges

Regardless of the actuation method or the dimensional nature of the structure, self-folding faces numerous challenges, as listed below:

- Universality,
- Scale and torque,
- Addressability and collisions,
- Reversibility and reprogrammability,
- Remote actuation,
- Sensors, communication and computation.

Universality describes the ability of a structure to morph into another structure. For example, a universal origami pattern allows a plane to be folded into 3D shapes. Scale and torque pertain to the physical ability of the hinges to self-fold considering the weight of the structure. Addressability concerns the controllability of the folding sequence of hinges along a self-folding structure. While self-folding, controllability allows a structure to avoid collisions to maintain the self-foldability of the structure. Reversibility pertains to the capacity of hinges to revert to their initial state after self-folding, and reprogrammability is the ability of the structure to morph into several shapes. For example, this enables trial-and-error in robotic functions owing to the reversibility allows a gripper to grasp and release an object and adapt to different objects. Remote actuation consist of triggering hinges from a distance, thus enabling the structure to perform freely in remote and constrained environments. This capability is often necessary for in vivo applications to perform with a certain form of autonomy. Finally, sensing provides a sense of self and of the world. Communication allows an external user to assess the situation, and computation enables decision-making. Together, these features would allow a self-folding robot full autonomy and adaptability to changes in contexts or environments.

1.0.2 Aims and Objectives

The aim of this thesis is to build crystalline, semi-crystalline and amorphous structures from self-folding strings by proposing design guidelines to tackle the challenges of universality, scale and torque, reversibility, reprogrammability and remote actuation.

To tackle these challenges, this thesis objectives include the proposal for:

- an alleviating environment and string design where the folding torque required is reduced to allow large strings to be self-foldable,
- a string design to form into multiple shapes upon reprogramming of the string to address the universality of shape-morphing structures,
- countering the reversible behaviour of some smart materials by locking the structure in place to maintain the structural capabilities without any input,
- design guidelines to self-fold remotely and sequentially strings to fold amorphous structures such as knots,
- a methodology translating continuous knots (based on bending of a string) into self-folding discontinuous knots to enable the self-folding of knots in the future,

1.0.3 Contributions

In my thesis, I propose approaches regarding the challenges of scale and torque, reprogrammability, reversibility, addressability and remote actuation, organised as follows:

- Chapter 3, the first technical chapter, describes my approach toward crystalline structures. This contribution relates to the scalability and torque requirements, along with the reprogrammability of self-folding chains. It presents a modular 1D chain self-folding in water at a neutral buoyancy temperature, and demonstrates the self-folding and reprogrammability of crystals/voxels from different chain configurations (Figure 1.1(a) and (b)).
- Chapter 4, the second technical chapter, details my approach to semi-crystalline structures. This contribution relates to the reversibility of self-folding robots. In this chapter, a novel fabrication process enabling the bonding of a hydrogel and a shape-memory polymer is presented and used to reversibly self-fold and permanently lock structures (Figure 1.1(c) to (e)).

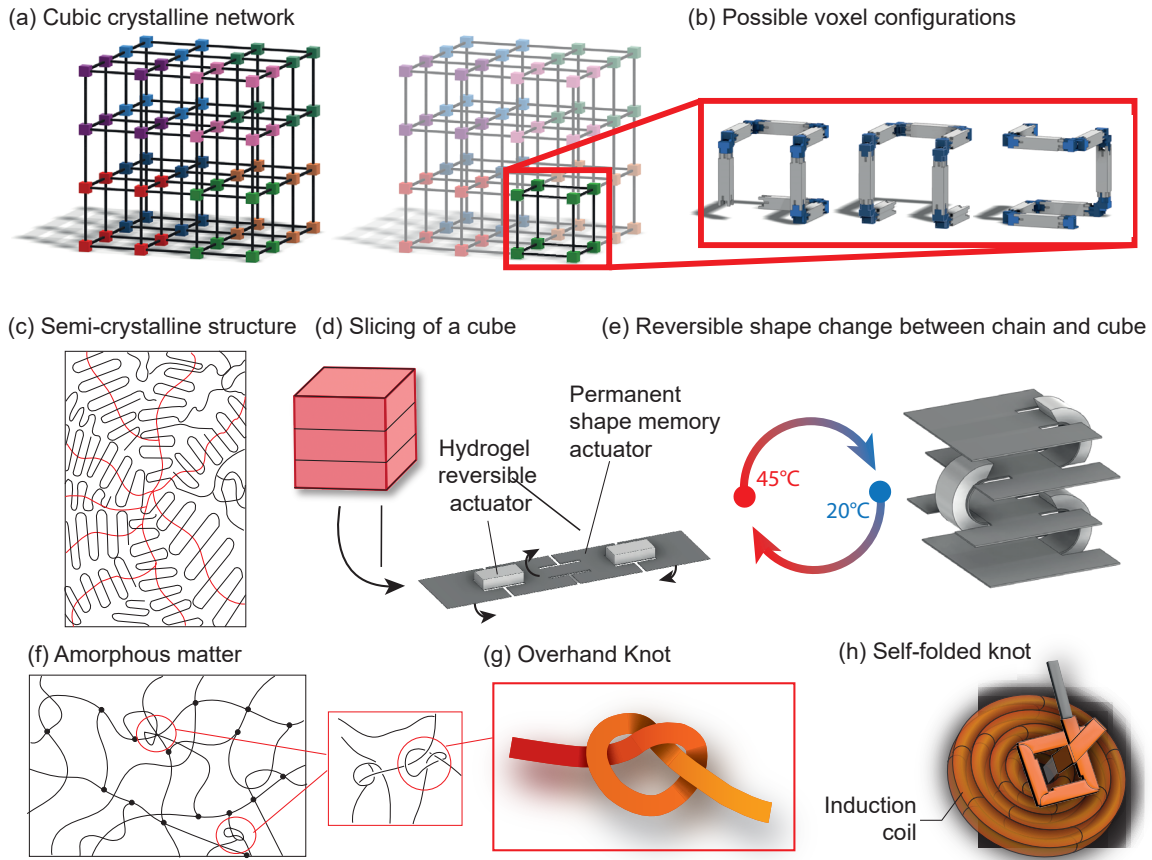


Fig. 1.1 Illustration of the aim of each chapter of the thesis. Chapter 1 aims to replicate crystalline structures that can be decomposed into cubes (a) using modular chain to fold into voxels (b). Chapter 2 aims to reproduce the zig/zag pattern seen in semi-crystalline structures (c) by slicing 3D shapes into many layers (d) that can fold reversibly between the flat and 3D shape (e). The objective of Chapter 3 is to output topological equivalent of amorphous structures (f) where knots appears (e.g. overhand knot) (g), by self-folding ribbons using magnetic induction (h).

- Chapter 5, the third technical chapter, presents my approach to amorphous structures. This chapter contributes to the remote activation and addressability of hinges in self-folding origami and ribbons (Figure 1.1(f) to (h)). I introduce magnetic induction to self-fold various structures, including a cube, a bio-inspired mimosa pudica leaf and an overhand knot .

1.0.4 Publications

This thesis includes the content of several articles I have written and participated to:

- "Origami-Inspired Micro-robots" is a review paper published in the Encyclopedia of Robotics in 2020 with the help of Alfred Wilmot and Shuhe Miyashita (doi: 10.1007/978-3-642-41610-1_195-1) whose content is partly used for the literature review. I helped devising a structure from the articles I read and gathered, and wrote the text which was corrected and adjusted by Alfred Wilmot and Shuhe Miyashita.
- "Temperature Driven Soft Reversible Self-folding Origami String" is a conference article published in RoboSoft conference in 2022 (doi: 10.1109/RoboSoft54090.2022.9762090) whose content is used in Chapter 4 of the thesis. The concept and technicalities originate from my work and the article was adjusted with the corrections of Shuhe Miyashita.
- "Reprogrammable Free-Self-Folding Strings" is an article draft made my Chapter 3 and aims to be submitted by the end of September 2023 in the journal named Smart Materials and Structures. Similar to the previous article mentioned, this article uses my technical work and concept, but was corrected as per the recommendations of Shuhe Miyashita.
- "Soft Hinge for Magnetically Actuated Millimetre-Size Origami" is an abstract published at the TAROS conference in 2020 (doi: 10.1007/978-3-030-63486-5_9), not used in the thesis based on the work done in the early stage of the PhD.

And also several papers of which I am a co-author:

- "Size Changing Soft Modules for Temperature Regulated Self-assembly and Self-disassembly" is a conference article published in Robosoft in 2022 (doi: 10.1109/RoboSoft54090.2022.9762093) whose main author is Junyi Han. I proposed the concept, design and fabrication of the modules used for the self-assembly while Junyi Han performed the experiments, proposed an experimental setup and redacted the article with Shuhe Miyashita.
- "Origami Robot Self-folding by Magnetic Induction" is a conference article published in IROS in 2022 (doi: 10.1109/IROS47612.2022.9981604). Here, I provided the theory behind the model and suggested the thermal experiments, while Kaan Esendag developed the experimental setup and Jialun Liu redacted the article along with Shuhe Miyashita and Dana Damian in addition of fabricating the origami robot with the help of Xiao Chen and performed the experiments.

Chapter 2

Literature Review

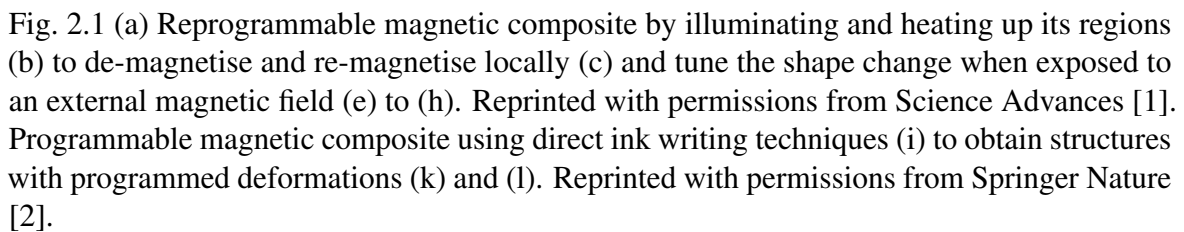
2.1 Overview of Programmable Materials

This section presents some of the most known programmable materials. They can be defined as materials whose physical properties such as mechanical, electrical, thermal, optical, can be altered by a given stimulus as an input. With a precise input variation and a good understanding of the material properties, the desired physical output can be accurately controlled resulting in a programmable material that can be introduced into a larger structure for added functionality. For example, the magnetisation of magnetic composites can be programmed to obtain certain shapes for robotic applications, while the thermal expansion of metals can be utilised to control the bending of bilayer to create thermometers.

2.1.1 Magnetic materials and composites

Magnetic smart materials have garnered significant attention due to their unique capabilities in responding to external magnetic fields. One fundamental approach to rendering a structure magnetic is by incorporating magnetic particles into a non-magnetic matrix [3] [4] [5]. This method allows for the programmability of the magnetisation profile, enabling the material to achieve specific shapes or behaviours when exposed to magnetic fields [6] [7] [8]. This results in an actuation where the programmable magnetic material does not change volume or mass but only position upon immersion into an external magnetic field. Such programmability is particularly advantageous in applications requiring precise control over the material's configuration and movement. This approach has notably been implemented by Li et al. to locomote miniature robots for drug delivery in the vascular system [9].

Furthermore, the reprogrammability of these materials adds a layer of versatility to meet changing requirements. The magnetisation can be modified by re-magnetising the material



The fabrication of magnetic structures typically involves advanced techniques such as lithography [12] [8] or 3D-printing [13]. This is demonstrated by Kim et al. and illustrated in Figure 2.1 (i) to (l), where a soft polymer dopped with permanent magnets micro-particles was extruded and magnetised during the extrusion process to obtain planar magnetisation, resulting in out-of-plane deformation of the printed structures when exposed to an external magnetic field [2]. These methods allow precise placement and patterning of magnetic particles within the matrix, leading to finely tuned magnetic properties. This precision is crucial for the creation of complex devices such as Hallbach arrays [7] or micro-robots [14], which require exact magnetic configurations to function correctly.

Another innovative approach is to use a smart material as the matrix itself, which can change its volume in response to induction or hysteresis heating [15] [16]. This method offers the dual advantage of remote heating, in addition to magnetic steering [17]. Such a combina-

tion has been demonstrated in applications like drug delivery, hyperthermia treatments [18] [19], water desalination [20] and self-folding structures [21], where both temperature and magnetic control are desirable.

Although not considered a smart material, integrating large permanent magnets onto a structure presents another viable method for achieving strong magnetisation. This approach generates more substantial magnetic forces than dispersed particles, making it suitable for larger-scale applications and providing greater magnetic force for tasks such as the locomotion of robots [22] [23] or actuation mechanisms [24] [25].

In conclusion, magnetic composites offer a versatile and dynamic solution for a variety of advanced applications. Their ability to respond quickly to external magnetic fields, combined with the flexibility to operate independently of environmental conditions, makes them particularly attractive for innovations in fields ranging from medical technology to robotics. The fast dynamics enabled by external magnetic fields, along with the precise control they offer, highlight the potential of magnetic smart materials.

2.1.2 Smart Materials

Smart materials, which can change their volume in response to environmental stimuli, play a major role in self-folding techniques [8]. When placed between materials that do not change volume, these materials can induce a folding motion. Shape memory alloys have also been utilised as linear actuators in robots, enabling the compression of foldable structures and the creation of worm-like robots [26]. Other notable examples include the integration of smart materials into structures through 3D printing of hydrogels [27] [28] or composite materials. Smart materials based structures are notably valuable in the biomedical field due to their ability to operate in remote and limited-access environments. Applications include wound patching [29], stents [30], or drug delivery [31].

Unlike traditional macro-scale methods requiring electrical power input, smart materials can be activated by changing environmental conditions, including temperature, pH and light, thus enabling direct contact, tethered or remote activation. Direct contact methods utilise pH [32] [9], hydration [33] [34], and temperature-sensitive [35] [36] smart materials by immersing the structure in environments with varying characteristics. Tethered energy supplies are commonly used for shape memory alloys [30] [37], piezoelectric actuators [38], and other high-power or high-frequency smart materials. To make self-folding structures less dependent on their environment and tethers, self-folding at user-defined times was achieved relying on remote activation methods including the use of photothermal effects [39] [40] or electro-magneto-thermal techniques [15] [41] [20] [42].

Hydrogels

Hydrogels are polymeric porous networks capable of retaining water within their structure which make them generally soft materials. One of the main characteristics of hydrogels is their volume change ability enabled by the change of hydrophilicity to release their water content. This volume change notably manifests upon changes to their hydration [34] [33], temperature [35] [43] or pH [44] [32]. With this volume changing and water release ability, during their actuations, hydrogels change mass and volume isotropically, resulting in a change of stiffness, opacity and position of the hydrogel boundaries. It is this ability to absorb water or biological fluids that enable their application to biomedical fields such as tissue scaffolds or in vivo robots.

Adding to their volume change ability, hydrogels benefit from a high tunability to their sensitivity and responsiveness. This tunability is achieved by modifying their polymeric network in particular by changing the crosslinker density [45] [32], by inserting copolymers [46] [47], or dispersing additives [48] [49]. These incorporations can enhance their functionality, allowing for added sensitivity or improved functionalities [50]. The addition of complementary materials can also improve the mechanical strength of hydrogels [51] [52], addressing one of their primary limitations, or also introduce new functionalities such as remote activation using infra-red absorbing particles [53] [54] or magnetic heating and locomotion using magnetic particles as mentioned in section 2.1.1.

The fabrication of hydrogel structures can be achieved through various advanced techniques, each offering unique advantages. Photolithography allows for precise patterning and microstructuring of 2D structures [55] [56], while 3D printing enables the fabrication of volumetric structures [57] [28]. Other fabrication methods providing fine control over the micro- and nanostructure of hydrogels include ionoprinting where ions from an oxidation reaction are trapped in a hydrogel sandwiched by two electrodes and then reduced within the hydrogel [51], and electrospinning consisting of a flow of hydrogel fibres delivered by a syringe and driven by a voltage towards a substrate [58].

Hydrogels find extensive applications in the biomedical field, owing to their tunability and biocompatibility. They are used in drug delivery systems, where their ability to respond to environmental stimuli ensures controlled and sustained release of therapeutic drugs [31] [53]. Self-folding hydrogel structures can be designed to change shape in response to specific triggers [35] [55], making them useful for minimally invasive surgeries. Such example includes the work of D'Argente et al. where a hydrogel patch self-deployment was demonstrated for the patching and plugging of wounds in the stomach (Figure 2.2)[34].

In conclusion, hydrogels represent a highly versatile and promising class of smart materials with significant potential in biomedical and self-folding applications. Their tunability,

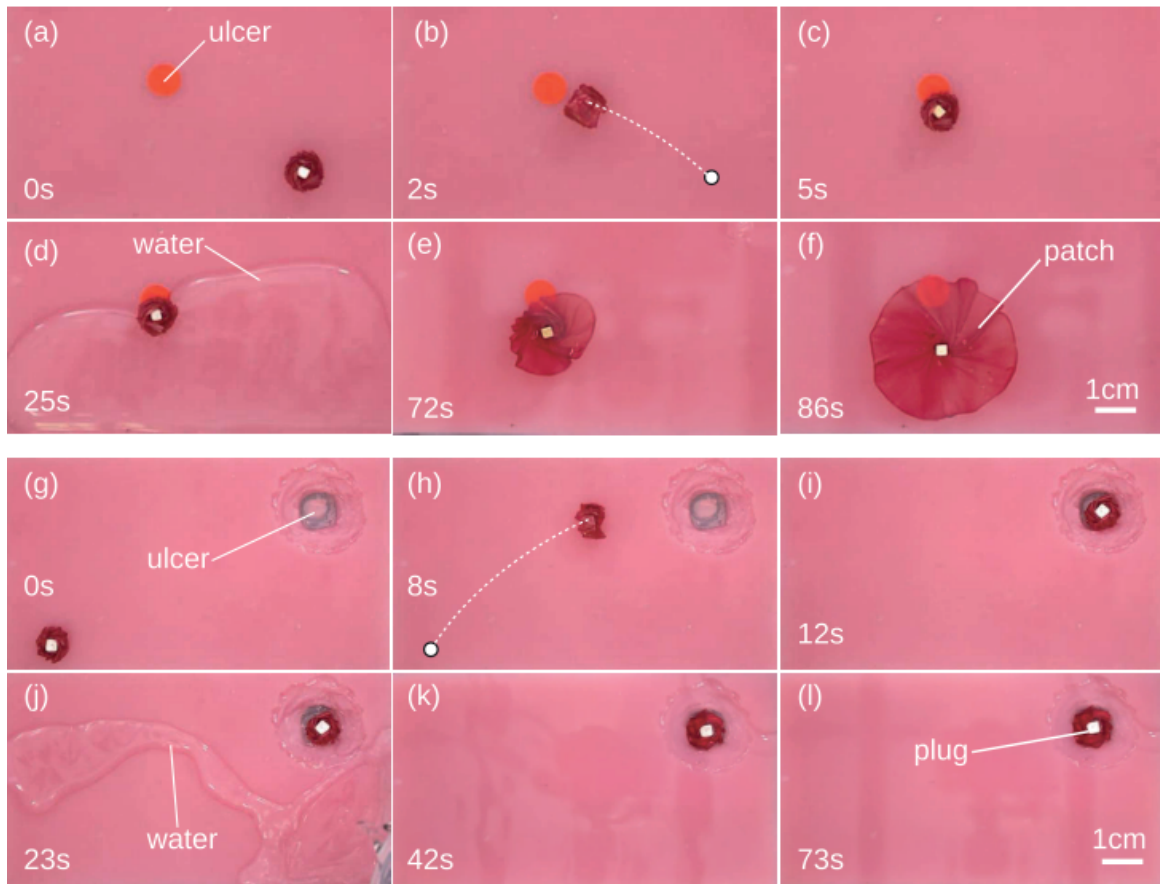


Fig. 2.2 Hydrogel patch navigating within a stomach phantom using an external magnetic field and deploying to patch or plug a wound. Reprinted from IEEE 2018 [34].

softness, and biocompatibility make them ideal for interfacing with biological tissues and developing advanced therapeutic systems. However, despite their many advantages, hydrogels also present some drawbacks. One significant limitation is their relatively slow dynamic response compared to other smart materials. Although not necessarily the case of hydrogels with particles additives, the volume change dynamic of bulk hydrogels depend on the dynamic of their environment, meaning the response speed of a hydrogel in a bath is tied to the heating rate of that bath. Additionally, while hydrogels are generally soft, some applications may require higher stiffness or mechanical strength, necessitating network modifications or the incorporation of reinforcing materials. For these applications, Shape Memory Alloys (SMA), Shape Memory Polymers (SMP) or piezoelectric materials are generally preferred.

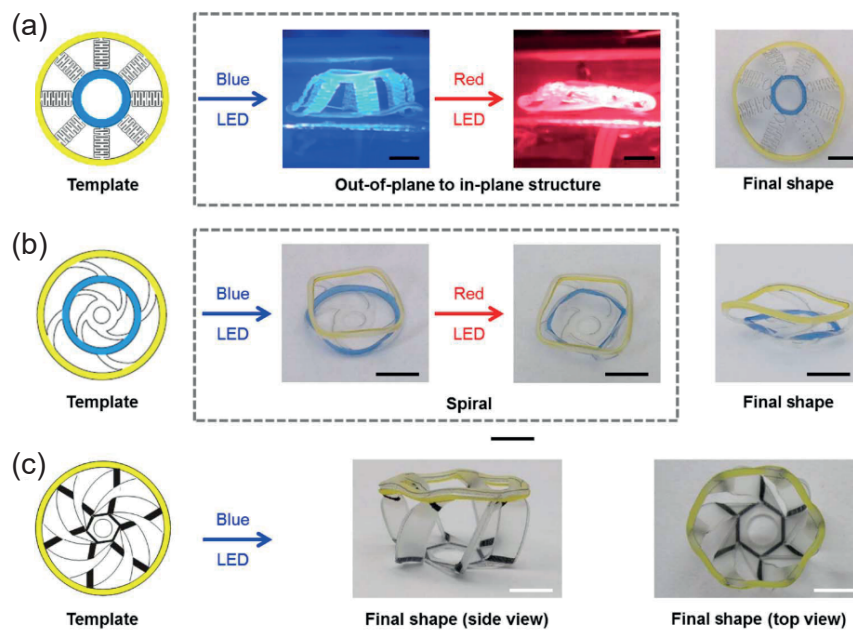


Fig. 2.3 Volume change of polystyrene shape memory polymer using light absorption of different colours to activate regions locally and enable multiple shape change (a) (b) and (c). Reprinted with permissions from Science Advances [39].

Shape Memory Polymers (SMP)

Shape memory polymers (SMPs) are a class of smart materials able to revert to a pre-defined shape after deformation when subjected to an external stimulus. The programming process generally involves deforming the polymer into a temporary shape at a temperature above its glass transition temperature (T_g) or melting point (T_m), then cooling it down to fix this temporary shape and, upon exposure to a temperature exceeding T_g or T_m , the SMP revert to its initial and permanent shape [59] [60]. Once their anisotropic volume change triggered, the SMP changes stiffness, opacity and position of its boundaries but keep a constant mass. This ability to be programmed and reprogrammed by thermal and deformation cycles is fundamental to the versatility and wide applicability of SMPs.

The main activation trigger for SMPs is temperature related [61]. Depending on the polymer used, this activation temperature varies greatly from 30°C for poly-vinyl acetate to 70°C for polyethylene-terephthalate SMPs [62]. This temperature can be adjusted using plasticizer, which can be added during the synthesis [63] or immersed in a solvent [64]. For example, water behaves as a plasticizer for certain polymers, lowering their activation temperatures and allowing their use for biomedical applications [65]. Additionally, SMPs are widely available in the market, facilitating their utilisation. Their lightweight nature

(compared to other smart materials), flexibility and the ease with which they can be processed and fabricated into complex shapes further enhance their appeal.

SMPs have found applications across a wide range of fields because their unique properties. In the biomedical sector, they are used for minimally invasive surgical devices, such as stents [66] [67] and sutures [68], while in robotics, their volume change can be incorporated to regulate the stiffness of actuators [69] or be functionalised for self-folding [70]. As demonstrated by Liu et al., the shape deformation could be accurately programmed and controlled by placing color patches onto the regions to trigger and, illuminating them with specific wavelength, resulted in an addressable shape deformation (Figure 2.3 (a) to (c)) [39].

In conclusion, shape memory polymers offer a number of advantages such as sensitivity, affordability, and market availability. However, SMPs main limitation lies in their need for manual reprogramming through mechanical deformation to reset their shape memory capabilities after each use, which limit their practicality for repetitive processes. Recent advances overcame this limitation by introducing copolymers within the initial polymer network, resulting in a material with several transition temperature and fully reversible behaviour [71] [72], but their availability on the market is still limited.

Shape Memory Alloys (SMA)

Similarly to reversible SMPs, Shape memory alloys (SMAs) share the same property of being able to morph back and forth between their mechanically programmed shapes upon exposure to a certain temperature.

SMAs exhibit two distinct phases: martensite, present at lower temperatures and easily deformed, and austenite, which appears at higher temperatures and is responsible for the material's original shape [74]. There are two types of SMAs depending on their activation methods: thermal SMAs and magnetic SMAs [73] (Figure 2.4 (a)). Thermal SMAs are activated by temperature changes, transitioning between their martensitic and austenitic phases. In contrast, magnetic SMAs undergo a crystal reorientation in response to a magnetic field, which can induce a shape change without significant temperature variation. In both cases, the volume change is anisotropic due to the crystal orientation and results into a change of mechanical properties and position of the material boundaries, which can be programmed using the memory effect.

One of the most remarkable properties of SMAs is their ability to undergo large strains, which allows for significant deformation and precise actuation, and can generate substantial forces during their phase transformation, making them capable of performing mechanical work [73] [75]. Unlike SMPs, SMAs exhibit a reversible volume change during their

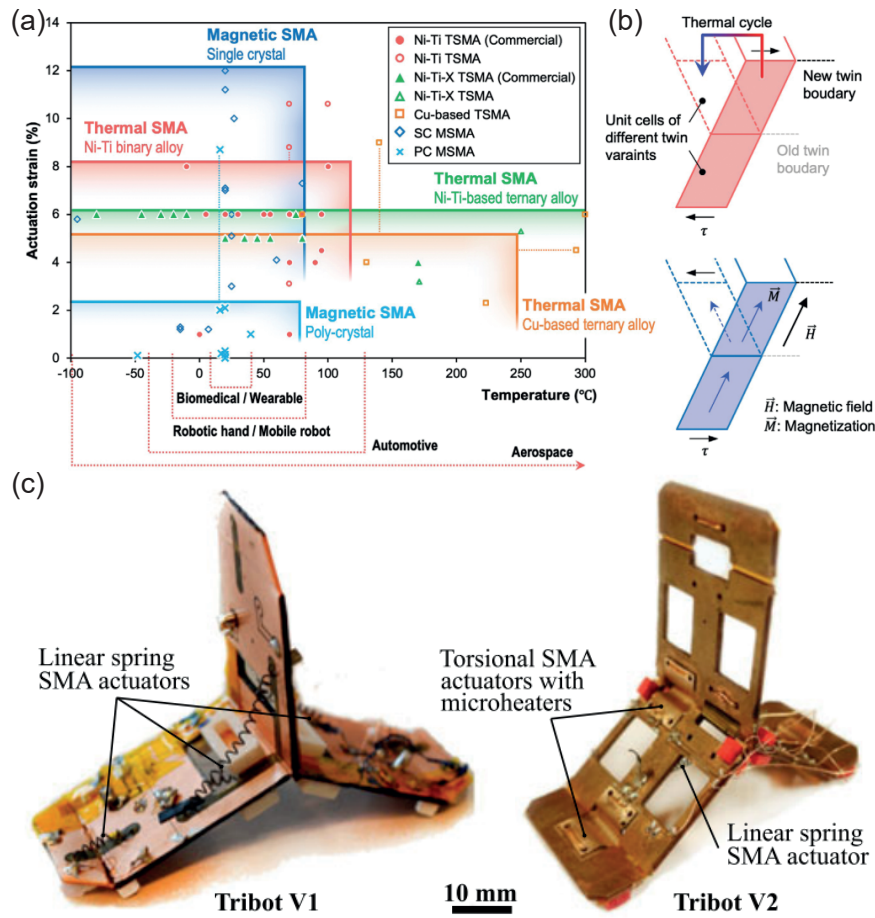


Fig. 2.4 (a) Summary of the strain developed by thermal and magnetic Shape Memory Alloys as a function of the temperature. (b) Physical principle of the shape memory effect of thermal and magnetic Shape Memory Alloys. Reprinted with permission from Wiley-VCH GmbH [73]. (c) Application of 1D Shape Memory Alloys springs and plates for a jumping robot. Reprinted with permission from IEEE 2018 [37].

phase transitions, ensuring consistent performance over repeated cycles without the need for reprogramming.

Despite their advantageous properties, SMAs face several limitations. One of the main challenges they face is their processing difficulty. Although the activation temperature of the SMA can easily be tuned by changing the proportion of metal in the alloy, small variation can exhibit large shift of activation temperature. For example, changing the proportion of Nickel to Titanium in Nitinol SMA by 1%, varies the activation temperature by 83°C, thus making the fabrication of SMAs delicate. SMA actuators cannot be given any shape as they are generally sold as wires that can be introduced in structures directly as actuators [76] [77], woven [78] [79] or knitted [80] [81] or as a filler in a matrix [82] [83]. In addition,

SMA are limited to temperature and magnetic activation, which deprives the responsiveness adaptability of other smart materials such as hydrogels. Finally, the actuation dynamics of SMA, which involve heating and cooling cycles, is slower with increased volume due to heat dissipation limitations, affecting their responsiveness in high-speed applications notably at larger scales [73].

The unique properties of SMA make them highly suitable for various robotic applications. For robotic grippers, SMA actuators provide large opening amplitude and high prehension force both at micro- [84] [85] and macro-scale [86] [87]. SMA are also implemented within robotic stents and endoscope for biomedical applications [30] [88] or robots for locomotion purpose (Figure 2.4 (c)).

Despite these limitations, SMA are widely studied and integrated in innovative applications thanks to their ability to undergo large, reversible deformations and generate large forces. Future research focusing on improving the manufacturability, increasing the deformation of magnetic SMA, and enhancing actuation dynamics will further broaden the scope of SMA in advanced robotic systems.

Piezoelectric

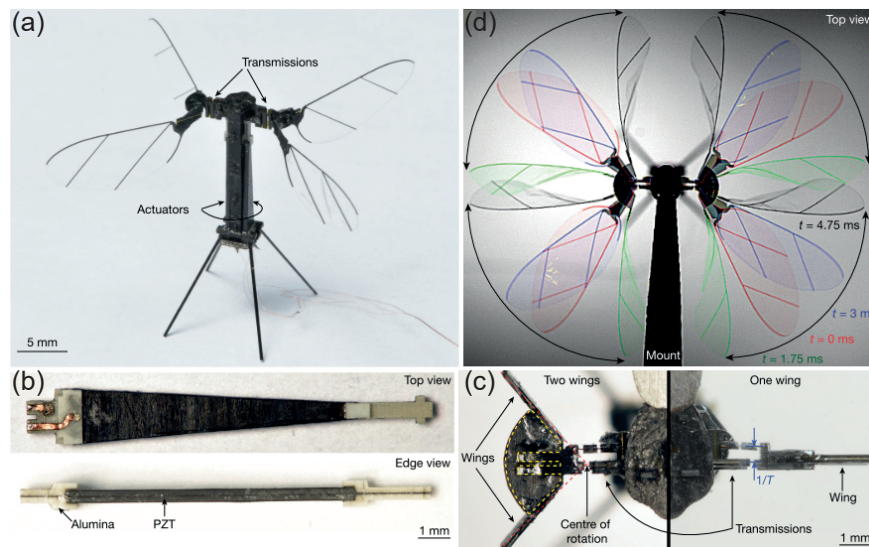


Fig. 2.5 Flying robot (a) with embedded piezoelectric smart material (b) that actuate a push/pull mechanism (c) oscillating the wings for lift up (d). Reprinted with permission from Springer Nature [89].

Piezoelectric materials are smart materials that generate an electric charge in response to mechanical stress and conversely, deform when an electric field is applied. The piezoelectric effect is intrinsic to certain crystalline structures where the centres of positive and negative

charges do not coincide, creating an electric dipole. This results in an anisotropic deformation where the volume change is predominant in the direction of the electrical offset, resulting in an elongation in that direction and compression in the others. The reversibility of the electromechanical coupling is the foundation of their use in sensing, actuation and energy harvesting applications [90].

Piezoelectric materials exhibit several key properties that make them highly suitable for robotic applications. They have a high-frequency response, which enables them to react and adjust quickly to changes [91]. Additionally, piezoelectric materials can operate in a wide range of environments including aquatic [92] [93] and aerial [94] [95]. Their ability to produce precise and fine movements with high accuracy further enhances their suitability for applications requiring delicate manipulations and adjustments notably at micro-scale when applied to micro-tweezers [91] [96].

Despite their advantageous properties, piezoelectric materials have limitations that affect their broader application. One significant limitation is their brittleness, especially in ceramics, which can lead to fracture under high mechanical stress or impact. This brittleness necessitates careful handling, especially when applied to energy harvesting where the environment interaction can be erratic [97]. Furthermore, piezoelectric materials often require high driving voltages to achieve significant actuation [98], which can complicate their implementation in low-power or compact systems.

In robotics, piezoelectric materials are employed in various applications including actuation and sensing due to their precise control capabilities and responsiveness. They are used in micro-robots actuators [99] [89] as illustrated in Figure 2.5. In addition, they can be utilised in medical devices for single neuron activation [69] and cell sorting [100], but also to achieve sensors for mechanical characterisation of tissues [101] or the detection of pesticides [102] within microfluidic systems. Their use in energy harvesting systems also enables the development of self-powered sensors and devices [103] [104] [105], improving the autonomy and efficiency of robotic systems.

Piezoelectric materials play an essential role in the advancement of robotics, offering unique capabilities such as high-frequency response, precise control and ability to operate in various environments that enhance the functionality and precision of robotic systems. However, challenges such as brittleness, high driving voltages, and manufacturing complexities must be addressed to fully exploit their potential. Continued research and development efforts are focused on improving the durability, efficiency, and integration of piezoelectric materials, paving the way for more sophisticated and capable robotic systems.

2.2 Functionalised deformations enabled by smart materials

To functionalise smart materials, their deformations are utilised to create specific motions and accomplish tasks.

These deformations can be described as continuous deformations, defined as deformation taking place along every point of the object which can be obtained by stretching, torsion, bending, buckling or a combination of these. For example, when bending an object, the curvature can be calculated on every point of the deformation as the ratio of the bending angle over a given length portion of the object. Conversely, discontinuous deformations are characterised by articulations or hinges, specific points of rotation that can be pinpointed along the object. For instance, a shoelace or a flag deforms continuously whereas a chain or an origami deform in a discontinuous way. Regardless of the continuity or discontinuity of the deformation, the resulting motion from the smart material volume change can be in a 1D (linear), 2D (planar) or 3D (volumetric) space. These motions can be achieved directly from the volume change of smart materials or by their introduction in more complex structures.

The various mechanisms to induce the aforementioned deformations are presented in this section to illustrate the utility of smart material and their capabilities in such mechanisms.

2.2.1 1D Motion

1D linear actuators convert energy into linear motion through various mechanisms, regardless of the specific smart material used. To effectively utilise these mechanisms for 1D linear actuation, various configurations are employed. The choice of configuration depends on factors such as the desired range of motion, the force output, and the application-specific requirements. Their motion can be designated as one way actuation or two way actuation. One way actuation refers to the capability to translate linearly in one direction upon activation and return to its original position either passively or through an external resetting mechanism. On the opposite, two way actuation involves the system's ability to move back and forth along a single axis under cyclic stimuli without the need for an external resetting mechanism. In both cases, common configurations include wires and springs, however this actuation is generally limited to smart materials with large deformations and large forces, notably SMA.

Straight wires or rods made from smart materials can be used as linear actuators. When the underlying mechanism is activated, the wire length changes, resulting in linear motion. This simple configuration is often used in applications where precise and predictable linear movement is required, such as in microactuators and precision positioning systems. Alter-

natively, springs, particularly helical springs, can amplify the stroke length of the actuator allowing for greater deformation than a straight wire [73].

In the work of Onal et al., such mechanisms were implemented to enable the walking of a robotic worm. Using the high compressibility and negative Poisson's ratio of various origami pattern including the box pleat or Yoshimura crease patterns, SMA springs were placed from edge to edge of the crease patterns and their shrinking, activated through Joule effect, enabled the robot to move on linearly [26]. Another example of linear actuator has been utilised in microfluidic systems where the remote activation of a magnetically sensitive hydrogel beam could control the flow of the pipe by shrinking using hysteresis heating [15]. Applied to a robotic endoscopic capsule, SMA wires are able to control the release of drugs when reaching targeted locations [106] [107]. Such linear actuation were also functionalised as self-suturing systems as exemplified by A. Lendlein and R. Langer who engineered an SMP to match in vivo conditions for biocompatibility and biodegradability, and, once made into a string, was sutured around a wound which closed gradually with heat [68].

Although, the deformation and the force produced by single wires can be limited, their bundle reinforce their force and stiffness potential [108] [109]. It is also noteworthy to mention that the implementation of such wires inside composite structures or woven enable bending, or torsion to be produced.

2.2.2 2D Motion

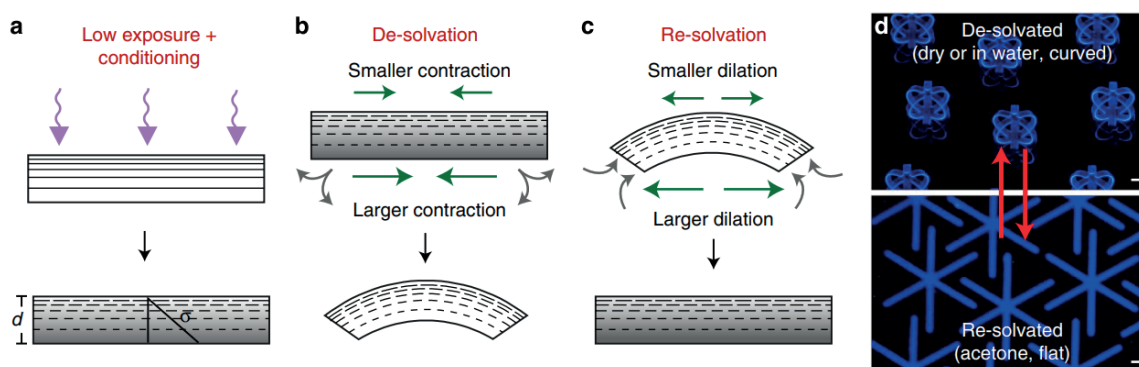


Fig. 2.6 Frontal photocuring of SU-8 epoxy resin (a) to induce a crosslinking gradient to bend reversibly micro-structures in acetone (b) and (c), demonstrated on the bending of stars into spheres (d). Reprinted with permission from Springer Nature [110].

Bending

Bending motion involves the deformation of a material into a curved shape around an axis. Mechanically, this process entails differential strain across the material's cross-section: one side experiences compression, while the opposite side undergoes tension, with an unstressed neutral axis in between. This type of deformation can be induced by applying mechanical loads such as moments or distributed forces, or conversely, can be induced when one of the portion of the cross-section experiences compression or tension. To do so, the most common approach consists of laminating two materials with different mechanical properties such as Young's modulus, thickness or volume change when exposed to a certain stimulus. The result is a bilayer structure that bends when subjected to stimuli (thermal [84] [28] [87], pH [9], hydration [33] [111], ...) due to the differential expansion/contraction, stiffness or strain mismatch around the neutral axis, creating a bending moment and causing the composite structure to curve. This approach is notably used with piezoelectric smart materials which, due to their miniaturisation potential, can be fabricated at small scale. A famous example is that of Jafferis et al. who integrated two of these actuators, each actuating a wing of a flying robot through a push/pull mechanism [89] as shown before in Figure 2.5. Another approach is to implement the wire actuator within a matrix. Depending on the stiffness profile of the matrix and the placement of the SMA wire actuator, various deformations can be obtained ranging from a simple one way bending to two way torsion and composition of bending and torsion when exposed to temperature [82]. Such behaviour can also be obtained using hydrogels with different stiffness. The mismatch in the activation and swelling range, result in one layer being "more" passive than the other and in the bending of the structure [31] to the point where several limbs can be activated sequentially in different temperature ranges [47].

Another approach consists of altering locally the stiffness of a monolayer smart material partly through the thickness. As a result, the gradient of stiffness within the monolayer and hence, gradient of strain upon volume change of the smart material induces a bending motion. This can be accomplished either by inserting particle additives within the smart material monolayer [112] [113] or by increasing locally the crosslinking density of the material [114], both resulting in reinforced stiffness. As illustrated in Figure 2.6, the gradient of crosslinking can result in the reversible bending of SU-8 micro-structures when immersed or removed from their acetone bath [110]. When the material is activated, regions with different densities expand or contract at different rates causing the structure to shift between different shapes depending on the exposure time to the stimuli [113]. To introduce a gradient of density, He et al. inserted graphene oxide within a liquid uncured hydrogel which they centrifugated, and depending on the rotation speed, created a gradient of dispersion of the particles within

the thickness of the material, thus resulting in the bending of their actuators [112]. Another approach was utilised by Zhao et al. where a polymer was cured gradually using a directed light. For example, when cured from the bottom, the polymer had an increased stiffness at the bottom and low stiffness at the top, resulting in a bending motion when exposed to its stimulus. Continuing on this concept, they introduced photomasks with gray regions to tune the crosslinking density along the structures, leading to fully crosslinked region bending small extents and lightly crosslinked regions able to fold [114].

An alternative approach consists of programming a monolayer into a bent shape. This can be accomplished during the programming of memory effect of SMAs and SMPs but can also be introduced in magnetic composites by programming their magnetisation [4]. This phenomenon was studied theoretically to predict the deformation of a magnetic composite subjected to a magnetic field [6] and applied to robotics enabling multiple shape deformations for drug delivery [2] or multimodal locomotions [14]. Extending on this methodology, folding could be achieved by magnetising a magnetic composite with sharp deformations, enabling the production of origami-like structures [11] and Halbach arrays [7].

Folding

Folding motion, from a mechanical standpoint, involves a rotational deformation of a material along predetermined axes resulting in joints forming a crease pattern. This type of deformation differs from bending, since it typically involves sharp angular changes rather than smooth curves. Consequently, the line between bending and folding can be subject to a person's perspective. Here, my definition of folding relies on three arguments: (1) a deformation introducing a discontinuity in the overall deformation of an object, (2) an overall deformation corresponding to a section with a curved deformation (the hinges) separating non-deforming sections and (3) the curving sections are much shorter than the non-deforming sections.

Smart materials can achieve controlled folding through various approaches, including bilayer and trilayer structures, as well as articulated systems. These methods leverage the intrinsic properties of smart materials to create hinge-like regions or differential strains that result in folding along specific lines.

Bilayer structures for folding consist of two bonded layers, one being a smart materials and the other being a rigid non-deforming layer split in two in the width. Upon activation of the smart material layer, the rigid sections collide and induce a rotation motion along the split line, designated as a hinge. This principle was used by Iwata et al. to self-fold a roll, a croissant and a helix owing to the interface between poly(N- isopropylacrylamide-co-acrylic acid hydrogel (pNIPAM-AAC) and rigid SU-8 pallets (Figure 2.7 (a) to (c)) [35]. Although

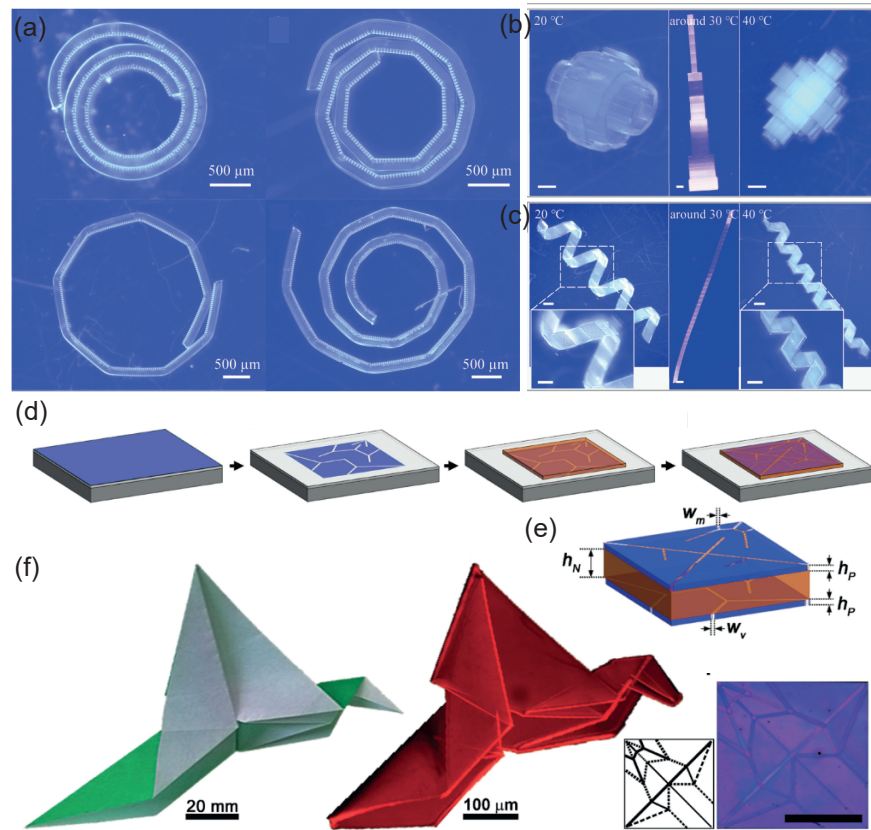


Fig. 2.7 pNIPAM hydrogel/SU8 bilayer inducing folding to create a rolling strip (a), a croissant (b), and a helix (c). Reprinted and modified with permissions from Journal of Micromechanics and Microengineering [35]. pNIPAM hydrogel/PpMS trilayer patterned using photoexposure (d) to induce folding at different angles by tuning the thickness of the layers and width of the gaps (e) and fold a crane (f). Reprinted with permission from Wiley-VCH GmbH [115].

a simple approach at folding, this design folds until the smart material expanded its stored energy, implying that the only solution to regulate the folding angle is to control accurately the smart material energy stored or its activation extent. Such implementations include the miniature star shaped robot by Breger et al. which consists of a rigid layer of polypropylene fumarate and poly(N-isopropylacrylamide-co-acrylic acid) (pNIPAM-AAc) smart material [5]. Boncheva et al. cut topological equivalent of 3D shapes out of an elastomeric sheet and embedded thick magnets around the hinges [12] and, upon folding, these magnets collided, limiting the folding to a predetermined angle.

Allowing for more complex and controlled folding patterns, and enabling precise angular changes, a tri-layer configuration can be used. Tri-layer structures are build upon the bilayer concept by adding a third rigid layer resulting in a sandwich rigid - smart material - rigid layer. While the first rigid layer split is as thin as possible to create the hinge, the second rigid

layer is shorter on each side of it, creating a gap of a certain width. Using this configuration, Tolley et al. and An et al. demonstrated that the thicknesses of the layers as well as the gap width were directly influencing the maximum achievable angle [116] [117]. As a result, Tolley et al. could self-fold accurately by programming the target folding angle within the structural design of their origami. A notable development upon this configuration is that of Na et al. who created a self-folding tri-layer structure less than $20\mu\text{m}$ thick and patterning rigid and smart material using photolithography techniques and demonstrated the folding of a crane (Figure 2.7 (d) to (f)) [115]. Expanding upon the trilayer configuration, Miyashita et al. developed a self-folding origami robot with an embedded magnet [118]. Coupled with the self-folded origami functionalities, this robot was able to swim and walk while being remotely controlled by an external magnetic field.

A third approach to folding systems pertains to articulated systems [119]. In these systems, embedded actuators are used to induce the structure folding along prefabricated joints. This approach mainly relies on the miniaturisation of rotational joints and their actuation. Ma et al. demonstrated the rotation of millimetre-sized rigid panels using the pull force of magnets to fold a box [25] and, Iwase et al. demonstrated the sequential actuation of pivoting rigid panels $400\mu\text{m}$ by tuning the amount of magnetic material enabling the actuation one panel at a time [24]. This type of rotating mechanisms is notably seen at micro-scale for micro mirrors and their use for optical filters and MEMS [120] [121] [37].

A fourth approach to induce the folding consists of selectively activating regions of a monolayer material. This activation, if not complete through the thickness of the material, creates a strain mismatch between the activated and inactivated region causing the material to fold. This method notably include pop-up techniques shown to fold micro-scale structures such as meta-material optical filters reactive ion etching [122]. At a centimetre scale, Ryu et al. introduced programmed the stress gradient within a shape memory polymer by activating specific area and applying a given strain. Baker et al. introduced particles within the hydrogel using redox chemistry. By inserting a hydrogel network between two electrodes and applying a voltage, the electrode ions migrated inside the network and remained trapped within. This resulted in a gradient of stiffness within the thickness of the hydrogel tuned with the voltage, the electrode metal and time of exposure [51]. Consequently, the storage of stress is programmed in the crosslinked area and, upon activation of the SMP, the stress release leads to the folding of the structure [59].

Overall, folding is a popular methodology with significant thrust at an algorithmic level to help further the designs of origami folding structures [123] [124] and embed electronic components for the functionalisation of self-folding structure [125]. This thrust is also seen at a technical level with the implementation of self-folding structures at small-scale for

biomedical applications to create deployable structures patching wounds in vivo [34] [29] or to deploy solar panels in space [126].

2.2.3 3D Motion

Torsion

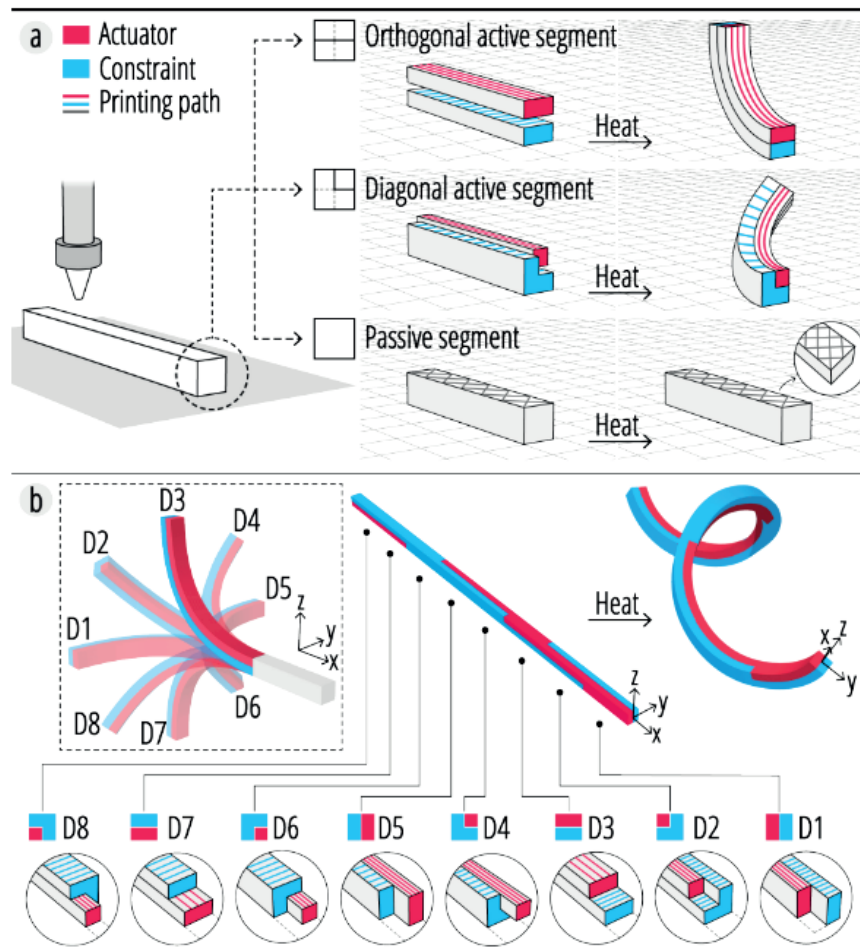


Fig. 2.8 3D printing of a beam of PLA which can be discretised in four quadrants to enable the deformation of the beam (a). The resulting bending and torsional deformations from various patterns (b). Reprinted with permissions from ACM Press [127].

Torsion motion involves the twisting of an object around its longitudinal axis, resulting in rotational deformation. This type of deformation is characterised by shear stress distributed along the material's cross-section, with one end of the material typically held fixed while the other end is rotated. Smart materials can achieve controlled torsion through several methods, including angled smart materials, offset actuators, and shape programming techniques. These

approaches exploit the intrinsic properties of smart materials to create differential strains or programmed responses that result in torsional deformation.

Angled smart materials are designed with internal structures oriented at specific angles to the longitudinal axis [39] or lateral axis [128]. When these materials are activated by stimuli, the differential expansion or contraction along these angled structures induces a twisting motion. The precise orientation and design of these internal angles determine the degree and direction of the torsion. An noteworthy example is that of Wang et al. who used the anisotropic thermal expansion of PLA, programmed the orientation of PLA lines using a 3D printer and produced various shape changing structures (Figure 2.8) including forceps, springs, a jumping frog toy [127].

Offset actuators achieve torsion by having regions of the material displaced relative to each other the neutral axis of the deforming object [73]. When activated, these offset actuators expand or contract differently, generating a twisting force around the longitudinal axis. This method can be implemented by embedding responsive elements within a matrix material [129] or by structuring the material itself to have non-uniform properties along its length [82].

Pre-programmed torsion deformation involves the use of shape memory smart materials, or magnetically responsive smart materials to achieve controlled torsional movement. SMAs and SMPs can be programmed to undergo specific shape changes, including torsion, upon activation [130]. Similarly, magnetically responsive smart materials, can be programmed to twist under the influence of a magnetic field during the magnetisation process [10].

These deformations are often presented in small robotics related work, as demonstrated by Song et al. who embedded an SMA actuator in a matrix of rigid fibres to mimic turtle flipper movement and enable a turtle robot to swim [131]. Similarly, Huang et al. developed a hydrogel/magnetic helical-tailed robot, showcasing the ability to blend different smart materials to combine bending and torsion to enable the reprogramming of the tail's shape to adjust the locomotion speed [132].

Buckling

Buckling motion refers to the sudden change in shape of a structural element under compressive stress, resulting in a failure mode characterised by lateral deformation. From a mechanical standpoint, buckling occurs when compressive forces exceed a critical threshold, causing the material to lose stability and deform perpendicular to the applied load. Smart materials can achieve controlled buckling through several methods, including tessellation of smart materials, gradients of stress, gradients of stiffness, and deformation of an elastic support.

Tessellation involves arranging smart materials in a repeating pattern or lattice structure. When subjected to compressive stress, the geometric arrangement and the responsive nature of the smart materials induce controlled buckling along the tessellated lines. Owing to the lattice structure and accuracy of photolithography fabrication techniques, tessellated designs can be optimised to achieve specific mechanical responses, enabling complex and tunable buckling behaviours [56].

A gradient of activation involves selectively activating different regions of the smart material cross-section to induce controlled buckling. This can be achieved by varying the intensity or type of stimulus across the material's cross-section. Regions with higher activation levels will respond more strongly, creating differential strains that result in buckling. This method allows for precise spatial control over the buckling deformation, enabling complex and programmable shape changes. The reprogrammability of this technique was demonstrated by Guo et al. on a hydrogel sheet by using a redox reaction and stamping technique to introduce locally, and remove globally, the iron oxide particles used to buckle the hydrogel through infrared light [54].

A gradient of stiffness within a smart material can be used to control buckling by varying the material's rigidity across its structure. By designing regions with different stiffness, the material can be made to buckle preferentially in the less stiff areas when subjected to the compressive forces of shrinking smart materials. This method can notably be implemented through material composition, as did Therien-Aubin et al. who copolymerised several polymers using photolithography to tessellate stiffness and volume change response of the smart material [55].

Using an elastic support to induce buckling involves integrating smart materials with elastic substrates or frameworks. When the smart material is activated, it interacts with the elastic support, causing the combined structure to buckle. This method allows for controlled and reversible buckling, as the elastic support can return to its original shape when the activating stimulus is removed [49].

2.3 Assembly of smart structures

Artificial materials can be fabricated using either a bottom-up or a top-down approaches. The bottom-up approach builds matter, whereas the top-down approach deconstructs matter into a specific shape. Depending on the scale and process, one approach or a combination of both can be used. This section reviews the different bottom-up techniques for creating artificial matter.

2.3.1 From 0D elements

Concept of module

Modular assembly consists of the manual or self-assembly of small elements (comparable to unit cells in matter) into 1D, 2D, or 3D structures. These small elements could be considered as 0D; however, the literature suggests that 0D refers to the self-assembly of nanoparticles.

Manual Assembly

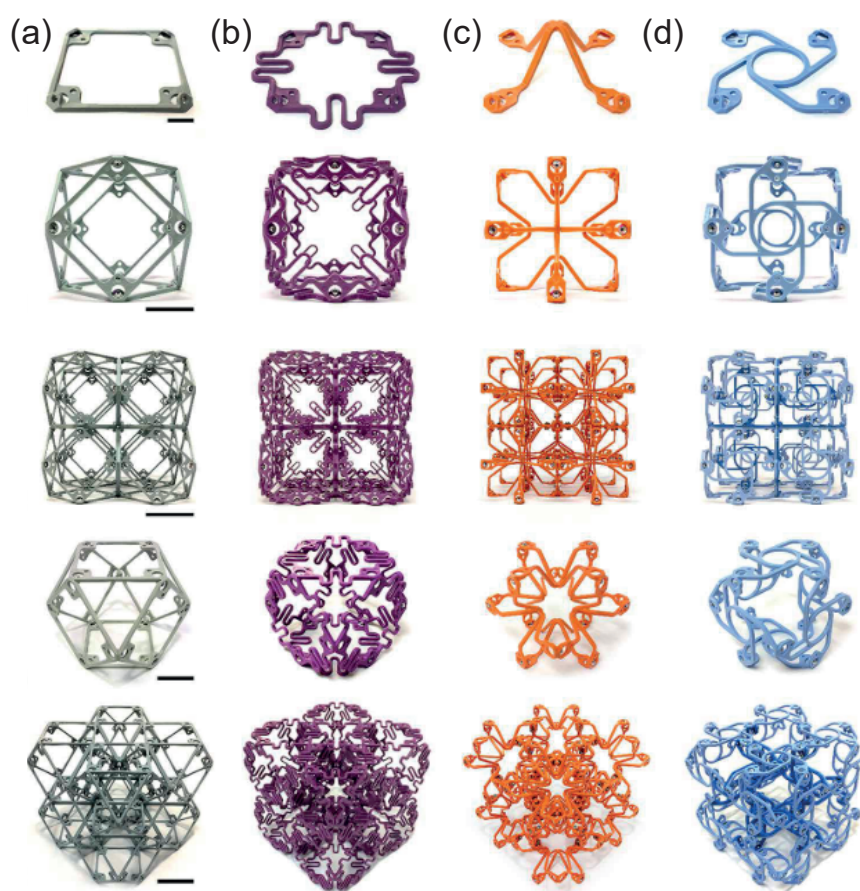


Fig. 2.9 Manual assembly of patterned plates to form voxels with mechanical metamaterial properties ranging from rigid (a), compliant (b), auxetic (c) and chiral (d). Reprinted with permission from Science Advances [133].

Most constructions are formed from building blocks (unit cells) that are manually piled up and linked. At smaller scales, one approach is to manually assemble blocks using magnets, such as Geomag toys, or nested blocks, such as Lego and Meccano. Utilised by Jenett et al., blocks can be assembled together to form larger structures whose mechanical response

depends on the characteristics of the blocks used, thus demonstrated the potential to create structures tunable properties (Figure 2.9) [133]. Another approach involves folding origami-based unit cells to create polyhedrons [134] [135]. This approach results in periodic lattices that can exhibit metamaterial properties. Such is the case study of Overvelde et al. who developed multistable and foldable units able to imbricate into one another to assemble into large morphable structures [136]. To increase the yield, lower the cost of construction and enable construction in dangerous or confined environments, robots have been brought into the picture of construction. Both Formoso et al. and Cheung et al. designed and built a robot whose aim is to navigate along a modular structures and expand it unit after unit. [137] [138]. Such modular structures have been used to extend the capabilities of the existing robots. Kim et al. [139] used an extending and retracting modular origami arm to grip objects out of the reach of a drone, and Zhong et al. [140] stacked several rotary modules controlled by only one motor to extend the range of motion of the robotic arm. These modular structures were brought to the micro scale notably to create robots. An example of such a robot is Xenobot [141], a self-assembled cellular bio-robot manually carved to provide an optimum shape to navigate in its environment. Later, this cellular robot self-assembled and demonstrated self-replication and self-healing owing to the natural ability of cells to agglomerate into wide structures [142].

Self-Assembly

Self-assembly is a subfamily of modular assemblies, in which the final structure is assembled autonomously without human intervention [145]. Two approaches are possible: the modules attract each other naturally and bond (static assembly), and/or the environment is agitated to force the modules to bond [146] (dynamic assembly). Mechanisms driving the assembly of modules notably include magnetic forces where the magnets polarities attracts or repel certain modules [118] [147], or capillary effects between module which can be patterned between positive and negative meniscus [144] [148] to achieve certain topologies. One of the main objective of self-assembly is to obtain the targeted structures. For this purpose, the modules can be made smart by implementing electronics that enables them to communicate and memorise target shapes [149]. These modules can be equipped with specific dockings that enable selective self-assembly [150]. Alternatively, the environment can guide the self-assembly into different structures [144]. An example of a smart module is the work of Goldstein et al. [151], in which robotic modules capable of communicating and bonding with each other using magnetic coils were developed. Similar to jigsaw puzzles, selective self-assembly consists of an assembly where the modules have encoded ports to promote the bonding of modules with a matching pattern. Based on the concept of jigsaw puzzle, Tasoglu

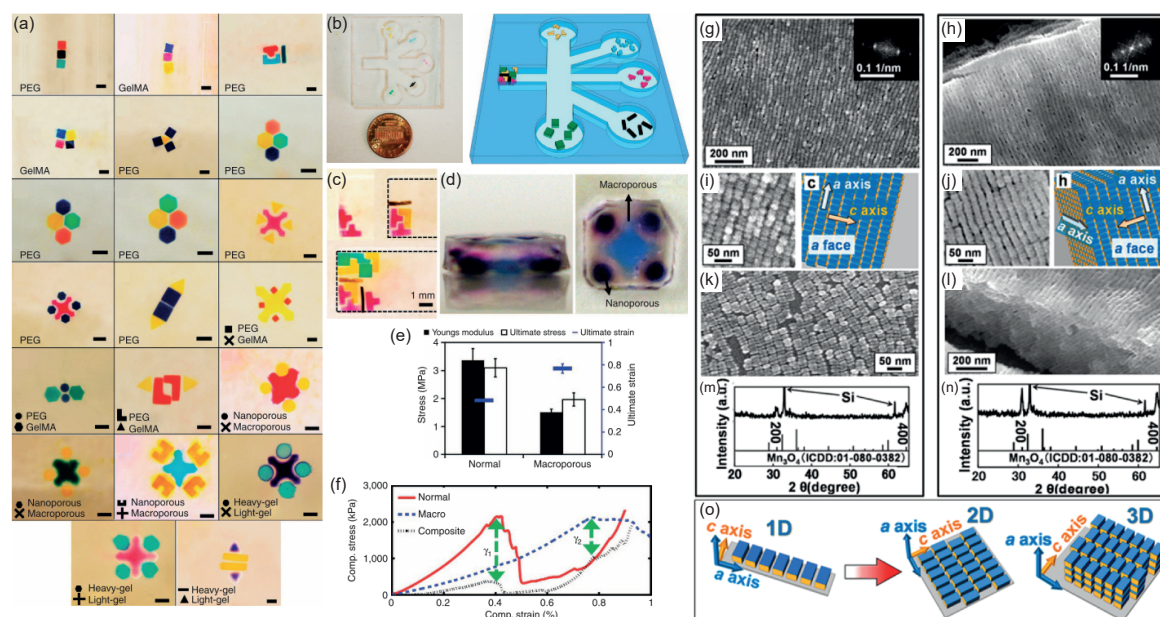


Fig. 2.10 Self-assembly directed by a magnetic field to gather together modules of different size and shape, all coming from different pools at a speed matching their magnetic content (a) to (c). These structures can be bonded together resulting in different mechanical properties depending on the modules used (d) to (f). Reprinted with permission from Nature Communications [143]. Self-assembly of 1D, 2D and 3D structures obtained by varying the evaporation speed of the medium of magnesium oxide crystals (g) to (l) along with their XRD patterns (m) and (n), and representations (o). Reprinted and adapted with permission from [144]. Copyright 2014 American Chemical Society.

et al. designed magnetic sensitive modules with matching shapes whose magnetically driven assembly could be fused together to obtain a monolithic structure instead of an agglomeration of modules (Figure 2.10 (a) to (f)) [143]. Selective assembly can also be encoded as magnetic pattern on the docking sides of the modules, enabling only modules with opposite magnetic patterns to bond [152]. Another type of self-assembly is the environmentally driven self-assembly where environmental changes dictate the assembly output and throughput. This is well highlighted by Nakagawa et al. where nanocrystals were thrown into solutions evaporating at different rates. This forced the crystals to arrange in different orientations and structures, ranging from 1D chains to multilayered 3D lattices (Figure 2.10 (g) to (o)) [144]. Another notorious example of environmental self-assembly is the study of Gracias et al. . Agitated in a heated flask, modules equipped with low melting point tin could bond together owing to capillary effects. Upon cooling, the solidifying tin guaranteed a monolithic structure able to light up due to the presence of embedded LEDs and inter-module bonding with electrical connections [153]. Several factors play important roles in the self-assembly

process. These include the density of the modules in their environment and the forces applied to the modules from each other and from the environment.

Limitations

The first and main challenge to date is yield. In the case of manual assemblies, manual work may require expert hands [141] [50] and numerous hours, making it an expensive and slow approach. In the case of dynamic self-assembly, agitation of the environment provides the energy required to properly assemble the structure but also to detach improperly attached modules, thus proofreading the assembled structure [43]. Consequently, the self-assembly time may increase drastically depending on the initial conditions and the number of elements for the target shape. The second challenge is the scaling of the self-assembly from 2D to 3D structures. Because of the nature of self-assembly forces, some approaches are inherently 2D, such as capillary effect-driven self-assembly and have not been used to demonstrate 3D structures. Some research groups have combined the principles of origami and self-folding with modular assembly to enable 2D lattices to form 3D structures [154] [155]. Modular assemblies have been applied in several fields, such as biomedical [156] and metamaterials [136] [133], but owing to the specificity of the assembling environments, their applications are limited. In addition, the probabilistic nature of self-assemblies makes them prone to misassembly, which further limits their implementation in real environments.

2.3.2 From 2D structures

Concept of origami

Origami, the art of folding square 2D paper sheets into 3D structures, has applications ranging from table-napkin decorations to the deployment of solar panels in space [126]. Although a consensus has never been reached on its definition, the meaning of origami has evolved over time to account for technological developments. In addition to the concept of folding in origami, kirigami complements folds with cuts, as demonstrated on the micro-scale by Li et al. [40], whose kirigami optical filters are cut in different shapes by locally etching the material, which induces strains and folding. The techniques employed for realising folds in origami robots depend on the length scale of the origami. At the macro scale, the incorporation of electronic components directly within origami pattern was exploited to produce origami robot faster [157]. However, this is not possible at the micro scale, which requires specific considerations regarding the embedment of actuators and their powering.

Folding algorithms

Various algorithms have been proposed for hinge patterns and folding sequences for 3D shapes observed in the literature. Robert J. Lang [158] illustrated the potential of his approach that consisted in creating a crease pattern from any volume and combining different patterns to introduce details to the structure. One example is his origami fish in which scales were added by combining the fish pattern and the scale patterns. Tachi [123] developed a program splitting the surface of a 3D shape into polygons and interconnecting them with “tucking molecules” so that, when folding, the “molecules” enter the inner volume leaving only the polygons on the surface. Benbernou et al. [124] linked together the two schools of thought by implementing an algorithm that proposed a folding sequence from universal patterns for any orthogonal 3D shapes. Rus and Tolley [159] highlighted discrepancies between algorithms and origami robots because algorithms ignore the difficulty of fabricating complex crease patterns, the material thickness during modelling and the necessity of electromechanical components, resulting in origami that are too difficult to fabricate, fold or actuate. Smart materials at the sub millimetre scale tend to be used instead of electromechanical components for remote folding. Given that complex origami shapes require large sheets and sequential actuation of numerous creases, the smart materials driving these creases must exhibit tightly controlled performance characteristics such that the creases fold in the desired manner. This was demonstrated by Iwase et al. with micro-scale panels interlocking sequentially by tuning the magnetic field strength and the sensitivity of each panel [24]. Such well-defined performance characteristics are difficult to achieve because of the challenges associated with the modelling and fabrication of smart materials, particularly in terms of the precision and accuracy of the placement of the materials and uniformity of their response.

Applications

Various origami structures have been used to address robotics tasks. Onal et al. [157], demonstrated an origami robot that exhibited multiple behaviours whose implementation was capable of both legged locomotion and manipulation via grasping. Given that manipulation can be applied in a vast number of applications, there is a corresponding plethora of research on origami manipulators. For example, Ongaro et al. [160] fabricated a star-shaped thermally activated microgripper that could locomote via an external magnetic field, and Hughes et al. [161] developed an origami microgripper that could exert a firm yet delicate grasp by incorporating multiple pneumatic sensors.

Folding structures show promising potential for applications in difficult-to-access spaces, owing to their versatile reconfigurability. For in vivo clinical applications, origami robots

have demonstrated encouraging results with regard to accessing constrained environments such as blood vessels [9]) or gastrointestinal tracts [30] [29], thus paving the way for the development of a range of minimally invasive clinical methods. Although various techniques for microrobot navigation exist, many utilise external magnetic fields. Using these navigation techniques, researchers have demonstrated wound patching [34], drug delivery [31] and biopsy [5]. Breger et al. [5]) developed a star-shaped hinged micro robot driven by an external magnetic field that could be reversibly opened or closed when cooled or heated to extract and carry cells. D'Argentre et al. [34] developed a water deployable ingestible origami hydrogel patch that could be navigated to the location of an ulcer in a phantom stomach using an external magnetic field. However, most in vivo micro robots are still undergoing preclinical trials.

Limitations

Crease folding is a complex process requiring a high degree of dexterity and precision. One of the major technological advancements in origami systems in recent years is the self-folding of creases, which began with the development of folding-machines. These machines vary in complexity, ranging from simple paper plane folding/launching devices [162], to systems capable of fabricating a wider range of origami designs. For instance, Balkcom and Mason [163] devised a system that consisted of a four degree of freedom (DOF) robotic system that would orient a piece of paper over a clamp, which, in combination with a metal blade and a suction cup, was able to introduce a crease pattern through sequential folding. Namiki and Yokosawa [164] developed a bimanual robot capable of manipulating paper in a manner similar to a human; and Ishikawa et al. [165] used the DaVinci medical robot to fabricate a 2×2 cm origami crane via teleoperation. Both at large and small scales, folding origami structures remains difficult due to the delicate and dexterous manipulation of flexible materials which require robotic hands with precise motion and firm handling.

Macro-origami robots tend to rely on mechatronics for sensing and actuation and on micro controllers for computation. Onal et al. [157] developed an eight-legged origami robot actuated by tendons connected to servomotors and controlled by an onboard Arduino. In contrast, micro-origami robotic systems rely on various folding mechanisms, including smart materials, induction of strain and micro-joints. When an origami folds, torques are induced at the creases, resulting in a folding motion. For example, Tolley et al. [116] achieved simultaneous self-folding of basic structures (such as cubes, icosahedrons and Miura patterns) by uniformly applying heat. However, in addition to the limited folding torque of smart materials which limits the size of foldable origami, simultaneous folding alone limits possible folding sequences, thus resulting in a narrow range of achievable

foldable structures. This can be addressed by selectively tuning the sensitivity of the origami smart material to realise disparate folding rates by considering the crease design parameters and loading conditions. Such sequential folding, also under a globally applied stimulus, was demonstrated by Iwase and Shimoyama [24], whereby sub-millimetre-sized origami creases had different magnetic sensitivities, and Kobayashi et al. [47] who controlled the activation threshold of a heat-sensitive hydrogel to demonstrate the sequential self-folding of structures.

2.3.3 From 1D structures

Concept of chains, strings and ribbons

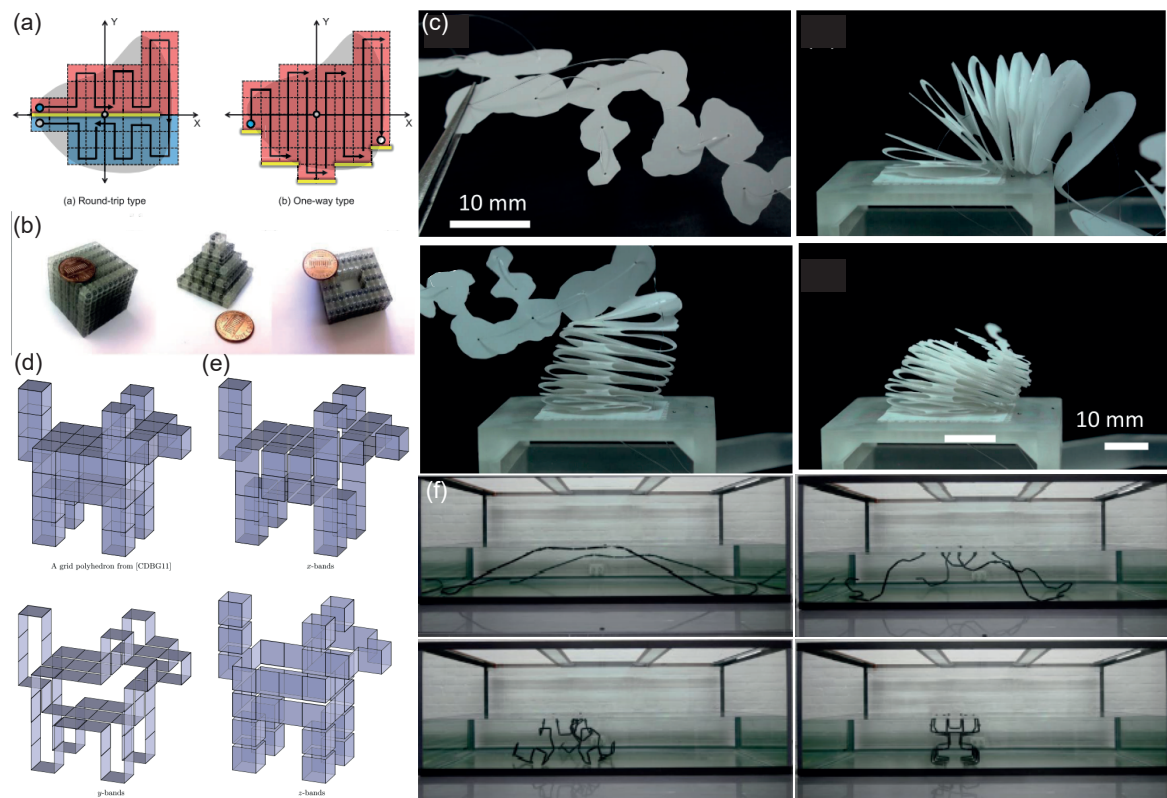


Fig. 2.11 Hamiltonian path describe to fill a given shape by putting different boundaries (a) and the resulting structures (b). Reprinted with permission from Copyright Sage Publications 2014 [166]. Chain in a straight configuration folding by superposition of the slices and of a rabbit design (c). Reprinted with permission Copyright Sage Publications 2018 [167]. Decomposition of a dog design (d) into ribbons depending on the direction of discretisation (e). Reprinted from [168] with permission from Elsevier. 4D printed hydrogel based string to fold into a cube made of eight voxels (f). Reprinted with permission from Wiley-VCH GmbH [169].

Strings, chains, and ribbons, although all classified as 1D structures, differ conceptually in their geometry. Strings are typically cylindrical and highly flexible, deforming continuously along their length. Chains consist of interconnected links, providing overall flexibility with semi-rigid or rigid links. Similarly, ribbons can deform continuously along their length, but their flatness, designated as their width-to-thickness ratio, limit their lateral deformation. Each type of 1D structure can be found in specific applications. For example, sewing, knitting and weaving rely on strings, while ribbons and chains are utilised for the transmission of force in mechanical systems.

Owing to the entanglement and the subsequent formation of knots, 1D structures can create long-lasting structures. In practice, a knot forms a stable structure either by entangling itself or around an object. However, in mathematics, a knot is “Any closed non-self-intersecting polygonal line in Euclidean space $\mathbb{R}^3$ ” [170]. Thus, a knot is an infinitely thin line that forms a loop without a start or an end. Although abstract, this definition is of importance to understand and compare the different topology of knots and the functions they enable.

Algorithms for the building of 3D shapes from 1D structures

To create any shape from a 1D structure, the first step consists of identifying how the 1D structure should fill the target shape. This concept is defined as the Hamiltonian path and consist of discretising the target shape into pixels for 2D shapes, or voxels for 3D shapes. After the discretisation of the space follows the path identification, meaning how the 1D structure is folded and intertwined in the discretised space without overlap (passing more than once by the same pixel/voxel). This methodology is an extension of G. Peano’s approach to fill of square lattices using a single line and without any overlap [171]. Although the work of G. Peano proposes a functional approach to fill squares, generalising the space filling to any shapes bring a new problem: “dead ends”, trapping the space filling line into a corner. For this purpose, S. Griffith demonstrated that increasing the resolution by dividing the square lattice into smaller sub-lattice allows the line to turn around in the “dead ends” [172]. To approach the collision problem, Cheung et al. developed a simulation method called reverse explosion. The center of mass of the target shape receives a radial force pushing the elements of the 1D structure away from the center of mass. As the target shape deploys into a line, the motion of each joint is recorded and can be reversed to fold a straight string into the final structure [173]. Despite its contribution, this study presents two main disadvantages, one being the difficulty to replicate the theoretical motion outputted by the algorithm with smart materials, the second being the complex path followed by the line which leads to a collision prone folding trajectory. Simplifying the path to reduce the probability of collisions was

achieved by Yim et al. who, in addition to discretising the space in a lattice, discretised the target shape into layers, prioritising the filling layer by layer (Figure 2.11 (a) and (b)) [166]. Continuing his work with 1D structures, S. Yim proposed to simplify again the problem of line filling spaces by slicing 3D shapes into many layers and assembling them into foldable chains. Expanding on this concept, S. Yim et al. introduced the possibility for the chain to branch into several sub-chains to allow shapes that are splitting to be reproducible with chains (Figure 2.11 (c)) [167]. On a more conceptual approach, Benbernou et al. proposed an algorithm to split the surface of a given structure into pixels, each connected to their neighbour in the desired cartesian axis. This chain of pixel could then be folded using an origami pattern proposed by the algorithm (Figure 2.11 (d) and (e)) [168].

The folding of strings, chains or ribbons are designed to avoid the collisions necessary to create knots. This can be explained by the empirical and random nature of collisions and the serial nature of 1D structures causing the propagation of unplanned motion along the whole string. Nonetheless, several field of research focus specifically on the study of collisions and self-assembly of 1D structures. This notably include the research on artificially made DNA structures [174] [175], on chain molecules to understand the mechanisms leading to the knotting of molecules [176] [177], or the simulation of knots tying to train surgeons for their sutures [178].

Type of strings and their actuation

As mentioned before, strings are mainly long and thin circular objects with uniform section, chains are long modular assemblies and ribbons are thin and long flat objects. Due to their difference in geometry, their actuation mechanisms strongly differ depending on their aspect ratio, defined as the ratio of width to thickness.

Regardless of the scale, and as depicted in Figure 5.17, the most common type of 1D structures are the chains. Owing to their limited degrees-of-freedom and the patterning of non-deformable sections and flexible joints, the trajectory can be predicted to obtain a final structure that requires the precise positioning of the modules within [166] [172]. At nano-scale this is particularly relevant for DNA strands self-assembly, where the pattern of nucleotide, namely adenine, cytosine, guanine, and thymine, should be carefully selected to guarantee their bonding and the creation of the target shapes [175]. Regarding the deformation of joints, where at small scale, the folding is mainly achieved through the use of smart materials in tri-layer configuration [179] or the shape programming by tuning the magnetisation profile [4], at larger scales, the folding can rely on more traditional elements such as motors twisting the joints of the chain to fold in a given shape [180] or rely on

rotation joints whose movement is enabled by the placement of strong magnets along the module faces and the chain [181] [43].

Strings can be found in many systems as pulleys or wires working as tendons to actuate rotational joints as seen in the work of Chen et al. where the actuation of a platform was achieved by pulling strings compressing an origami spring structure, resulting in the inclination of the platform [182]. In many macro scale applications, these strings are actuated by motors [167], but for finer structures, the strings may be directly made of a smart material, as requires Noh et al. study, where a robotic jumping flea, less the 30 mm in length, utilises an SMA spring to store and release the energy required for the flea to jump [76]. However, this type of actuation is heavily reliant on the shape memory effect, the high stiffness of the smart material and energy storage. Another approach consists of patterning different materials across the section of the string. This was achieved at micro scale by Uchida et al. [28] who developed a 4D printer able to print hydrogel bilayers through a single nozzle. For this purpose, two syringes supply the extruder simultaneously, and the flow from the syringes dictates the proportion of one hydrogel to the other, resulting in string able to shrink or bend at different extents.

On the opposite of strings, ribbons benefit from an enlarge width compared to their thickness, limiting their deformations to out-of-plane rotation perpendicular to their width. Despite their limited deformation, Kamrava et al. demonstrated that complex structures could be obtained with bistable and auxetic properties emerging from the collective work of interlocked ribbons [183]. Owing to their enlarged width, ribbons benefit from the integration of bending and folding techniques detailed in Section 2.2.2. Kamrava et al. expanded on their previous work by demonstrating that their single degree-of-freedom origami ribbon could be folded with a single actuator and applied this principle to a robotic gripper and robotic hand [184]. In the work of Janbaz et al. where they integrated hyperelastic polymer and SMP as ribbons to obtain their bending or twisting and could insert their ribbon actuator into rigid guides to enable folding [185].

Strings, ribbons, and chains: their applications

Whether a string, a ribbon or a chain, 1D structure find applications in various fields including programmable matter, robotics, and medical surgeries.

Regarding programmable matter, 1D structures possess a wider range of possible configuration, dictated by the number of viable Hamiltonian path, each of which produce different physical properties of the final shape. Although distant from 1D structures, this can be seen in the work of Holobut et al. who demonstrated how the stress within modular structures evolve with the release of inter-module connections and how it can affect robotic functionalities

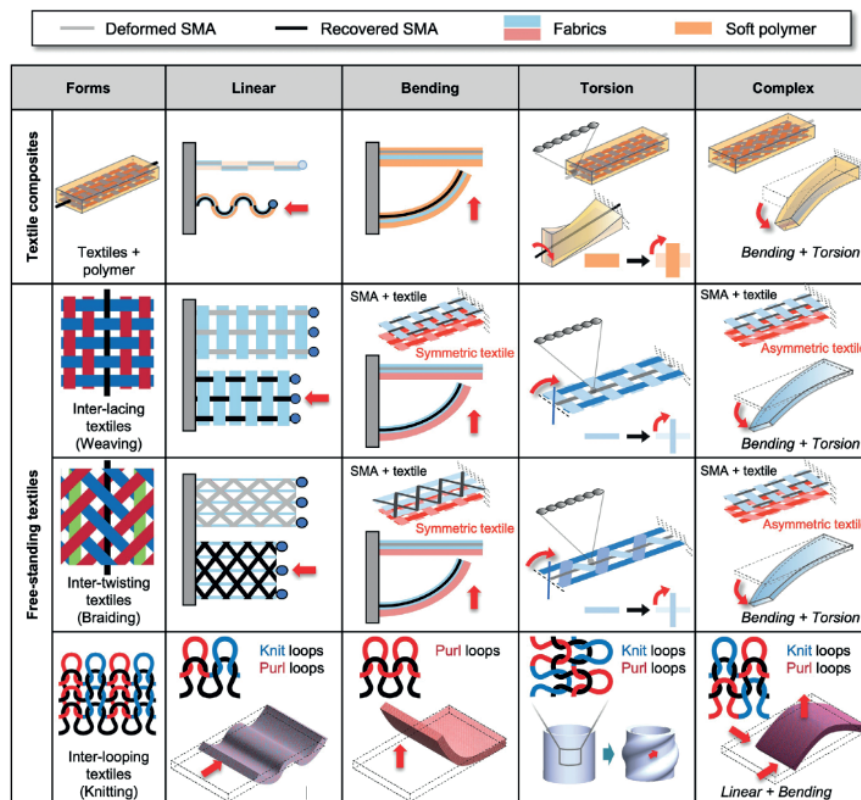


Fig. 2.12 Weaving and knotting of SMA within passive material to enable bending or torsion which can be adjusted by configuring differently the interlacing of the passive and active material. Reprinted with permission from Wiley-VCH GmbH [73].

[186]. Expanding on this, Piranda et al. studied the stress within 3D structures obtained from a modular chain and highlighted experimentally the challenge of structural integrity within modular assembly [187]. Rather than discretising the whole target shape and filling it voxel by voxel, Yim et al. [167] and Southern et al. [181] proposed to slice the target shape, form a chain from the slices and reconstruct the target shape by folding the chain, leading to semi-crystalline or spring-like structures. Another approach to make smart structures from strings consists of weaves and knits. Knitting involves creating fabric by interlocking loops of strings with needles, resulting in a flexible and stretchable material as seen in Figure 2.12 [73]. When knitting smart fabric, the material properties along with the loop geometry and the stitch style alter the mechanical properties and the shape change, resulting in anisotropic deformations, bending or even buckling [81], thus highlighting the potential of SMA smart fabric as light yet energy dense actuators [80]. Weaving involves interlacing two sets of strings at right angles (warp and weft) on a loom, producing a fabric that is generally more rigid and less elastic. To develop smart fabrics, Skatitzky et al. used a narrow fabric weaving machine to weave together SMA wires enabling the fabric to change shape depending on its

programming and change mechanical properties depending on the temperature [78]. Where large planar SMA are not viable due to their fabrication method, SMA fabrics enable the SMA to be used as large actuators. This is notably demonstrated on the deformation of wings of aerial vehicles to tune the drag coefficient and adjust the winged object's flight [79].

Within robotics, 1D structures can be found as grippers whose twisting, folding and prehension force and motion can be tuned by adjusting the folding pattern as demonstrated by Orlofsky et al. who developed different designs for folding tweezers able to grab various objects including pens, blueberries or cans and untwist bottle caps [188]. As small robots, 1D robots attract attention owing to their potential to enter narrow passages. One such example is the work of Downs et al., where a multi-responsive hydrogel robot could navigate through a narrow maze and transport a payload owing to the magnetic control and the selective responsive-ness of the hydrogel beads constituting the robot [50]. With a similar locomotion approach, Hu et al. developed a ribbon robot whose magnetisation profile could be programmed to enable multimodal locomotion such as rolling, swimming, diving and jumping.

With the ability to enter narrow openings, Gu et al. applied a self-folding chain for in vivo surgery. Their chain possesses flexible joints and rigid part embedded with magnets which fold and assemble the string into a tweezer once out of the catheter [189]. A notable application of strings in vivo pertains to suturing. With the development of new smart materials, able to carry drugs or antibacterial for example, novel sutures were developed [190]. This includes the usage of shape memory polymer engineered to be biocompatible, biodegradable and self-tightening [191] for suturing applications [68]. By programming the magnetisation of a needle and tailoring a wire behind it, Mair et al. drove the robot around to tie a knot and penetrate biological tissues for future applications for in vivo surgery [192].

Main parameters of 1D structures and their limitations

While 1D structures are diverse and their applications are especially relevant to the robotic and biomedical field, 1D structures can be severely limited. Regarding the scalability, self-folding strings suffer from limited length or number of modules of chains and self-folding torque enabled by the embedded smart materials that result in a limited length of string and volume of target shape. The motion range of 1D structures is also limited, ranging from the single degree-of-freedom of ribbon to the infinite degrees-of-freedom of strings reducing their applicability. The structural integrity of final structure along with the lack of study on physical properties enabled by the Hamiltonian path leads to final structures whether lacking the functions to enable their usage for any applications or lacking the sufficient stiffness and resilience required for their applications. Finally, the inherent

limitations from the smart materials and driving mechanisms used to fold the 1D structures is to be considered when designing the Hamiltonian path as avoiding self-collisions is a challenge both on the algorithmic side and practical side. Nonetheless, many methodologies and designs for self-folding 1D structures have been proposed tackling or approach the challenges aforementioned.

Reference	Type of string	Module Size	Module number	Torque	Medium	Mass in medium	Power Source	Folding in	Actuation Method	Assembly time	Remote folding	Un-thered	Revers-ability	Addres-sability	Reprogram-mability	Sensing	Assembly and Deformation	Application field(s)
Yim et al. [167]	Chain	3x3x3 mm	999*	$3.2e^{-2}$ Nm [†]	Guide exit	36 g	Magnets	3D	Magnetic pull/push	108 s	Yes	/	Yes	/	/	/	Self-assembly and self-folding	Medical implants
Fass et al. [189]	Chain	20x20x10 mm	6	$2.5e^{-1}$ Nm [†]	Planar surface	$>>0^{\dagger}$	Magnets	2D	Magnetic pull/push	0.2 s	Yes	Yes	Yes	/	/	/	Self-assembly and self-folding	Minimally invasive surgery
Griffith et al. [172]	Chain	25x25x3 mm	20*	$2.1e^{-1}$ Nm [†]	Air cushion	$\sim 0^{\dagger}$	Magnets + Air	2D	Magnetic pull/push onto air cushion	30 s	Yes	/	Yes	/	/	/	Self-assembly and self-folding	Programmable matter
Han et al. [43]	Chain	14Øx9 mm	20*	$2.9e^{-2}$ Nm [†]	Agitated hot water	$\sim 0^{\dagger}$	Magnets + Air + Heat	2D	Magnetic pull/push into bubbling water	10 s	Yes and No	Yes	Yes	/	/	Yes	Self-assembly and stretching	Programmable matter
Gu et al. [185]	Chain	15x5x5 mm	9*	$3.4e^{-3}$ Nm	Guide exit	$>0^{\dagger}$	Magnets	3D	Magnetic pull/push	13 s	Yes	/	Yes	/	/	/	Self-assembly and self-folding	Minimally invasive surgery
Li et al. [7]	Ribbon	4x5x0.5 mm	5	$1.42e^{-2}$ Nm [†]	Planar Surface	320 mg [†]	Magnets	2D	Magnetic push/pull	/	Yes	Yes	Yes	/	/	/	Self-folding	Robotics and programmable matter
Ijäs et al. [194]	Chain	3x2x2 nm	8064	/	Solution	~ 0	Atoms	3D	Atomic forces	/	Yes	Yes	/	/	/	Yes	Self-assembly and self-folding	Drug delivery
Douglas et al. [195]	Chain	3x2x2 nm	7308	/	Solution	~ 0	Atoms	3D	Atomic forces	12h	Yes	Yes	/	/	/	Yes	Self-assembly and self-folding	Drug delivery
Bai et al. [191]	Chain	3x2x2 nm	7249	/	Solution	~ 0	Atoms	3D	Atomic forces	12h	Yes	Yes	/	/	/	/	Self-assembly	Programmable matter
Cheung et al. [173]	Chain	77x38x38 mm	7*	1.5 Nm	Planar surface	>371 g	Electricity	3D	Servo Motors and Pivot	3 s	Yes	/	Yes	Yes	Yes	/	Torsion	Robotics and programmable matter
Knaian et al. [176]	Chain	1x1x1.5 cm	4*	$4.5e^{-4}$ Nm	Planar surface	14 g	Electricity	3D	Servo Motors and Pivot	5 s	Yes	/	Yes	Yes	Yes	/	Torsion	Robotics and programmable matter
Yim et al. [166]	Chain	1.3cm x cm x 0.1 mm	$>30^*$	/	Planar surface	$>0^{\dagger}$	Electricity	3D	Servo Motors and Pivot	15 s	Yes	/	/	/	/	/	Pulling and Folding	Robotics and programmable matter
Sung et al. [175]	Chain	$>cm$ x cm x <0.5 mm	57	$4e^{-2}$ Nm	Hot water	$>0^{\dagger}$	Heat and Magnetic field	3D	Smart Materials Magnetic steering	32 s	/	Yes	/	/	/	Yes	Self-folding	Robotics
Tibbits et al. [168]	Chain	$>cm$ x 6.4Ø mm	63*	/	Hot water	$>0^{\dagger}$	Heat	3D	Smart Materials	>40 mn	/	Yes	Yes	/	/	Yes	Self-bending	Programmable matter
Uchida et al. [28]	String	$>cm$ x Ø1 mm	/	/	Hot water	$\sim 0^{\dagger}$	Heat	3D	Smart Materials	/	/	Yes	Yes	/	/	Yes	Self-bending	Medical
Wang et al. [128]	String	$>cm$ x $>Ø10$ mm	/	/	Hot air or water	$>0^{\dagger}$	Heat	3D	Smart Materials	$>60s$	/	Yes	/	Yes	/	Yes	Self-bending and Torsion	Programmable matter

Fig. 2.13 Summary of the literature on self-folding 1D structures. The asterisc in the module number column depicts the 1D structures' number of module demonstrated. The cross in the "torque" and "mass in medium" correspond to calculated values inferred from the information within the respective studies.

Within chains, the magnetic assemblies introduce an inter-module force creating a cohesion inside the assembly. This self-assembly relies on the patterning of magnets polarisation along the modules surface to program the self-assembly and trajectories of the modules. To reprogram the chain and obtain a different structure, a re-patterning can be performed manually by changing the magnets orientations [193], or remotely by using the Curie temperature of magnets and remagnetising them [1]. Owing to the precision of microfabrication techniques, magnetic 1D structures can be scaled down below millimetre scale as shown by Kim et al. who used photolithography techniques for the fabrication and external magnetic field for the actuation [4] and shown by Li et al. who fabricated a magnetic ribbon folding into Halbach arrays using moulding techniques [7]. Benefiting from the high force that magnets exhibit when in proximity of each other, magnetic string is the approach with the largest scale range, spanning from the milli- to centrimetric scale, and demonstrating the fastest assembly time due to the monostable behaviour of the chain (Figure 5.17 column module size, mass in medium, actuation time). In addition, the high magnetic force and inter-module interaction allows for the structure to be highly resilient to deformations and thus, can behave more as a monolith than an assembly, as shown by Yim et al. where the self-assembled magnetic chains were dropped from a 50 cm height and resisted in 85% of cases [166]. Although magnetic chains show strong advantages, notably on the scale, the main limitation pertains to the strength/weight ratio of magnets which limits the size of assemblies to a few modules [193] without the implementation of alleviating environments (i.e. air cushion [172] or free fall [43]) or a close proximity between modules [166]. To obtain the folding of large structures, this exception implies that an optimum can be found between strength/weight ratio of magnets, proximity between magnets, and magnet strength/flexibility of the chain (Figure 5.17 column module size and module number). However, and as demonstrated by Fass et al. [193], the assembly occurs immediately after release of the string from its anchoring and at fast speed (0.2 second). The speed of assembly in such cases can be detrimental and can cause misassemblies when the magnets are close from one another [166]. On a side note, this problem is specific to 3D assemblies where the range of motion required for the string to assemble is larger than for 2D assemblies. Current designs of magnetic strings are monostable with the stable state being any assembled state, be it the targeted structure or any misassembly, thus highlighting the necessity to implement more control to the folding speed, sequence or release method of self-folding strings as shown by Gu et al. [189] or Griffith et al. [172] where the string is released from a tube preventing uncontrolled assembly. In conclusion, exploring the possibilities of bistable magnetic strings (straight or assembled) or understanding the possible stable states of multistable magnetic strings are crucial aspects to build large structures. Coupled with the advance of reprogrammable

magnetic composite [1] and the addressability of magnetic structures [24], magnetic strings could benefit from intermodule interaction stiffening the final structure, while being able to be reconfigured into different shapes at will, or see the overall stiffness of the structure reduced by applying global heat to decrease the magnetisation and thus, the intermodule force.

The forming of DNA structure is obtained from the self-assembly of nanometre size DNA strands whose sequence of nucleotide bases (Adenine, Cytosine, Guanine, Thymine) can be programmed to bond with other strands. Recent advances in the sequencing of the DNA structures allows a high degree of confidence in the programming resulting in the parallel assembly of large DNA structures counting several thousands of elements (Figure 5.17 column module number) [194] [195]. It is this high precision of patterning that enables structures to be functionalised with stimuli sensitive strands to perform robotic tasks such as sensing or actuation [196]. Currently applications of DNA robots mainly target in-vivo tasks such as drug delivery to remote areas that surgeons or current miniature robot struggle to access. Examples include the development of a nanosyringes to deliver drugs directly into cells [197], or cargo nano-robots able to deliver drugs upon detection of an alkaline pH change in the body, a marker of cancerous tissues [198] [199]. Although to date, DNA assembly is the smallest structures obtain from chains with only few nanometres in size (Figure 5.17 column module size), scaling up has not been achieved beyond a micrometre, thus limiting the controllability and localisation of the nano-robots in less predictable environments. In addition, the large number of building elements inflates the assembly time to several hours (Figure 5.17 column assembly time), but considering that numerous structures are fabricated in parallel, this leads to questions regarding the controllability not of a single robot, but of a swarm at a small scale and with materials difficult to track.

Self-folding chains relying on motors and joints exhibit a large motion range and high degree of controllability as each motor can be addressed separately in terms of position and rotation speed. When folding, this enables a precise trajectory to be followed and thus, limits collisions and entanglements resulting in the possibility to fold structures with complex Hamiltonian Paths [173]. In comparison to other self-folding 1D structures approaches, motors are the only method currently proposing torsion within the chain, which broadens the range of motion of the hinges to 360° around the chain length axis with a single hinge (Figure 5.17 column assembly and deformation), instead of a maximum of $\pm 180^\circ$ around the hinge axis using self-folding techniques. This enables the string better programmability and adaptability during the shape morphing from string to structures. In addition, and depending on the size, motors can deploy large torque (Figure 5.17 column torque) which allow for large structures to be folded but, two drawbacks limit the use of motors: their weight and

the required power. This limitation was highlighted within the study by Knaian et al. where the miniaturisation of the motor used to fold the chain reached 1 cm but the output torque could only actuate a four hinge chain [180]. Since the power consumed in a motor is directly related to the torque it outputs, this means the larger the chain, the larger the power required. Currently, chains with motors only relied on direct energy supply through a cable which limits the independency of possible robotic applications using this method [167]. Hence, new powering implementations are required and have already been demonstrated. As seen in the work of Yang et al., a sub centimetre scale robot was powered using the combustion of methanol to heat up the SMA actuator enabling the robot to walk [200]. In the work of Ogata et al. the folding of their origami structures was achieved by selectively powering the SMA hinges using magnetic induction [201]. Although scale and torque is the main challenge for motor-based strings, with the wide variety of electronics module available on the market, such systems could be implemented to achieve wireless communication and sensory feedback to provide proprioception and exteroception (a sense of self and sense of the environment) to communicate the shape of the environment, or the status of the deformed 1D structure to tackle the challenge of structural resilience of self-folding chains and communication.

On the opposite of motors or magnets used for self-folding 1D structures, smart-materials with volume change properties possess the main advantage to be both actuators and sensors (Figure 5.17 column sensing). Consequently, a change in the environment is detected by the smart-materials which results in a transformation from 1D to 2D or 3D. Although this is only limited to detection and not sensing, in both cases there is no form of communication that would indicate the folding extent or state of the 1D structure to an external user e.g. a surgeon. Currently, a large range of smart-materials exists with various sensitivities to external stimuli such as pH, electric field, temperature, hydration, etc. In addition, these smart-materials benefit from an adjustable synthetisation allowing the tuning of their properties (i.e. mechanical properties, sensitivity range) and hence, enabling them to be tailored for the envisioned applications or environment [202]. With the recent introduction of 4D printers, and the progress regarding printing at small scale, structures powered by smart-materials can be produced, thus expanding the range of applications and ease of fabrication whether at micro- [28] or macro-scale [127] with complex patterning and deformation as demonstrated by (Figure 2.11 (f)) [169]. This makes smart-materials the most versatile materials in terms of sensitivities and size. However, smart-materials currently implemented within self-folding 1D structures exhibit low actuation torque to length ratio (Figure 5.17 column torque), thus implying a strong necessity for smart-materials to be implemented into lightweight 1D structure or requiring alleviating environments [179]. Also, when considering the main features a smart-structure can demonstrate, a large range of smart-materials exhibit reversible

deformation, thus requiring a constant energy supply to maintain the structural state. This justifies the need and use of smart-materials with shape memory properties such as SMP, SMA and shape memory hydrogels [203] to maintain the structure's shapes without power input. Lastly, even though triggering the volume change of smart-materials remotely and untethered has been demonstrated in the past [16] [130], these technologies have not been implemented to self-folding 1D structures more complex than a ribbon bend or twist. A possible explanation pertains to the size and uniformity of the input field (whether thermal, magnetic or photonic) for long 1D structures which is paramount for the folding to be predictable, be it sequential or simultaneous, or the aspect ratio of 1D structures and the complex deformations that can be produced.

To date, the information related 1D structure is limited although most data e.g. torque and module size can be estimated, thus limiting the evaluation of the capabilities of 1D structures to basic properties. In the future, 1D structure studies and development could add plethora of parameters. This include the joints stiffness, maximum load of the structure (in x, y and z), the intermodule interactions along with the force related to the modules distances as well as the strain for various loading conditions to demonstrate the structural integrity. The degrees-of-freedom of the hinges could be advertised with their possible range of motion to evaluate the possible trajectories achievable by the 1D structure. Complementarily, the alleviating environment should be described by its alleviation time (in the case of a hot bath cooling, up to an hour for example), the alleviation dimension (1D, 2D, 3D), and the force reduction percentage (100 % in the case of neutral buoyancy ignoring the drag force and internal hinge friction for example).

2.4 Conclusion about smart structures and their requirements

2.4.1 Universality

The concept of universality refers to the ability of a structure to morph into arbitrary shapes. This was achieved with origami using aperiodic tessellation [123] or periodic tessellation [77] and with chains using Hamiltonian paths [172] or slicing methods [167]. Whatever approach is used, large target structures may require numerous elements and hinges with a specific folding sequence. In addition, these large structures may not be foldable due to the limited strength of smart material actuators.

2.4.2 Sequential folding and addressability

To be foldable some self-folding structures require a folding sequence, for which each crease or set of creases must be addressed specifically. The need for folding sequences is generally highlighted in structures with a locking aspect [204]. Examples of sequential folding include creases electrically connected to an on-board microcontroller [37] and exploiting the differing stimuli sensitivities of wirelessly powered creases [86] [21]. Selective crease folding can also be achieved by incorporating different types of smart materials, as done by Baker et al. [51], who sensitised the creases of a hydrogel origami by imprinting metal ions into the hydrogel surface (using redox reactions) to be activated in the presence of specific chemicals such that the folding of individual creases could be triggered. However, in the sequential folding approach using smart materials, the number of folding steps is limited by the variety of compatible smart materials.

2.4.3 Position control

There are currently three main approaches to regulating the angle of self-bending creases: mechanical stops, curved creases and shear stress control. Mechanical stops refer to the intentional prevention of folding by the implementation of physical obstructions. This technique has been used by T. Tachi [205], Tolley et al. [116], and others, who have used origami with thick panels functioning as folding end-stops, so that panel thickness and crease width determine the extent of the folding angle.

Curved creases are creases whose elastic potential energy is minimised when their folding angle reaches a critical value, as demonstrated by Miyashita et al. [118] who achieved the folding angle regulation of a self-folding sheet using curved creases. This is due to the accumulation of shear stress (i.e. internal mechanical forces that can result in deformation). Miyashita et al. [206] demonstrated a precise control of a folding angle by kinematically coupling two creases in the origami pattern. Using a nonlinear relation of the folding speeds, one crease is crudely actuated by uniform heat, while the other crease folds in a more controlled manner. Liu et al. [70] showed that the beam-width and power of a laser could be used to tune the folding angle of a shape memory polymer strip (as this would determine the internal stresses developed at target locations).

2.4.4 Reversibility and Reprogrammability

The reversibility of the folding is an important feature when a self-folding structure is required to transform into different configurations. One of the most successful implementations is

based on antagonistically coupled shape memory alloys, where reversible self-folding is realised by exploiting the heating/cooling cycles of the two shape memory alloys [207]. Shape memory polymers, on the other hand, face a difficulty in this regard given that a pre-stretched strain stored in the material is released when the material is activated, resulting in mono-stable structures [39]. Conversely, some hydrogels, such as pNIPAM, can be activated repeatedly, as this class of stimuli-responsive polymers exhibit reversible volume changes [35]. An alternative approach to reversibility is to replace a part of an origami-made body. Based on the fact that reconfiguration through folding is not commonly observed in nature, Miyashita et al. [23] demonstrated that a miniature origami robot can self-fold, walk, and be equipped with a new self-folding body part to exchange the locomotion modalities such as gliding or swimming.

2.4.5 Remote actuation

Actuators can be powered by power lines, onboard generators or external remote sources. With power lines, the robot is tethered to the external power supply on which it depends, which limits the environment of the robot but, theoretically, the robot receives unlimited energy. Examples include fluidic pressure [69] or electric power [208]. In contrast, onboard or remote power supplies enable the robot to be independent, but the power must originate from onboard generators with limited autonomy (battery [149] or chemical reactions [209]) or from external equipment with unlimited energy (induction [86], microwave [210], etc.). In the case of remote supply, the supply can be global when the robot is smaller than the supply zone [39] or local when the robot is larger than the supply zone [70].

2.4.6 Sensors, communication and computation

Sensors are one of the most important parts of a robot, as they are the sensory organs that provide the robot with a sense of self (proprioception) or the sense of the world (exteroception). Similar to actuation, sensing can be external [211] or on-board with external communication [69] [208]. This sensing and communication is directly linked to the computation that allows decision-making. At a small scale, computation is generally achieved externally after the data has been communicated, as in Huang et al. [132] where a camera was used to assess the position of the robot to adapt the actuator inputs. On macro scale robots, computation can also be performed onboard [149] or embedded within the cells of the robot [142].

2.5 Summary of smart structures

Building smart structures require programmable materials which are able to store their programmed deformations in their molecular structure (shape memory effect, magnetisation profile adjustments) or within their design (tri-layer, dual-layer) to deform in specific ways. To achieve the shape programming, smart materials are used which can actuate with linear, planar, or 3D deformation. Owing to their fabrication method, enabling smart actuators of various scales to be produced, smart structures can be 2D, 1D or 0D, each of which being able to assemble and construct 3D shapes notably by the means of folding techniques. Regardless of their initial nature (2D, 1D or 0D), smart structures all possess common requirements which would make the smart structure the holy grail of robotic systems. Ideally, the grail of miniature robotics is a system that is able to morph reversibly and indefinitely into any shapes relying on the reprogrammability of the system's structure or materials, whose deformations and displacements can accurately be controlled and assessed. With sensory feedback and communication abilities, information can be relayed internally for the robot to adapt itself autonomously but also relay information externally to any user. As a result, the robot is intelligent by its adaptability, autonomous by its programming and reprogramming, resilient structurally to perform the tasks reliably, and can be powered remotely to navigate its environment unrestrained.

In the context of 1D structures, the challenges of self-folding structures and their technical limitations leads to the following research questions regarding self-folding strings:

1. How can a self-folding string without embedded electronics communicate its state?
2. How the limited energy that smart-materials provide be circumvented for strings to self-fold into large 3D structures?
3. How to make self-folding strings reprogrammable to widen the range of structures and grant them a universal shape change capability?
4. How does the Hamiltonian path relates to the final mechanical properties and what kinds of mechanical properties can be expected from string-based structures?
5. How to turn off the reversibility of smart-materials to maintain a shape or functionality without having to maintain the power input?
6. How can untethered strings be self-folded remotely in a sequential manner in order to avoid collisions and entanglements?

7. How can the non-uniformity of input fields be used to the advantage of self-folding strings?
8. Amorphous structures being born of disorder, is the folding accuracy and sequence as crucial as for highly ordered arrangement such as crystalline or semi-crystalline structures?
9. As strings are serial in nature, how does the position error propagates and how can it be compensated for?
10. How can the kinematic of the string be restrained to achieve the self-folding in tight environment?
11. How can an amorphous string-based structure, whether semi-crystalline or fully amorphous, be made reprogrammable if the essence of the disorder in said structure originates from the infinite degrees-of-freedom and flexibility of the string?

In this thesis, research questions 1 to 4 are broached in Chapter 3, research questions 2 and 5 are approached in Chapter 4, elements of response for research questions 6 to 8 are brought in Chapter 5 and research questions 9 to 11 are discussed in Chapter 6.

Chapter 3

Reprogrammable Self-Folding Chains for Crystalline Structure

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3.1 Summary of Chapter

In this chapter, I present my approach to mimicking crystalline structures from a string (Figure 5.1 (a) and (b)). Current approaches to fabricate crystalline structures use modular assemblies [212] [136] or self-folding strings. As of today, self-folding mechanisms for crystalline structures include magnets [166], hydrogels [169] and motors [173] which exhibit the most potential to morph into various structures due to their reprogrammability.

To compete with motors infinite range of rotation and high torque, which allows self-folding and reconfiguration into a different crystal, here I propose a manually reprogrammable modular chain self-folding in a cubic crystal/voxel using smart-materials.

A voxel comprises 7 edge modules interconnecting 6 self-folding corner modules serving as the vertices of the cube (Figure 5.1 (b)). The main feature of the edge modules is their air chamber that balances the weight of the chain at 65°C under water, thus providing the neutral buoyancy required to reduce the required folding torque to a minimum. The

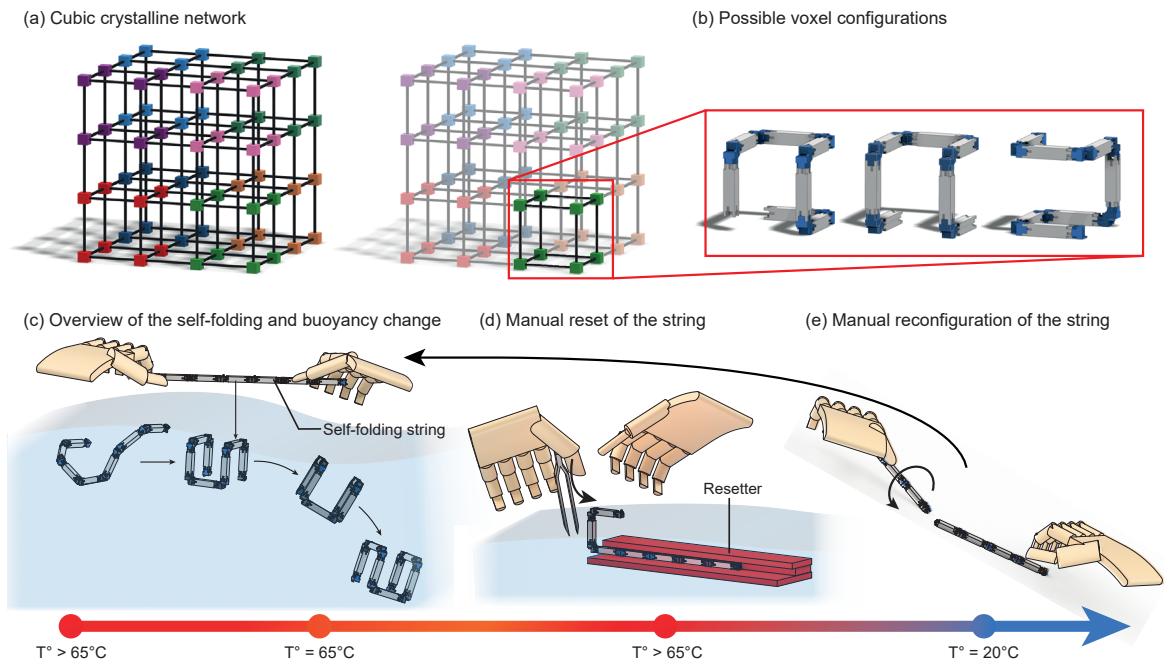


Fig. 3.1 Overview of Chapter 1. (a) Crystalline structures are highly ordered networks that form crystal after crystal. (b) These crystals are equivalent to voxels that, in the case of cubic crystals, can be self-folded in three different configurations. (c) The developed string self-fold into a voxel in water above 65°C before slowly sinking at the temperature cools below 65°C. (d) After folding, the string can be reset in its initial straight configuration and (e) reconfigured by manually changing the hinges axis of rotation before being self-folded once more.

corner modules of the chain are equipped with thermoresponsive shape memory polymer self-folding at temperatures greater than 65°C. Because of the shape memory feature of the smart material, the self-folded corner modules could be reset by dipping them in hot water, manually unfolding them and letting the water cool. Coupled with the modularity of the chain, the resetability and the re-orientatability of the corner modules around the edge modules enable the chain to be reprogrammed, and (Figure 5.1 (c), (d) and (e)) allows different voxels to be self-folded from the same chain.

Facing the limiting torque of the smart material, the buoyancy is a crucial feature of the chain. In air, the corner modules fail to lift one edge module, whereas entire voxels can be folded in water with a minimum torque requirement at the temperature of neutral buoyancy. For this purpose, a buoyancy model is proposed to help adapt the design to different smart materials and medium, and help estimate the required folding torque for the corner modules to fold depending on the temperature. Adding to the limiting torque required contribution of the buoyancy, detection of the end of self-folding is enabled by the change of buoyancy of the chain upon cooling below 65°C.

Overall, corner modules were successfully folded in 96.5 % of the trials and self-folding voxels reached a success rate of 84.6 %. Though the self-folding was successful, the inaccuracies in the folding of the corner modules affected the final shape of the crystals. I propose to assess the difference between the targeted crystal shape and the self-folded voxel via photogrammetry: a method reconstructing objects in 3D using numerous pictures from different angles. This was complemented by a model calculating the deformation of the structure depending on an external torque and its location, thus allowing one to estimate the deformation of a chain structure.

In summary, this chapter contributes to self-folding chains by :

- Proposing a reprogrammable self-folding corner module design using a shape memory polymer smart material,
- Enabling long chains to be foldable by reducing the effect of gravity using neutral buoyancy,
- Proposing a theoretical model to assess the structural integrity and resilience to external forces,
- Proposing a method to assess at a structural level the accuracy of the folding.

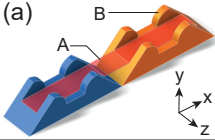
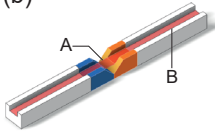
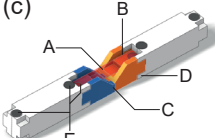
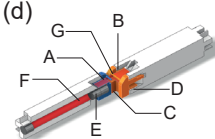
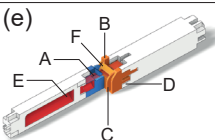
Module Designs	Features	Size	Mass	Degrees Of Freedom	Buoyancy Control	Folding Environment	Reprogrammability			Maximum Folding Angle
							Orientability	Resetability	Modularity	
(a) 	A: PVC Bridge B: PVC Guide	15x5x4mm	EM: 181 g	$\begin{Bmatrix} 0 & 1 \\ 0 & 1 \\ 0 & 1 \end{Bmatrix}$	No	Air	No	No	No	40°
(b) 	A: PVC Bridge B: PVC Guide	EM: 15x4x3mm CM: 10x4x3mm	EM: 181 g CM: 108 g	$\begin{Bmatrix} 0 & 0 \\ 0 & 1 \\ 0 & 1 \end{Bmatrix}$	No	Air	No	No	Yes	40°
(c) 	A: PVC Bridge B: PVC Guide C: Indents D: Connectors E: Magnets	EM: 15x5x5mm CM: 12x5x5mm	EM: 406 g CM: 140 g Magnets: 6x47 g	$\begin{Bmatrix} 0 & 0 \\ 0 & 0 \\ 0 & 1 \end{Bmatrix}$	No	Salted water below 100°C (360g/L)	No	Yes	Yes	65°
(d) 	A: PVC Bridge B: PVC Guide C: Indents D: Connectors E: Magnets F: Chamber G: Fairlead	EM: 20x5x5mm CM: 12x5x5mm	EM: 480 g CM: 140 g Magnets: 4x47 g	$\begin{Bmatrix} 0 & 0 \\ 0 & 0 \\ 0 & 1 \end{Bmatrix}$	Yes (Low)	Fresh water below 95°C	Yes	Yes	Yes	90°
(e) 	A: PVC Bridge B: PVC Guide C: Indents D: Connectors E: Chamber F: Fairlead	EM: 20x5x5mm CM: 10x5x5mm	EM: 472 g CM: 96 g	$\begin{Bmatrix} 0 & 0 \\ 0 & 0 \\ 0 & 1 \end{Bmatrix}$	Yes (Medium)	Fresh water below 95°C	Yes	Yes	Yes	90°

Fig. 3.2 Sum-Up of the designs of corner and edge modules (a) Design 1, (b) Design 2, (c) Design 3, (d) Design 4, (e) Design 5 (Final design).

3.2 Methods

3.2.1 Modules design

As highlighted in chapter 1, several challenges surrounds self-folding strings including universality, scale and torque, self-collision, and reprogrammability. Each of these challenges funnel the choice of smart-material, the environment for the self-folding and the design features of the string (see Figure 3.2).

Regarding the challenge of universality, the string should be able to self-fold into any 3D structure. A first possibility relies on the string's ability to fold and re-fold in any direction into a different structure, or the possibility to manually reprogramm the string. The first scenario requires a high degree of controllability of the self-folding, which needs to be reversible. On the other hand, the second scenario requires the string to be modular, the self-folding modules to be orientable and their folding to be reversible. The first approach was already considered by implementing servo-motors inside the string [173] and faces difficulties regarding miniaturisation and scale. The second approach requires the implementation of a smart material with reversible volume change properties. For this purpose, Polyvinyl-Chloride (PVC) shape memory polymer was selected. PVC as a shrink film is able to shrink when exposed to temperatures superior to 65°C , its glass transition temperature (T_g). The volume change ability can be reset by stretching the PVC again above T_g and cooling it down while maintaining the stretching. This behaviour can be explained at a material level. Inside the PVC polymer network, two phases exist: frozen phase and active phase. Above T_g , the PVC is mainly constituted of active phase which allows the PVC to deform. When exposed

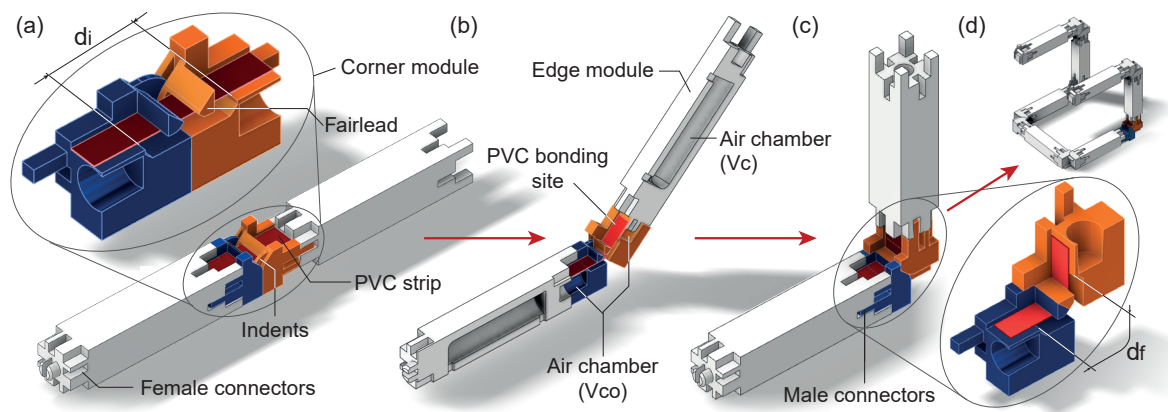


Fig. 3.3 Illustration showing the main traits of two crystal edges: a hinge module connected to two link modules. (a) Straight configuration with the PVC strip inactivated bridging the two components of the hinge. (b) On-going folding caused by the contraction of the PVC when exposed to temperatures exceeding 65°C . (c) Completed folding. (d) Completed folding showing the 3D structure.

to a mechanical strain while cooling below T_g , the active phase transitions to the frozen phase keeping in memory the strain. Once re-exposed to temperatures exceeding T_g , the frozen phase transitions to active phase and release the mechanical strain that was stored [213] [68]. This gives the shape memory ability to the PVC and enable the reprogrammability of a PVC actuator, thus the suitability of PVC for the second approach.

In regards to the challenge of scale and torque, since actuators based on smart materials provide a finite energy as volume change, the required folding torque should be within the range the smart material can provide. This implies that the modules of the string should be as compact and as light as possible to limit the torque required. To alleviate this torque, a counter force to gravity can be introduced, for example buoyancy force, by changing the self-folding environment. Moreover, the limited volume change of the smart material actuator constrain the design of the self-folding modules such that the volume change enables the folding to the target angle. To reach the target angle, the following condition must be satisfied: $(1 - \alpha) \geq d_f/d_i$, where d_i and d_f represent the initial and final lengths of the PVC separating the two components of the self-folding module (Figure 3.3 (a) and (c)). If this condition is not respected, the hinge will not attain the target angle, resulting in a mismatch between the final and targeted shapes, as well as a loose hinge lacking stiffness.

At a structure level, matching the targeted shape requires (1) the hinges to self-fold accurately to their target angle, (2) the string to avoid collisions while self-folding and Once self-folded, (3) the structural integrity must be sufficient for the string-based structure to resist its weight in addition to the external forces that may be applied; in other words, the self-folding modules must exhibit sufficient stiffness.

The first design in Figure 3.2 (a) aimed at evaluating the main difficulties of designing a self-folding module. It consisted of two long modules bridged by a PVC strip and enabling the modules to fold around the contact line. However, due to the low contact surface area and softness of the PVC, the module could deform in other directions than the folding axis. In addition, the required folding torque exceeded the torque provided by the PVC and folded to only 40° . The main shortcomings of this design is its lack of modularity and integration towards more complex structure, and its folding around a single axis and direction.

The design in Figure 3.2 (b) compensate for the lack of modularity by introducing self-folding modules and structural modules. They allowed the string to fold in the xy-plane and to be configured with multiple non folding sections or with multiple self-folding modules to fold beyond the maximum folding angle. Also, under the assumption that the more compact the modules of the string are, the more likely the string is to collide and entangle, the structural modules were elongated compared to the previous design to limit the likelihood of collisions during the self-folding. Despite succeeding in removing a degree-of-freedom,

and improving the assembly capability of the string to fold in 2D, the lack of connectors between the module made the PVC strip the only component responsible for the structural integrity of the string. In addition, the self-folding modules were only able to fold to 40° .

For this purpose, the third design was adjusted with male and female connectors relying on magnets to hold the modules together and eventually enable self-assembly at a later development stage. Adding to the improvement, the degrees-of-freedom were limited further by implementing indents in the self-folding module which prevent the rotation around the x-axis. However, with this design, the magnets represent more than 40% of the total weight and drastically increase the required folding torque. Although the maximum folding angle reachable increased to 65° from the previous design, the self-folding modules systematically failed to fold more than 10° due to the excessive torque even though the environment was changed from air to water saturated with sodium chloride to introduce a buoyancy force.

As a consequence, the fourth design in Figure 3.2 (d) was amended with an air chamber connecting each side of the structural module which was plugged with a magnet on each side binding structural and self-folding module. In addition, a fairlead was implemented in the self-folding module which guided the deformation of the PVC and thus, guaranteed that the volume change of the PVC was sufficient for the self-folding module to reach 90° . In spite of the decreased number of magnets (from 6 to 4) and the cavity improving the buoyancy, in the experiments the air cavity leaked due to 3D printing inaccuracies. However, with tightly shut chambers, cubic voxels structures were able to fold in 30% of trials in a bath of fresh water. Here, the cubic voxels are composed of 7 structural modules called edge modules in this case, and 6 self-folding modules designated as corner module due to their position in the cubic voxel (see Figure 5.1).

Finally, for the last design (seen in Figure 3.2 (e) and illustrated in more details in Figure 3.3, a watertight air chamber was introduced in the edge module, the magnets were removed and their chambers were repurposed as an additional air chamber. In this way, the chambers could hold air more reliably and provide sufficient buoyancy for voxels to fold in more than 80% of trials. With the removal of the magnets, the connectors were amended to imbricate tightly to maintain the structural integrity of the string so that the loads could be transferred effectively. Additionally, the fairlead was thickened to improve its printability and its resistance to the force exerted by the PVC. As a result, out of the 30% maximum shrinkage the PVC provides, only 12% are required to fold this corner module design to 90° in water. Figure 3.4 highlights this result by demonstrating the importance of fairleads by comparing the extent of shrinkage that is required on a self-folding corner module with and without fairleads.

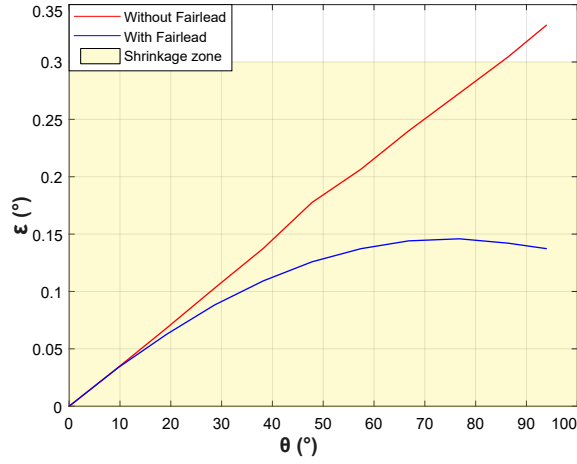


Fig. 3.4 Plot of the bridge length shrinkage in percentage versus the folding angle of the hinge.

Without a fairlead, the shrinkage extent of PVC does not allow the corner module to fully self fold. On the contrary, the design with a fairlead folds to 75° in air as the fairlead placement requires the PVC to extend beyond 75° . However, due to the buoyancy, the corner module with a fairlead folds to 95° in water. Any unused shrinkage can compensate for any mismanufacturing, thus ensuring correct folding and guaranteeing the robustness of the design. Opposite to other self-folding approaches relying on mechanical contact to limit the folding angle, here the design of the corner module can be altered to prevent the folding beyond the targeted angle. As soon as the design requires the PVC to extend, the folding should stop since the PVC can only shrink. This statement stays true if the inertia is neglected. As shown in Supplementary Video SV1, the corner module experiences a bounce back after hitting the buttresses implying that the PVC extended while the module was bouncing. Limiting this bouncing can be achieved by increasing the drag force or limiting the speed of folding. The drag force given as :

$$F_d = \frac{1}{2} \rho_w v^2 C_D A \quad (3.1)$$

where v is the speed of folding, C_D is the drag coefficient and A is the cross sectional area.

Increasing the drag force implies increasing the viscosity of the environment or changing the design of the modules to increase the cross sectional area. However, limiting the speed of folding can be achieved by tuning the curve of PVC shrinking against folding angle with the help of the plot in Figure 3.4.

3.2.2 Fabrication & assembly processes

All modules of the string were printed out of the clear resin from Formlabs and in their Form 2 stereolithography 3D printer. After printing, the parts were washed in clear isopropanol, and the corner modules were colored by dipping the parts in dyed isopropanol. After curing, the corner modules were manually placed onto the glueing platform, as shown in Fig 3.12 (a), and were compressed against a spacer to maintain the proximity between the parts. This proximity is crucial because a small spacing can dislocate the indents, whose aim is to prevent

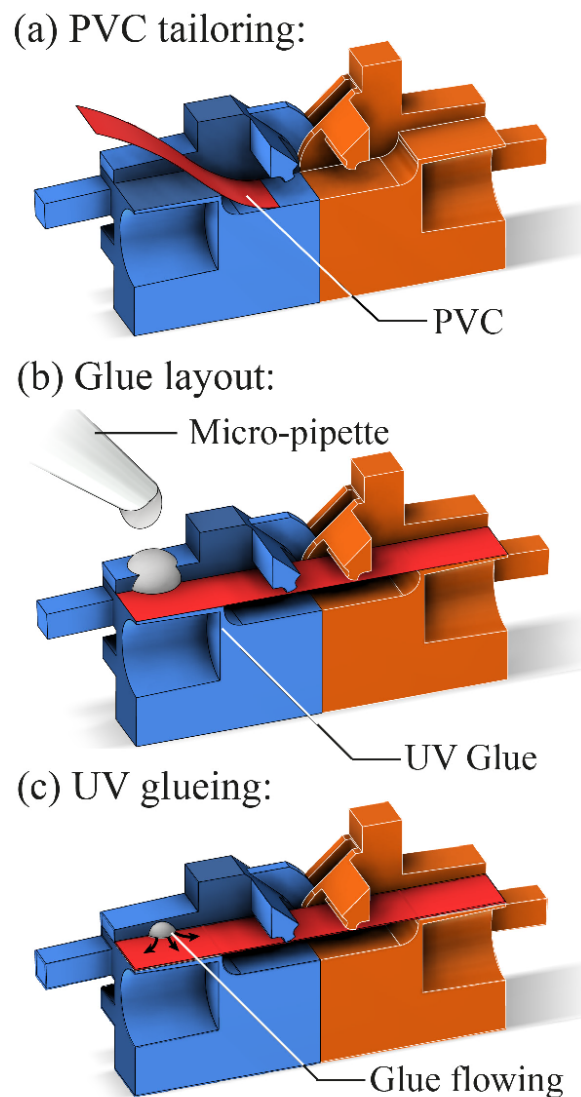


Fig. 3.5 The fabrication process of the modular string. (a) Tailoring process of the PVC below the fairleads and (b) deposition of the UV glue on the side of the bonding sites using a micro-pipette. (c) Spreading of the glue and UV curing.

roll rotation of the corner modules. Then, the $20\mu\text{m}$ thick PVC (reference B08YDDG5QX on Amazon) was cut into 3 mm wide and 9 mm long strips in a vinyl cutter Silhouette Cameo 3, cleaned in a bath of isopropanol, and rinsed in a bath of deionized water. After drying, PVC strips were tailored below the fairleads of the corner modules, as shown in Figure 3.5 (a). Dispensed from a micropipette, the UV glue Loctite AA 3491 was dropped on the side of the PVC strip, allowing the glue to flow below the PVC and only onto the clear resin, as illustrated in Figure 3.5 (b) and (c). After the glue was laid out, the glueing platform was exposed to UV light using the UWave UV torch, until the glue solidified. Each corner module was manually checked to determine if any glueing was defective.

3.2.3 Characterisation

PVC folding torque

The folding torque is required to validate the foldability of the string in water for a given cavity size. Here, we used a corner module with one side anchored, and loads applied on the other side. The corner modules were then placed in an oven at 75°C , and the folding angle was measured. The graph in Figure 3.6 represents the folding angle as a function of the torque applied to 5 corner modules. As shown, only the corner modules folding to the target angle of 95° experienced a torque of $13\mu\text{Nm}$. This result is supported by the single corner module folding experiment in subsection 3.3.1 and Supplementary Video SV(1), where the voxel edge with the highest chamber size is unable to fold due to the torque created by the buoyancy.

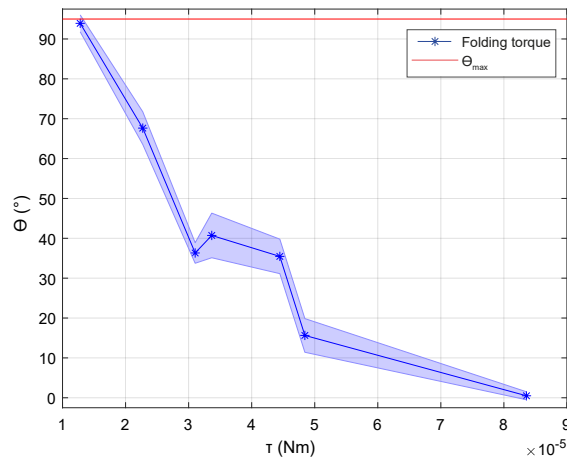


Fig. 3.6 Graphic showing the average folding angle on 5 hinges depending on the torque applied.

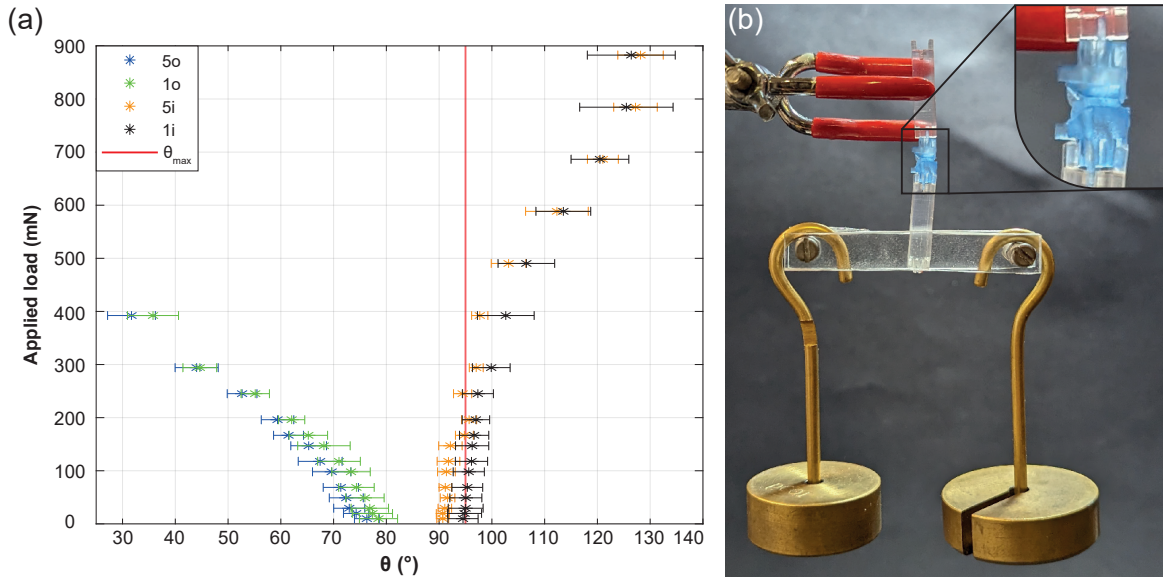


Fig. 3.7 (a) Plot of the folding angle of the corner module recovered once the load was removed. (b) Picture of the maximum load on a corner module before the indents dislodge from the ridges and let the corner module experience parasitic rotations.

Loading

In our design, the PVC is the only element holding together the two parts of the corner module. Like any material, the PVC has an elastic (reversible) deformation and plastic (irreversible) deformation. Similarly to Figure 3.11 (a), the corner modules were loaded inward and outward, and for one reset and 5 resets. As indicated in Figure 3.7 (a), for each case the curves can be split into two lines with different slopes. The junction between these two lines highlight the elastic and plastic deformation. For example, when loaded inward, the corner module could sustain up to 250 mN before entering plastic deformation; however, outward, the corner module could receive around 125 mN. When scaling up or loading the structure, the voxels should remain in elastic deformation for the structure to recover its original shape and maintain its mechanical properties. In addition, scaling up strains the connections between the modules when unfolded. This requires the PVC of the corner module to be able to sustain the weight of the string when unfolded. Figure 3.7 (b) shows the connections and the PVC sustaining 220 g, however, with this load, the PVC already entered plastic deformation, and has extended to its limit before the indents of the corner module exit the slits. This means that, beyond this load, the corner modules has additional degrees-of-freedom and the self-folding is not reliable.

3.2.4 Buoyancy model

As mentioned in Chapter 2, one of the main challenges of the string approach is the required folding torque created by the weight of the string, which scales up as the string is longer. However, folding in water provides a counterforce: buoyancy. In the case where the buoyant force (F_b) is equal to the weight force F_w , the string enters neutral buoyancy, thus simulating microgravity, meaning that theoretically, an infinitesimal torque is required to fold any length of string or, in other words, an infinitely long string is foldable. To attain neutral buoyancy, the buoyancy of the edge module must counter its weight in addition to the weight of the hinge module and the PVC on it. For this purpose, the edge module features a sealed inner chamber filled with air of volume V_c . The equation for the neutral buoyancy in Eq. 3.2 details the balance between buoyancy and weight, where V_{pf} is the volume of the edge module without V_c , V_h is the volume of the hinge, V_{co} is the volume of air contained within the connections between the edge and corner modules, m_{SM} and V_{SM} are the mass and volume of the smart material, respectively. Here, the smart material is a Shape Memory Polymer whose mass and volume remain constant across temperatures, unless the smart material experiences a change in phase.

$$\rho_w(T^\circ) \left(V_{pf} + 2V_h + V_{Co} \right) (1 + \epsilon) \Delta T^\circ + V_{SM}(T^\circ) = g \left(\rho_{CLR} (2V_h + (V_{pf} - V_c)) + m_{SM}(T^\circ) \right) \quad (3.2)$$

The density of water, ρ_w , is given in [214] as a function of temperature. According to the manufacturer, the resin used to print the string has a density $\rho_{CLR} = 1.208 \text{ g/cm}^3$

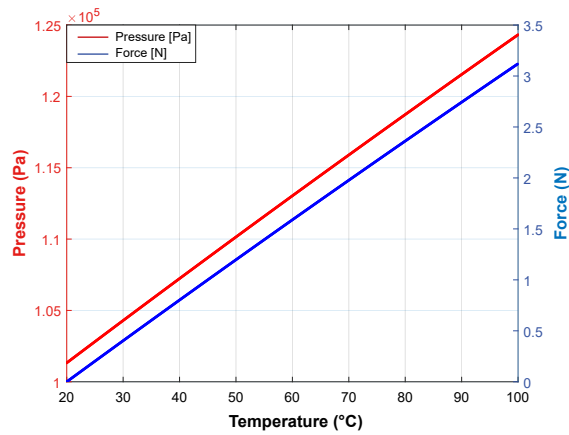


Fig. 3.8 Graph of the inner pressure and the inner force inside the cavity depending on the temperature.

but was measured to be 1.225 g/cm^3 . The linear thermal expansion of the clear resin ϵ is $124 \mu\text{m/m}/^\circ\text{C}$ as per the datasheet; however, measured experimentally, it was found to be $156 \mu\text{m/m}/^\circ\text{C}$ which gives a volumetric thermal expansion of $468 \mu\text{m}^3/\text{m}^3/^\circ\text{C}$ assuming that the resin expands isotropically. However, this value does not consider any pressure change within the module due to the air expanding.

With an increase in heat, the air expands in a manner similar to the thermal expansion of any material. Air was assumed to expand at a constant rate of $3.4 \text{ mm}^3/\text{m}^3/^\circ\text{C}$ between 20°C and 100°C . This expansion in the cavity generates an increase in pressure, which can be calculated as follows, assuming that air is a perfect gas:

$$P(T^\circ) = P_i \frac{V_c}{V_{th}} \frac{T^\circ}{T_i} \quad (3.3)$$

where $P(T^\circ)$ is the pressure in the cavity as the temperature increases, P_i is the atmospheric pressure, V_{th} is the volume of the cavity expanding thermally, and T_i is the ambient temperature. According to the plot in Figure 3.8, the pressure increases by 22% which generates a force inside the cavity of up to 3 N at 100°C . As the clear resin softens when exposed to high temperatures, the thermal expansion should be compensated by adding mechanical expansion. Thus, to estimate the overall expansion, we measured the change in the length of the edge module which resulted in a corrected thermal expansion of $854.6 \mu\text{m}^3/\text{m}^3 \cdot ^\circ\text{C}$ instead of $456 \mu\text{m}^3/\text{m}^3 \cdot ^\circ\text{C}$. This validates the hypothesis that the inner pressure within the cavity of the edge module impacts significantly the module expansion.

Because the equation for neutral buoyancy depends on the temperature and V_c , the volume of the chamber can be altered to select the temperature at which the string meets the neutral buoyancy, allowing one to adapt the platform to a different smart material. This is given in Eq. (3.4), where $X = V_c/V_{pf}$ and is the proportion of the edge module that is occupied by V_c . In the present case, the neutral buoyancy temperature was chosen to be 65°C so that the string would naturally sink below T_g . This indicates that the corner modules will not fold further, and that the string is ready to be handled without affecting the angles of the corner modules. Therefore, X is selected as 19.06%.

$$X = 1 - \frac{2V_h}{V_{pf}} \times \frac{\rho_w(T^\circ)((V_{pf} + 2V_h + V_{Co})(1 + \epsilon\Delta T^\circ) + V_{SM}(T^\circ))}{\rho_{ClR}V_{pf}} \quad (3.4)$$

To estimate and predict the buoyancy changes depending on the water temperature, we compared the average density of the edge module connected to a hinge on each side (Fig 3.9 (a)). Three cases were considered for this study. Using the manufacturer's data, buoyancy was estimated with V_{Co} containing air in Case 1 and with V_{Co} containing water in Case 2. Using the experimental data, Case 3 reflects buoyancy changes according to

the experiments, assuming V_{co} contains air. In Case 1, the theory indicates that the voxel edge is buoyant between 50°C and 65°C, and that in Case 2, the voxel edge sinks across all temperatures between 20°C and 100°C. However, in Case 3, the voxel edge experienced only one neutral buoyancy temperature above which, the voxel edge is buoyant and under which, the voxel edge sinks. This behavior indicates that the thermal expansion under experimental conditions (voxel edge underwater) is at least 5 times the measured thermal expansion. Initially, at 20°C, the air in the cavity of the edge module is at atmospheric pressure, but the pressure increases as the module experiences heat (Figure 3.8). Coupled with the ductility of the material at high temperatures, the pressure in the cavity shown in Figure 3.8 may increase the expansion of the edge module, thus increasing the buoyancy force and explaining the behavior observed during the experiments.

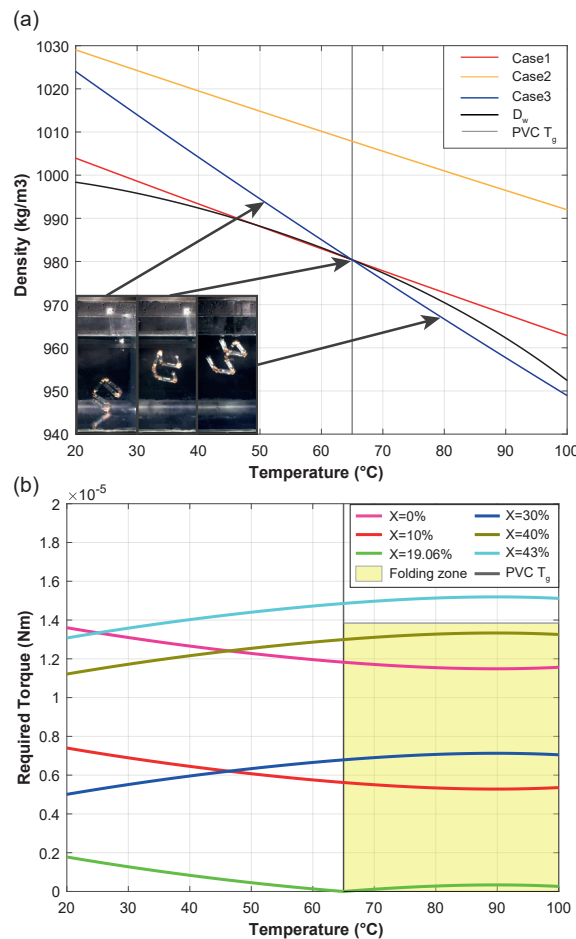


Fig. 3.9 Theoretical modelling of (a) the buoyancy change depending on the temperature experienced by a voxel edge, and (b) the absolute value of the folding torque required to fold a hinge depending on the volume of the chamber within the edge module.

However, an overly buoyant edge module creates a torque that the PVC must counter. The maximum folding torque that the PVC can provide was measured experimentally on 5 samples and was estimated to be $\tau_{PVC} = 13\mu\text{N}$ as explained in Supplementary Materials. The required folding torque of a voxel edge for different X is plotted and compared to the maximum folding torque in Figure 3.9 (b) using the experimental data. As can be seen, an oversized chamber induces a torque that is too great for the PVC to fold, thus reversing the problem of the foldability created by the weight.

3.2.5 Hamiltonian Path

A 3D object can be converted into a string after it is discretized into elements and sub-elements, called voxels and sub-voxels. The string must then follow a path passing through each subvoxel without overlapping. Depending on the size of the structure and the details of the discretization, that is, the size of the voxels, the string will be shorter or longer. There may be an infinite number of Hamiltonian paths for a given structure; however, the self-folding aspect requires the string to not entangle or collide with itself during the transformation from 1D to 3D. In addition, once removed from the water, the mechanical properties of the

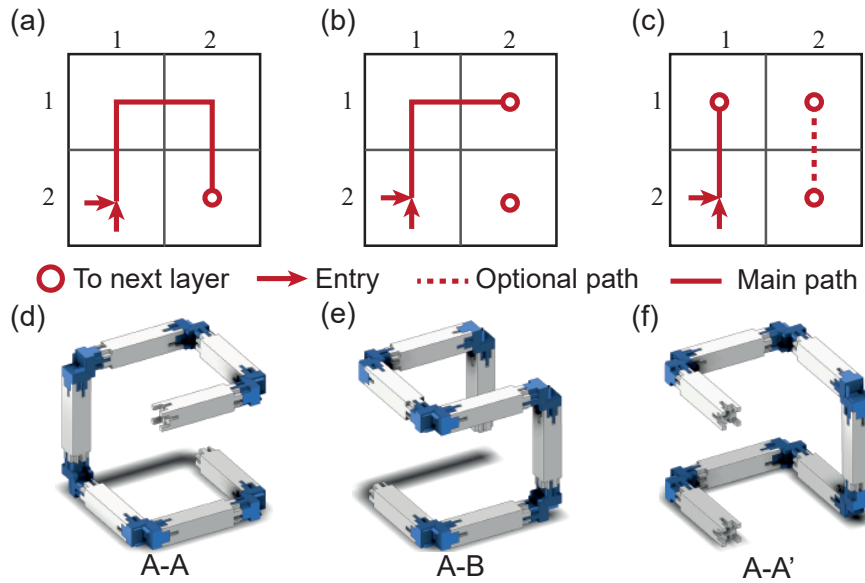


Fig. 3.10 Illustrations of the configurations for the voxels layer and the entire voxels. (a) to (c): Depiction of the three possible Hamiltonian paths filling one layer of the subvoxel with two turns (a), one turn (b), and no turns (c). (d) to (f): CAD views of the three existing Hamiltonian paths within a voxel using the configurations (a), (b), and (c) that guarantee no overlap and only one entry and exit. Rotations and mirror configurations are not taken into account.

structure are dictated by the Hamiltonian path and stiffness of the hinges. Thus, one can conclude that there exists a trade-off between the number of hinges within the string, which will increase the complexity of the motion risking entanglements, and the length of arms (distance between two connected hinges) within the structure, which will induce increased torques making deformations greater for lower forces. Therefore, the Hamiltonian path should consider the targeted stiffness of the structure, its targeted shape, and its self-folding motion. Because there may be an infinite number of Hamiltonian paths for a given shape, we only studied the Hamiltonian path within a voxel containing eight subvoxels, as shown in Figure 3.10. To do so, we split the voxel into two layers comprising four subvoxels each. There exist only three possible Hamiltonian paths that can fill a layer without overlapping, ignoring mirror paths and 90° rotations. These Hamiltonian layer paths were determined by counting the number of turns required to fill the layer, that is, two turns (Figure 3.10 (a)), which is named *A*, one turn (Figure 3.10 (b)), denoted as *B* or zero (Figure 3.10 (c)), and named *C*. Two of the three layers combined lead to six possible configurations, however, once inserted in a 3D perspective, the Hamiltonian path *C* appears on other configurations when viewed from a different axis. Taking only a valid Hamiltonian path reduces the possible number of configurations to three, as shown in Figure 3.10 (d), (e), and (f), respectively. The voxel configurations are designated by composing the names of the two layers complemented with an apostrophe for a mirror configuration (*A'*) or a suffix “ 90° ” for a clockwise rotation (*A90°*). To avoid confusion, we decided to assign a name to the voxel starting from the entry of the voxel so that only one name could be given. For example, the configuration *AA'* in Figure 3.10 (f) can be understood as *A'A* from bottom to top or *CC90°* from front to back.

3.2.6 Model of the stiffness and deformation

This subsection proposes a model that evaluates the deformation of a voxel depending on its Hamiltonian paths when an external force is applied to a given point on the string, and predicts the effect of the number of reset cycles on the mechanical response. Thus, the mechanical properties of the voxel can be understood, and the mechanical response of a larger structure can be predicted. The string behaves as a serie of torsional springs following the equation: $\tau = k\Theta$, where τ is the torque applied to the corner module, k is the stiffness coefficient, and Θ is the folding angle. To later assess the global deformation of a string structure, the stiffness k of the corner module and how the folding angle deviates with resets needs to be characterised. The experimental process is as follows. First, the corner module was folded in a water bath, then dried and connected to two edge modules. A string with weights ranging from 1 g to 90 g was hung onto a connected module. The resulting torque deformed the corner module and, using a camera, the angle of deformation was

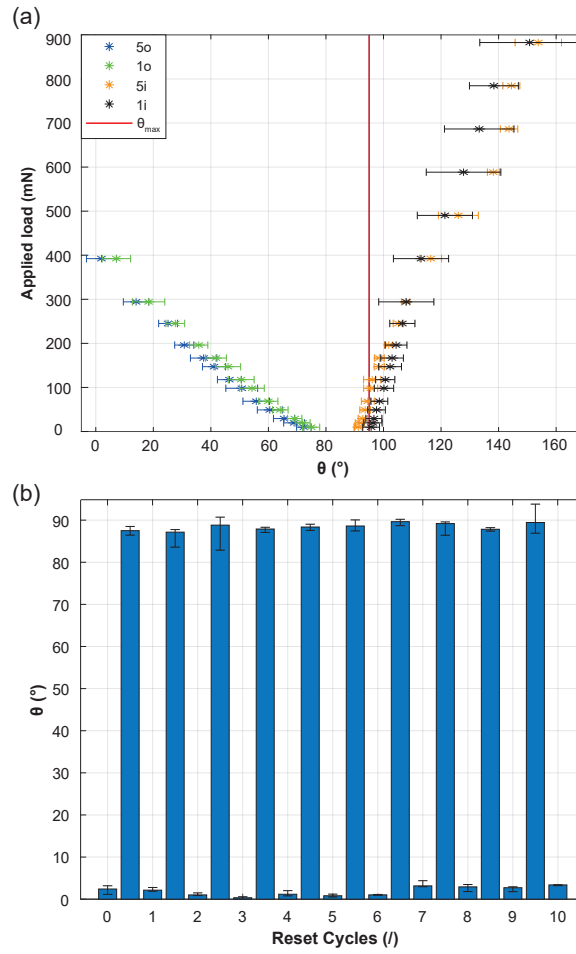


Fig. 3.11 Characterisation of the hinges. (a) Graph depicting the deformation of the hinge when loaded towards the internal or the external direction and depending on the number of resets. (b) Folding accuracy of the hinges once taken out of the water over 10 reset cycles.

measured. The corner module was then resetted by maintaining it in a straight position in the water bath (above 65°C) using weights and folded 5 times before repeating the same loading. The same process was applied for 5 corner modules, in both directions around the folding axis. The experimental setup is illustrated in Supplementary Figure 3.12(b). The numbers in the legend of Figure 3.11 (a) correspond to the number of resets, and the letter indicates the direction of the loading, depending on whether the load deformed the corner module inward "i" (compressing the corner module to $\Theta > 90^\circ$) or outward (pulling the corner module to $\Theta < 90^\circ$). From Figure 3.11 (a), the average stiffness of the corner module inward is $k_{1i} = 23 \times 10^{-4} \text{Nm}/^\circ$ and after five cycles had deviated to $k_{5i} = 20.9 \times 10^{-4} \text{Nm}/^\circ$. However, the outward stiffness is approximately twice as low as the inward loading with only $k_{1o} = -8.47 \times 10^{-5} \text{Nm}/^\circ$ but deviates to only $k_{5o} = -8.15 \times 10^{-5} \text{Nm}/^\circ$. When loaded

inward, the two components of the corner module rest on each other resisting around four and a half times more than in the outward direction, where the PVC strip is the only component holding together the hinge module. In addition, when free of any load, corner modules show a gap between the inward and outward Θ , which is caused by the PVC flexibility, ultimately creating a range of free motion of approximately 20° below the target angle, despite the shrinking of the PVC and the tension it creates (see Figure 3.4). This free motion ultimately induces shifts between the targeted shape and the self-folded voxel; however, as can be seen in Figure 3.11 (b), the folding angle does not vary significantly over ten resets, and no visible deviation occurs, making the corner module reliable in the face of the resets. At the structural level, the load propagates along the string affecting all connected corner modules, but those in contact with the ground are designated as grounded corner modules; we assume that all the load is transferred to the ground rather than to other hinges. Two torques are applied to the string: the PVC counter torque (τ_c) and the torque applied by an external force τ_{in} where i and n are the first and last corner module that one considers. The general equation for the external torque is:

$$\vec{\tau}_{in} = \frac{\sum_i^n \vec{F}_i}{2-a} \cdot \vec{O_iG} \quad (3.5)$$

where $\vec{F}_i$ is the force applied on a given corner module i , $\vec{OG}_{in}$ is the center of mass from corner module i to n , O is the position of corner module i , and a is an index with a value of 1 if the corner module n is not grounded or 0 otherwise. This ties to the assumption that if the corner module n is grounded, then the external forces apply equally to corner modules i and n , thus dividing the force in two. In the opposite case, the forces create a torque solely on the corner module i making $a = 1$ and therefore not dividing the force. The second assumption assumes that the corner modules behave as unidirectional torsional springs with infinite stiffness in directions other than the folding axis. For the structure to hold, the corner module i must not enter plastic deformation, leading to the condition that the holding torque $\tau_{CM_{max}}$ must be greater than or equal to the external torque.

$$\tau_{h_{max}}^{\rightarrow} \geq \vec{\tau}_{in} \quad (3.6)$$

The maximum elastic torque $\tau_{c_{max}}$ is assumed to be k_{1i} and k_{1o} depending on the loading direction. Combined with the free-motion range, the position of the corner modules of the folded voxel can be estimated, as shown in Figure 3.11.

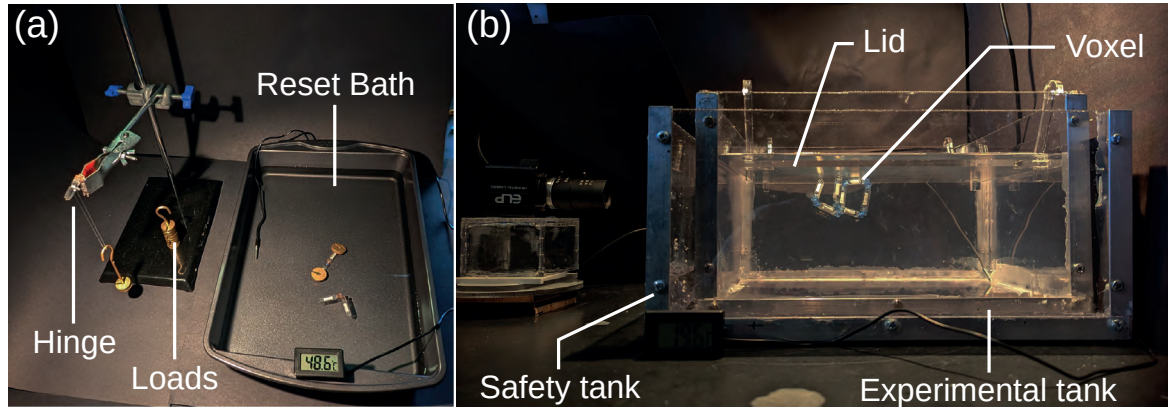


Fig. 3.12 Photography of the different setups: (a) corner module loading setup, and (c) Voxel self-folding setup.

3.2.7 Experimental setup

For our experiments, we required a water tank capable of containing a sufficient volume of water to allow the voxels to fold without interacting with the boundaries of the environment. For this purpose, the experimental tank measured $27 \times 27 \times 16$ cm while the length of an unfolded voxel was 25 cm. For safety reasons, the experimental tank was placed in a wider tank to contain any leak and protect the user in case of catastrophic failures. The tanks were made of 5 mm-thick polycarbonate: a transparent material resisting temperatures of up to 150°C without deformation. The tanks were reinforced with angle aluminium to maintain structural integrity, as it could hold 11.5 L of boiling water, and was sealed using clear silicone sealant. In addition to the tank, a lid was fabricated from acrylic to push down the voxel below the water/air interface.

3.3 Experimental Results

In this section, we test the self-folding of a single corner module attached to two edge modules with different cavity sizes, the self-folding of the voxels, and the detection of the end of the experiment via buoyancy change.

3.3.1 Self-folding of a hinge

The aim of the experiment was to verify the foldable zone highlighted in Figure 3.9 (b) by having voxel edges with different cavity sizes self-fold and compare their folding speeds. Here, we compared the folding speeds of three sets of voxel edges with link modules: (1) without cavities ($X = 0\%$), (2) with the optimum cavity ($X = 19.06\%$), and (3) with the

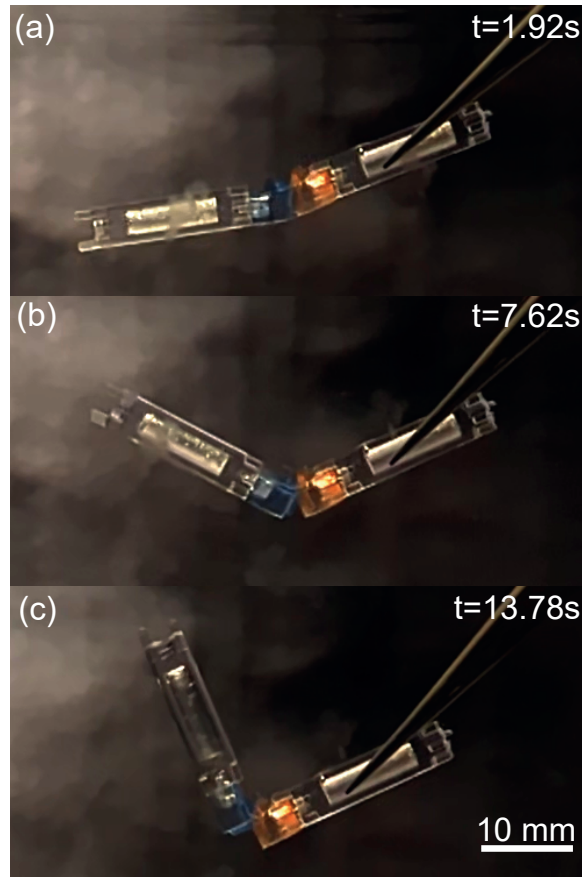


Fig. 3.13 Snapshots of two voxel edges self-folding in a bath of water at 67°C with a cavity of 19.06%.

maximum cavity size ($X = 43\%$). During the experiment, the voxel edges were held in water until folding was completed, and self-folded in the orientation that was the least favorable to folding. In Figure 3.13, the voxel edges with the optimum cavity can be seen folding in around 14 s while, in the Supplementary Video SV1, the voxel edges without cavity succeed to fold in 48 s and the crystal edge with the maximum cavity size fails to fold. Overall, the trend of the model was validated via the speed of folding, as faster folding indicates a lower effect of gravity on identical hinges and under identical environmental conditions.

3.3.2 Self-folding, Sinking and Reprogramming of the voxels

The self-folding process of each voxel configuration was recorded, as shown in Supplementary Video SV2, whereas Figure 3.14 (a) to Figure 3.14 (d) and Figure 3.14 (m) to Figure 3.14 (p) depicts the self-folding of configurations AA and AA'. According to the results in the video, configuration AA' folded in only 4 s, configuration AB in 6 s, and config-

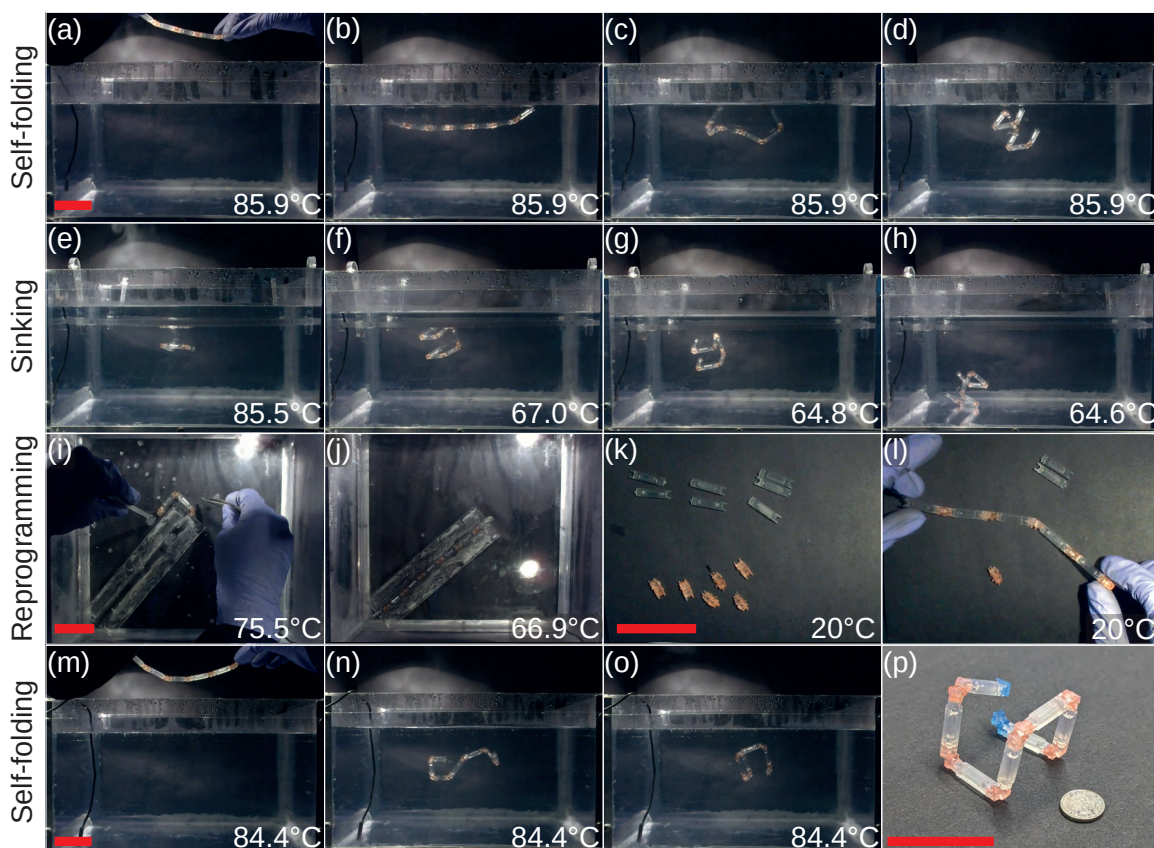


Fig. 3.14 Snapshots of the reprogrammable and reconfigurable self-folding voxel. The string self-folds from its 1D state to the 3D voxel AA' ((a) to (d)) before sinking gradually as the temperature passes from (e) 85.9°C to (f) below 65°C. The tank was emptied and filled with boiling water before the voxel was straightened (i) to be inserted into the resetter (j), while the temperature remained above 65°C. The modules of the string are detached and reassembled into the configuration AA ((k) and (l)), and finally the string is dropped again in water at 89°C to self-fold ((m) to (n)). See Supplementary Video SV2 for a video of the entire process. All scale bars are 5 cm in length.

uration AA in 10 s. The reason for the dispersion in folding time among the configurations is the presence or absence of air within the female connectors between the hinge and link modules. In the absence of air, the buoyancy force was reduced, causing the string to be virtually heavier. As it is dropped in water, the string hits the bottom of the tank, where the folding is constrained before rising again and finishing the folding. In comparison with the folding speeds of the two crystal edges shown in Figure 3.13, folding of the entire voxel is faster by a factor 3. Here, the voxel is free folding as no external force acts upon any of the constituting modules, but the single hinge was held with tweezers, which, in a sense, required the PVC to bring the modules in contact where the string folds around the PVC in free fold. Once self-folding is completed, the voxel remains at the water/air interface owing to the surface

tension of the water, which maintains the voxel afloat. We countered the surface tension by placing a cover that pushed the voxel in the water away from the interface (Figure 3.14 (e)). As long as PVC actuators are exposed to a temperature greater than their glass transition temperature (65°C), they retain the memory of any incurred deformation, which may alter the volume and mechanical properties of the voxel. Therefore, the voxel was left untouched until the change in buoyancy occurred at 65°C . The buoyancy change acts as a detection of the end of the experiment and is encoded directly in the string design via the cavity size in the link module. The buoyancy change occurs owing to the change in the water density with temperature and the thermal expansion of the clear resin. These two effects together let the voxel sink gradually as the temperature dropped below the glass transition temperature of the PVC (65°C), which was validated experimentally in Figure 3.14 (f) to (h). Although the model can predict when the change in buoyancy occurs, a slight difference in the volume of air in the string results in significant changes in the neutral buoyancy temperature. After 8 trials, the neutral buoyancy temperature was determined to be $70.7 \pm 8.1^{\circ}\text{C}$. The presence or absence of air trapped in the female connectors, and possibly air bubbles adhering to the surface of the modules, strongly affect the neutral buoyancy temperature. These three parameters depend on the design, roughness of the material, and printing quality, making the volume within the connections V_{co} more or less airtight. To reduce dispersion, one would need a material with high hydrophilicity so that random bubbles would be pushed away by the water and propose a design containing only tightly sealed chambers. The assumption of the presence of air in the connections was validated by completely drying the modules before the second self-folding. As a result, the neutral buoyancy point was measured to be $63.0 \pm 3.0^{\circ}\text{C}$ in 4 trials.

3.3.3 Voxels and folding accuracy

The orientation of each voxel is illustrated in Figure 3.15. One of the assumptions in the loading model is that the grounded corner modules prevent the excessive deformation of the voxel. This was validated using the bar chart shown in Figure 3.17 but can be graphically identified here. Configurations $AA - 010$ in Figure 3.15 (e), $AA' - 010$ Figure 3.15 (f) and $AB - 010$ Figure 3.15 (k) each possess limited grounding causing the voxel to deform significantly. In contrast, configurations $AA - 001$ in Figure 3.15 (a), $AA' - 001$ in Figure 3.15 (b), $AB - 100$ in Figure 3.15 (g), and $AA' - 100$ in Figure 3.15 (j) deforms significantly less because of the number of grounded corner modules.

To obtain the results shown in Figure 3.17, photogrammetry was used to reconstruct the voxel shape in 3D and extract the corner modules positions. As photogrammetry requires many pictures from various locations, markers around the voxel serves as references, as

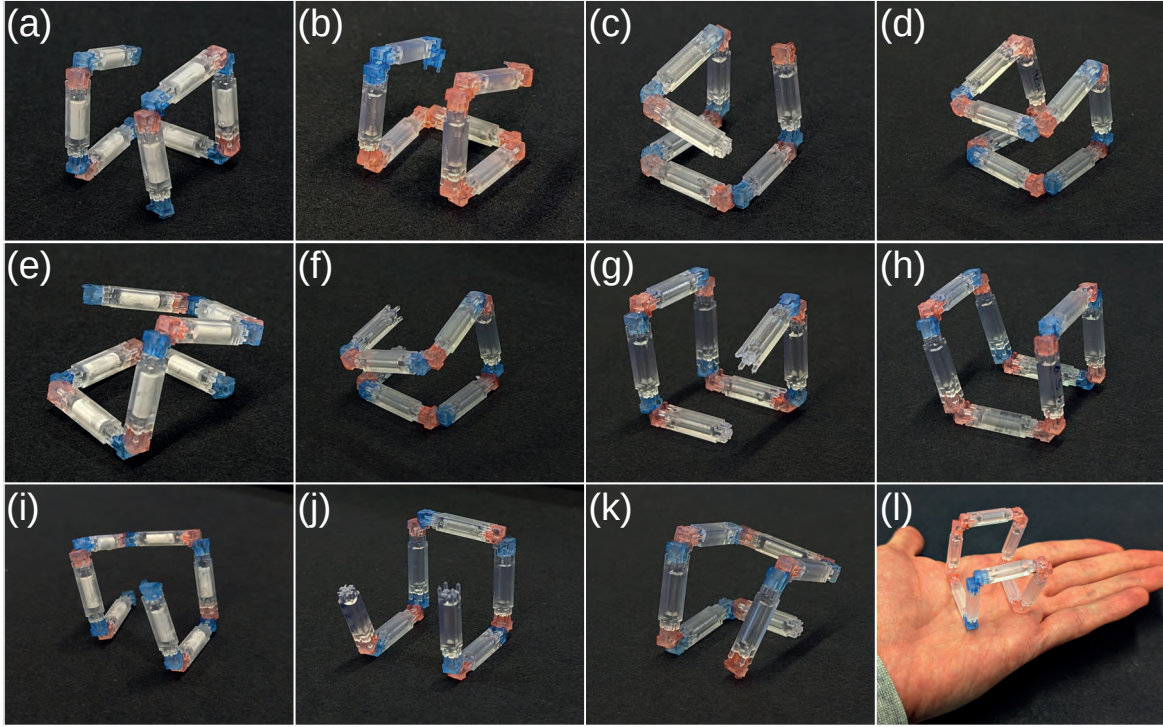


Fig. 3.15 Photographs of the voxels: (a) $AA - 001$, (b) $AA' - 001$, (c) $AB - 001$, (d) $CL - 001$, (e) $AA - 010$, (f) $AA' - 010$, (g) $AB - 100$, (h) $CL - 010$, (i) $AA - 100$, (j) $AA' - 100$, (k) $AB - 010$, (l) $CL - 010$ in hand for scale.

shown in Figure 3.16(a). An example of raw reconstruction of voxel AA is shown in Figure 3.16(b), from which the volume can be assessed in Figure 3.16(c) and the voxel shape can be rebuilt in the CAD software in Figure 3.16(d).

The result of the modelling are exposed in Figure 3.17(a) and (b) where the photogrammetry of voxel $CL-001$ (in blue) is compared to the result interpreted from equation 3.5 seen in gray. In Figure 3.17(a), the voxel is unloaded meaning only the weight of the modules is considered. In this scenario, the model outputs a voxel with a volume of 41.3 cm^3 while the experimental volume was estimated to be 40.3 mm^3 . This represents a mismatch of 2.45 % which was exacerbated to 8.17 % when experiencing a 10 g load in the middle of its hanging perimeter (In Figure 3.17(b)). The main reason for the model mismatch is due to the assumption that the corner modules have infinite stiffness in other axis than the folding axis. The inaccuracy in the assumption is caused by the asymmetry in a corner module indent and slit, which allows it to deform out of plane. On the other hand, the additional degree-of-freedom renders the voxel structurally softer and more resilient to external external forces before breaking.

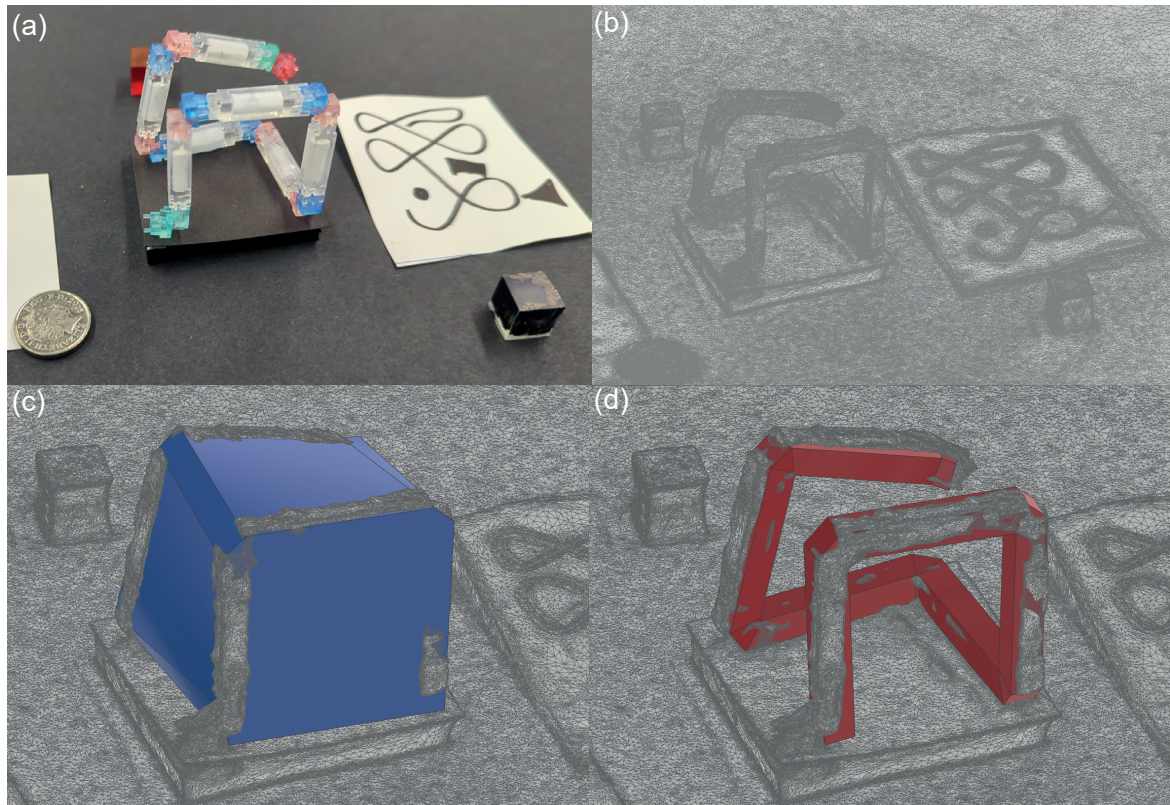


Fig. 3.16 Voxel AA oriented in 001 after photogrammetry (a), meshing (b), volume estimation (c) and voxel reconstruction (d).

Folding accuracy is an important factor for any self-folding structure; however, in the case of self-folding strings, any inaccuracy is propagated along the entire structure. The self-folding accuracy of the voxels was derived by comparing the average error of the position of the hinges between the target voxel and the experimental results. The different orientations of the voxels are designated similarly to crystals with numbers 001, 010, and 100 where each number represents the x-, y-, and z-axes, respectively. Obtaining the positions required the use of photogrammetry, a method for reconstructing 3D objects using multiple photographs of the object (usually more than a hundred) from different angles and distances. With 3D reconstruction, the coordinates of the hinges were used to rebuild the voxels in the CAD software and estimate their volume. The entire process is illustrated in Figure 3.16 in the Supplementary Material. The results of the folding accuracy study are plotted in Figure 3.17 for the different voxels configurations and orientations. Because the voxels can deform significantly with free ends, the configurations AA' and AB were grouped under the same category named closed-loop voxel “ $CLvox$ ” in such a way that the two ends of the voxel are connected. Thus, the influence of hand manipulation can be limited, and dispersion is reduced. Based on the results in Figure 3.17 (c), $CLvox - 001$ and $AA - 010$ are the most inaccurate

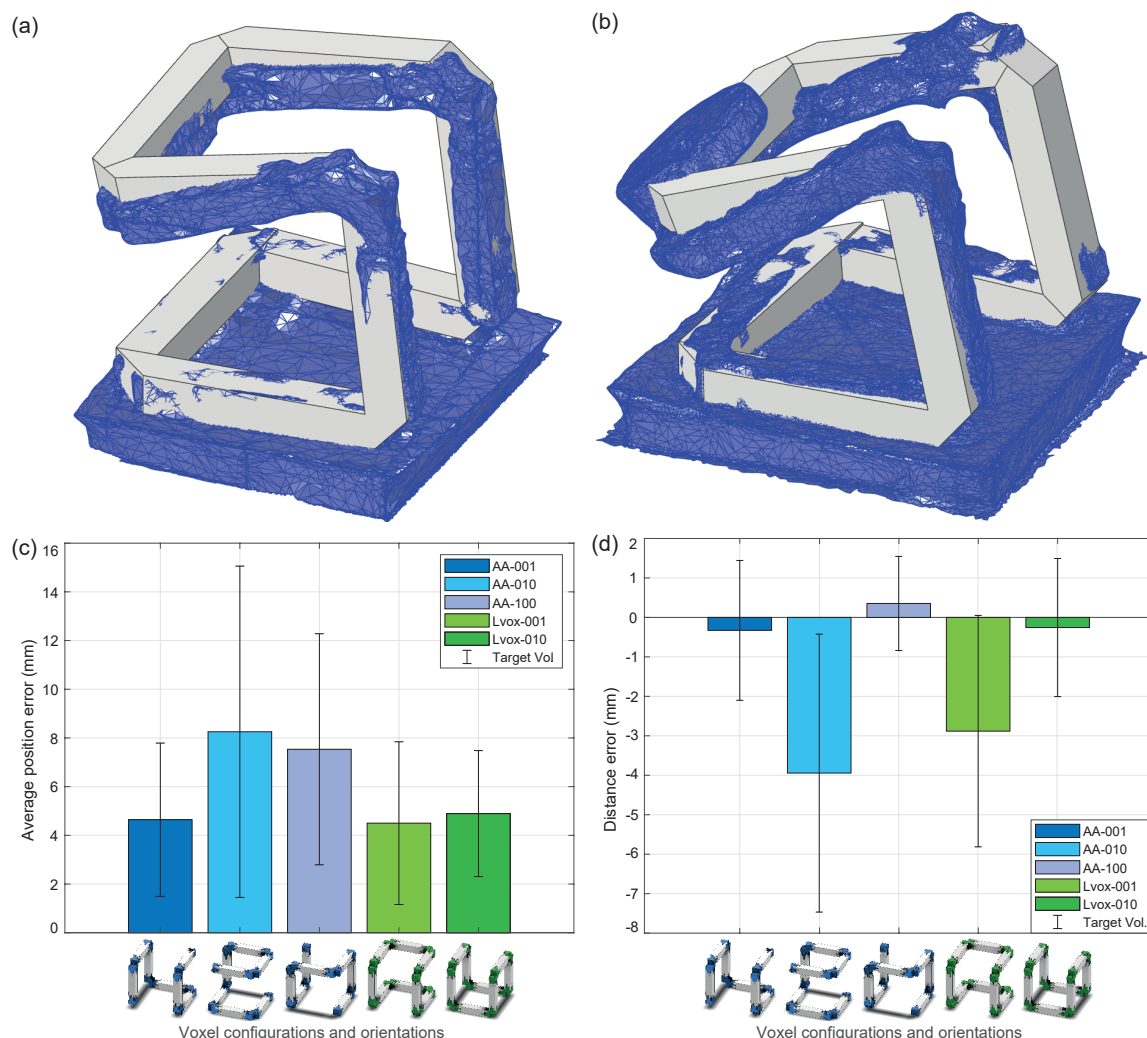


Fig. 3.17 Deformation of voxel CL-001 outputted by photogrammetry in blue and by the model in gray when unloaded (a) and loaded with 10 g in the center of its hanging perimeter (b). Histogram illustrating the average position error of the corner modules within a self-folded voxel depending on its orientation and configuration (c), and average distance error between the corner modules to one taken as reference.

configurations, with average position errors of 4 mm and 3 mm, respectively. However, configurations AA – 001 and CLvox – 010 were the most accurate with only $250\mu\text{m}$ of average position error. In our study, the main factors involved in the accuracy of voxels are: the free motion of the hinge observed around the target folding angle, as shown in Figure 3.11 and the number of grounded hinges directly supporting the hanging edges of the voxel. CLvox – 001 and AA – 010 have 4 and 3 supporting grounded hinges, respectively, whereas CLvox – 001 and AA – 010 have only 2 and 1. Thus, the accuracy of the final shape is directly linked to the number of supporting grounded hinges and looseness of the hinge

modules. With such inaccuracies, scaling up the number of voxels would prove difficult, especially for tall structures. The current hinge design leads to a great discrepancy between the target shape and the experimental result and would certainly cause the collapse of the resulting structure. However, the current corner module design exhibits low stiffness, which makes voxels loose and deformable. While this property could be of interest for obtaining a low-stiffness structure, a rigid structure would require a different hinge design or a stiffer smart material.

3.4 Discussion

3.4.1 Comparison with state-of-the-art

Reference	Type of string	Module Size	Module number	Torque	Medium	Mass in medium	Power Source	Folding in	Actuation Method	Assembly time	Remote folding	Untethered	Reversibility	Addressability	Reprogrammability	Sensing	Assembly and Deformation	Application field(s)
Yim et al. [167]	Chain	3x3x3 mm	999*	$3.2e^{-2}$ Nm [†]	Guide exit	36 g	Magnets	3D	Magnetic pull/push	108 s	Yes	/	Yes	/	/	/	Self-assembly and self-folding	Medical implants
Fass et al. [189]	Chain	20x20x10 mm	6	$2.5e^{-1}$ Nm [†]	Planar surface	>>0 [†]	Magnets	2D	Magnetic pull/push	0.2 s	Yes	Yes	Yes	/	/	/	Self-assembly and self-folding	Minimally invasive surgery
Griffith et al. [172]	Chain	25x25x3 mm	20*	$2.1e^{-1}$ Nm [†]	Air cushion	~0 [†]	Magnets + Air	2D	Magnetic pull/push onto air cushion	30 s	Yes	/	Yes	/	/	/	Self-assembly and self-folding	Programmable matter
Han et al. [43]	Chain	14Øx9 mm	20*	$2.9e^{-2}$ Nm [†]	Agitated hot water	~0 [†]	Magnets + Air + Heat	2D	Magnetic pull/push into bubbling water	10 s	Yes and No	Yes	Yes	/	/	Yes	Self-assembly and stretching	Programmable matter
Gu et al. [185]	Chain	15x5x5 mm	9*	$3.4e^{-3}$ Nm	Guide exit	>0 [†]	Magnets	3D	Magnetic pull/push	13 s	Yes	/	Yes	/	/	/	Self-assembly and self-folding	Minimally invasive surgery
Li et al. [7]	Ribbon	4x5x0.5 mm	5	$1.42e^{-2}$ Nm [†]	Planar Surface	320 mg [†]	Magnets	2D	Magnetic push/pull	/	Yes	Yes	Yes	/	/	/	Self-folding	Robotics and programmable matter
Ijäs et al. [194]	Chain	3x2x2 nm	8064	/	Solution	~0	Atoms	3D	Atomic forces	/	Yes	Yes	/	/	/	Yes	Self-assembly and self-folding	Drug delivery
Douglas et al. [195]	Chain	3x2x2 nm	7308	/	Solution	~0	Atoms	3D	Atomic forces	12h	Yes	Yes	/	/	/	Yes	Self-assembly and self-folding	Drug delivery
Bai et al. [191]	Chain	3x2x2 nm	7249	/	Solution	~0	Atoms	3D	Atomic forces	12h	Yes	Yes	/	/	/	/	Self-assembly	Programmable matter
Cheung et al. [173]	Chain	77x38x38 mm	7*	1.5 Nm	Planar surface	>371 g	Electricity	3D	Servo Motors and Pivot	3 s	Yes	/	Yes	Yes	Yes	/	Torsion	Robotics and programmable matter
Knaian et al. [176]	Chain	1x1x1.5 cm	4*	$4.5e^{-4}$ Nm	Planar surface	14 g	Electricity	3D	Servo Motors and Pivot	5 s	Yes	/	Yes	Yes	Yes	/	Torsion	Robotics and programmable matter
Yim et al. [166]	Chain	1.3cm x cm x 0.1 mm	>30*	/	Planar surface	>0 [†]	Electricity	3D	Servo Motors and Pivot	15 s	Yes	/	/	/	/	/	Pulling and Folding	Robotics and programmable matter
Sung et al. [175]	Chain	>cm x cm x <0.5 mm	57	$4e^{-2}$ Nm	Hot water	>0 [†]	Heat and Magnetic field	3D	Smart Materials Magnetic steering	32 s	/	Yes	/	/	/	Yes	Self-folding	Robotics
Tibbits et al. [168]	Chain	>cm x 6.4Ø mm	63*	/	Hot water	>0 [†]	Heat	3D	Smart Materials	>40 mn	/	Yes	Yes	/	/	Yes	Self-bending	Programmable matter
Uchida et al. [28]	String	>cm x Ø1 mm	/	/	Hot water	~0 [†]	Heat	3D	Smart Materials	/	/	Yes	Yes	/	/	Yes	Self-bending	Medical
Wang et al. [128]	String	>cm x >Ø10 mm	/	/	Hot air or water	>0 [†]	Heat	3D	Smart Materials	>60s	/	Yes	/	Yes	/	Yes	Self-bending and Torsion	Programmable matter
Chapter 3	Chain	30x5x5 mm	6*	$1.4e^{-6}$ Nm	Hot water	~0	Heat	3D	Smart Materials	4 s	/	Yes	Yes	/	Yes	Yes	Self-folding	Metamaterials and Programmable matter

Fig. 3.18 Comparison of the literature on self-folding 1D structures and the characteristics of the chain developed in this chapter. The asterisc in the module number column depicts the 1D structures' number of module demonstrated. The cross in the torque and mass in medium correspond to calculated values inferred from the information within the respective studies.

As mentioned in Chapter 2, the current challenges of self-folding 1D structures include tether-less systems, reversability of the actuation, reprogrammability of the 1D structure and sensing/communication (Figure 5.17). Along with these are research questions regarding the structural integrity, scaling and self-folding torque and state communication.

In this chapter, the chain is made reprogrammable owing to the reversibility of the deformation using shape memory effect of the PVC, and the reconfigurability of the chain by using male and female connectors to build the chain. Moreover, the corner module design ensures a guided deformation during the reprogramming to prevent any unplanned motion during the subsequent folding. Although, occurrences of reprogrammable magnetic ribbons already have been demonstrated [215] and tethered reprogrammable chains have been developed using motors [173] [180], the chain design within this chapter makes, to my knowledge, the first tether-less and reprogrammable self-folding chain using smart materials.

In addition, the chain's modules were made neutrally buoyant to negate the effect of gravity, the main limitation to scale-up the number of modules. As a result, the chain was able to fold in less than five seconds, being the fastest self-folding chain using smart materials under water.

Adding to the design contributions, the hollowness of the voxels allow for Hamiltonian path dependant deformation to appear. This highlights the potential of the work of this chapter for metamaterials which is achieved here using hollow voxels, a similar approach seen in the work of Overvelde et al. [136] and Jenett et al. [133], thus pertaining to the structural integrity research question that is eluded in the field of 1D self-folding structures for programmable material.

Finally, the chain developed here indicates the state of the environment by its change of buoyancy, thus achieving a form of communication that other 1D structure within the literature does not consider.

3.4.2 Success rate

The self-folding of voxels was attempted 26 times, of which 4 failed, resulting in a success rate of 84.6%. In most cases, failures occurred when a single hinge had its PVC detached because of a lack of glue and only once because the PVC snapped in the middle of the bridge length. However, when estimating the success rate of the hinge in all experiments, this accounts for a total of 195 successful hinges out of 202 hinges, giving a far higher success rate of 96.5%. With the success rate of a single corner module, the theoretical success rate of a voxel is 80.7% which closely matches our results. Consequently, the longer the structure, the lower the folding success rate, which can be estimated to be as low as 34.3% for a shape composed of 4 voxels (30 hinge modules). This low success rate can easily be addressed by

replacing the defective hinge with a new one owing to the modularity and reprogrammability of the string.

3.4.3 Buoyancy

The design of the edge module is flexible, as one can easily change the size of the cavity to adjust the buoyancy changes and neutral buoyancy temperature. Coupled with the buoyancy model, our method can be adapted for different liquids and smart materials.

However, this model does not consider any phase transition of the liquid and relies on the theoretical values of thermal expansion. It was seen in Figure 3.9 (a) that an additional factor, which we hypothesize is the pressure within the cavity, heavily affects the volume change of the edge module. Therefore, the mechanical properties of the material when exposed to varying temperatures should be well understood to accurately predict the buoyancy changes or expansion of the edge module validated under the experimental conditions.

3.4.4 Scaling-up

The main challenge regarding the scale-up of the structure is the size of the environment. Currently, the tank can hold approximately one voxel in the string state while holding 10 L of water. With the current method of dipping, increasing the size of the tank would require a large amount of water, challenging the accuracy of temperature control. When scaling up, the holding strength of the connections becomes more relevant to the integrity of the structure. As depicted in Supplementary Figure 3.7 (b), the current connections of a voxel edge can hold around 330 times its weight of 665 mg before the elongation of PVC due to the load unhinges the indents and slit, causing the corner module to gain an additional degree-of-freedom. Hence, the longest string theoretically reach almost 10 m (41 voxels).

3.4.5 Hamiltonian Path

In this article, the range of possibilities of the Hamiltonian path for a structure on a greater scale was not addressed. Future work should emphasize different strategies to avoid collisions during the self-folding of a crystalline-like structure, but also consider how the targeted stiffness for a given structure can be attained by altering the Hamiltonian path so that a user can obtain the desired mechanical response. Similar studies have already been conducted [186] [187] but not on self-folding strings.

3.4.6 Application

String-based structures are currently applied in the biomedical field because of their capability to access narrow environments and perform operations in vivo [216] [156]. However, in a constrained environment, the self-folded structure risks collapse, and the string may entangle. Therefore, the folded structure must be sufficiently stiff and remain unfoldable; two aspects that future work should emphasize.

3.5 Conclusion

In summary, in this chapter a reprogrammable string that free-self-folds into voxels was presented. We proposed a new method to construct voxels from a string, which can self-fold in water, passively detect the end of the experiment by sinking, and can be reprogrammed using the shape memory aspect of PVC and by interchanging the position of the modules of the string. We designed a new self-folding corner module, characterized its stiffness, in addition to the folding angle deviation of the hinge. Divided into 8 subvoxels, 3 Hamiltonian paths were identified within the voxel, and each of the 3 configurations was self-folded. We proposed a method to assess the accuracy of self-folding from a structural point of view and proposed a model to estimate the torque propagation along the string. Finally, we studied buoyancy at a modular level to provide an environment simulating microgravity and enabling long strings to fold with limited torque.

Here, the following research questions were approached:

1. How the limited energy that smart-materials provide be circumvented for strings to self-fold into large 3D structures?
2. How can a self-folding string without embedded electronics communicate its state?
3. How to make self-folding strings reprogrammable to widen the range of structures and grant them a universal shape change capability?
4. How does the Hamiltonian path relates to the final mechanical properties and what kind of mechanical properties can be expected from string-based structures?

Here was presented a reprogrammable string that free-self-folds into voxels using smart-materials as actuator. To render the string self-foldable, the design of the string was adapted to be neutrally buoyant so that the required self-folding torque was reduced to near zero. This method contrasts from existing alleviating environments such as air cushion [172] and bubbling water [43], where the folding or assembly is two dimensional only.

Concerning the electronic-less sensing, the problem was approached by adjusting the temperature of neutral buoyancy. This lets the string change from buoyant to sinking when the temperature of activation of the smart-material is passed. As a result, the string gives a visual and positional feedback within the tank indicating the end of the self-folding process regardless of the success or failure of the self-folding. Further work should be accomplished in this direction to separate the successful and failed structures depending on the main topological difference between success and fail: the volume.

To further the capability of the strings regarding the challenge of shape morphing and universality, the string was designed to be reprogrammable. For this purpose, the hinges design were adapted to be manually resetable after re-exposure to the stimulus, and the string was made reconfigurable by the modular aspect helping in adapting the direction of the hinges and change the morphology of the structure after self-folding. Hence, the necessity for a structure to be resetable and reconfigurable in addition to be modular to be reprogrammable. Although, this is only true for a highly ordered structure such as crystals where all units are of the same volume and dimensions.

The current focus of the literature regarding self-folding strings has mainly be shape-morphing, leaving a significant gap as to what can be expected in terms of mechanical behaviour of string-based structure. Here, the folding torque and the stiffness of the hinges are characterised with the aim of demonstrating the deformation of voxel when loaded. This aspect of the field could be deepened by simulation work relating the Hamiltonian path to mechanical response or testing in practice the resistance of different crystal shapes and Hamiltonian path within it.

Chapter 4

Strings Reversibly and Permanently Self-Folding into Semi-Crystalline Structures

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4.1 Summary of Chapter

In this chapter, I present my approach to mimicking semi-crystalline structures from a chain. The lamellae in semi crystalline structures is similar to a tower with several layers, where each layer is self-folded in a zigzag pattern. This configuration presents a significant advantage compared to crystalline and amorphous structures: the structure is self-foldable, regardless of the folding sequence. Currently, only two approaches have attempted to replicate semi-crystalline structures. Yim et al.'s approach consisted of a servomotor pulling tethers to fold the flat chain into semi-crystalline-like structures [167], while C. Sung's approach removed the tethers to directly use a thermoresponsive smart material to self fold a remotely actuated robot[179]. However, both approaches exhibited permanent folding. Though reversibility in the self-folding grants the structure several functions via shape reconfiguration, the removal of the stimulus brings the structure back to its initial state, thus losing its shape after activation.

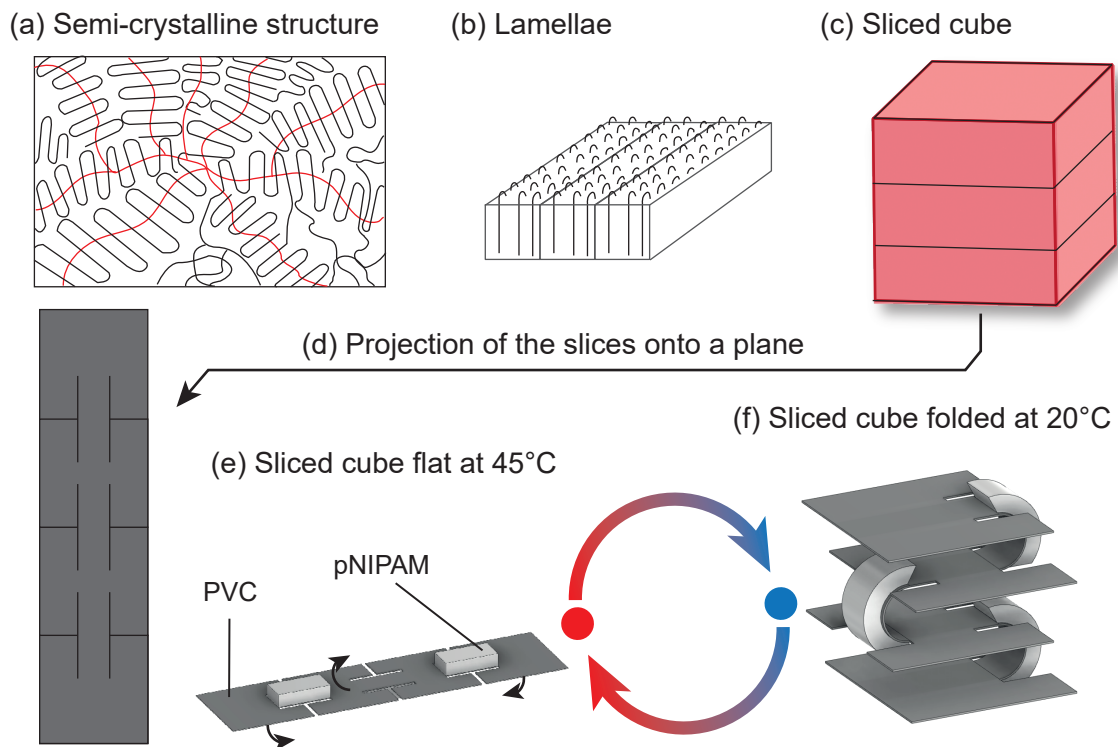


Fig. 4.1 Illustration of the concept of the asemi-crystalline structure (a) and of lamellae (b). To replicate the accordion structure of the semi-crystalline structure, 3D shape are sliced (c). The slices are projected onto a plane, and spread to obtain a pattern (d). Using the pattern, pNIPAM is fabricated against the PVC pattern and the structure can transform from a flat state at 45°C (e) and a folded state at 20°C.

Herein, I present a tether-less semi-crystalline structure capable of reversibly self-folding from a flat state to a semi-crystalline state and of permanently deforming to turn off the structure. To make the self-folding reversible, I propose a novel fabrication process for a bilayer made of a thermo-responsive PVC shrink film and a thermo-responsive hydrogel named Poly(N-isopropylacrylamide) abbreviated as pNIPAM. The hydrogel swells and shrinks reversibly between 20 and 45°C and is bonded to a layer of PVC shrink film that is passive below 65°C but shrinks beyond. In this way, the volume change of the pNIPAM enables the bilayer to be flat at 45°C and bent at 20°C, while the deformation of the PVC locks the bilayer in a bent state. The process consists of four main steps:

- PVC cutting and cleaning,
- PVC surface activation by plasma treatment,
- Synthesising the pNIPAM and bonding,
- Cleaning the pNIPAM string.

The bilayer performance was assessed to optimise the size of the actuator and understand the behaviour of the bilayer depending on the two main parameters: the thickness of the pNIPAM and its stiffness tuned via the cross-linking density. As a result, the bilayer was able to perform 180° bending in 12.5 mm, but it was also capable of exhibiting three bending behaviours between 20°C and 45°C: from bending outward to flat for low crosslinking density, from bending outward to inward for medium crosslinking density, and from bending outward to flat for high crosslinking density. PVC being a shape-memory polymer that shrinks at temperatures above 65°C, the duality pNIPAM/PVC allows the structure to change shape both reversibly and permanently. I characterised this behaviour on single hinges and demonstrated on a magnetically actuated robot, in which the locomotion performance was a function of the shape. All the materials used for the semi-crystalline structures were bio-compatible, except for the magnet, which can be coated with another bio-compatible material. Thus, the robot and structures developed in this chapter can perform safely within the body.

In summary, this chapter contributes to self-folding strings by:

- Proposing reversibly self-folding semi-crystalline structure,
- Introducing dual thermal activation to obtain reversible folding and to turn off the reversibility,
- Introducing a novel fabrication process.

4.2 Methods

4.2.1 Mechanism and principle

One approach to fabricate an origami self-folding structure is to laminate a “passive” material to a volume changing smart material and create a bilayer structure. When exposed to a stimulus, a passive material maintains its volume, whereas the smart material undergoes a volume change. Depending on whether the smart material swells or shrinks, the volume change of the active material induces tensile or compressive forces on the passive material, resulting in a different bending direction of the bilayer.

Here, the hydrogel pNIPAM and the shape memory polymer PVC are used. Poly(N-isopropylacrylamide) (pNIPAM) exhibits a thermo-responsive behaviour in aqueous solutions, undergoing a phase transition at its lower critical solution temperature (LCST) of approximately 32°C. Below this temperature, pNIPAM is hydrophilic due to its amide group ($-CONH-$), but becomes hydrophobic beyond due to the weakening of the hydrogen bonds of the amide group and water molecule, thus the hydrophobicity of the propyl group ($-CH(CH_3)_2$) takes over and due to the shrinking of the molecular chain, release the water from the polymer network [48].

In this study, between 20°C and 32°C, pNIPAM swells, causing the bilayer PVC/pNIPAM to bend. Between 32°C and 45°C, pNIPAM shrinks and the bilayer becomes flat. While, once exposed to 65°C, the PVC volume shrinks permanently, locking the bilayer in a bent state. Hence, a shape memory bilayer can be produced with reversible bending and permanent deformation enabled by the pNIPAM and the PVC shape memory.

4.2.2 Fabrication

Plasma treatment of PVC

Replaced by: Plasmas are commonly used in microfabrication to treat materials surfaces, deposit thin layers of material or enable bonding between two materials. Here, the aim is to make the PVC surface hydrophilic to bond with the pNIPAM and prevent the PVC hydrophobicity to influence the kinematic of our structures.

The PVC was treated with an air plasma (78% nitrogen and 21% oxygen) at 200 W at a pressure of 0.45 mbar and the duration of the treatment was varied to study the extent of activation of the PVC surface, as shown in Figure 4.2 (a). The surface activation of the PVC was characterised by depositing a 10 μ L de-ionised water droplet onto a 100 mm² PVC square activated surface and measuring the surface coverage of the droplet using a microscope.

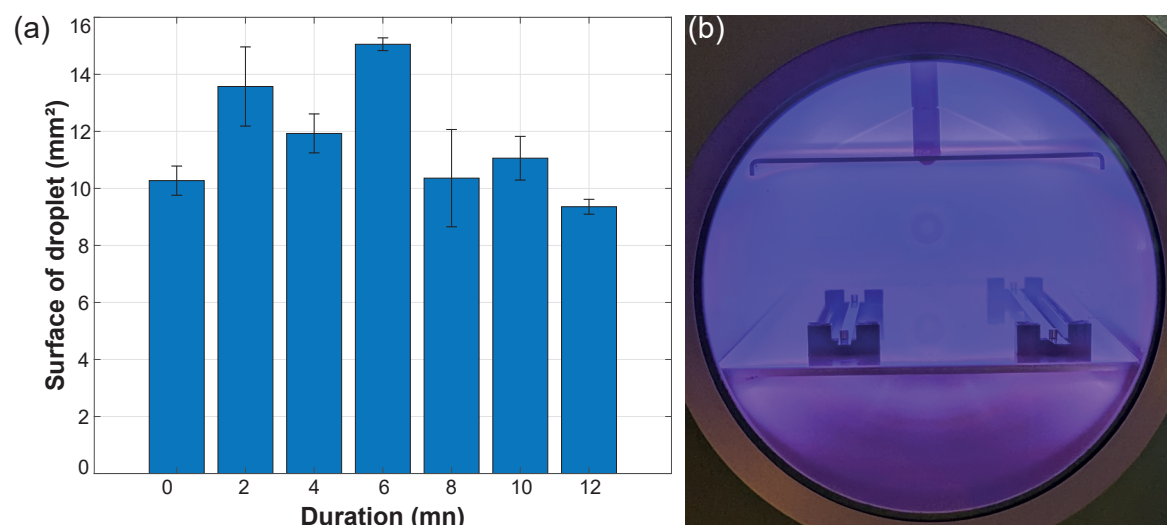


Fig. 4.2 PVC treatment of the shrink film PVC. (a) Graphic of the surface coverage of a $10\mu\text{L}$ water droplet onto PVC exposed to a 200 W air plasma for various durations. (b) Photograph of the lyre and PVC strips in the plasma.

Initially hydrophobic, the droplet forms a pearl on the PVC covering a surface of approximately 10mm^2 . As the duration of the treatment is increased to 6 mn, the droplet spreads on the surface up to 15mm^2 indicating a higher surface energy. Above 6 mn of treatment, the treatment must be split into two cycles as the temperature within the chamber of the plasma coater increases beyond 65°C , resulting in the PVC shrinkage. Between the two cycles, the plasma coater chamber was left to cool and the activated PVC was protected under a box to prevent dust from interacting with the surface. However, after more than 6 mn of plasma treatment, the water coverage decreases. This is most likely due to the generation of “ashes” onto the surface because the atoms of the broken bonds cannot be removed anymore by the airflow within the barrel, and thus leading to a de-energised and hydrophobic surface.

Bilayer fabrication

The production of a PVC/pNIPAM bilayer consists of five main steps: moulding, pNIPAM synthesis, PVC preparation, UV exposure, and cleaning. First, a rigid mould is 3D printed with an SLA printer (Formlabs Form 2). Then MoldStar-15, a silicone resin, is poured in and left to cure for 12 hours (Figure. 4.3 (a)) resulting in a soft mould for easy moulding and demoulding of the pNIPAM due to the hydrophobicity of silicone. The PVC (reference L119Y80C162AM110OBS7 from SupVox) is $24\mu\text{m}$ thick and is manually cut into 3 mm wide strips, the length of which varies depending on the designs (see Section 4.3). After cutting, the strips were washed in a bath of ethanol and in a bath of de-ionised water.

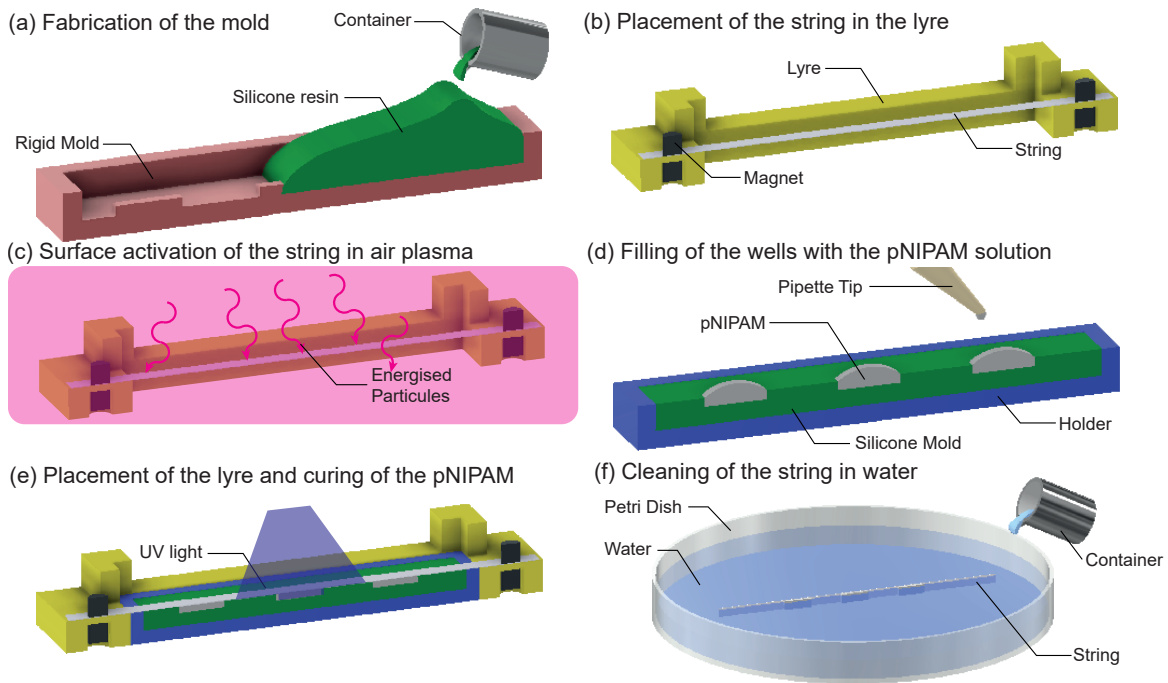


Fig. 4.3 Main steps of the fabrication of pNIPAM/PVC bilayer. (a) Fabrication of the soft mold by pouring MoldStar-15, a silicone resin, in a SLA 3D printed mold. (b) Placement of the PVC string on the FDM printed lyre and trapping of the string between the magnets. (c) Plasma treatment of the string mounted on the lyre. (d) Filling of the wells of the Moldstar mold with pNIPAM solution using a micropipette. (d) Placement of the lyre onto the mold and UV curing of the pNIPAM hinges. (e) Cleaning of the pNIPAM in distilled water at 45° C.

Once dried, the string is placed on a lyre, to hold the strip in position (Figure. 4.3 (b)). Then, the string is treated in the plasma coater (HPT-200) at 200 W for 6 min at 0.45 mbar (Figure 4.3 (c) and Figure 4.2(b)) to promote the adhesion of pNIPAM onto PVC. A holder, in poly-lactic acid (PLA), is 3D printed with an FDM printer to prevent the soft mould from deforming out of shape (Figure 4.3 (d)). If too large, the deformation of the soft mould can alter the shape of the wells and prevent the alignment of the string onto the mould. The pNIPAM synthesis consists of mixing three reagents: the monomer N-isopropylacrylamide (NIPAM-200 mg/mL), the crosslinker N,N'-Methylenebisacrylamide (BIS-8 mg/mL) and the photo-initiator Irgacure-2949 (4 mg/mL). The reagents were diluted in a 50 : 50 solution of ethanol and de-ionised water. The pNIPAM solution was then deposited in the wells of the silicone mould (Figure 4.3 (d)). The lyre was fitted onto the holder such that the PVC string sealed the wells and was in contact with the pNIPAM solution. To cure the pNIPAM, a UV Torch (UTarget-365 nm) at $20\text{ W}/\text{cm}^2$ was used to cure each pNIPAM panel. For this purpose, the wells were exposed twice to 60 s (Figure 4.3 (e)). As PVC is a heat-shrinkable

material, attention must be given to the heat received by the string. During curing, pNIPAM absorbs the energy irradiated by the torch and heats up. Splitting the exposure into two allows the pNIPAM and the PVC to cool down and consequently prevents the PVC from shrinking. The string was then demoulded and placed in a bath of DI water at room temperature until the pNIPAM panels were fully hydrated (Figure 4.3 (f)). Then, the string was successively placed into a bath at room temperature to re-hydrate and in a bath of hot DI water to ensure the extraction of unused reagents which may negatively affect the volume change of pNIPAM.

4.2.3 Characterization of the bilayer

pNIPAM volume change

The aim of the first characterisation is to quantify and evaluate the trend in the volume change of pNIPAM. In a soft mould, 3 pNIPAM panels were synthesised to be 7 mm long, 5 mm wide, 3 mm thick and with a hole in the centre to anchor the panels to the Petri dish. A bain-marie was used to measure the volume change in the pNIPAM panels, which were placed in a Petri dish and immersed in DI water. The Petri dish was then placed in a bowl filled with DI water and the bowl was heated slowly to 45° C using a hotplate. The bain-marie guarantees a slow and uniform temperature rise which allows for a more accurate temperature measurement. The volume change in Figure 4.4 (a) is the average of 3 panels upon heating (red area) and cooling (blue area) of the bain-marie. The LCST can be defined as the point of volume change with the sharpest slope and was evaluated by differentiating the swelling ratio against temperature. The global LCST was calculated as the average LCST for heating and cooling and was found to be 32.1° C. The volume change can be approximated as a sigmoid function [202] occurring between 20° C and 45° C. The sigmoid equation applied to the volume change of pNIPAM is :

$$SW(T(^{\circ}C)) = \frac{A}{1 + e^{-c_1 \times (T+c_2)}} + SW_{min} \quad (4.1)$$

where A is the amplitude of the volume change (%), c_1 is the slope of the sigmoid (%/° C), c_2 is the LCST (°C) and SW_{min} is the maximum shrinkage of the pNIPAM panels (%). Coefficient c_1 was calculated by inverting the sigmoid function and was evaluated to be 0.467%/° C. The trend, plotted in Figure 4.4 (a), fits the measurements except on the “shoulder” of the sigmoid below the LCST. The measurements, the measured LCST, the range of activation and the trend are similar to those obtained by Ehrenhofer et al. [202]. This validates the experimental setup and the use of a sigmoid function for further interpretations.

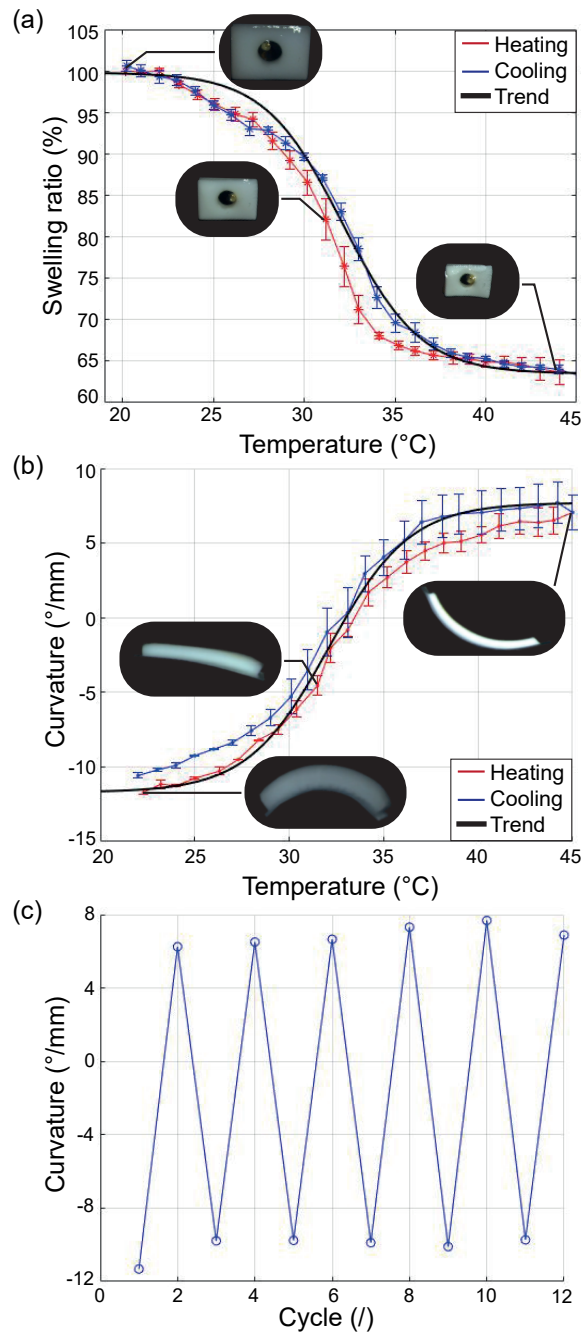


Fig. 4.4 Characterization of the pNIPAM and the pNIPAM/PVC bilayer: (a) Average on 3 samples of the swelling ratio of the pNIPAM panels upon heating (red) and cooling (blue). (b) Average of the curvature of 3 bilayers upon heating (red) and cooling (blue). (c) Repeatability of the curvature of a bilayer for 12 cycles measured on one sample.

Reversible bending

The second characterisation aims to provide a reasonable estimation of the hinge length for a targeted angle and its variation with respect to temperature. The bilayers were fabricated

in a manner similar to that for the strings. The wells in the mould were 7 mm long, 3 mm wide and 0.72 mm thick. The pNIPAM/PVC bilayer has 3 domains: an inward bending below the LCST, a zero bending point at the LCST and an outward bending beyond the LCST. According to the results plotted in Figure 4.4 (b), the bilayers exhibit an average maximum curvature of $11^\circ/\text{mm}$ when bending inward and an average maximum of $7.5^\circ/\text{mm}$ when bending outward. The explanation for the inversion of the curvature lies in the volume change of pNIPAM on PVC. pNIPAM is cured in a mould and shares the same volume noted V_M . Below the LCST, pNIPAM swells to a volume V_{LCST-} greater than that of the mould ($V_{LCST-} > V_M$). This causes the bilayer to bend inward, implying that pNIPAM exerts a compressive force on the PVC. When the pNIPAM reaches the LCST, the bilayer is straight indicating that the volume of pNIPAM is similar to that of the mould ($V_{LCST} = V_M$). Above the LCST, pNIPAM shrinks to a smaller volume ($V_M > V_{LCST+}$) than that of the mould. This causes the bilayer to bend outward, implying that the PVC receives tensile forces from the pNIPAM. Therefore, the bending orientation is governed by the change in the reaction forces induced by pNIPAM in the PVC. The sigmoid trend on the curvature κ (see Figure 4.4 (b)) and on the swelling ratio exhibit similar inflection points and slopes at the inflection point, but with different amplitudes and minimum value as shown in the equation below :

$$\kappa(T(^{\circ}\text{C})) = \frac{B}{1 + e^{c_1 \times (T + c_2)}} + \kappa_{min} \quad (4.2)$$

The resulting trend fits the measurements and indicates that the entire range of the volume change of pNIPAM is used for bending (see Figure 4.4 (b)). The volume change of the pNIPAM and its stiffness are closely related; when swollen, the pNIPAM has a low Young's modulus, and when shrunk, pNIPAM has a higher Young's modulus. The Young's modulus changes accordingly to the concentration of crosslinker to monomer and curing conditions [48]. If the Young's modulus of the pNIPAM were too low, pNIPAM would not be able to bend the PVC and a drop between the trend and the measurement of the curvature would be observed. In our case, the entire range of the volume change of pNIPAM was used to bend the PVC indicating that the stiffness of the pNIPAM is optimal. Equation 4.2 can be used to control the bending of one hinge. However, in Section 4.3, the strings are straight and loose when they are bent in the other direction. This is explained by the change of reaction forces in the string. When swollen, all the hinges are tensing the string, which provides stiffness to the structure; however, when shrunk, the hinges exert compressive forces, that loosen the string.

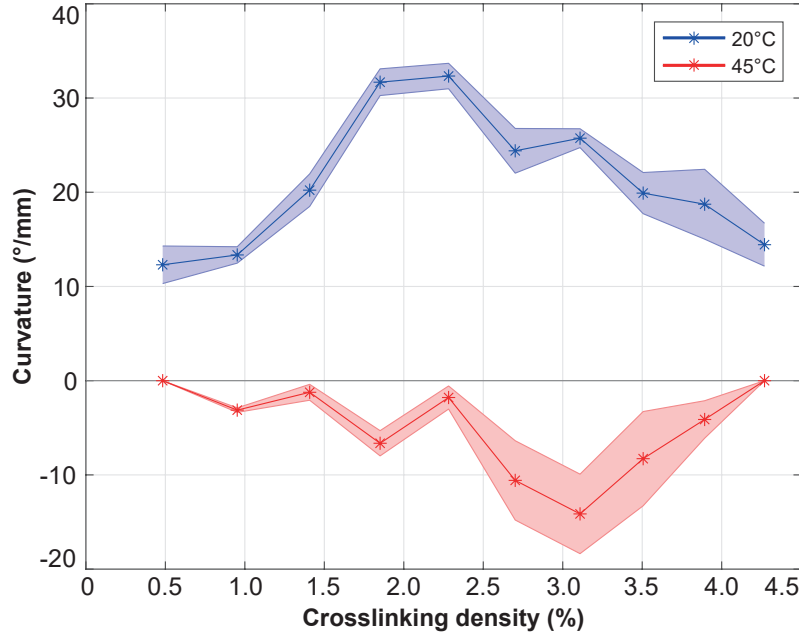


Fig. 4.5 Graph of the curvature of the bilayer depending on the crosslinking density of the pNIPAM and the temperature.

Curvature and pNIPAM stiffness

Derived from Bernoulli and Timoshenko beam theory, the bilayer curvature κ is a function of the layers height t , stiffness E and volume change extent ε as shown in 4.3.

$$\kappa = \frac{6E_1E_2(t_1 + t_2)t_1t_2\varepsilon}{E_1^2t_1^4 + 4E_1E_2t_1^3t_2 + 6E_1E_2t_1^2t_2^2 + 4E_1E_2t_2^3t_1 + E_2^2t_2^4} \quad (4.3)$$

The third characterisation consists of studying the stiffness of the pNIPAM via its crosslinking density and measuring the extent of bending at 20°C and 45°C.

In a sense, the crosslinker in a polymer acts as a crossroad for other monomers to branch from. As the density of the crosslinker increases, the stiffness of the polymer rises. In hydrogels, this is also highlighted by their capacity to swell in water. The polymer chains being more connected, swelling is limited and less water can flow in or escape from the polymer network. Consequently, the bilayer should bend to a lesser extent as the crosslinking density increases. Similarly, the curvature of the bilayer should be reduced if the pNIPAM layer is too thin as it should not develop sufficient torque to bend the PVC.

Bilayers with identical features to those in the previous characterisation experiment were fabricated except that the crosslinking density was varied from 0.5% to 4.3%, calculated as

the mass ratio of crosslinker to monomer and photoinitiator. For each crosslinking density, 5 bilayer curvatures were measured under a microscope at both 20°C and 45°C. The results, plotted in Figure 4.5, highlight three main characteristics of the pNIPAM/PVC bilayer. First, at a low crosslinking density, pNIPAM bends the PVC the least at 20°C and 45°C. Second, the bending behaviour can be controlled with only inward bending at crosslinking densities below 0.5% and above 4.2%, and inward and outward bending at crosslinking densities between 0.5% and 4.2%. Third, the optimum crosslinking density for the maximum inward bending is situated at approximately 2.0% whereas the maximum outward bending is measured at 3.1% crosslinking density.

Repeatability of the bending

The fourth characterisation verifies the repeatability of the self-folding and self-unfolding of the bilayer, as can be seen in Figure 5.3 (c). The bilayer is the same as that used for the characterisation of the curvature. The bilayer was immersed in a bath of DI water at room temperature, and then dipped in a bath of DI water at 45°C. The average curvature at 45°C is $-10.1 \pm 0.3^\circ/\text{mm}$, and the average curvature at room temperature is $6.9 \pm 0.26^\circ/\text{mm}$. The low standard deviation of the curvature averages indicates a high repeatability of self-folding and self-unfolding.

Permanent bending

The pNIPAM volume change results in the bending of the bilayer, but PVC is a shrink film that experiences a volume change when exposed to temperatures higher than 65°C. Hence, the exposure of the bilayer to high temperatures should result in a deformation different from that of pNIPAM. This hypothesis was tested on 5 bilayers of the same dimension as the one used for the characterisation of the bending of the pNIPAM in Section 4.2.3 but 1 mm thick instead of 0.72 mm thick. The resulting deformation is shown in Figure 4.6. First, the bilayers were immersed in water at 20°C and, as shown in Figure 4.6 (a), the bilayers bent inward. The water was gradually heated to 85°C and the bilayers became flat before bending inward again as shown in Figures 4.6 (b) and (c). The water was then left to cool to 20°C, and the bilayers maintained their shape Figure 4.6 (d). To validate that the deformation was permanent, the bilayers were heated again to 45°C and the bilayers kept their shapes despite the shrinking of the pNIPAM in Figure 4.6 (e). While this behaviour was observed on the 5 bilayers tested, the same activation resulted in different deformations in the following experiment shown in Section 4.3 and 4.6. This lack of repeatability may be explained by numerous factors related to the fabrication of the bilayer and the smart

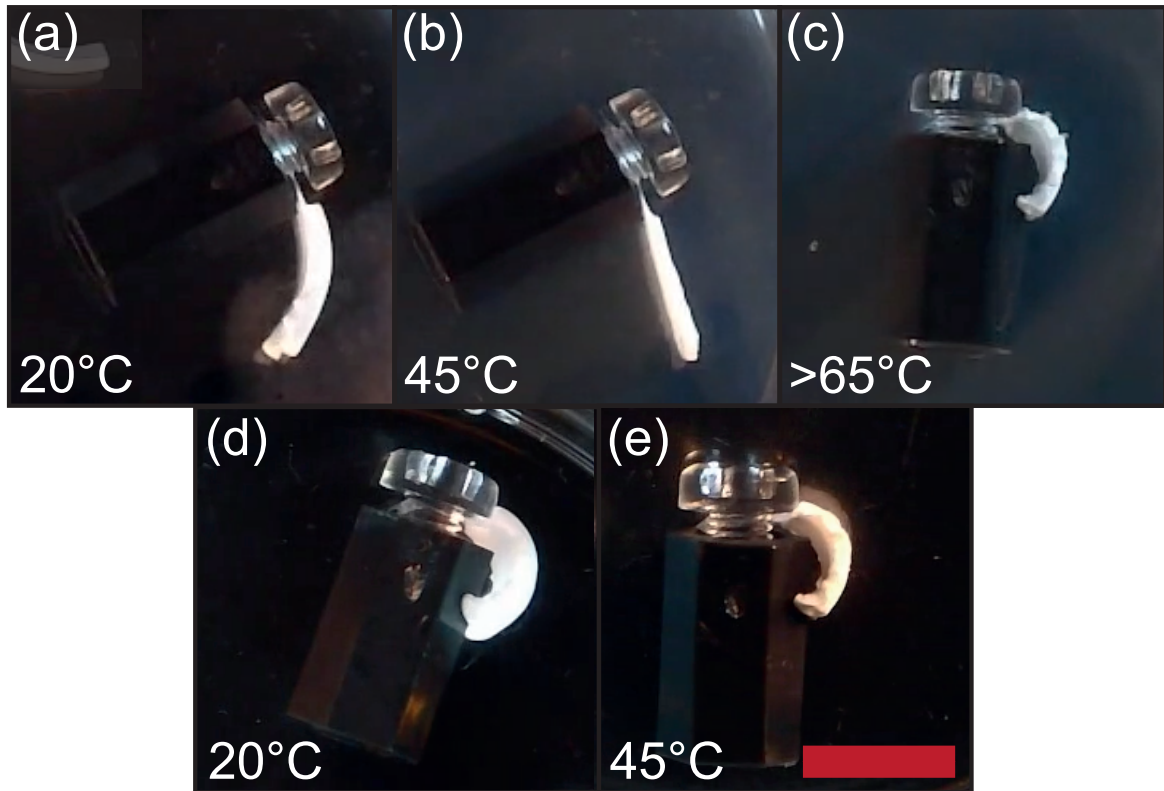


Fig. 4.6 Deformation of the bilayer shown from 20°C in the bent state (a) to 45°C in the flat state (b) due to the pNIPAM volume change, and then exposed to 65°C triggering the PVC shrinkage (c), cooled down to 20°C (d) and re-heated to 45°C (e) to show how the reversibility of the bilayer was turned off. Scale bar is 7 mm

materials. In the case where the PVC strip is not parallel to the pNIPAM well, the pNIPAM induces a lateral deformation of the PVC. Coupled with the anisotropic shrinking of PVC, the lateral deformation may result in a bistable bilayer whose potential energy may influence the permanent deformation of the bilayer. In addition, the permanent deformation seems to be affected by the bilayer initial position and orientation in the environment, as shown in Figure 4.18 and Figure 4.11 (b). When the pNIPAM solution is poured into its well and PVC is deposited on top, the capillary effect allows the excess pNIPAM solution to spread under the surface of the PVC. Thus, if overfull, the nearby empty wells start filling up with the pNIPAM solution, which is cured and bonded to the PVC during UV exposure. If this pNIPAM chunk in the neighbouring well is next to the anchoring when the PVC shrinks, this pNIPAM chunk hits the anchoring and acts as a buttress, thus inducing folding similar to origami folding mechanism, as can be seen in Section 2.

4.2.4 Slicing for Semi-Crystalline Structures

Slicing is an important part to replicate semi-crystalline structures. As mentioned in Chapter 2, semi-crystalline structures are neither crystalline nor amorphous, as they exhibit some form of ordering. This ordering takes the form of a lamellae that can be described as a structure of a regularly stacked chains of polymer.

Translating lamellae or semi-crystalline structures into foldable structures requires the hinges to fold 180° in successive mountain and valley fold so that each layer is above the previous one. This arrangement can be viewed as a zig/zag or an accordion. The algorithm replicating a semi-crystalline structure at the macro-scale has already been proposed by Sung et al. [179] and Yim et al. [167] and consists of slicing a 3D shape into many layers and interconnecting them edge to edge afterwards. Compared to previous works, which relied on folding, here, the slicing algorithm accommodates bending. The only difference between bending and folding is the acuteness of the deformation, with folding having a sharper deformation than bending. For example, a fold could exhibit a curvature of $90^\circ/\text{mm}$ whereas a bend could exhibit a curvature of $10^\circ/\text{mm}$. Because of the low curvature and the length of the hinges required to perform a 180° bending, the hinges should be embedded within the shape. If not done so, the hinge appear on the outside of the shape, thus misforming the targeted shape. Since the shape of an object and its function are closely related, embedding the hinges is an essential predicament to maintain the functionality of the targeted structure.

The slicing algorithm is detailed in Figure 4.7 and below

1. Define the width of the hinge W_h , the length of the hinge L_h and height of the slice Z_s
2. Import STL object (Figure 4.7 (a))
3. Calculate the height of the STL object Z_{max}
4. Calculate the number of slices $N = Z_{max}/Z_s$
5. For each slice from $i = 0$ to N ,
 - 4.1 Create a plane Pl at $Z = i \times Z_s$
 - 4.2 Find the intersection of Pl and the polygons of the STL file
 - 4.3 Create the perimeter P_i
 - 4.4 Project P_i onto the plane $Z = 0$ (Figure 4.7 (b))
 - 4.5 Offset P_i in x axis by the width of P_{i-1} and P_i (Figure 4.7 (c))
 - 4.6 Identify the segment of contact S between P_{i-1} and P_i
 - 4.7 If S is shorter than W_h , then
 - 4.71 Offset P_i in -x axis until the points of contact between P_{i-1} and P_i are equal or above W_h

6. Trace on either side of the contact points and on either side of the x axis lines of length L_h separated in y axis by W_h (Figure 4.7 (d))
7. Erase any points of the perimeters P_i and P_{i-1} between the contact points (Figure 4.7 (e))
8. Join together all points of P_{i-1} , P_i and the lines in one matrix (Figure 4.7 (f))
9. Export all points coordinates

The algorithm has four inputs: the STL file of the 3D shape, the width of the hinge, its length and the layer height. The hinge length L_h is determined by the curvature of the bilayer which is function of the mechanical properties, volume change and thickness of the materials as depicted in equation 4.3. It directly impacts the slice height Z_s which is calculated as the distance between the start and end of the bilayer bending at 180° . Consequently, characterisation of the bilayer bending abilities is necessary to output a semi-crystalline structure using the algorithm, which outputs a semi-crystalline chain as a series of points, translatable as a DXF file, and usable with a cutting machine such as the vinyl cutter used. First, the shape is sliced using the Matlab Library "Slice_stl_create_path" [217] (Figure 4.7 (a)). Then, the slices are projected onto a plane perpendicular to the slicing direction and juxtaposed next to each other such that the next layer is on the right-hand side of the previous one (Figure 4.7 (b) and (c)). In the case of objects with their right-hand sides smaller than the width of the hinges, the layers are overlapped to accommodate the hinges at the cost of deforming the targeted shape (Figure 4.7 (d)). The intersecting segments are then erased as shown in Figure 4.7 (e) and all the segments are fused into a single object in Figure 4.7 (f). Although the algorithm is able to slice and propose folding patterns, it possesses several limitations. If the hinge width and length are too large for the size of the targeted structure, the algorithm outputs an improper pattern. In addition, the slices are juxtaposed in one direction continuously, which can be detrimental if the semi-crystalline chain is longer than the material to be cut. Finally, at the current date, the algorithm does not slice the lowest layer nor the highest layer of the shape; however the shapes can be adjusted to obtain the missing slices.

4.3 Results

4.3.1 1D to 2D shape transformation

This section presents the results of reversible self-folding and self-unfolding of the pNI-PAM/PVC strings into the following 2D structures: triangular, square, and rectangular coils. The experiments aim to demonstrate the shape reconfiguration of simple structures owing

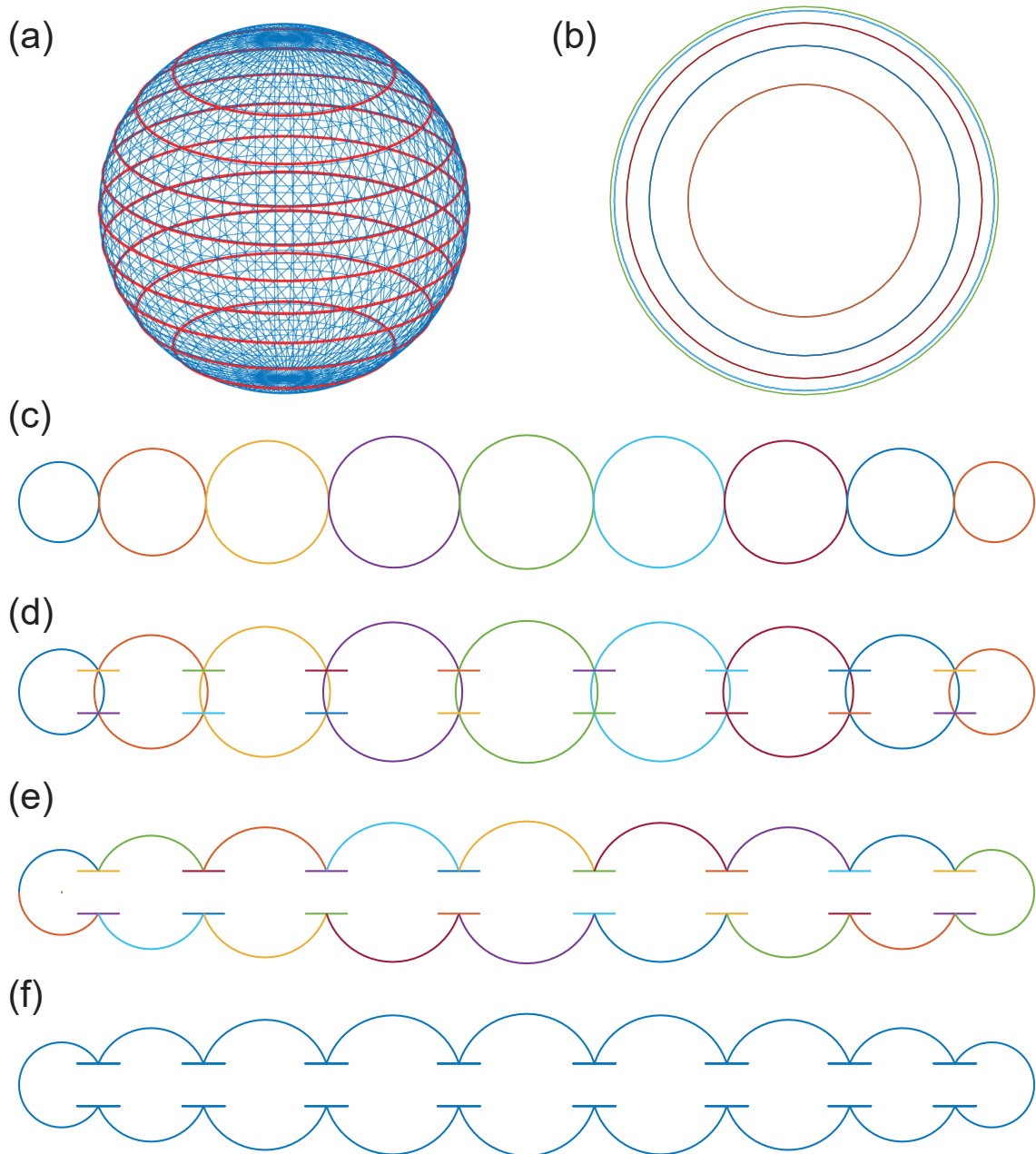


Fig. 4.7 Illustration of the slicing. (a) The STL of a sphere (in blue) is sliced, then all slices are projected onto a perpendicular plane to the slicing axis (b) and the slices are juxtaposed next to each other (c). In the case of round objects, the slices are superimposed to connect the slices with hinges (d). The intersecting edges are erased (e) and all the edges are made into one single object (f).

to the reversibility of folding. The same experimental setup and conditions were used as in Section 4.2.3, and the pNIPAM hinges were designed to be 1 mm thick in the mould and 2 mm wide; however the hinge lengths were adapted to obtain the desired angles.



Fig. 4.8 Shape transformation of the pNIPAM/PVC soft strings: Triangle structure (a) self-unfolding into a string (b) and (c), and self-folding into a triangle (d) and (e). Square structure (f) self-unfolding into a string (g) and (h) and self-folding into a square (i) and (j). Shape transformation of the rectangular coil (k) into a string (l) and (m) and reversed transformation (n) and (o). Self-unfolding of the Acordeon string from (p) to (r) and self-folding from (r) to (t). For all pictures, scale bars are 10 mm

String to triangle

The triangular string possesses 3 hinges 7.7 mm long, which are 20 mm distant from one another, making the string 76.3 mm long. When folded, the structure covers a surface $28 \text{ mm} \times 16.5 \text{ mm}$. The bending angles of the pNIPAM hinges were measured as $101.2^\circ \pm 22.8^\circ$ at

room temperature. In Section 4.2.3, the inward curvature was found to be $11^\circ/\text{mm}$, although the hinges of the triangle were measured to curve at $13.1^\circ/\text{mm}$. This discrepancy originates from the fabrication process. The manual placement of the string on the lyre can lead to misplacement of the string onto the well of the mould. Thus, when curing, the pNIPAM is in contact with the edge of the string. Once swollen, pNIPAM ends up surrounding the edge of the PVC and rubs the surface of the Petri dish. In addition, printing inaccuracies of the wells varied their depth by approximately $300\ \mu\text{m}$ around the targeted depth causing the pNIPAM to bend the PVC further.

The folding speed of a string can be evaluated using baths at different temperatures. As shown in Figure 4.8 (a) to (c), the string reconfigures from a triangular structure into a string in only 19 s, and folds back to its initial state after 23 s (Fig. 4.8 (c) to (e)). The soft characteristics of the string and the robustness of the 2D structure were assessed by manually pulling and poking the string (Supplementary Video).

At 45°C , the string is flexible and behaves like a ribbon, following the motion given by the tweezers. At 20°C , the string resists poking and the hinges maintain their bending. The variation in the flexibility of the string follows the change in the reaction forces into the PVC. Because pNIPAM and PVC are bonded to one another, the stress applied on one is transmitted to the other as a reaction force. If pNIPAM expands, it applies tensile forces onto the PVC below. Therefore, two hinges pull and tense the PVC in between them. The resulting internal stress makes the 2D structure less flexible than its string counterpart. After the removal of the external forces, the pNIPAM folds back the string into a triangle. In contrast, if the pNIPAM shrinks, the reaction forces “push” the PVC, which in turn loosens the string. This mechanism allows the flexibility of the string to be adapted via temperature.

String to square

The square string is composed of 4 hinges that are 5.75 mm long and 15 mm distant from one another, leading to an overall length of 85 mm. After folding the structure, the string covers a $25\ \text{mm} \times 25\ \text{mm}$ surface. The resulting hinges are $876 \pm 29\ \mu\text{m}$ thick, providing a bending at $88.5 \pm 8.7^\circ$. The bending angle deviation causes the string to fold into a parallelogram instead of a square pattern.

When placed in the hot bath, the self-unfolding took 4 s, and when placed in the bath at room temperature, the self-folding spanned 45.5 s which is almost twice as long as the triangle string. The difference of folding time may be explained by the drag force caused by water. In Figure 4.8 (i), the string can be seen with its “arms” spread. This means that when folding, the volume of water displaced is greater, thus making the moment of the drag force

on the hinges greater and the square pattern folding slower. Similar to the triangle, the string is observed to be stiffer at room temperature than at 45° C (Supplementary Video).

String to rectangular coil

The coil string is 122.5 mm long, spreads over a surface 28 mm × 23 mm and is composed of seven hinges spaced to avoid collision when folding. Every hinge is 5.7 mm long and bends at approximately 90° except for the central hinge which is twice as long and bends at 180°. Once the string is placed in the hot bath, the coil self-unfolds in 16 s (Figure 4.8 (l) to (m)) and self-folds back to 25 s when exposed to 45° C (Figure 4.8 (o) and (p)).

In Figure 4.8 (p), the rectangular coil structure misfolds at the outer edge. When the string folds, each hinge has its own rotational axis. The linear speed (V) of the hinge in the Petri dish is a function of the angular velocity around the axis of rotation (ω) and the distance between the hinge and the rotational axis (R) ($V = \omega \times R$). To a certain extent, the pNIPAM hinge can compensate for this drag force; however, if it is too large, the PVC deforms. Despite the correct folding of the pNIPAM hinge, this leads to its misplacement and therefore, mistransformation of the origami (Supplementary Video). In the case of the triangle, the rotation axis is situated at the centre of the structure; however, in the case of the coil, the rotation axis of the hinges is offset from the centre of the structure for each segment. This leads to a greater distance between the hinge and the rotation axis, and hence to a greater drag force. Finally, the success rate of the reconfiguration of the rectangular coil structure was proved to be approximately 10 %. To prove that the drag force was responsible for the misreconfiguration, the last hinge of the structure was cut, which made the string 20 mm shorter and resulted in a successful reconfiguration in 100 % of the cases (Supplementary Video).

String to accordion

An accordion structure is characterised by a succession of inward and outward foldings, leading each panel to overlap the previous one and be overlapped by the following. The Accordion string is an extension of the fabrication process illustrated in Figure 4.3. Here, after synthesis of pNIPAM against the PVC on one side, and the pNIPAM demoulded from the silicone mould (4.3 (e)), the lyre is turned upside down, thus bringing the opposite side of the PVC against the wells in the mould. The synthetisation of pNIPAM is performed once more, making a string with pNIPAM panels on each side. The accordion string is 56 mm long and has four hinges, each 3 mm apart. The hinges are 6 cm long and bend at approximately 90°C. The methodology for self-folding of the Accordion string was different from that for

the other strings. Previously, the string was transferred from a (45°C) water bath and a 23°C bath to highlight the dynamics of pNIPAM alone, but the Accordion string was kept in the same bath that was heated up and left to cool. In the second case, the pNIPAM volume change is highly dependent on the dynamics of its environment, whereas in the other cases, the pNIPAM response is independent of its environment. This highlights that if the pNIPAM can be directly heated instead of its environment, the hydrogel could provide fast responses.

4.3.2 Robotic application

The aim of this section is to showcase the possibility of robotic development using the PVC/pNIPAM bilayers. To enable locomotion, a magnetic actuation was selected for its precision, directionality and its high frequency of actuation. For this purpose, a magnet glued onto the PVC has its orientation controlled by an electromagnetic coil system whose specifications are described in more details by Miyashita et al. [23]. This implementation result in the magnet attempt in following the rotation magnetic field which causes the robots to roll around their environment.

Dual arm robot

The dual-arm robot is a preliminary test of the pNIPAM/PVC capabilities for robotic applications, which was developed in collaboration with Junyi Han and presented at the Exeter International Symposium 2022. The aim of this experiment is to demonstrate the shape transformation of a structurally simple robot and how the various shape transformations tune its navigation and overcome obstacles.

Using the external electromagnetic field, the robot is steered remotely under water over a stairway with 3 mm-thick stairs. The robot experiences a 1.5 mT strong rotating magnetic field at 60 Hz that enables the robot to roll around the workspace.

The robot is made out of one PVC strip with a 3 mm cubic magnet glued at the centre separating the two 12 mm long pNIPAM arms. The arms bend and form a U-shape at 20°C, are flat at 45° and lastly, according to the permanent deformation experiment above, the robot arms can be disabled after exposure at 65°C. The magnet was glued to the PVC using UV glue Loctite AA 3491 and had its magnetisation pointing perpendicular to the arms of the robot as shown in Figure 4.9, so that the robot did not roll on its arms.

While the robot is observed to rotate at 60 rpm, at 20°C, the robot navigate at a speed of 20 mm/s. This difference between the theoretical rolling speed (9.45 mm/s) and the actual speed is due to the resistance of the arms and friction enabled by pNIPAM, which also provides the gripping required to climb the stairs in 33% of the trials. In Figures 4.9 (d) to (f), at

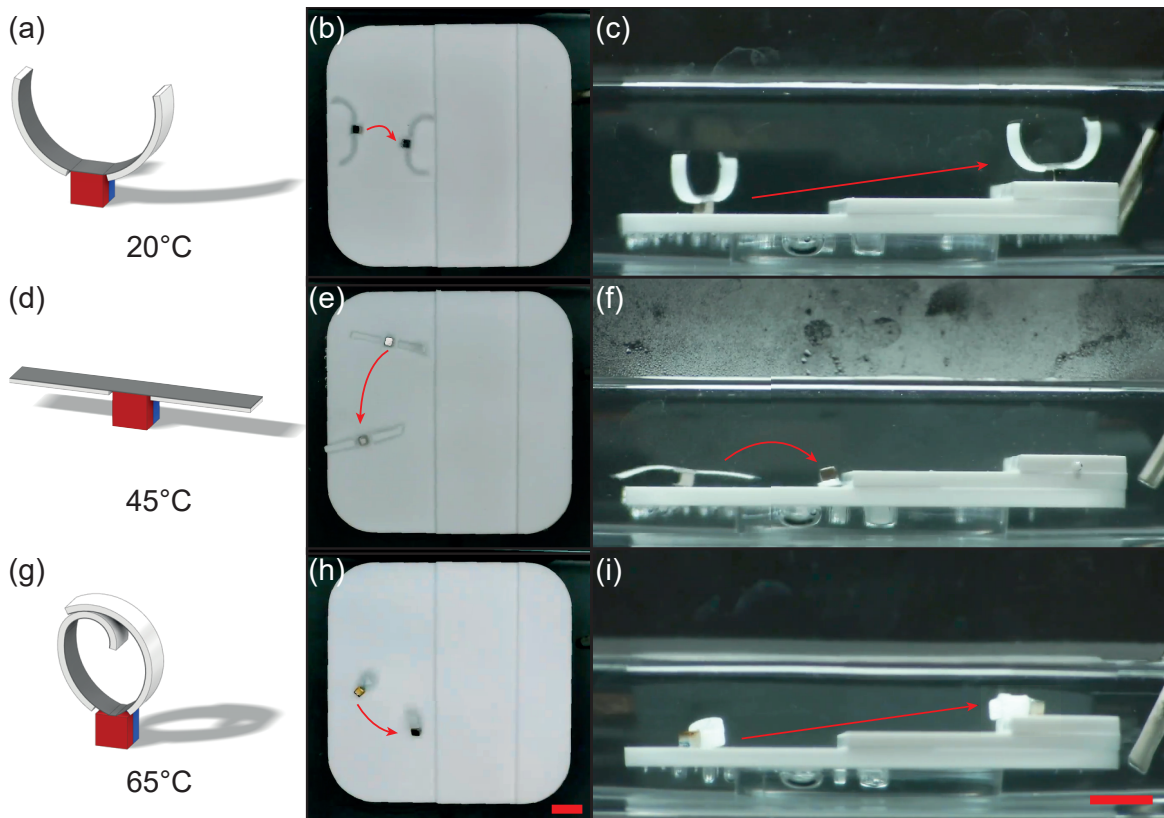


Fig. 4.9 Locomotion of the dual arm robot onto a stairway in: (a)(b)(c) a U-shape at 20°C, (d)(e)(f) a flat state at 45°C, and (g)(h)(i) after shape lock (when exposed to 65°C).

45°C, the robot rolls at a speed of 12 mm/s; however, because of the lack of grip, it is unable to climb stairs, thus highlighting the performance enabled through shape transformation. When exposed to temperatures above 65°C, the robot arms started bending inward until both arms interlaced. The water was then left to cool to room temperature and, as can be seen in Figures 4.10 (a) and (b), the arms remained in the same position. This shape enabled the robot to navigate on its side compared with the original design. Because of the asymmetry of the robot, the success rate of the climbing was highly dependent on the orientation of the robot and its angle of attack. In addition, unlike other shapes, this configuration was performing two trials per rotation, thus increasing the chances of success. However, as the robot rolls around, pNIPAM is subjected to collisions with the environment.

Figure 4.11 (a) shows the impact of delamination on the dual arm. As the navigates around the workspace and climbs stairs repeatedly, pNIPAM enters into contact with the environment. This interaction competes with the bonding strength of pNIPAM on PVC until it delaminates and the robot arms unfold owing to the reduced contact area between PVC and pNIPAM. The failure rate is exacerbated when the arm possesses only a single

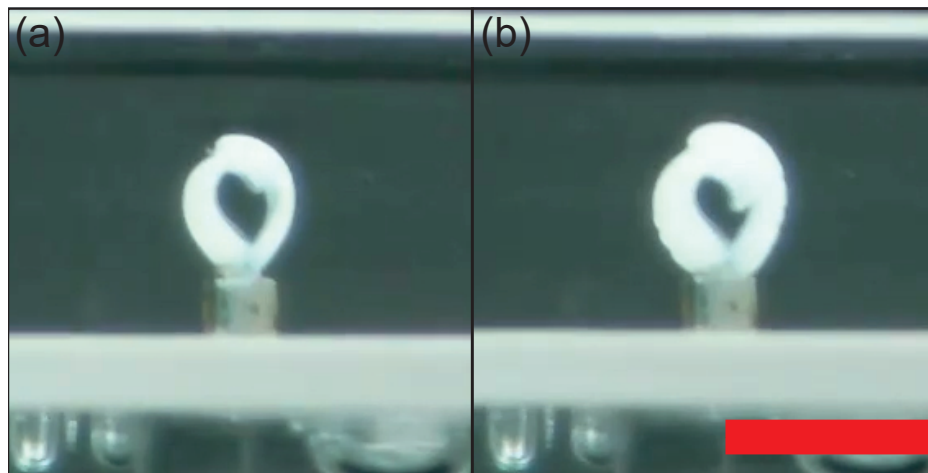


Fig. 4.10 Dual arm robot after permanent deformation at (a) 45°C and (b) 20°C

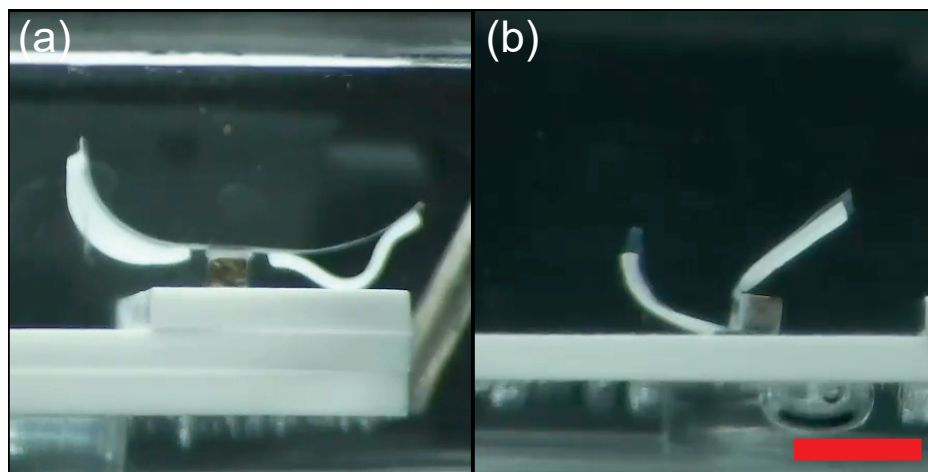


Fig. 4.11 Pictures of (a) the delamination of the pNIPAM after navigation, and (b) misfolding after the PVC activation

pNIPAM actuator because the local delamination will spread to the entire actuator, hence the requirement for multiple pNIPAM actuator to ensure a longer lifespan. Figure 4.11 (b) illustrates PVC misfolding when exposed to 65°C and above. The reasons for such misfolding are not easily visualised. One possible explanation is the initial position of the robot when exposed to 65°C or a misfabrication. When the magnet was glued onto the PVC, the capillary forces extended slightly farther than the edge of the magnet. As the glue was cured, one side of the PVC was glued while the other was free to shrink. Similar to the origami folding mechanisms, this may lead to the PVC folding.

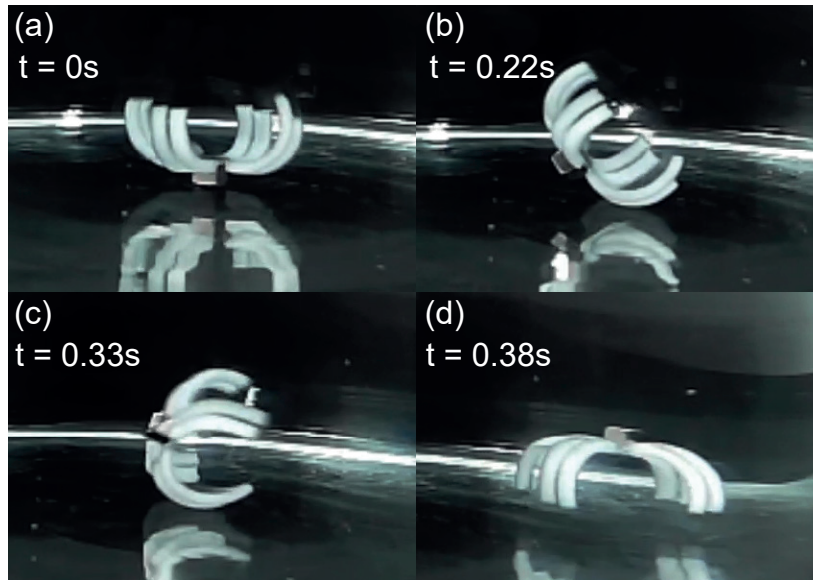


Fig. 4.12 Quadruped robot operating a rolling motion at 20°C when immersed into a rotating magnetic field.

Quadruped robot

The quadruped robot is an extension of the dual arm-robot demonstrating that the robotic functionalities can be translated from 2D to 3D. It was fabricated in a similar way to the dual-arm robot; however, instead of a single actuator on the arms, the robot arms were equipped with two 1 mm-wide actuators to increase the robot lifetime. Locomotion was achieved using two different methods. First, the rolling motion at 20°C allowed the robot to roll at 26.8 mm/s and move forward owing to the gripping of pNIPAM as shown in Figure 4.12. Second, when the robot was positioned with its magnet was under the PVC, a stick-and-slip motion was achieved at 45°C allowing the robot to move at 36.4 mm/s as depicted in Figure 4.13. During the first half of the rotating magnetic field, the magnet tries to follow the magnetic field, but the flat arms of the robot come into contact with the environment and resist the magnetic force (Figure 4.13 (b)). As a result, the arm bends and lifts the robot, which in the second half of the rotation of the magnetic field, glides down as the magnet pulls the resisting arm off the environment surface (Figure 4.13 (c)). However, at 45°C, when the magnet was above the PVC, the magnetic field was insufficient to bend the arms outward and let the robot flip upside-down. This resulted in a quasi-static robot that was still observed to move but only at a speed of 1.45 mm/s, thus almost disabling locomotion.

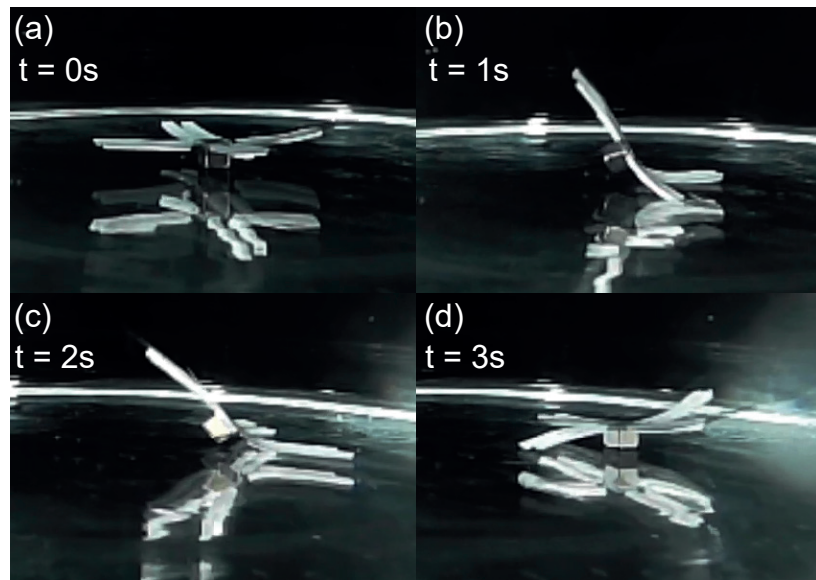


Fig. 4.13 Quadruped robot operating a stick and slip motion at 45°C when immersed into a rotating magnetic field.

4.3.3 String to sliced structures

1D to 2D

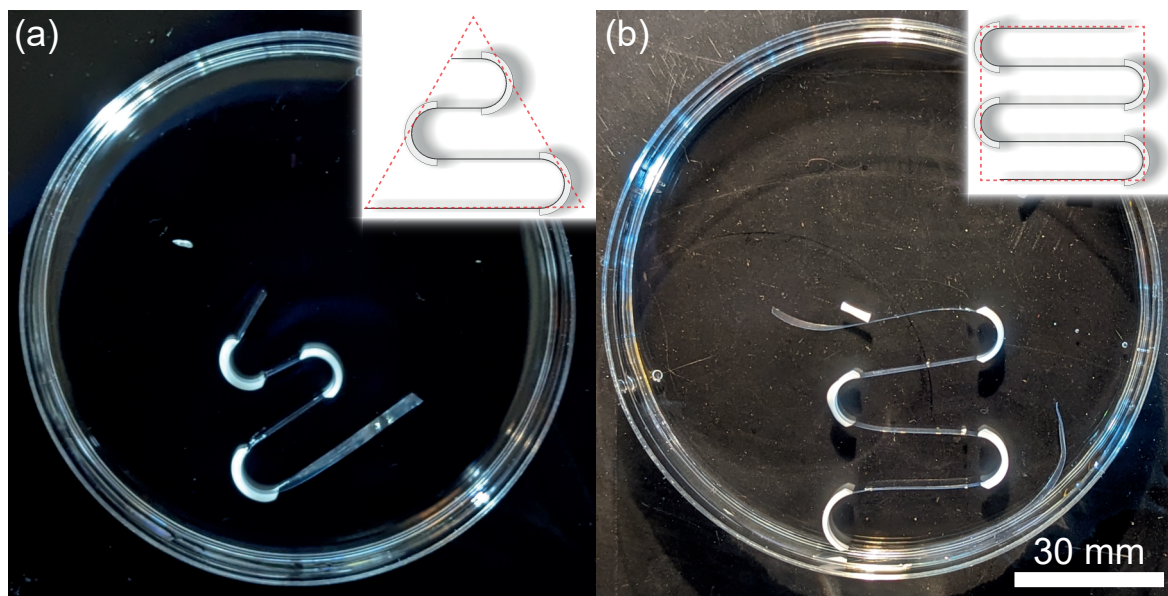


Fig. 4.14 Photographs of self-folded sliced triangle (a) and rectangle (b) with their CAD equivalent.

The slicing principle was first applied to a 2D triangle. A triangle can be described by its base and its height which both describe its surface. The targeted triangle base is 33.2 mm, the height is 28.8 mm and should cover 478 mm². This triangle was sliced in 3 layers approximating the triangle to a trapezoid shape which, in theory, should cover 456 mm² and have a height of 22.9 mm. Since on the lyre, the PVC ribbon is longer than required to form the triangle as can be seen on Figure 4.3 (e), the edges of the ribbon are manually cut to remove the unnecessary length of PVC. In this way, the resulting self-folded triangle covered a surface of 489.7 mm² with a height of 29.5 mm and a base of 34.5 mm.

The slicing principle was first applied to a 2D triangle. The geometry of a triangle can be described by its base and height, both of which describe its surface. The targeted triangular base is 33.2 mm, the height is 28.8 mm and it should cover 478 mm². This triangle was sliced into 3 layers approximating the triangle to a trapezoidal shape with a height of 22.9 mm, and a theoretical surface of 456 mm². As the ribbon length is longer than required to fit on the lyre (Figure 4.3 (e)), the edges of the ribbon were manually cut to remove the unnecessary PVC length. Thus, the resulting self-folded triangle covers a surface of 489.7 mm² with a height of 29.5 mm and a base of 34.5 mm.

In the case of the rectangle, the targeted shape is 28.8 mm wide and 32 mm long, covering a surface of 921 mm². The resulting rectangular strips failed to fold owing to the partial delamination of the third hinge from the bottom in Figure 4.14 (b) and the inconsistency of the folding owing to fabrication inaccuracies. However, the rectangular ribbon could be pushed into a corner of the Petri dish to form the correct shape covering a surface of 1316 mm². If the pNIPAM block were removed, the rectangle would extent on 2738 mm². From these dimensions, it can be concluded that the accuracy of the shape is a function of the cutting accuracy of the PVC on the lyre and the summed accuracies of the self-folded hinges.

2D to 3D

Slicing was extended to 3D structures to demonstrate the algorithm output in practice . A parallelepiped of dimensions $18 \times 16 \times 15$ was sliced into 3 layers. Figure 4.15 (a) depicts the resulting sketch whose sides in length were extended to fit on the lyre as represented passed the dashed lines. The results of self-folding are shown in Figure 4.15 (b) at 20°C and Figure 4.15 (c) at 45°C. Although the sliced cube is seen to become a flat chain at 45°C, its folded state does not resemble the targeted parallelepiped. Apart from the folding inaccuracies due to fabrication, here, the slicing algorithm cuts the size of the arc length exactly as inputted. This implies that the PVC of the sliced cube must be positioned perfectly onto the mould or that the pNIPAM wells will extend onto one of the panels. If it is

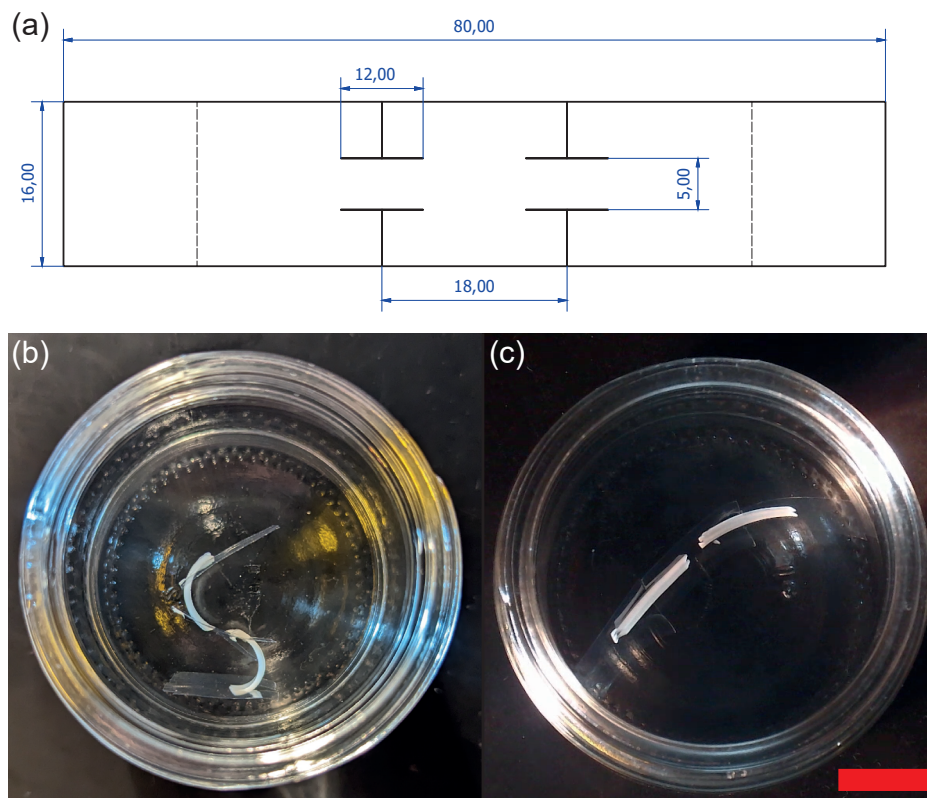


Fig. 4.15 Photographs of a self-folded sliced cube. Sketch of the PVC cutting pattern (a). Self-folded cube at 20°C (b) and flat at 45°C. Scale bar is 10 mm.

misaligned, the bilayer cross section suddenly increases and the stiffness of the bilayer with it. Therefore, the curvature will be reduced, leading to inaccurate folding. This limitation of the algorithm can be compensated by inputting a longer hinge dimension or by manually extending the length of the hinge with a cutting knife.

4.4 Discussion

4.4.1 Comparison with state-of-the-art

Reference	Type of string	Module Size	Module number	Torque	Medium	Mass in medium	Power Source	Folding in	Actuation Method	Assembly time	Remote folding	Untethered	Reversibility	Addressability	Reprogrammability	Sensing	Assembly and Deformation	Application field(s)
Yim et al. [167]	Chain	3x3x3 mm	999*	$3.2e^{-2}$ Nm [†]	Guide exit	36 g	Magnets	3D	Magnetic pull/push	108 s	Yes	/	Yes	/	/	/	Self-assembly and self-folding	Medical implants
Fass et al. [189]	Chain	20x20x10 mm	6	$2.5e^{-1}$ Nm [†]	Planar surface	>>0 [†]	Magnets	2D	Magnetic pull/push	0.2 s	Yes	Yes	Yes	/	/	/	Self-assembly and self-folding	Minimally invasive surgery
Griffith et al. [172]	Chain	25x25x3 mm	20*	$2.1e^{-1}$ Nm [†]	Air cushion	~0 [†]	Magnets + Air	2D	Magnetic pull/push onto air cushion	30 s	Yes	/	Yes	/	/	/	Self-assembly and self-folding	Programmable matter
Han et al. [43]	Chain	14Øx9 mm	20*	$2.9e^{-2}$ Nm [†]	Agitated hot water	~0 [†]	Magnets + Air + Heat	2D	Magnetic pull/push into bubbling water	10 s	Yes and No	Yes	Yes	/	/	Yes	Self-assembly and stretching	Programmable matter
Gu et al. [185]	Chain	15x5x5 mm	9*	$3.4e^{-3}$ Nm	Guide exit	>0 [†]	Magnets	3D	Magnetic pull/push	13 s	Yes	/	Yes	/	/	/	Self-assembly and self-folding	Minimally invasive surgery
Li et al. [7]	Ribbon	4x5x0.5 mm	5	$1.42e^{-2}$ Nm [†]	Planar Surface	320 mg [†]	Magnets	2D	Magnetic push/pull	/	Yes	Yes	Yes	/	/	/	Self-folding	Robotics and programmable matter
Ijäs et al. [194]	Chain	3x2x2 nm	8064	/	Solution	~0	Atoms	3D	Atomic forces	/	Yes	Yes	/	/	/	Yes	Self-assembly and self-folding	Drug delivery
Douglas et al. [195]	Chain	3x2x2 nm	7308	/	Solution	~0	Atoms	3D	Atomic forces	12h	Yes	Yes	/	/	/	Yes	Self-assembly and self-folding	Drug delivery
Bai et al. [191]	Chain	3x2x2 nm	7249	/	Solution	~0	Atoms	3D	Atomic forces	12h	Yes	Yes	/	/	/	/	Self-assembly	Programmable matter
Cheung et al. [173]	Chain	77x38x38 mm	7*	1.5 Nm	Planar surface	>371 g	Electricity	3D	Servo Motors and Pivot	3 s	Yes	/	Yes	Yes	Yes	/	Torsion	Robotics and programmable matter
Knaian et al. [176]	Chain	1x1x1.5 cm	4*	$4.5e^{-4}$ Nm	Planar surface	14 g	Electricity	3D	Servo Motors and Pivot	5 s	Yes	/	Yes	Yes	Yes	/	Torsion	Robotics and programmable matter
Yim et al. [166]	Chain	1.3cm x cm x 0.1 mm	>30*	/	Planar surface	>0 [†]	Electricity	3D	Servo Motors and Pivot	15 s	Yes	/	/	/	/	/	Pulling and Folding	Robotics and programmable matter
Sung et al. [175]	Chain	>cm x cm x <0.5 mm	57	$4e^{-2}$ Nm	Hot water	>0 [†]	Heat and Magnetic field	3D	Smart Materials Magnetic steering	32 s	/	Yes	/	/	/	Yes	Self-folding	Robotics
Tibbits et al. [168]	Chain	>cm x 6.4Ø mm	63*	/	Hot water	>0 [†]	Heat	3D	Smart Materials	>40 mn	/	Yes	Yes	/	/	Yes	Self-bending	Programmable matter
Uchida et al. [28]	String	>cm x Ø1 mm	/	/	Hot water	~0 [†]	Heat	3D	Smart Materials	/	/	Yes	Yes	/	/	Yes	Self-bending	Medical
Wang et al. [128]	String	>cm x >Ø10 mm	/	/	Hot air or water	>0 [†]	Heat	3D	Smart Materials	>60s	/	Yes	/	Yes	/	Yes	Self-bending and Torsion	Programmable matter
Chapter 4	Ribbon	122x3x1 mm	7	/	Hot water	~0	Heat and Magnetic field	2D	Smart Materials magnetic steering	41 s	/	Yes	Yes and No	Yes	/	Yes	Self-bending	Robotics and programmable matter

Fig. 4.16 Comparison of the literature on self-folding 1D structures and the characteristics of the ribbon developed in this chapter. The asterisc in the "max number" column depicts the 1D structures' number of module demonstrated. The cross in the torque and mass in medium correspond to calculated values inferred from the information within the respective studies.

As mentioned in Chapter 2, the current challenges of self-folding 1D structures include tether-less systems, reversability of the actuation, addressability of the hinges, and sensing (Figure 5.17). Along with these are research questions regarding the scaling and self-folding torque, the communication via structural state and turn off of reversible smart material.

Here, the 1D structure consists of pNIPAM hydrogel grafted to PVC shape memory polymer. Although bending bilayer are not a novel techniques (as explained in section 2), it is the fusion between smart materials with reversible and permanent deformation that enable a shape locking mechanism to appear and with it, a form of communication based on structural change. Consequently, through the shape locking, the 1D structure can indicate its encounter with an object or environment at 65°C or higher, achieving a form of communication. Overall, it is this shape locking mechanism and the performance that it entails for robotics that makes the novelty of this work.

In this chapter, the design of the hinges and rigid panels is kept to a minimum; instead, the work undertaken here focuses on material aspects. To approach the challenge of scale and torque, gravity forces are reduced as most as possible by using a hydrogel which is close to neutrally buoyant as it contains mostly water and a light shape memory polymer. As a result, the mass of the structures are optimised to reduce the folding torque as much as possible and allow for up scaling potential. On this aspect, this method is similar to approaches where robots are made entirely of hydrogel [50] [51] [28].

Finally, the bending sequence of the structure shown in this chapter can be controlled by progressively dipping the 1D structure in its medium. This is a similar approach shown by the folding chains developed by Sung et al. [179] or the magnetic chains assembling out of catheters [193] [189].

4.4.2 Slicing algorithm

Here, the slicing algorithm adapted from a previous study accommodates bending instead of folding. The algorithm was applied to a sphere and a cube, which after slicing, were outputted as an Excel file readable by Autodesk-Inventor. Currently, the algorithm was tested on a few selected shapes and further tests should be conducted to assess its robustness. In addition, a few issues appear in the algorithm, as the first and last layers are not sliced, and the position in space of the shape in the CAD file can generate errors. Although these defects can be problematic, especially for shapes with small number of slices, future work could address the slicing issue or increase the curvature of the pNIPAM/PVC bilayer to increase the number of slices possible in small structures and the resolution of the outputted shape. In comparison to the literature, other approaches mimicking semi-crystalline zig/zagging structures accomplish more hinge per unit volume. This is due to their use of folding instead

of bending which allows more compact structures to be produced as seen in the study of Yim et al. [167] and Sung et al. [179]. However, these two approaches utilise only permanent folding and lack the reversibility of actuation required to perform repetitive robotic tasks.

4.4.3 Fabrication process

In this chapter, a novel fabrication process was developed allowing pNIPAM to be bonded to a PVC shrink film. Despite its limited number of steps, the process suffers from two aspects: the precision of the alignment between the PVC and the mould, and the bonding force between the PVC and the pNIPAM. Although the alignment was controlled by fitting the lyre around the mould, a small tolerance must be implemented to allow the lyre to slide around the mould holder. Coupled with the manual positioning of the PVC strip onto the lyre, some misalignment may appear which leads to the accidental insertion of air bubbles between the PVC and the pNIPAM, or the introduction of an angle between the length axis of the PVC and the pNIPAM, which reduces the bending of the PVC. The bonding force pNIPAM/PVC is mainly a function of the PVC surface activation, as well as the care provided during the fabrication process to the pNIPAM. Usually, after plasma treatment, the contact angle of the droplet on the treated surface is very low, meaning that the surface becomes highly hydrophilic [218]. Here, after PVC surface treatment for 6 mn, the droplet onto the PVC is neither pearling nor spreading, thus the PVC is between hydrophobic and hydrophilic. In addition to the plasma itself, this low surface activation may be due to PVC and its protective coating. For better bonding, the use of a different PVC would be highly recommended. Finally, as proposed in equation 4.3, the bending angle is a function of the hinge length, height and stiffness. This implies that the pNIPAM should be carefully prepared to obtain the right stiffness, that the moulds should be fabricated with good repeatability and that any delamination drastically impacts the bending angle. In summary, even though the process appears simple, the precision required for each step is crucial making it difficult to obtain repeatable results. Although, the bonding success rate reaches 95 % for a hinge, the success rate of a string with 10 hinges drops to 60 %. Consequently, fabricating complex structures with numerous hinges would be difficult.

4.4.4 Reversible and permanent deformation

The coupling of reversible and permanent deformations offers new features for applications such as robotics and, potentially, fluidics. The volume change of the pNIPAM is fully reversible, thus allowing a robotic gripper to try and retry until success when grasping. The permanent deformation offers a shape lock, enabling a robotic gripper to hold more firmly

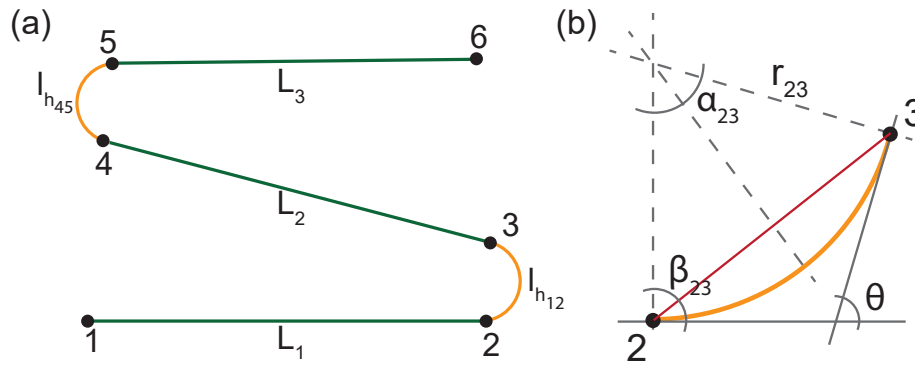


Fig. 4.17 Photographs of a self-folded sliced cube. Sketch of the PVC cutting pattern (a). Self-folded cube at 20°C (b) and flat at 45°C.

than with the pNIPAM and maintain the grasp without constant temperature control of the environment. In fluidics, this technology could be used as a valve capable of detecting temperature variations with open or closed states depending on the pNIPAM and PVC activation. However in this respect, experiments are currently limited and further work is required. In addition, the preliminary results indicate that the permanent deformation is heavily influenced by the initial position of the robot; in Figure 4.6 and Figure 4.10, the arms bend inward, but bend outward in Figure 4.18 (b).

4.4.5 Bending angle and inaccuracy propagation

The folding angle, as for any self-folding strings, is crucial as the folding inaccuracies propagate along the string. To calculate the inaccuracies, the aim is to link the position of the points of the string to the bending angle θ . Figure 4.17 (a) depicts this propagation for bending semi-crystalline structures. Using the following equations and the characterisation of the bending angle, one can assess the speed and position of each point during folding and estimate how inaccuracy affects the rest of the semi-crystalline structure.

$$\text{If } i \text{ is even: } \begin{cases} X_i = \sum_{i=1}^{n-1} X_i + L p_{i-1,i} \cos(\theta_{i-2,i-1}) \\ Y_i = \sum_{i=1}^{n-1} Y_i + L p_{i-1,i} \sin(\theta_{i-2,i-1}) \end{cases} \quad (4.4)$$

$$\text{If } i \text{ is odd: } \begin{cases} X_i = \sum_{j=1}^{n-1} X_j + \frac{Lh_{i-1,i}}{\theta_{i-1,i}} \sin(\theta_{i-1,i})^2 \\ Y_i = \sum_{j=1}^{n-1} Y_j + \frac{2Lh_{i-1,i}}{\theta_{i-1,i}} \sin(\theta_{i-1,i})^2 \end{cases} \quad (4.5)$$

The points in Figure 4.17 (a) are labelled with odd numbers at the start of a panel and with even numbers at the start of a hinge segment. There are a total of n points starting from index $i = 1$. The length of the hinge Lh is as designed, remains constant, and is related to the radius of the bend noted r and the angle between the two edges of the bend α via the equation $Lh = r\alpha$. The height between two slices is known as h and the length of a slice is Lp . The direct line to each edge of the bend has an angle β between the previous and the subsequent flat segment.

4.5 Conclusion

This chapter presents a novel fabrication process that enables the bonding of the pNIPAM hydrogel, which changes its volume reversibly between 20°C and 45°C, onto a PVC film shrinking permanently above 65°C. By using this combination of heat-responsive materials, the structure could reversibly self-fold and lock its shape through permanent self-folding. By having multiple hinges at different locations of a PVC string and encoding the bending angles by changing the length of the hinges, ribbons succeeded in self-folding into 2D structures such as a triangle, a square, a coil, an accordion and 3D structures. The capability of the reversible/permanent deformation was applied to a 2D dual-arm robot whose locomotion speed was tuned and the ability to climb stairs was disabled by changing the shape of the robot, and to a 3D quadruped robot whose locomotion types and speed were tuned. Owing to the reversibility of the folding processes enabled by pNIPAM, the structures could be unfolded with an increase in temperature. The reconfiguration was successful in 100 % of the cases for the triangle, the square and the coil with 6 hinges, indicating that the string could transform back and forth into a 2D pattern. Regarding semi-crystalline structures, the process was applied to a 2D triangle, a rectangle and a cube that was sliced using the algorithm developed to accommodate bending in sliced structures.

Here, the two following research questions were approached:

1. How the limited energy that smart-materials provide be circumvented for strings to self-fold into large 3D structures?

2. How to turn off the reversibility of smart-materials to maintain a shape or functionality without having to maintain the power input?

The actuator developed here, made of PVC and pNIPAM hydrogel, pertains to the challenge of scale and torque, and reversibility. In the present case, the pNIPAM is mainly composed of water rendering this part of the structure close to neutrally buoyant while the PVC film is only $20\mu\text{m}$ thick making the structural layer lightweight. However, non-zero weight under water suggests that the structure sinks across all temperature. On the opposite of previous chapter where buoyancy was controlled by adjusting the design, here the object is in free fall, another state with diminished gravity, where the required folding torque is reduced. Not demonstrated here, this approach benefits mostly from the use of vertical reactor to the cost of temperature change rate (due to the slower cooling of a vertical reactor in comparison with a flat reactor). In addition to the lightweight aspect, the softness of the PVC allows the structural layer to deform when facing high drag force causing the surface area of the structural layer onto which the drag force applies to reduce, hence the reduced counter torque onto the hinges of the structure.

The other main aspect of this chapter is the mixing of shape memory polymer with permanent deformation and hydrogel with reversible deformation. This allows the structure to shape change at will in the range of 20°C and 45°C and permanently once exposed to temperature exceeding 65°C . Consequently, structures based on such actuator can benefit from numerous trials to self-fold correctly, until the structure is locked in its folded shape permanently. In the case of in-vivo applications, biological tissues are delicate as their temperature cannot exceed 40°C without damage. Compared to other approaches, the locking allows to turn off the input triggering the shape change, which in turns, reduces the energy consumption and possibly the exposure of tissues to the input, thus increasing the biocompatibility of the approach. Overall, such actuator could contribute to robotic functions, especially in-vivo (upon adjustment of the activation temperatures), where many self-folding trials may be required due to ever changing environment.

In contrast to other approaches, the semi-crystalline structures, as well as the actuator developed here, suffer from low stiffness. Although for robotic applications, the softness of the actuator can be used for delicate applications in-vivo; as a structure, the actuator will let the structure collapse onto itself because of its own weight. There is therefore a trade-off between the number of layers to obtain the structure with sufficient detail and its weight/size compared to the stiffness of the hinges. To this date, numerous research articles are published on bistable actuators which can sustain higher loads even from low stiffness actuators. The implementation of such feature within PVC/pNIPAM semi-crystalline structure

could drastically increase the number of layers within the structure without technological changes.

4.6 Annexes of Chapter 2

4.6.1 Initial position and PVC activation on dual arm robot

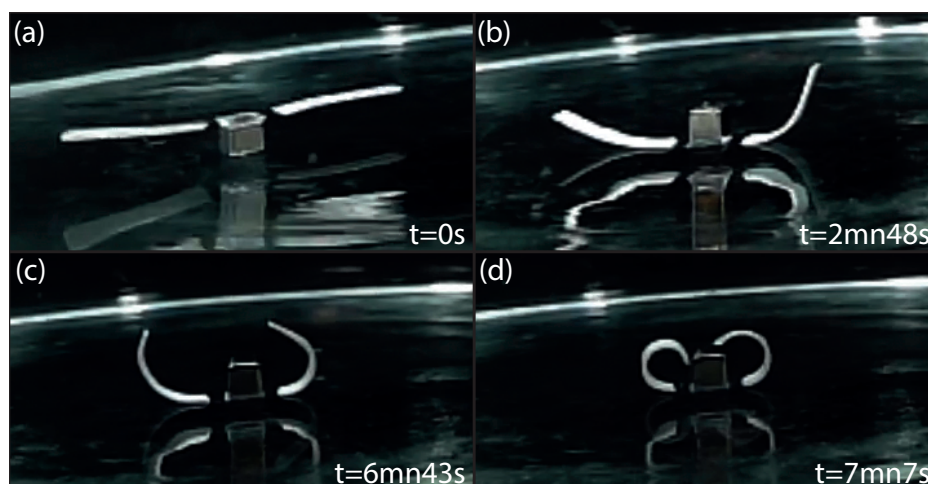


Fig. 4.18 PVC activation on bilayer robot when immersed in water heated beyond 65°C . The robot starts first in an activated pNIPAM/Inactivated PVC state (a) to a still activated PNIPAM and gradually activated PVC (b) and (c) and ends with the bilayer completely curled, PVC fully activated (d).

Here, in Figure 4.18, the deformation of the PVC is shown on the dual arm robot. Over the course of 7 mn, the robot transformed from a flat state to a curled shape. In previous experiments, the bending of the bilayer through PVC activation occurred in the same direction as the bending enabled by the pNIPAM. However, here, the dual arm robot had its arms directly in contact with the surface of the environment, which seems to have forced the PVC to bend in the other direction.

Chapter 5

Addressible and Remotely Self-Folding Strings via Magnetic Induction for Amorphous Structures

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5.1 Summary of Chapter

In this chapter, I present my approach to mimicking amorphous structures from a ribbon.

In an amorphous structure, the chains within a material lack order, are arranged randomly and, similar to cooked spaghetti, agglomerate and intertwine into knots. A knot is a 3D structure obtained from a 1D flexible string with infinite degree-of-freedom and deforming in all directions of the Cartesian space, inter-twinning to form a shape. In polymers, the spontaneous formation of various knots, including the overhand knot and the trefoil knot, was observed in the simulation of self-folding chains [176], while at macro-scale, the formation of knots is driven by mechanical agitation [219]. This is achieved by inter-chain bonding [220] and friction [221], but the resulting agglomerations are unstable [176] as the holding forces of the knot can be weakened by providing more energy to the chains, promoting further assembly or disassembly. Regardless of the scale, it is knots that secure the building of larger structures as R. Graumont and J. Hensel explained in their book: "It (the overhand knot) has one chief function: to serve as base or part of other knots" [222].

However, to reproduce the same structures, the intertwining of the strings should be deterministic, and therefore the self-folding of such strings should be precisely designed. Hence, the fabrication of artificial amorphous structures requires sequential folding to avoid

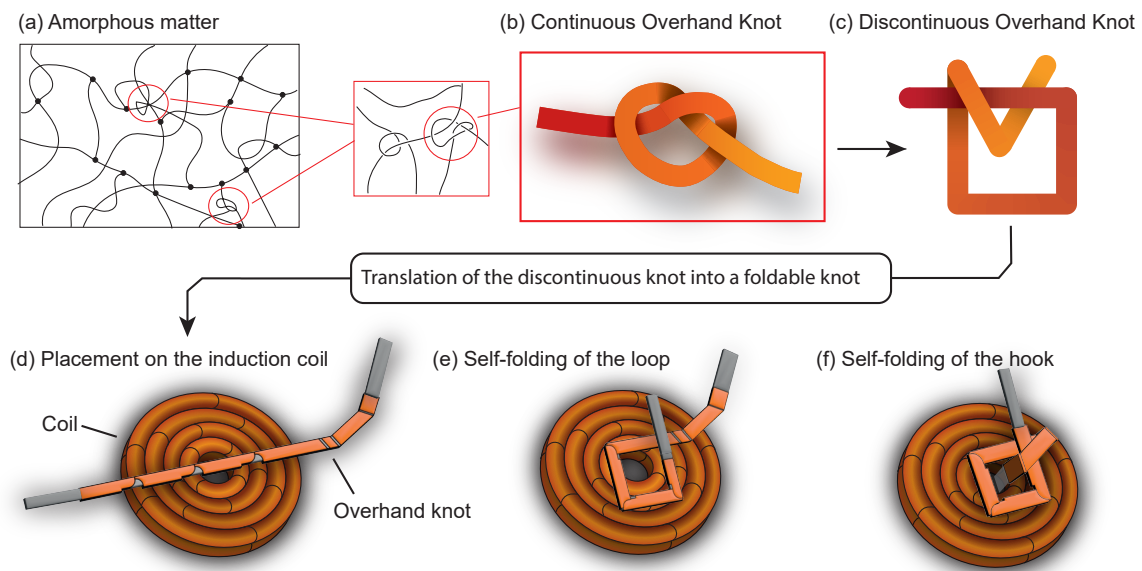


Fig. 5.1 Overview of Chapter 3. Amorphous structures are characterised by their random arrangement within their structure, doing so knots may appear (a). Knots are obtained from the continuous deformation of a string (b), but can be translated into a discontinuous deformation (c). After translation of the knot, a self-folding knot can be produced (d), and self-folded sequentially by folding the loop (e), then the helix (f) via magnetic induction.

self-collisions and fold into the desired knot [178]. This led to two questions: How can a knot be translated from a probabilistic deformation to deterministic deformation? What are the geometric design rules for self-folding strings into knots?

Here, the aim of this chapter is to demonstrate the self-folding of an overhand knot, the basis of many amorphous structures. For this purpose, a methodology is proposed to design an overhand knot and introduce sequential self-folding using magnetic induction along with its theoretical modelling.

Magnetic induction is a phenomenon consisting of an electrically conductive and magnetically susceptible material receiving an alternative magnetic field, which generates electric currents within the material and produces heat due to the Joule effect. The induction coil generated a 140 kHz alternative magnetic field with an amplitude of 3.5 mT and was assembled by Kaan Esendag. Due to the span of mobility in polymer chains, the formation of knots is probabilistic. To form artificial amorphous structure, the process should be deterministic, thus requiring a reduction in the degree-of-freedom in the string. For this purpose, instead of a string, I used an articulated ribbon with local and unidirectional hinges separated by rigid sections. However, methods to tie knots rely on string flexibility, which is not applicable to rigid ribbons with fixed articulations, therefore implying geometric constraints. The first step in forming knots is to create a loop for the main body, and helices crossing the loop to stabilise the knot. [223] Therefore, the methodology requires the loop to be any polygonal shape whose corner is a hinge folding in-plane. By disconnecting one hinge of the loop, the helix can be added. The hinge of the helix should bring the section parallel to the plane of the loop, position the end of the helix above the loop surface and fold through without colliding with the edges of the loop.

This brings to the design of the hinges. Admitting the loop is positioned in the xy-plane, the hinges of the loop should remain in the xy-plane. As for the helix, the hinges must fold out-of-plane (xz-plane or any rotation around the z-axis), thus requiring straight hinges or angled-hinge folding. For this purpose, I propose the design of an in-plane hinge to enable the folding of the loop. An important characteristic of magnetic induction is the angle between the magnetic field and the surface of the object. If parallel, the coupling of the coil to the magnetic receiver is maximal but reduces to zero as the angle approaches 90° meaning that the heat generation vary with the angle. In the case of self-folding enabled by a thermo-responsive material, this implies that the magnetic receiver perceives a smaller amount of energy from the induction coil thus slowing down or stopping the folding at an angle called stalling angle. This participates in creating a folding sequence as the in-plane hinges remain at maximum coupling regardless of their folding angle. The heating rate of a receiver is a function of its placement on a magnetic coil as well as its geometry. Here, I

propose a method based on a theoretical model to sequentially fold structures by tuning the receiver design. This method was demonstrated by sequentially self-folding the flaps of a box, with the hollowest receiver folding first and the plain copper receiver folding last, and by self-folding the leaves of a bio-inspired mimosa pudica origami. Overall, the placement of the ribbon on the induction coil, the design of the hinges and the design of the receivers were all implemented onto a ribbon to self-fold into an overhand knot.

In summary, this chapter contributes to the self-folding by:

- Achieving sequential folding using induction,
- Proposing a methodology to translate a knot into a self-foldable string,
- Proposing a model to predict the heat increase rates using magnetic induction,
- Sequentially self-folding structures such as a box, a biomimetic mimosa pudica leaf and an overhand knot.

5.2 Methods

5.2.1 Modelling

Induction heating is a method to heat remotely magnetic materials using an alternative magnetic field. When a metal receiver is immersed in an alternative magnetic field, electric currents form and flow in the metal which, in turn, heats up gradually the metal receiver due to Joule effect. However, with alternative current, the moving electrons are “centrifugated” at the edge of the conductor, thus heating the periphery of the receiver first. This effect is called the skin effect and depends on the frequency of the magnetic field.

The aim of the model is to predict the folding angle and the position of the structure depending on the magnetic field received and the initial position of the receivers.

The model is split in three parts as shown in Figure 5.2. In light blue, the first part predicts the power induced in the receiver P_{ind} as a function of the amplitude of the magnetic field B_{max} , its pulse ω and the initial position Pos of the receiver as coordinates x, y, z and folding angle θ . In red, the second part of the model computes the temperature of the receivers T_{Cu}° and the temperature of the PVC T_{PVC} depending on the heat loss within the system. In green, the third part of the model estimates the folding angle of the hinge depending on the heat the PVC receives Q_{PVC} which provide information on the position of the structure and the new surface of induction.

The aim of the first part of the model is to calculate the power induced in the receiver. The calculation of the induced power requires an estimation of the magnetic flux Φ :

$$\Phi = \int B_{rec}(d) \cdot dS_{ind} \quad (5.1)$$

where the magnetic field received B_{rec} depends on the distance d from the centre of the induction coil to the receiver and S_{ind} is the surface of induction, which can be seen as the projection of the receiver onto the horizontal plane shown as shown in Figure 5.2 (b) and (c). To simplify the previous equation, the magnetic field is assumed to be uniform along the receiver for a given position during folding. Thus, d corresponds to the distance separating the centre of mass of the receiver from the centre of the coil and the equation 5.1 can be rewritten as:

$$\Phi = B_{rec}(d)S_{ind}(\theta)\cos(\theta) \quad (5.2)$$

Although the assumption of a uniform magnetic field along the receiver helps to simplify the model, it limits the accuracy of the model to receivers with short lengths. Admitting an infinitely long receiver be on the coil, only the part of the receiver onto the coil should heat up, but the model predicts that the whole receiver will heat up.

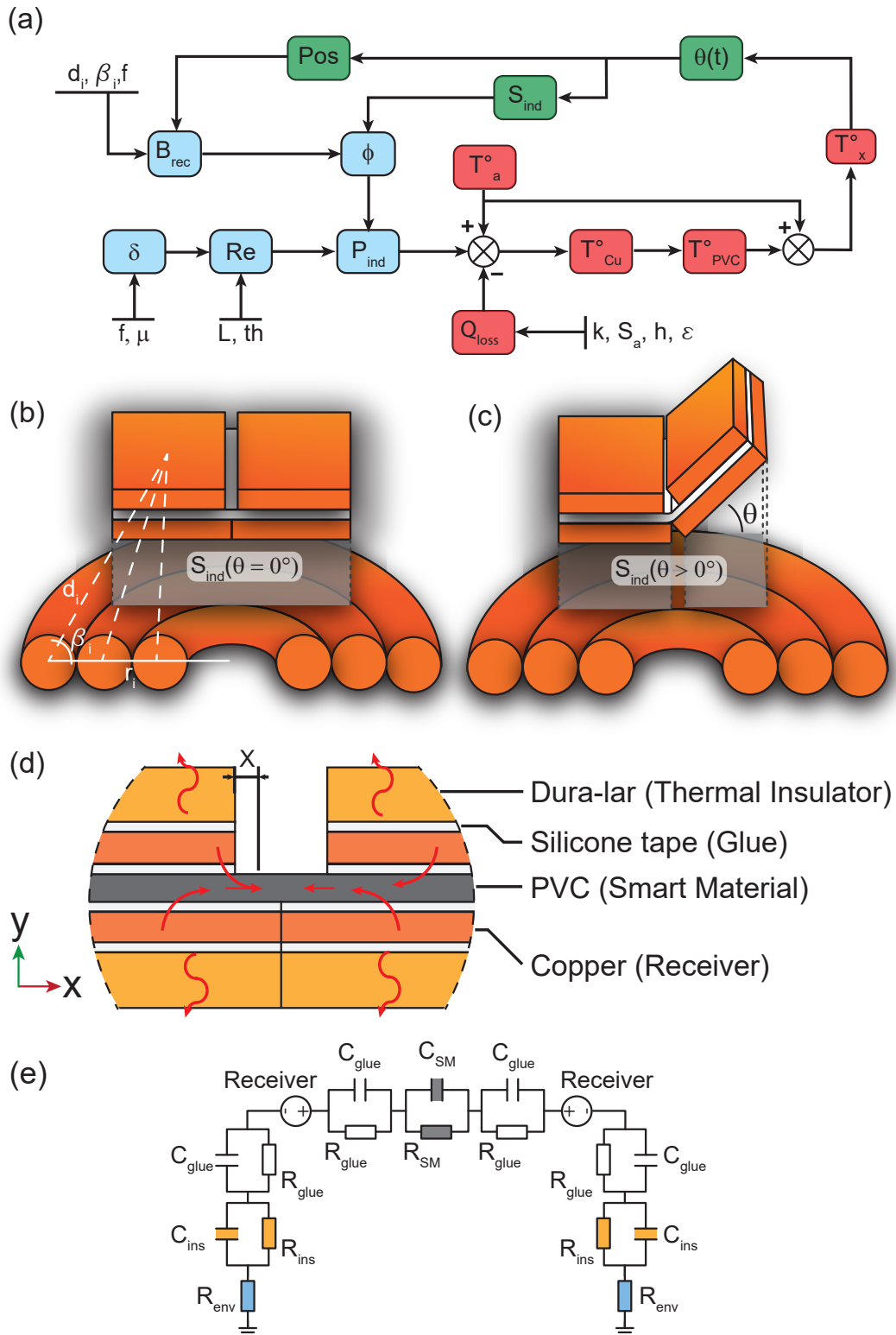


Fig. 5.2 Illustration of: the overview of the model (a), the change of surface of induction as the hinge folds (b) and (c), the cutview of the multi-layer self-folding structure (d) and the electric equivalent of the heat exchange.

The next step is to estimate the power induced in the receiver which is given as:

$$P_{ind} = \frac{-\frac{d}{dt}\Phi^2}{R_e} \quad (5.3)$$

where R_e is the electric resistance of the receiver which is given as:

$$R_e = \rho_e \frac{P_e}{S_e} \quad (5.4)$$

where ρ_e is the resistivity of the receiver, S_e is the section of the receiver that conducts the current and P_e is the path length followed by the current. P_e is considered the perimeter of the receiver. At this stage in the model, P_{ind} is a function of the folding angle θ and the distance coil/receiver d , both of which depend on time t .

The second part consists solely of thermal aspect and aims to estimate the temperature of the receiver T_{Cu} , the temperature of the smart material T_{PVC} and the energy transferred to the smart-material Q_{PVC} .

As shown in Figure 5.2 (d), the smart material is sandwiched between the receiver and the thermal insulation, all glued together with double-sided silicone tape. From the receiver, the heat can escape towards the smart material, the environment, and directly from the side of the receiver. The thermal problem can be represented as an electrical equivalent (Figure 5.2 (e)). Several methods can be developed to simplify this problem. First, the problem is symmetrical; therefore, only half of the problem can be considered, thus neglecting the difference in power induced between the top and bottom receivers. Second, the thermal conduction is unidirectional, meaning that the convection between the environment and the bend of the blanket of the receiver is neglected. From the calculation of the induced power, the heat generated in the receiver can be quantified in the structure by using the following equation:

$$m_{tot} C_{peq} \frac{dT_{Cu}}{dt} = P_{ind} - \dot{Q}_{loss} \quad (5.5)$$

The heat generated in the receiver is calculated by estimating the total mass of the layers of the system and the equivalent specific heat capacity C_{peq} . $\dot{Q}_{loss}$ symbolises the heat losses including the conduction through the many materials of the structure and the convection with the environment, hence its dependency on k the thermal conductivity of each material, S the surface of the receiver considered, h the thickness of each material and h_{conv} the heat transfer coefficient.

At this stage, two differential equations describe the system: one from the point of view of the receiver and one from the point of view of the PVC, as shown below:

$$\left\{ \begin{array}{l} m_{tot} C_{peq} \frac{dT_{Cu}}{dt} = P_{ind} - T_{Cu} \left(\frac{1}{\frac{h_{Si}}{Sk_{Si}} + \frac{h_{Ins}}{Sk_{Ins}} + \frac{1}{Sh_{comv}}} + \frac{1}{\frac{h_{Si}}{Sk_{Si}} + \frac{h_{PVC}}{2Sk_{PVC}}} \right) + T_a \frac{1}{\frac{h_{Si}}{Sk_{Si}} + \frac{h_{Ins}}{Sk_{Ins}} + \frac{1}{Sh_{comv}}} \\ \quad + T_{PVC} \frac{1}{\frac{h_{Si}}{Sk_{Si}} + \frac{h_{PVC}}{2Sk_{PVC}}} \end{array} \right. \quad (5.6)$$

$$\left\{ \begin{array}{l} m_{tot} C_{peq} \frac{dT_{PVC}}{dt} = P_{ind} - (T_{PVC} - T_{Cu}) \frac{1}{\frac{h_{Si}}{Sk_{Si}} + \frac{h_{PVC}}{2Sk_{PVC}}} \end{array} \right. \quad (5.7)$$

where T_a is the ambient temperature. The equation is sorted such that the terms in front of $\frac{dT_{Cu}}{dt}$, T_a and T_{PVC} are denoted by A, B and C respectively. This gives the following solutions:

$$\left\{ \begin{array}{l} T_{Cu} = \frac{P_{ind}}{B+C} (1 - e^{-t \frac{(B+C)}{A}}) + T_{PVC} \frac{C}{B+C} (1 - e^{-t \frac{(B+C)}{A}}) + T_a \frac{B}{B+C} (1 - e^{-t \frac{(B+C)}{A}}) \\ \quad + T_a e^{-t \frac{(B+C)}{A}} \end{array} \right. \quad (5.8)$$

$$\left\{ \begin{array}{l} T_{PVC} = \frac{P_{ind}}{C} (1 - e^{-t \frac{C}{A}}) + T_{Cu} (1 - e^{-t \frac{C}{A}}) + T_i e^{-t \frac{C}{A}} \end{array} \right. \quad (5.9)$$

Equations 5.9 and 5.9 consider that the temperature of the PVC and the temperature of the copper receiver are constant in their own equations; therefore, in the algorithm, both solutions need to be computed separately, and the solution of each should be inserted for the calculation of the following step. This is a necessary method because the calculation of the differential equation requires knowing the variation in P_{ind} in time, and cannot be calculated until the folding angle has been computed. With the data of the induction coil (described in Section 5.2.6 and of the various materials, the model does not predict meaningful temperature output. However, with a reduced magnetic field amplitude, Simscape outputs the green graph, as shown in Figure 5.3 (a). At this stage, the model can only predict the temperature rise of a receiver without insulation or folding, although it exhibits inaccuracies and needs to be adjusted further. The difference in the heat rate between the model and the experiments highlights a faster temperature increase from the model than the experiments, meaning that the parameters of the heat loss should be adjusted. Also, the robustness of the model was not tested on different receiver design and the skin effect is not considered. Using a more complete and accurate model, the heat losses and the stalling angle could be more clearly assessed. As the receiver folds and approaches 90° , it decouples from the coil ($\Phi = B_{rec} S \cos(\theta)$). Consequently, the heat generated in the receiver reduces gradually as the hinge folds and the heat losses stop further temperature increase; thus, the hinge may stop folding at an angle called stalling angle. This angle also depends on the power required to trigger the smart material. The higher the activation temperature, the sooner the hinge should encounter its stalling angle. Furthermore, the stalling angle is affected by the conduction losses and the heat capacity, as they impact the heat rate of the receiver. Once the receiver

starts heating, the heat must circulate through the insulation layers, each storing the heat and dispensing it to the next layer. This creates a small delay between the starting time of the heating and the start of the folding during which, if sufficient heat is supplied, the hinge should fold beyond its stalling angle. On the contrary, this delay could negatively impact the hinge, as it prevents the heat dispersion. Overheating PVC to its melting point ($> 100^{\circ}\text{C}$) creates highly toxic fumes made of Hydrogen Chloride. Precautions must be taken in order to avoid reaching this temperature.

5.2.2 Receiver design and configuration

The main challenge of structures self-folding with magnetic induction is the miniaturisation of the hinges because, as the surface of the metallic receivers is reduced, the energy they receive from the induction coil is decreased. This subsection describes the design and configuration of receivers to improve the power received from the induction coil, thus enabling a wider range of structures to be self-foldable.

Design of receivers

When exposed to high frequencies, the electrons from the induced current are subjected to the skin effect, causing them to travel only at the periphery of the conductor. Because electrons generate heat via the Joule effect, the sides of the receiver heat more than its centre, causing the central part of the receiver to function as a heat sink. The first improvement consists of removing the heat sink, which slows the temperature rise. For this purpose, two square receivers are cut with 5 mm and 10 mm square holes at their centre and are referred to as hollow 5 mm and hollow 10 mm. These receivers were exposed to an alternating magnetic field, and their temperature rise was averaged over 5 trials and compared with that of a plain receiver, as shown in Figure 5.3 (a). Over the course of 9 s of heating, the temperature of the plain receiver increased to 130°C , whereas the temperature of the hollow receivers increased to 140°C in 9 s for hollow 5 mm, and 145°C in 5 s for hollow 10 mm. Because the temperature rise is a function of the surface of the receiver, this amounts to a temperature rate of $0.232^{\circ}\text{C/s/mm}^2$ for the 10 mm hollow receiver compared with a rate of $0.11^{\circ}\text{C/s/mm}^2$ and $0.09^{\circ}\text{C/s/mm}^2$ for the 5 mm hollow and plain receivers at 5 s, hence reducing by a factor of two the surface by making the receiver hollow double the heating efficiency.

Configuration of the receivers

There are several possible designs for creating self-folding hinges using smart materials. The most basic hinge consists of a dual layer made of smart material laminated onto a structural

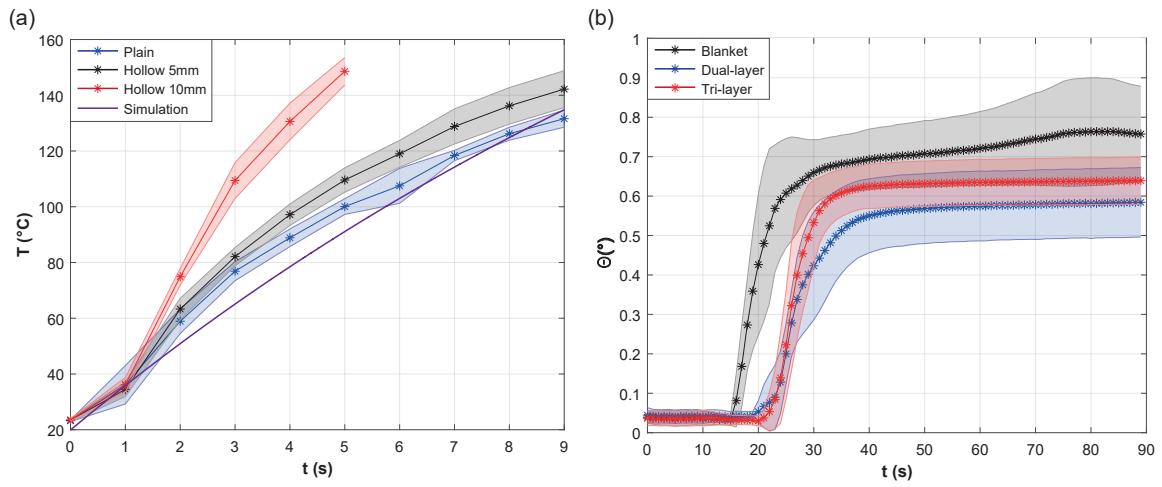


Fig. 5.3 Temperature rise of 15 mm side square receivers: plain and hollow with 5 mm and 10 mm hole (a), and completion of the folding of the hinges Dual-layer, Tri-layer and Blanket (b).

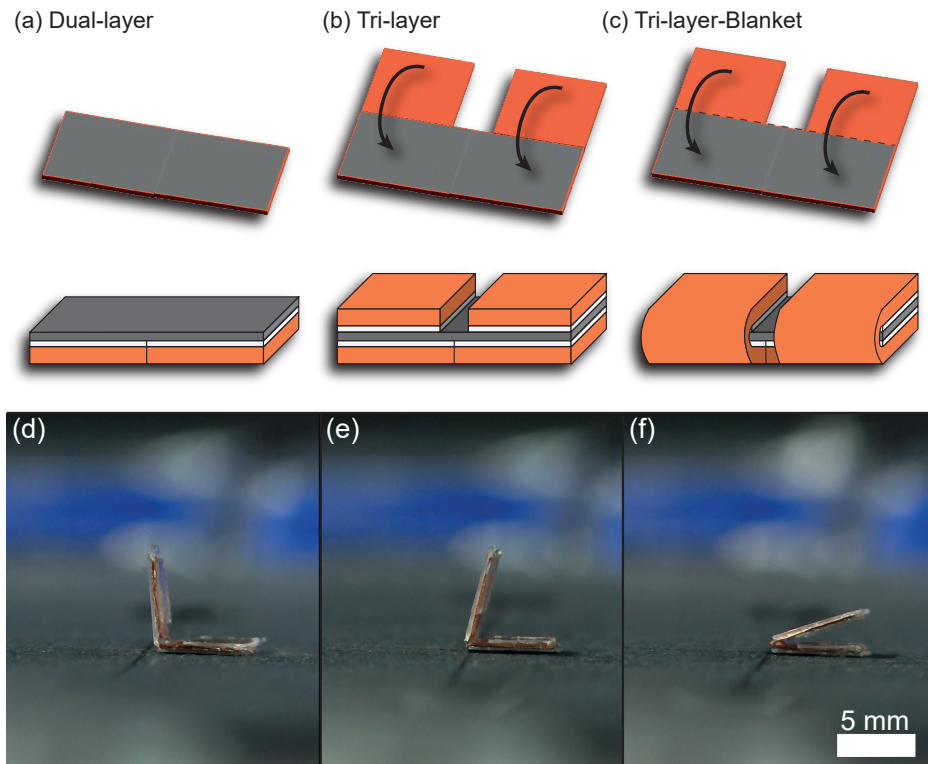


Fig. 5.4 Illustration of the hinges dual-layer (a), tri-layer (b) blanket (c), and a picture after folding of the dual-layer (d), tri-layer (e) blanket (f).

layer (Figure 5.4 (a)). Here, the hinge is made of copper, which serves as a receiver for the magnetic field. With a tri-layer approach, the smart material is sandwiched between the structural layers that serve as folding angle limiters (Figure 5.4 (b)). Although the number of receivers is increased, and thus the volume of magnetic material, the receivers are independent of each other. Because of the larger size of receiver, a greater amount of energy is received, which lead to a higher heat generation. Extending the tri-layer configuration, a third design is proposed: the tri-layer blanket hinge referred as blanket hinge. As shown in Figure 5.4 (c), instead of separating the top and bottom receivers, they are interconnected by folding the receiver on top of the smart material. Additionally, the blanket hinge also simplifies the fabrication process, as one only needs to fold the structural layer back on itself, instead of placing and aligning the top receivers manually. Here, 6 dual-layer, 7 tri-layer and 8 blanket hinges are self-folded, each with square receivers 5 mm wide. They were insulated with 127 μm -thick Dura-Lar, which limited their folding angle to a maximum of 160° . Their folding angles were tracked a video camera and plotted in Figure 5.3 (b) as a percentage of folding completion. The blanket hinge required 15 s to start folding, followed 5 s later by the tri-layer and dual-layer hinges. With respect to the folding angle, initially folding at 6.5%/s, 3.6%/s, 2.7%/s for the blanket, tri-layer and dual-layer configurations respectively, the folding speed was reduced to 0.16%/s, 0.053%/s, 0.059%/s after reaching the stalling angle. Although, the dual-layer folds the least due to its lower heat capacity corresponding to the amount of energy it stores within its structure, the dual-layer and tri-layer fold simultaneously. This implies that both generate a similar amount of energy via magnetic induction, which is coherent since the bottom layer behaves as a magnetic shield, preventing electromagnetic waves from passing through. On the opposite, the blanket configuration does not behave similarly due to the electric connection between top and bottom layers which doubles the electrical resistance, thereby increasing the power induced. Overall, the blanket hinge demonstrated self-folding at higher speed and angle than other configurations, and succeeded in passing the stalling angle in 71% of the trials, thus indicating a better resilience to the stalling and potential to be further miniaturised. Although the hinge capabilities are improved, the physical explanation to the improvement are only hypothesised.

5.2.3 Self-folding overhand knot design

Amorphous structures are composed of intertwined chains that spontaneously form knots. These chains are objects that can deform at any point in the length (Figure 5.5); however, replicating such structures with self-folding techniques requires the reduction of the degrees-of-freedom. Here, such a methodology is proposed. A knot can be decomposed into helices with different pitches, revolutions and directions. The direction dictates whether the helix

coils turn right or left, which correspond to the chirality of the knots. The pitch depicts the height of the helix for a complete turn, and the revolution corresponds to the number of turns.

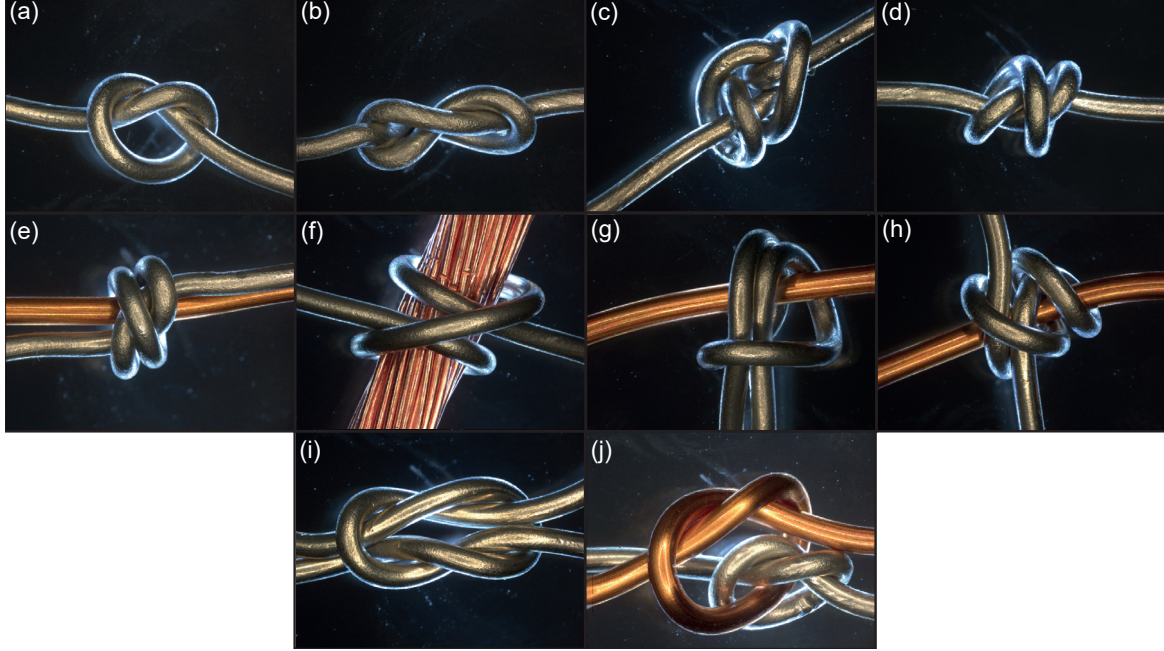


Fig. 5.5 Microscope pictures of the: Overhand knot (a), Figure 8 (b), Ashley stopper (c), Double Overhand stopper (d), Fishermen's bend (e), Clove hitch (f), Cow's hitch (g), Rolling hitch (h), Half Knot (i), Double Overhand Knot (j).

To translate a knot into a folding pattern, the knot is split into helices and loops which are, in a sense, a helix with near-zero pitch (Figure 5.6 (a)). Both the loop and helix sections can be discretised into a certain number of hinges (Figure 5.6 (b)). Thus, one can imagine the loop as any l -side polygon (Figure 5.6 (c)), where the continuity of the first edge is the beginning of the string, and the last edge l links the loop and the helix (Figure 5.6 (d)). At this point, the folding pattern consists of only $l - 1$ in-plane hinges and of one out-of-plane hinge to prevent the collision of the start and end of the loop (Figure 5.6 (e)). Then, the first hinges of the helix should fold out-of-plane to bring the helix parallel to the loop (Figure 5.6 (f)). This requires two hinges to fold out-of-plane as a single hinge cannot fold 180° without colliding with itself. In this manner, as the helix is parallel to the loop and the induction coil, the receivers maintain their coupling with the coil and can be heated more easily. Then, the helix must fold out-of-plane and pass through the surface of the polygon (Figure 5.6 (g)). This implies a need for an additional out-of-plane hinge. To accommodate the passage of segment h through the loop without collision, the length of segment $l + 2$ and its angle can be adjusted. For this purpose, the angles of the hinges around segment $l + 1$ should be modified.

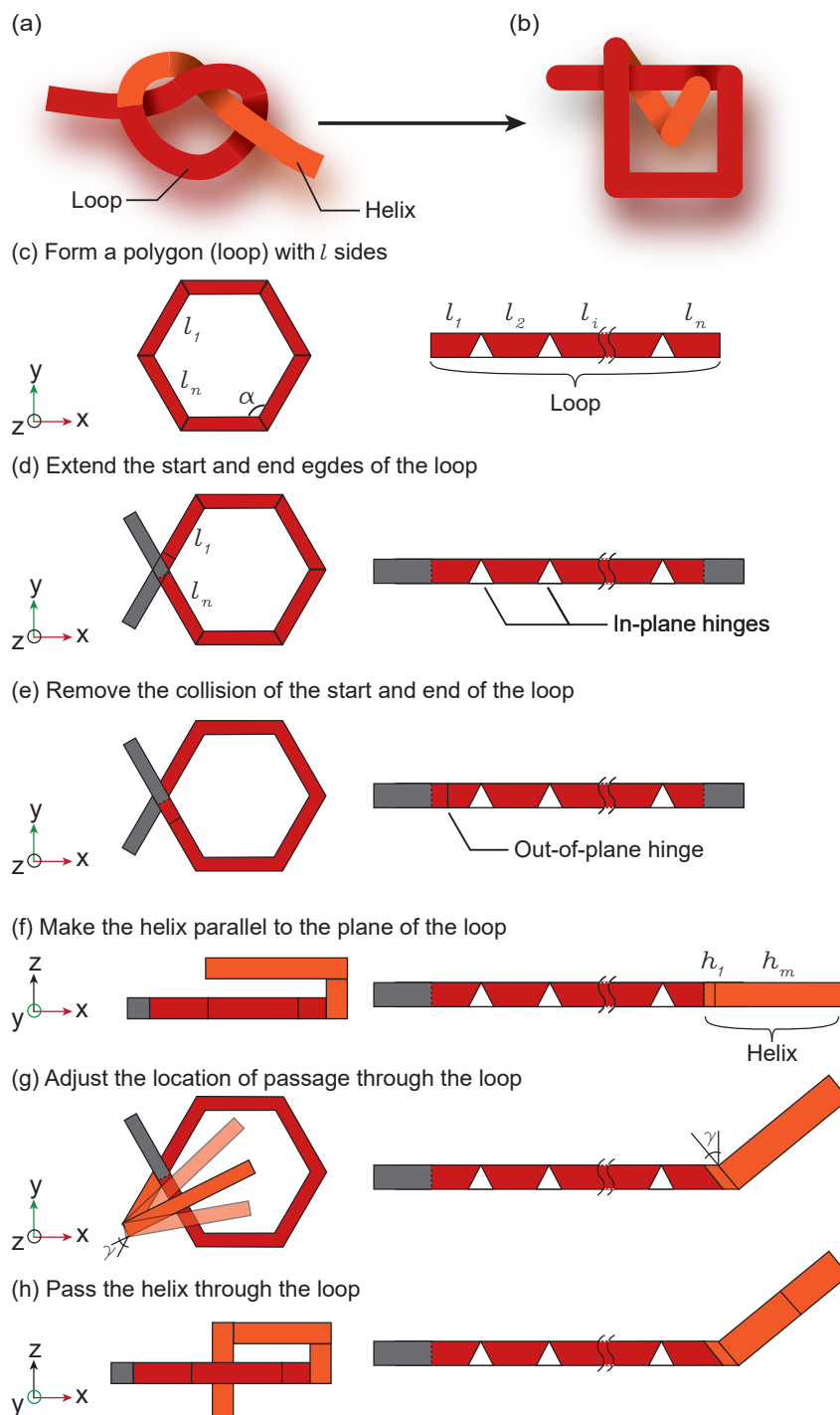


Fig. 5.6 Illustration of the methodology to design overhand knot. An overhand knot with infinite (a) and with limited degrees-of-freedom (b). The loop is formed from a polygon (c), of which a corner is split in two ends that are extended (d). An out-of-plane hinge is placed to avoid collisions between the two ends of the loop (e). The helix is folded parallel to the plane of the loop by placing two out-of-plane hinges on the helix (f). The end of the helix is folded through the loop by adding another out-of-plane hinge (g). The location of the passage of the helix through the loop is adjusted by tuning the angle γ of the first hinges of the helix (h.)

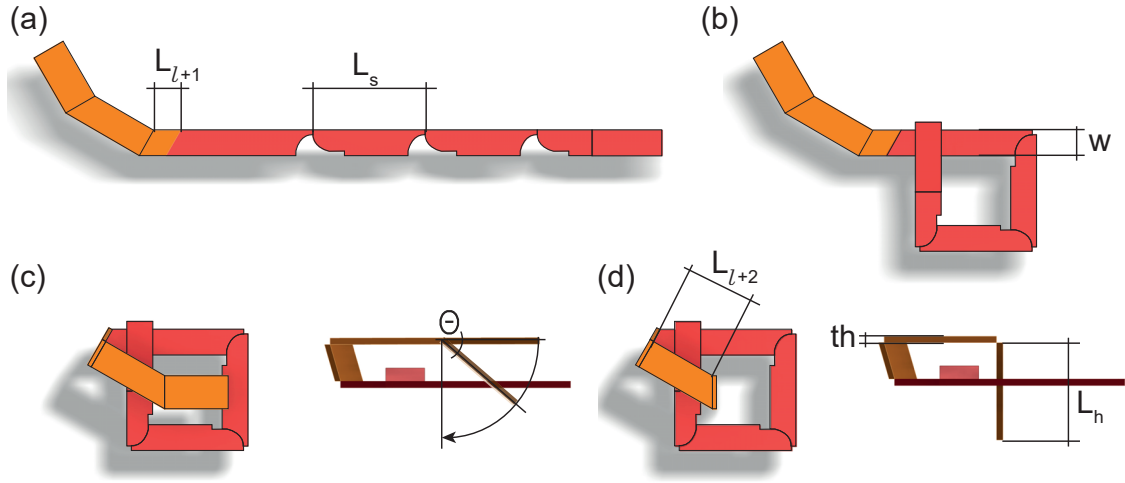


Fig. 5.7 Snapshots of the simulation of the knot folding from the pattern proposed by the knot design methodology. The string is initially in its flat state (a), then the loop folds (b), the helix places itself parallel to the loop (c), and passes through the loop (d).

This methodology was validated by simulating an overhand knot and extending the method to geometric constraints as illustrated in Figure 5.7. The string has a width w , a thickness th , and each segment length is denoted L_i where i is the index to identify the segment starting from 0 to h . The segments of the loop all have a length L_s corresponding to the length of the polygon side except for segments 0 and l which should be longer to extend beyond the knot. This length extension depends on the polygon used as a base, but considering that two segments of a polygon are at an angle α from each other, the lengths of segments 0 and l can be found as $L_0 = L_s + \frac{w}{\tan(\alpha)}$. For $\alpha = 90^\circ$, as shown in (Figure 5.7 (b)), segments 0 and l should only be longer than L_s .

Figure 5.7 (c) shows three geometrical constraints. The first constraint concerns the parallel condition of the helix and the loop. To ensure that the helix is parallel, L_{l+1} should be at least twice as long as the thickness th of the string. Consequently, the collision of the helix with the loop segments is prevented and they can be parallel. Additionally, L_{i+1} must remain in the self-foldable zone provided by the induction coil. The second constraint pertains to the passage of segment h through the knot. During folding, the projected length of segment h in the plane of the loop should not extend beyond the edges of the polygon; otherwise, the helix is ultimately not locked in the loop. In addition, L_h should be longer than L_{i+1} to pass through the loop. To fulfil the previous constraint, segment $h - 1$ of the helix must be long enough to position segment h above the loop but should take into account its rotation and potential collision. Figure 5.7 (d) highlights the last geometrical constraint: the collision of the loop edges with themselves (Figure 5.6 (e)). The out-of-plane hinge should

be placed at a distance such that it does not collide with the loop. This can be calculated as $L_x = \frac{th}{\tan(\theta_0)}$ where L_x is the minimum length between the closest point of segment l and the axis of the hinge of segment 0.

5.2.4 Design of the hinges

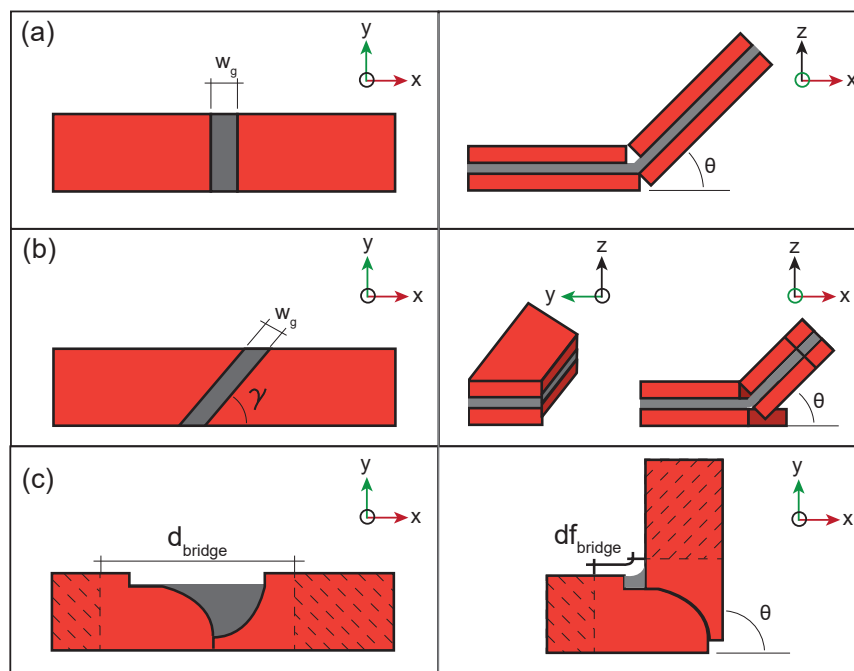


Fig. 5.8 Illustration of the straight out-of-plane hinge (a), the angled out-of-plane hinge (b), in-plane hinge (c), and the plot of the surface of induction as a function of the folding angle for the in-plane and out-of-plane hinges (d).

As mentioned in the Chapter 2, one of the main challenges of self-folding strings are the scale and torque, thus constraining the design of the knot to minimise the required folding torque on the hinges.

Since the loop is a structure in a single plane of space, the hinges should bring the entirety of the string in the plane. Considering the wide literature on origami, the first attempt consisted of a set of two angled out-of-plane hinges (Figure 5.8 (b)), which could bring the rest of the string in-plane. Although this approach is functional and manufacturable, its use can only be limited to short strings as both hinges are required to lift the entirety of the string above the workspace and therefore, loose coupling with the induction coil. Adding to the fact that the string lays on one side onto the workspace, the entanglement of the string is difficult to predict as hinges fold and decouple simultaneously to form the loop.

To improve the foldability, a new hinge was designed called "in-plane hinge" seen in Figure 5.8 (c). On the opposite of out-of-plane hinges seen in Figure 5.8 (a) and (b), in theory, this new design remains at maximum coupling through all folding angle) and hence, do not show any stalling angle. This allows the hinge to fold faster than its out-of-plane counter parts due to the constant energy provided to the receivers and, when implemented in a serie of such hinges, allows a more intuitive understanding of the folding sequence. However, the first version of the design in Figure 5.9 (b) failed to fold to its target angle (90°) due to the inadequate bridge length labelled d_{bridge} in Figure 5.8 (c) preventing the shrinking PVC to bring together the two receivers. The main weakness of this design was identified to be the lack of surface of contact between the receivers. This caused the in-plane hinge to deform out-of-plane, or the two receivers to slip beneath each other and overlap, thus only translating.

To improve the folding angle of the previous design, the silicone glue was partly removed from the surface of the receivers (dotted surface in Figure 5.9 (c)) in order to increase the bridge length of PVC. As a result, the hinge could fold to 85° if no out-of-plane motion occurred. The design was also altered to increase the surface of contact by adding a layer of Dura-lar (labelled E) above and below the receivers. This solved the translation issue, but the out-of-plane deformation remained in the case of misalignment of the Dura-lar layers. In addition, this design introduced a new challenge concerning the surface of induction. As the hinge fold, the two receivers surface of contact increases and with it, the surface of induction. Consequently, the two separate receivers became one larger receiver causing the hinge to overheat and vaporize the PVC, thus destroying the hinge.

As a final design in Figure 5.9 (d), the receivers were spaced from one another to prevent the increase of induction surface, and the Dura-lar guides were extended on one side and shortened on the other so as to ensure no out-of-plane motion could appear. In addition, the Dura-lar guides design was changed to gear in each other, thus easing their alignment during the fabrication. Lastly, the blanket (illustrated as a dashed line in Figure 5.9) was changed side so that the receiver could behave as a fairlead similar to the approach in chapter 3. All these modifications resulted in the in-plane hinge 90° folding and single degree-of-freedom. More details can be found in section 5.2.5 regarding its fabrication process.

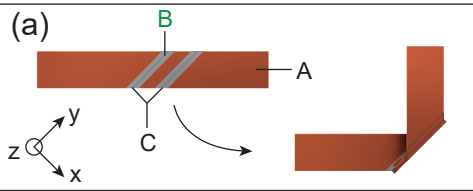
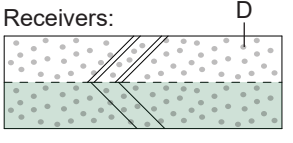
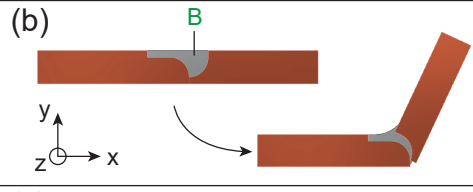
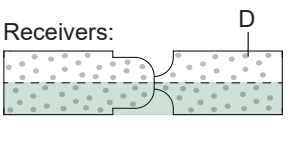
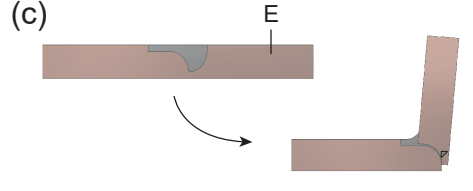
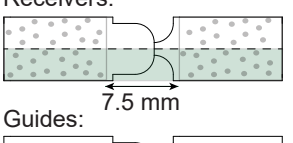
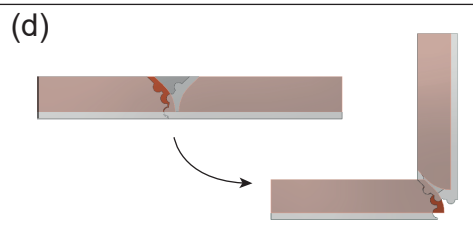
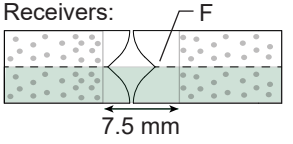
Hinge Designs	Sketch	Features	Size	Degrees Of Freedom	Maximum Folding Angle
(a) 	Receivers: 	A: Copper receiver B: PVC C: Out of plane hinges D: Silicone glue 	32x5x0.22 mm	$\begin{Bmatrix} 0 & 0 \\ 0 & 1 \\ 0 & 0 \end{Bmatrix}$	2x 90°
(b) 	Receivers: 	A: Copper receiver B: PVC bridge C: In-plane hinge D: Silicone glue 	44x5x0.22 mm	$\begin{Bmatrix} 1 & 0 \\ 0 & 1 \\ 0 & 1 \end{Bmatrix}$	65°
(c) 	Receivers:  Guides: 	A: Copper receiver B: PVC C: In-plane hinge D: Silicone glue  E: Duralar guide	44x5x0.52 mm	$\begin{Bmatrix} 0 & 0 \\ 0 & 1 \\ 0 & 1 \end{Bmatrix}$	85°
(d) 	Receivers:  Guides: 	A: Copper receiver B: PVC C: In-plane hinge D: Silicone glue  E: Duralar guide F: Fairlead	50x6x0.52 mm	$\begin{Bmatrix} 0 & 0 \\ 0 & 0 \\ 0 & 1 \end{Bmatrix}$	90°

Fig. 5.9 Table summarising the main designs of in-plane hinges investigated. (a) Initial design made out of two out-of-plane hinges. (b) Second design, (c) Third design, (d) Final design.

Figure 5.10 (a) to (d) illustrates the self-folding of the latest version of the in-plane hinge with a 90° target folding angle. This version of the in-plane hinge started folding after 30 s and took an average of 100 s on five samples to fold to 90° . During the initial stages of the folding, the folding angle discrepancy is of $3^\circ \pm 3.8^\circ$, reaches a maximum of $62^\circ \pm 21^\circ$ during the transient stage before stabilizing to $88^\circ \pm 8.5^\circ$ in the final stages of the folding as shown in Figure 5.10 (e). Overall, out of the 5 samples, only one failed to fold beyond 80° while the other samples folded between 90° and 100° .

Despite the modifications to improve the folding success rate of the in-plane hinges, the reliability of the hinges regarding their folding accuracy and smoothness of the folding is low. The main limitation being the flexibility of the Dura-Lar layer (due to the material mechanical properties and thickness), a stiffer material able to withstand temperatures reaching 100°C is required.

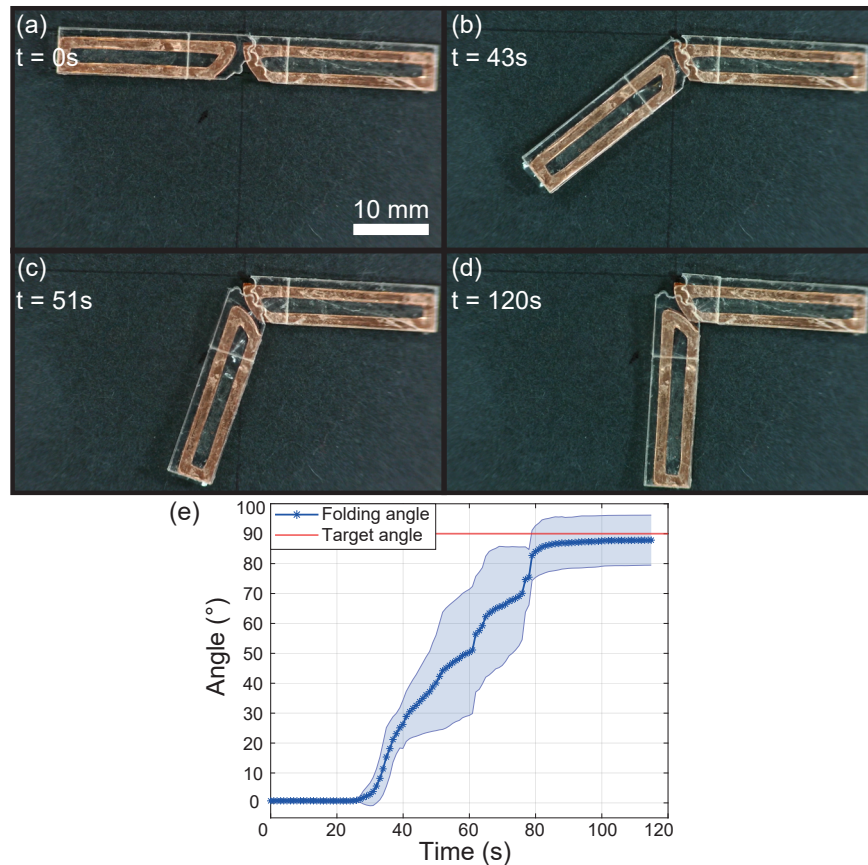


Fig. 5.10 In-plane hinge in its straight configuration (a), and self-folding progressively to 40° (b), 80° (c) and reaching its target angle: 90° (d). Average folding angle for five samples (e).

5.2.5 Fabrication process

Out-of-plane self-folding structure

The fabrication process comprises 7 steps. First, the copper sheet is carefully laid on a sticky mattress to avoid introducing air below the copper sheet. The mattress is then fed into the vinyl cutter (Silhouette Cameo 3) and the pattern is cut (Figure 5.11 (a)). Once cut, the unwanted sections of copper should be peeled off to avoid damaging the structure when removing the copper in later stages. Similarly, the silicone tape is laid and processed (Figure 5.11 (b)). Pressure is applied to the silicone tape to ensure that air is not trapped between the layers and the tape is properly glued onto the copper (Figure 5.11 (c)). Thus, the thermal conduction through the silicone tape is more uniform and the protective layer of the silicone glue can be removed without peeling off the glue. The PVC film is then placed onto

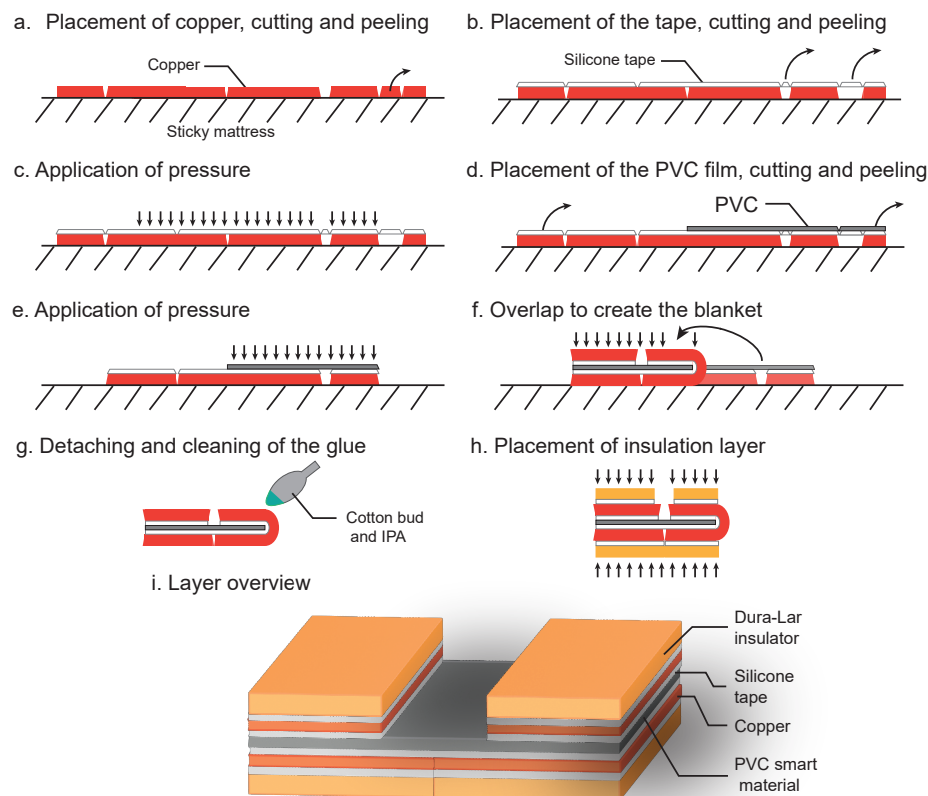


Fig. 5.11 Manufacturing process of the PVC/Copper blanket tri-layer. The copper sheet is placed onto a sticky mattress and cut (a), then the double sided silicone tape is layed onto the cut pattern, and pressure is applied to ensure correct glueing (c). The PVC shrink film is placed and unused pieces are removed (d), before pressure is applied again (e). The PVC is tucked underneath the copper blanket (f), and the structure is detached and cleaned using a solvent (g).

half of the pattern, cut by the vinyl cutter and the excess material is removed (Figure 5.11 (d)). Pressure is applied once again and the PVC film side of the pattern (Figure 5.11 (e)) is folded back onto the other half (Figure 5.11 (f)). To ensure that all layers are glued properly, pressure is applied once more and the sticky mattress is removed from the vinyl cutter. Finally, the PVC film/copper composite is peeled off and any tracks of glue from the silicone glue or the sticky mattress are dissolved using a cotton bud soaked in with isopropanol (Figure 5.11 (g)). As a result, there is no remnant of glue around the hinges that could act as a buttress and limit the folding angle.

In-plane self-folding structure

Although the fabrication is similar to that of out-of-plane hinges, some adjustments are required to maximise the performance of the in-plane hinges. As can be seen in Figure 5.10 (a) to (d), the receivers directly above the bridge are covered with Dura-lar. During the fabrication process depicted in Figure 5.11 (c), (e) and (h), pressure is applied on the whole surface to prevent delamination and improve thermal contact. The main function of the Dura-lar, other than insulation, is to act as a guide during the folding to prevent out-of-plane motion, to smooth the folding, and prevent the opposite receivers to tuck within one another. Even though these panels are thick enough to function as rigid guides, the material's flexibility is high enough above 65° C to occasionally bend, causing the Dura-lar to tuck between one another. For this purpose, two layers of Dura-lar were placed onto each other to reduce the bending. Nonetheless, these two Dura-lar layers requires pressure to be applied onto them while keeping the PVC film bridge free to shrink to ensure their glueing. Hence, a 50 μm thick spacer was slid between the receiver and the PVC film before applying the pressure to avoid pinching it. Although the spacing reduces the efficiency of the heat transfer to trigger the shrinkage of the PVC film bridge, this reduces the friction within the hinge and prevent premature stoppage during the folding. Lastly, to prevent the blanket to open during the folding, the top and bottom Dura-lar panels extend beyond the width of the receivers and connects to each other thanks to a thin Dura-lar spacer. Additionally, this width extension lowers the center of rotation of the hinge during the initial stage of the folding to increase the folding torque.

5.2.6 Setup

This section details the characteristics of the induction heating system built with the help of Kaan Esendag. The main output parameters for an induction coil are magnetic field strength, frequency and size of the magnetic field. The first two parameters are translated into the coil

via its inductance, system electrical capacity and current supplied. Inductance is function of the number of turns of the coil, its surface, and its length. Made up of 200 strands of magnetic wire (Farnell: ECW0.2) manually winded, the wire of the coil is formed. Here, the circular coil consists of 6 turns with two 4 mm-thick layers and spans over 95 cm², thus exhibiting 5.47 μ H inductance. The capacitance C is matched using the equation $f = \frac{1}{2\pi\sqrt{LC}}$ to obtain a resonance frequency at 140 kHz. The coil is held by two acrylic plates, each 6 mm-thick; hence, the coil produces a 3.2 mT magnetic field at the centre of its workspace.

5.3 Results

This section presents the newly developed methods for achieving sequential folding via magnetic induction. The methods are demonstrated by self-folding a bio-inspired mimosa pudica leaf, a box and an overhand knot. In addition, the self-shrinking of a string is demonstrated as a first step towards self-tightening self-folding knots.

Sequential folding via receiver size

The aim of this experiment is to highlight sequential folding by varying the heating rate of receivers of different sizes.

For this purpose, a leaf origami pattern was designed and fabricated to enable a thermal response from the origami leaf, similar to that of mimosa pudica, whose leaves naturally close sequentially from the point of physical contact. The overall leaf is 40 mm long with a maximum width of 30 mm, and the sub-leaf surfaces range from 33.8 mm² to 12.1 mm². The leaves are fabricated using the classic trilayer approach, and the stem of the leaf uses the blanket configuration, thus supplying heat to the sub-leaves along its length. Sub-leaves are symmetrical from each side of the stem, meaning that each level of the leaf has the same surface and should fold simultaneously. As shown in Figure 5.12 (a), the leaf is placed such that the second-largest subleaves are at the centre of the induction coil (between the two screw heads). In this way, the induction supplies maximum power to the second-largest subleaves and decreases for farther sub-leaves. The second and third levels of the subleaves fold first after 12 s (Figure 5.12 (b)) followed by the first and fourth levels after 14 s of induction heating, and the fifth level after 18 s (Figure 5.12 (c) and (d)). However, the smallest levels at the farthest edge of the leaf from the centre of the induction coil fail to fold even after 45 s because they receive too little power from the induction coil. Sequential folding using different receiver sizes is therefore validated. This approach to sequential folding may be limited to certain origami patterns; however a possible solution to generalise the receiver size

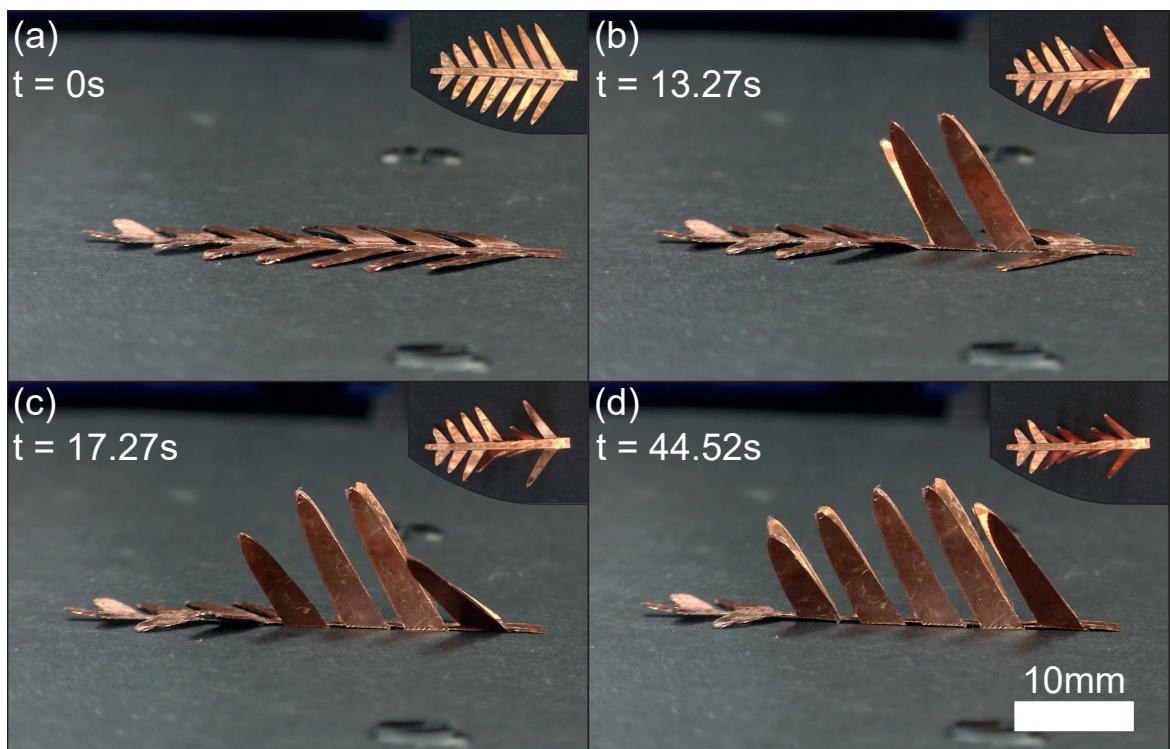


Fig. 5.12 Timestamps of the sequential self-folding of the mimosa pudica leaf. The leaf in its flat state (a) folds first the subleaves of layer 2 and 3 (b), followed by layer 1 and 4 and finally layer 5. Scale bar is 10 mm.

approach could be to insert several small receivers to fold a large angle. In this manner, a 180° fold could be split into three 60° folds.

5.3.1 Sequential folding via receiver design

The aim of this experiment is to achieve sequential folding by changing the receiver design.

To demonstrate the addressability of the folding, an origami box pattern was self-folded. The cubical box is 12 mm wide and possesses four flaps, of which, three flaps have a square hole at their centre 4 mm, 6 mm and 8 mm wide and the last flap is plain (without hole in the centre). To further decorrelate the heating of the flaps, the central receiver of the box pattern is replaced with paper; thus, no heat was generated at the centre of the box, indicating that each flap was independent in terms of heat generation. Additionally, to prevent the shrinkage anisotropy of PVC from influencing the result, opposite hinges share the same PVC strip that is cut in the direction of maximum shrinkage. The origami box was placed at the centre of the induction coil workspace so that each receiver could receive the same energy. As suggested in Section 5.2, the presence of holes in the receiver causes it heat at a higher rate. This is validated in Figure 5.13 where the 8 mm hole receiver folded in 6.5 s, followed by the 6 mm hole receiver in 7.37 s and 15 s for the 4 mm hole receiver and plain receiver. Although sequential folding was achieved by altering the design of the receivers, the sequence was not fully respected as the plain receiver folded slightly before the 4 mm hole receiver folded. Possible explanations include a potential misfabrication of the hinges (bent receiver or constrained hinge) or a misplacement of the origami box onto the induction coil as a similar folding rate would contradict the result in Section 5.2. The main advantage of the receiver design approach for sequential folding is its universal nature. Any receiver can be made hollow and, in the case of rigidity issues, be reinforced with a rigid insulator. Hence, this approach can be used for 1D self-folding strings and 2D origami patterns. Although the folding sequence can be tuned intuitively owing to the visibility of the holes in the receiver, the fabrication of the hinges becomes more difficult as the hole surface is increased and the receiver becomes more fragile.

5.3.2 Self-folding knot

Folding a knot requires the loop to fold first, followed by the helix passing through the loop. This can be achieved by altering the design of the receivers and their sizes in order to self-fold an overhand knot.

The methodology for translating knots from continuous into discontinuous deformation developed in Section 5.2 recommends forming a polygon with a strip whose ends should

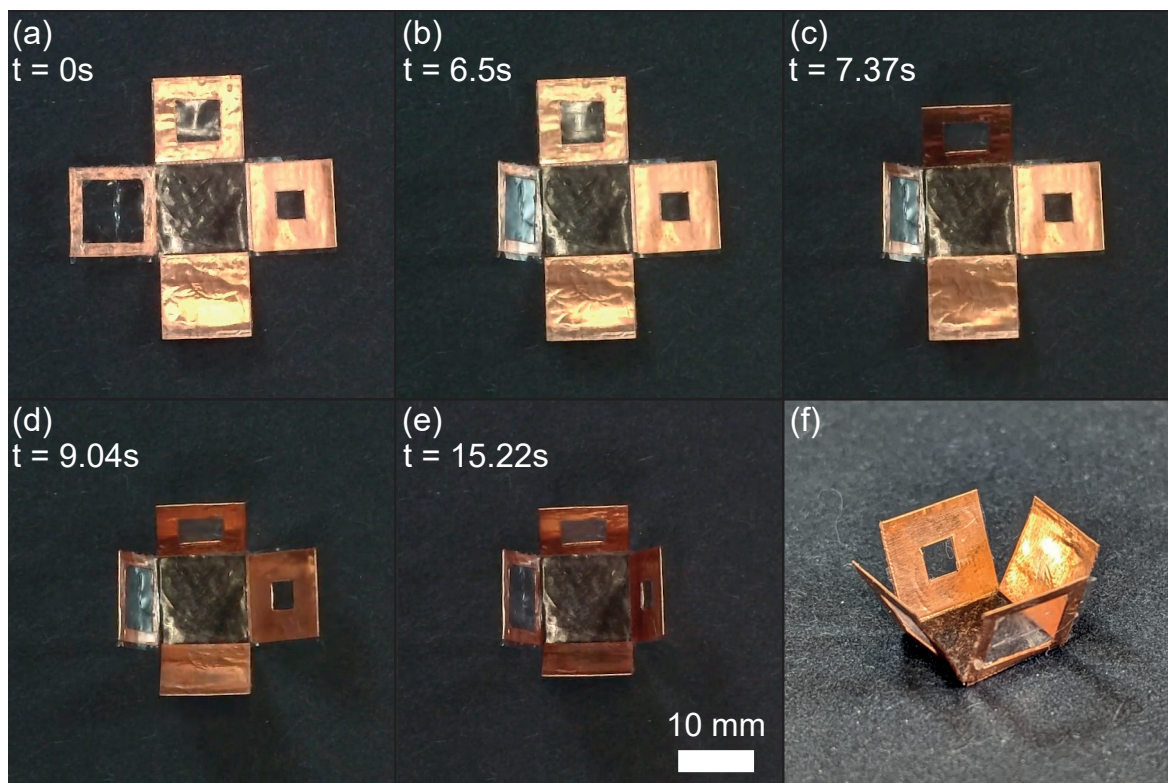


Fig. 5.13 Timestamps of the sequential self-folding of a cross pattern into an opened box. The structure in its flat state (a) folds first its receiver with an 8 mm wide hole, followed by the receiver with a 6 mm wide hole and simultaneously its plain and 4 mm wide hole receivers (d) until completion of the folding (e). Pictures from another perspective (f). Scale bar is 10 mm

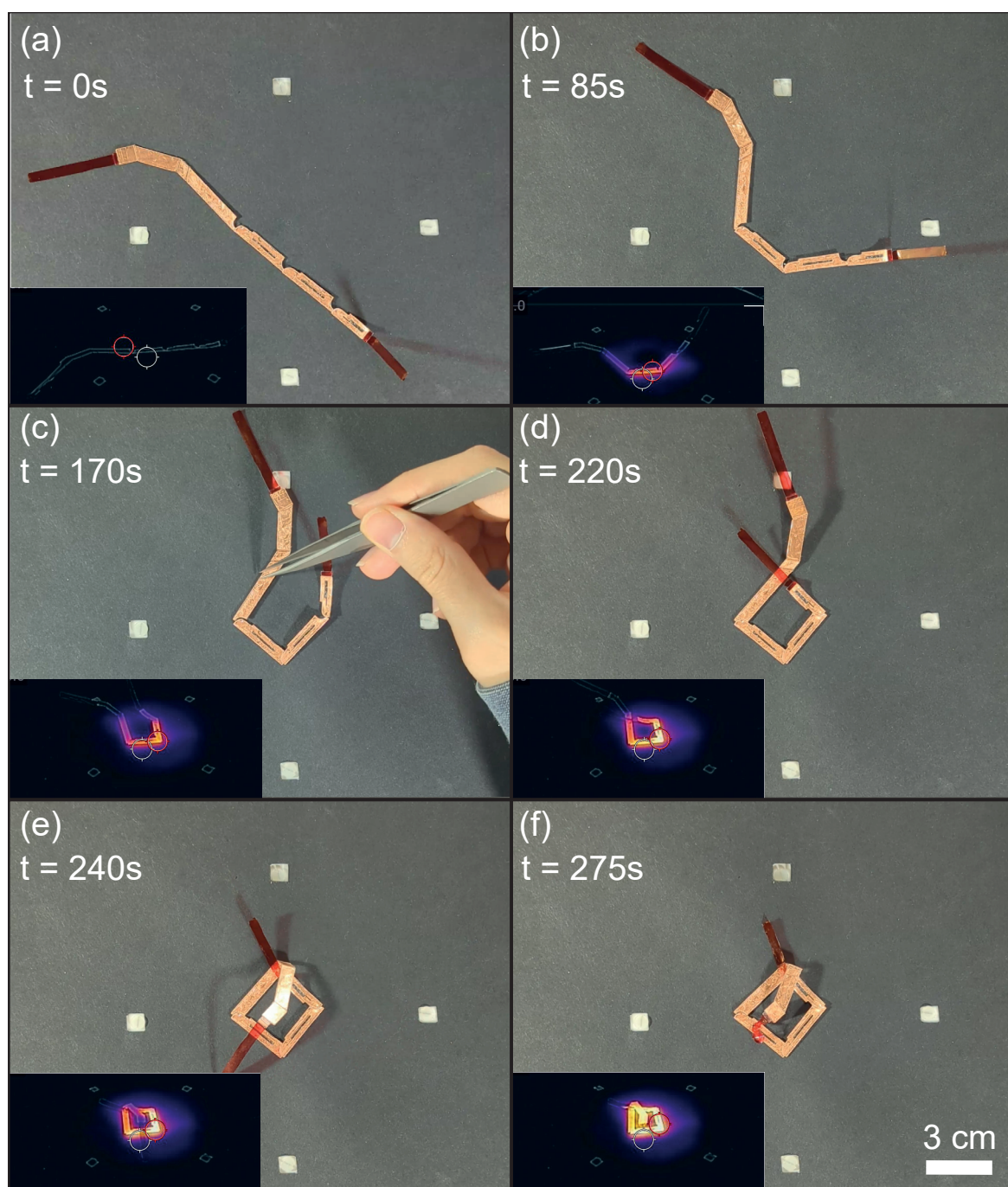


Fig. 5.14 Timestamps of the self-folding of the overhand knot. The knot is first in a flat state (a), starts folding its loop (b) before requiring the helix side of the loop to be pushed to complete the folding (c). The helix starts folding (d), has folded parallel to the loop (e), and passes through the loop (f).

not cross. Here, the loop forms a square with an internal length of 16 mm, thus covering 676 mm^2 around the centre of the induction coil. Instead of adding a hinge in receiver 0, it was manually bent upward so that the two ends of the string do not collide while self-folding. To fold the loop before the helix, the loop was made out of hollow receivers and was placed around the centre of the workspace. Because the placement of the structure is important for ensuring proper folding, the knot was anchored. For this purpose, the receiver $l - 1$ was glued to the workspace with an offset from the centre of the induction coil, so that the two ends of the knot, initially outside the self-folding zone, are brought in when other hinges self-fold. To further accentuate the discrepancy between the heating speed of the helix and loop, the receivers of the helix are plain. In addition, the helix is initially outside the self-folding zone and the first receiver of the helix is smaller than any other receiver to reduce the power it receives. Overall, when unfolded, the overhand knot string is between 5 mm and 7 mm wide, 135 mm long, and has six hinges split equally between the helix and loop which are equipped with $127 \mu\text{m}$ -thick Dura-lar to insulate the receivers and to limit the folding angle.

Figure 5.14 depicts the self-folding of the overhand knot along with its infrared thermal reading during folding. As shown in Figure 5.14 (b), the first section to heat up and fold is the loop that folded in 220 s with a slight manual intervention to push the receiver l that had failed to fold to its 90° target and the first receiver of the loop that had folded out-of-plane. From this stage onward, the rest of the knot folded without assistance. While folded, the loop brought the helix within the self-folding region in Figure 5.14 (c) which can be seen heating up on the infrared reading. From 180 s onward, the helix started to fold and completed its intended motion after 275 s, thus completing the self-folding of the knot. The self-folding of the overhand knot was conducted over four trials and for each, human intervention was required to fully self-fold the loop. When performed on a single hinge at the centre of the workspace, the in-plane hinges folded with a low success rate. The reason lies in the mismanufacturing of the hinges whose misalignment induces out-of-plane folding and thus, decoupling with the induction coil. In addition, if the insulation layer is not placed properly, once fully folded, the receivers of the in-plane hinges enter in contact, thus increasing the surface of induction. As a result, the temperature rises sharply and may cause the PVC to burn. Therefore, the in-plane hinge requires further design iterations and trials to improve its success rate and safety. Finally, the edge of the string being made of PVC, shrinkage occurs when the temperature of the receivers of the loop exceeds 65°C , resulting in the string failing to pass through the loop. While the methodology for translating knots recommends a receiver h longer than the receiver $l + 1$, glueing the knot to maintain its position while folding prevents the hinge of the receiver h from lifting the knot and passing the string through. In general, a knot is tied in space with both ends of the string free; however here,

the plane of the coil prevents the free motion of the string that bent out of the loop when it came in contact with the workspace surface.

5.3.3 Self-tightening

After self-folding, the knot should be tightened, thus increasing the friction within the knot as the string comes into contact with itself. This is a crucial step because it is the friction within the knot that gives it its mechanical integrity. This subsection pertains to the first steps towards a self-tightening knot.

This was performed using free receivers, called rings. Unlike the previously mentioned receivers, they are not glued to the PVC strip but instead are wrapped loosely around the strip. As a result, when the temperature increases above 65°C, the PVC surrounding the ring shrinks, bringing the other rings within the heating zone. The loose attachment of the rings also provide them with the necessary freedom of motion to move along the strip and let it shrink to a greater extent than if they were firmly attached. The rings are 8 mm long and 5 mm wide rectangles with rounded edges and whose arms, also rounded, hold onto the PVC strip. The round feature is essential for self-tightening because, two receivers entering in contact may fold out-of-plane, and the tightening should remain in-plane. In addition, the rings are insulated with the Dura-Lar layer, as mentioned previously. This second purpose of the insulation is to prevent the PVC from deforming out-of-plane. The exposure of PVC to a temperature gradient causes it to warp like bubbles. Consequently, if the PVC enters in contact with the hot receiver, it starts gripping to it, leading it to fold in an accordion. Thus, the insulation reduces the temperature of the PVC that can be in contact with the receiver and prevents out-of-plane deformation. In Figure 5.15, six rounded rings are placed on a PVC strip, each 10 mm from one another so that the two farthest rings are outside the workspace. While the rings are loose around the PVC strip, the leftmost ring is firmly attached to the PVC so that the rings can be placed on the strip, similar to the pearls on a necklace. This way, as the string shrinks, the rightmost ring should be brought within the workspace. As the ring temperature increases to 65°C, the PVC strip starts shrinking and the rings moves closer to each other. Initially 10 mm apart, the rings are 5.15 ± 3.9 mm closer after 96 s, thus shortening the PVC strip from 140 mm to approximately 115 mm. This is equivalent to 17.8% of shrinkage or almost 50% of the overall shrinkage capability of the string.

The self-shrinking experiment was extended to demonstrate how it could be used to close a gap in a 1.2 mm-thick soft membrane made out of Moldstar-15. The gap is 10×80 mm and covers a surface of 628 mm². The string is 300×5 mm long and manually pierced 6 times on each side of the gap to tailor the string around it. The gap closure is compared between a PVC-only string and a string with one ring between each hole around the wound.

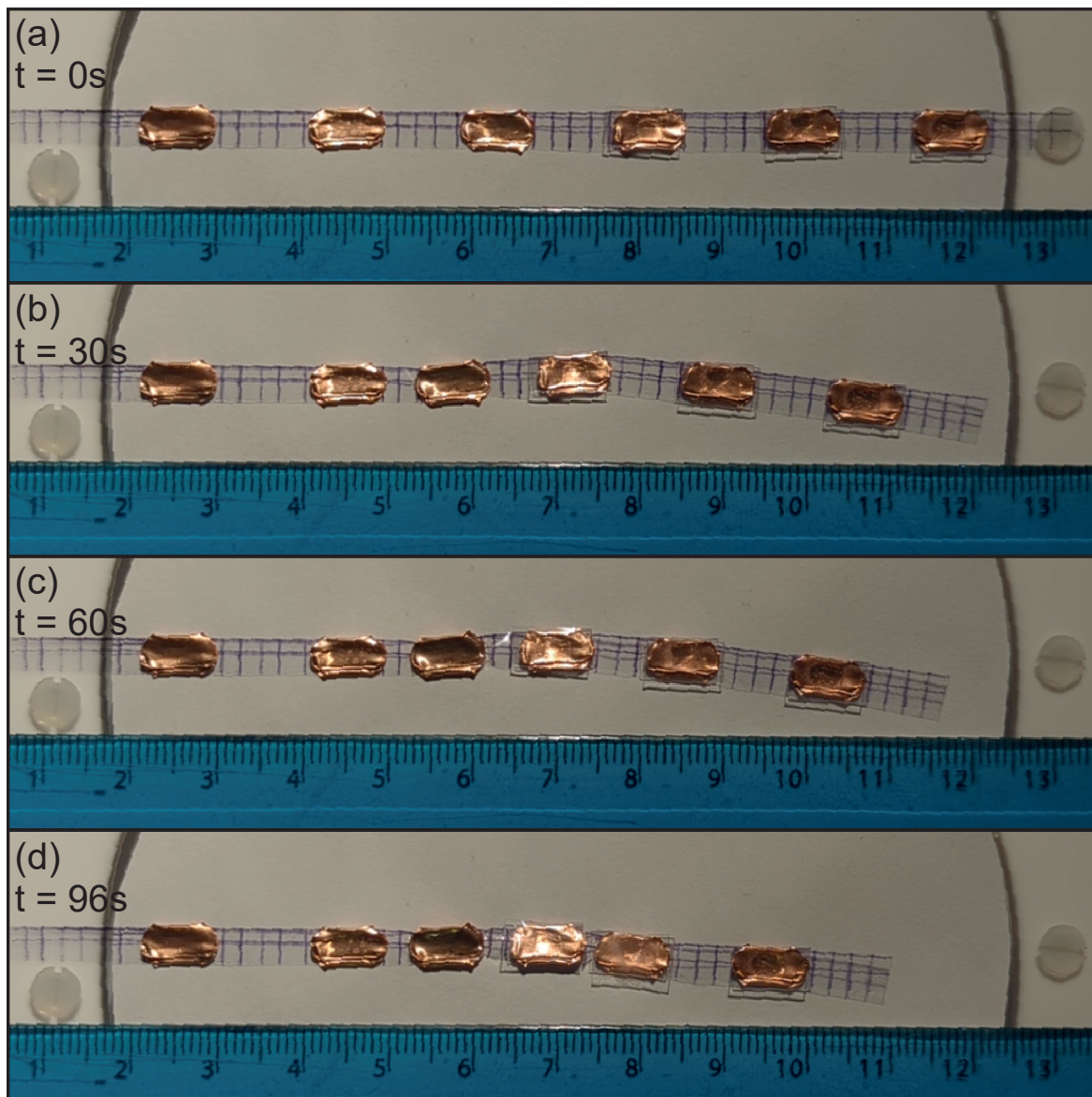


Fig. 5.15 Timestamps of the self-shrinking of a PVC strip. The strip in its initial state (a), after 30 s (b), 60 s (c) and 90 s of of induction heating

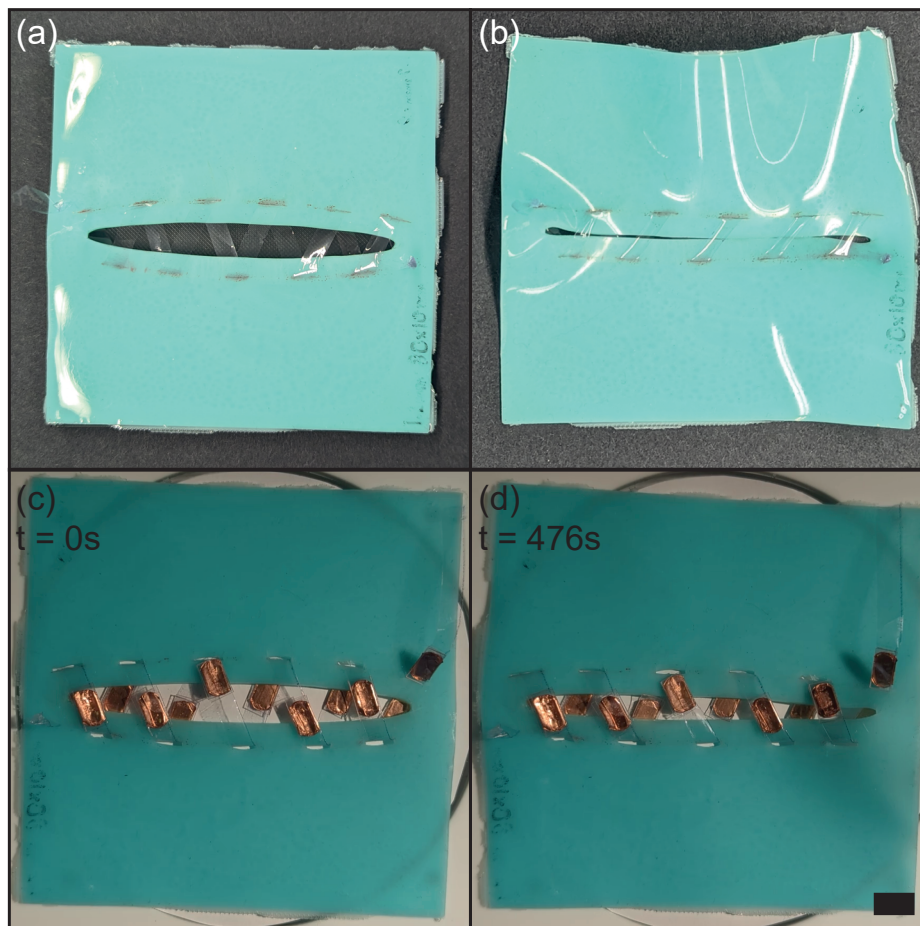


Fig. 5.16 Comparison of the closure of a gap in a soft membrane between a plain PVC strip before (a) and after being placed in an oven (b), and the ring string before (c) and after almost 5 mn of induction heating (d).

After exposure to magnetic induction for 476 s, the gap could be remotely tightened down to 368 mm^2 equivalent to 41% closure of the gap, whereas the PVC-only string closed 91 % of the gap. The main difference between the PVC-only string and the ring string is the actuation method. In contrast to the string ring that could only be triggered locally via magnetic induction, the PVC-only string was placed in the oven that could activate the entire string at once.

The actuation method coupled with friction and obstacles along the string prevents the rings from travelling and fully activates the PVC. In addition, silicone adhered well to the workspace surface, which implied that a strong force had to be applied to close the gap as the membrane can be seen warping in Figure 5.16 (b).

Overall, self-tightening is in its early stages, as it requires more design to be tested and experiments to confirm the basis of the preliminary results. For example, changing

the size and number of peripheral holes around the gap, changing the number of rings or changing their design to allow them to deform and slide could be a part of future experiments. Foreseeing such an application to self-suturing for in vivo applications, the closing force as well as the resorbability, biocompatibility and method to incise the tissue should be a priority along with the navigation and control of such a string. The concept of self-tightening could be applied to other fields, such as fluidics to control canal size or to assemble and strengthen matter by introducing strings that could rigidify the structure.

5.4 Discussion

5.4.1 Comparison with state-of-the-art

Reference	Type of string	Module Size	Module number	Torque	Medium	Mass in medium	Power Source	Folding in	Actuation Method	Assembly time	Remote folding	Untethered	Reversibility	Addressability	Reprogrammability	Sensing	Assembly and Deformation	Application field(s)
Yim et al. [167]	Chain	3x3x3 mm	999*	$3.2e^{-2}$ Nm [†]	Guide exit	36 g	Magnets	3D	Magnetic pull/push	108 s	Yes	/	Yes	/	/	/	Self-assembly and self-folding	Medical implants
Fass et al. [189]	Chain	20x20x10 mm	6	$2.5e^{-1}$ Nm [†]	Planar surface	>>0 [†]	Magnets	2D	Magnetic pull/push	0.2 s	Yes	Yes	Yes	/	/	/	Self-assembly and self-folding	Minimally invasive surgery
Griffith et al. [172]	Chain	25x25x3 mm	20*	$2.1e^{-1}$ Nm [†]	Air cushion	~0 [†]	Magnets + Air	2D	Magnetic pull/push onto air cushion	30 s	Yes	/	Yes	/	/	/	Self-assembly and self-folding	Programmable matter
Han et al. [43]	Chain	14Øx9 mm	20*	$2.9e^{-2}$ Nm [†]	Agitated hot water	~0 [†]	Magnets + Air + Heat	2D	Magnetic pull/push into bubbling water	10 s	Yes and No	Yes	Yes	/	/	Yes	Self-assembly and stretching	Programmable matter
Gu et al. [185]	Chain	15x5x5 mm	9*	$3.4e^{-3}$ Nm	Guide exit	>0 [†]	Magnets	3D	Magnetic pull/push	13 s	Yes	/	Yes	/	/	/	Self-assembly and self-folding	Minimally invasive surgery
Li et al. [7]	Ribbon	4x5x0.5 mm	5	$1.42e^{-2}$ Nm [†]	Planar Surface	320 mg [†]	Magnets	2D	Magnetic push/pull	/	Yes	Yes	Yes	/	/	/	Self-folding	Robotics and programmable matter
Ijäs et al. [194]	Chain	3x2x2 nm	8064	/	Solution	~0	Atoms	3D	Atomic forces	/	Yes	Yes	/	/	/	Yes	Self-assembly and self-folding	Drug delivery
Douglas et al. [195]	Chain	3x2x2 nm	7308	/	Solution	~0	Atoms	3D	Atomic forces	12h	Yes	Yes	/	/	/	Yes	Self-assembly and self-folding	Drug delivery
Bai et al. [191]	Chain	3x2x2 nm	7249	/	Solution	~0	Atoms	3D	Atomic forces	12h	Yes	Yes	/	/	/	/	Self-assembly	Programmable matter
Cheung et al. [173]	Chain	77x38x38 mm	7*	1.5 Nm	Planar surface	>371 g	Electricity	3D	Servo Motors and Pivot	3 s	Yes	/	Yes	Yes	Yes	/	Torsion	Robotics and programmable matter
Knaian et al. [176]	Chain	1x1x1.5 cm	4*	$4.5e^{-4}$ Nm	Planar surface	14 g	Electricity	3D	Servo Motors and Pivot	5 s	Yes	/	Yes	Yes	Yes	/	Torsion	Robotics and programmable matter
Yim et al. [166]	Chain	1.3cm x cm x 0.1 mm	>30*	/	Planar surface	>0 [†]	Electricity	3D	Servo Motors and Pivot	15 s	Yes	/	/	/	/	/	Pulling and Folding	Robotics and programmable matter
Sung et al. [175]	Chain	>cm x cm x <0.5 mm	57	$4e^{-2}$ Nm	Hot water	>0 [†]	Heat and Magnetic field	3D	Smart Materials Magnetic steering	32 s	/	Yes	/	/	/	Yes	Self-folding	Robotics
Tibbitts et al. [168]	Chain	>cm x 6.4Ø mm	63*	/	Hot water	>0 [†]	Heat	3D	Smart Materials	>40 mn	/	Yes	Yes	/	/	Yes	Self-bending	Programmable matter
Uchida et al. [28]	String	>cm x Ø1 mm	/	/	Hot water	~0 [†]	Heat	3D	Smart Materials	/	/	Yes	Yes	/	/	Yes	Self-bending	Medical
Wang et al. [128]	String	>cm x >Ø10 mm	/	/	Hot air or water	>0 [†]	Heat	3D	Smart Materials	>60s	/	Yes	/	Yes	/	Yes	Self-bending and Torsion	Programmable matter
Chapter 5	Ribbon	136x5x0.5 mm	6*	$2.8e^{-6}$ Nm	Planar surface	>0	Heat via Magnetic field	3D	Smart Materials induction heating	275 s	Yes	Yes	/	Yes	/	Yes	Self-folding	Programmable matter

Fig. 5.17 Comparison of the literature on self-folding 1D structures and the characteristics of the ribbon developed in this chapter. The asterisc in the module number column depicts the 1D structures' number of module demonstrated. The cross in the torque and mass in medium correspond to calculated values inferred from the information within the respective studies.

As mentioned in Chapter 2, the current challenges of self-folding 1D structures include tether-less systems, remote folding, sequential folding, reversability of the actuation and reprogrammability of the 1D structure (Figure 5.17).

In this chapter, the ribbon's folding is enabled remotely and without tether by relying on magnetic induction. This provides independancy of the self-folding structures and potential robots from the environment direct influence. While other studies have successfully achieved remote heating using magnetic fields [130], their methods typically involve embedding micro- or nanoparticles within smart material matrices, which generate heat through hysteresis heating. Hysteresis heating occurs in ferromagnetic materials when an oscillating magnetic field changes the magnetization of the material's crystals, causing friction and heat at their boundaries. In contrast, magnetic induction generates currents within a magnetically permeable plate due to the oscillating magnetic field, resulting in Joule heating of the plate. Due to the reduced particle size, hysteresis heating requires high working frequency ($>250\text{ kHz}$) [224], high magnetic field amplitude [225] resulting in high power (kW) flowing through the induction coil, thus the need for water-cooling. In addition, magnetic field with a working frequency beyond 100 kHz starts heating human tissue and prolonged exposure can cause hyperthermia, a state at which cell dies due to excessive temerature. Consequently, magnetic field with high frequency are not deemed safe for prolonged exposure, although the hyperthermia caused at these frequencies is utilised in the targeted treatment of cancerous tissues [226]. The only study that denotes from other hysteresis heating approaches is the work of Razzaq et al. [16]. In their study, a particle dopped shape memory polymer was bent in several minutes using hysteresis heating at 50 Hz by encasing it inside a bell under vacuum to reduce the heat loss to a minimum, which reduces drastically the field of application. In contrast our study relies on a relatively low frequency and low magnetic field compared to other methods relying on hysteresis heating, but in spite of these, enable a fast folding of structures above millimeter scale. It is noteworthy to mention that the frequency used here can be further reduced by increasing the magnetic material thickness and the magnetic permeability without amending the magnetic induction system or design of the structures.

Regarding sequential folding and addressability, the developed ribbon and structures heat up at different rate owing to design and structure alterations. To my knowledge, this approach has not been proposed in other studies, however sequential self-folding using magnetic induction to relay power between transmisting and receiving coils was achieved [86]. One such example is that of Zhu et al. [21], who embedded receiving coils within a composite paper/SMA actuators and triggered their activation selectively by tuning the resonance frequency of each coil, and with it the power received. While these systems are fonctionnal, they require the integration of electronic components directly within the

structure and require the actuator to be part of the electric circuit. Although based on the application, this implies that the electronic components require an encasing to protect them from the environment, and that the actuator is electrically conductive which limits drastically the range of possible smart material that can be used. Here the method proposed is more universal as it would require any thermo-responsive smart material to be tucked in between two metal receivers.

Compared to the approach in Chapter 3 which made the PVC reprogrammable owing to the modularity of the string, and reversible due to the hinge design, here the ribbon is neither reprogrammable or reversible. This is due to lack of modules along the string which is inherent to the unique-ness of knots and their ability to change length in addition to topology. Moreover, the thinness of the materials involved in this chapter makes the manual reprogramming challenging as the subsequent structures are fragile and the glue utilised is not resilient enough to prevent the delamination during the process.

5.4.2 Stalling angle and modelling

The aspect of the stalling angle and subsequent modelling to prevent premature stoppage of folding hinges have only been partially studied. More experiments and simulations are required to link all key parameters together to predict the size of the self-foldable region and the exact range of the stalling angle between which the hinge does not receive enough heat to continue its folding. Currently, the fabrication of the overhand knot requires approximately three hours and, unlike the method in Chapter 3, the PVC cannot be reset to its initial position easily, thus making mismanufacturing costly. A complete model could help design intricate shapes with numerous folding sequences and ensure their foldability. However, the model combines several physics with numerous empirical values that can easily differ depending on the make of the materials and quality of the induction coil. Consequently, the model may inaccurately predict the self-folding of the hinges and the kinematics of self-folding on wider structures.

5.4.3 Sequential folding

In this chapter, sequential folding was achieved by altering the design and size of the receivers, as well as the configuration of the layers. The tuning of the receiver size was only demonstrated on 1D elements, e.g. the subleaves of the *mimosa pudica*; however, the potential of this approach should be extended to origami patterns, which could be achieved by splitting a given receiver into smaller sections. Although, the folding speed of receivers is highly influenced by their size, the fabrication of such structure is challenging, either related

to the alignment of the receivers or the delicacy of manipulation of the structure especially while detaching it from the sticky mattress. In addition, as the glueing surface is reduced, the shrinkage of the PVC and collision between the receivers increase the risk of delamination, thus inducing self-folding failure.

Fabricating self-folding structures presents challenges, particularly in the precise positioning and alignment of top and bottom receivers and thermal insulators, which leads to incorrect folding angles and sequences. While the blanket configuration helps align the receivers, its applicability is limited to certain patterns. For instance, in the crease pattern of the box in Figure 5.18 (a), the central receiver cannot use the blanket method and must be manually positioned. However, modifying the crease pattern in Figure 5.18 (b) allows all hinges to use the blanket configuration. Although both patterns result in the same box structure, the axis of rotation for the amended receiver 3 depends on its attachment to receiver 4 (Figure 5.18 (c)). Assuming that the central receiver aligns with the coil center, reduced heat generation from increased distance and double rotation between receivers 4/1 and 3/4 causes receiver 3's coupling to decrease rapidly. Consequently, receiver 4 folds first, bringing receiver 3 perpendicular to the coil and stopping its heat generation. This highlights the limitations of the blanket configuration and crease pattern design for planar self-folding structures. To circumvent this limitation, a tri-layer configuration at the center can be introduced while adjusting the hollowness of the copper receivers to compensate for the heat rate discrepancy between tri-layer and blanket configurations.

5.4.4 Scaling

Compared to other Chapters, the workspace using induction is even more limited as the hinges require a certain amount of heat to fold, which can only be supplied in a region of the induction coil. So, unlike the other Chapters, where the ratio between the self-foldable region S_{SF} and size of the workspace S_W is equal to one, here, the self-foldable region is smaller and the ratio is less than one. A possible path forward could be to bring this ratio closer to one using induction without necessarily making the system more powerful. Such a method would increase the heat generated in the centre of the coil, and unless the hinges are further miniaturised, the receivers could reach temperatures that would evaporate the PVC or burn the surface of the setup. A potential approach could be to change the smart material with a lower activation temperature or a smart material that does not release harmful gases.

In addition, given the limited working space available, it is important to ensure that all the structure hinges enter the self-folding region. There are two ways of achieving this: either the structure contracts as it folds, allowing hinges located outside the self-folding region to enter the self-folding region, or the partial self-folding of the structure allows sufficient navigation,

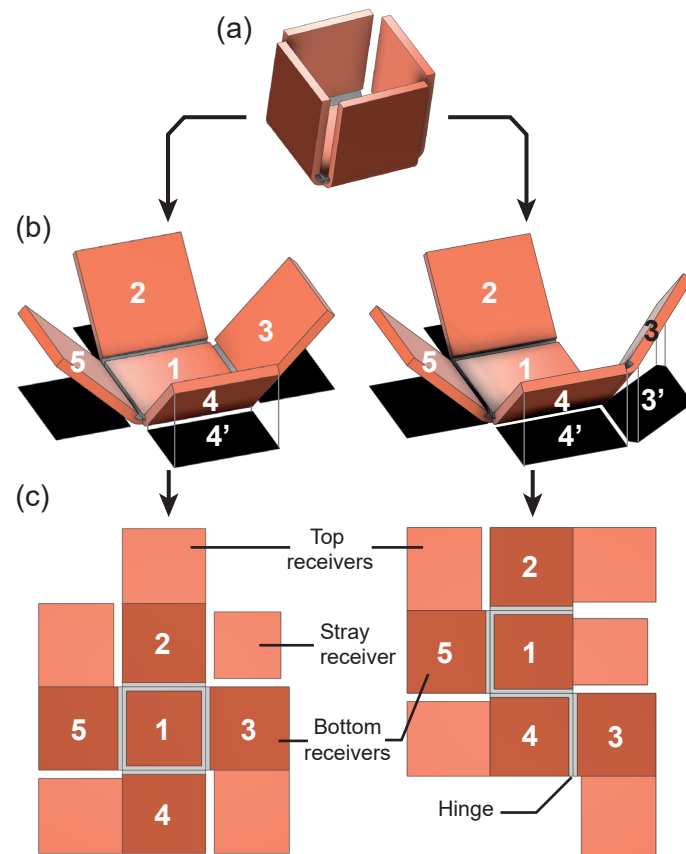


Fig. 5.18 Limitation of the trilayer-blanket configuration. The target box structure (a) has different possible folding pattern (b) (with numbered bottom panels in dark orange and top panels in light orange) which gives a folding configuration (c) with projected surface of induction in black.

for example with the aid of a magnet, so that the structure can be locomoted and all hinges introduced into the self-folding region. This way, the need for a self-foldability model is reduced as the existence and optimisation of a self-foldable region should be prioritised.

5.4.5 Amorphous structures

Unlike other crystalline or semi-crystalline structures, which require a precise bending angle to form the target structure, the bending precision of amorphous structures is lower. As long as the knot's key features (loop and helices) are formed and nested correctly by the folding sequence, folding the hinges beyond their angle should not affect the topology of the knot and would only tighten it further.

An important aspect of knots not mentioned in this chapter is the length of the knot ends that the hinges must allow to pass through the loop. The solution to this problem depends on the length of the knot ends and their stiffness. Assuming that the knot ends are stiff, hinges must be introduced to ensure kinematics that allow passage through the loop. In the case of a soft string, the methodology proposed in Section 5.2.3 already allows the knot end to pass through. Although the knot end will bend due to collision with the workspace surface, adding out-of-plane angled hinges to swirl around the loop edges should force the knot end to pass through the loop regardless of its length.

Polymer chains have several knots along their length, and other knots may appear in their structures. Here, only the overhand knot was considered in the simulation and demonstration. The methodology and implementation should be extended to other knots and longer knotting chains. Although collisions prevent the formation of the targeted structure, polymers are intrinsically stochastic in their formation. Therefore, randomising the direction of hinges along a string could allow a string to form knots naturally, or the simultaneous self-folding of several randomised strings could produce larger, more complex structures. Another, more deterministic approach to obtaining such structures might be to make a determined self-folding knot along several strings that could serve as entanglement points for more random strings. However, the main limiting factors would be the size of the self-folding region and the miniaturisation of the hinges, as well as the torque dispensable by the PVC.

5.5 Conclusion

In this chapter, by modifying the design of the hinges, the configuration of the layers and the size of the magnetic receivers, a method is presented to enable induction to sequentially self-fold origami and strings. A manufacturing process has been developed to ensure optimum,

reproducible fabrication of the structures. In addition, the first steps of a model to predict the folding sequence as a function of design parameters were proposed. Based on the thermal test and the folding speed test, sequential folding was demonstrated by the self-folding of a box and a leaf. To extend the test and demonstrate the level of control over the folding sequence, a methodology for translating knots from continuous into discontinuous deformations was proposed. Following the development of self-folding in-plane hinges, the methodology enabled self-folding of an overhand knot requiring 3 of folding sequences with limited assistance. Finally, the possibility of self-tightening a knot was investigated by self-shrinking a PVC strip that was applied to close a gap inside a soft membrane.

The following key research questions highlighted in chapter 2 were approached in this chapter:

1. How can untethered strings be self-folded in a reproducible manner?
2. How can the concept of Hamiltonian Path be applied to amorphous structures, and how can it be extended to other arrangements?
3. Amorphous structures being born of disorder, is the folding accuracy and sequence as crucial as for highly ordered arrangement such as crystalline or semi-crystalline structures?

The concept of amorphous structures relies on the random collisions and entanglements of a molecular chain, with other chains or with itself, creating structures susceptible of further topological changes upon thermal excitation. Here, the addressability and sequential folding is aimed at preventing collisions and entanglements which is opposite to the very concept of amorphous structures. As per the definition, amorphous structures would resemble more the "failed" self-folding attempts of a magnetic string demonstrated, where magnets misalign with each other and create a mesh instead of the well-defined structure initially envisioned by the designer [166]. This example illustrates the meta-stable nature of self-folding strings where any misfolding of the hinges creates a discrepancy between what is and what should be but remain relevant at a structural level. Naturally, the aim of a fabrication method is to obtain an output with the least number of steps and maximum possible success rate. Undenying the necessity of the efficiency of a fabrication process to near 100% success, the main challenge for strings self-folding into amorphous structure is the reproducibility of the final shape and its mechanical properties despite the randomness of the folding/assembly process. This can only be achieved with the well controlled folding sequence achieved in this chapter to propose a specific folding motion and avoid unplanned collisions.

In the cases of crystalline and semi-crystalline structures, the Hamiltonian path is well defined, whether filling unit cell by unit cell of the space lattice or zig/zag-ing through it.

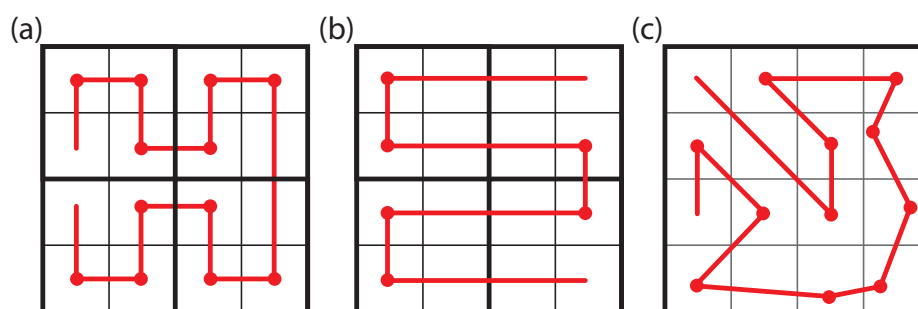


Fig. 5.19 Comparison of the the Hamiltonian Path that can be obtained for (a) crystalline structures, (b) semi-crystalline structures and (c) amorphous structures on a 2D pattern.

These crystalline structures have a repeating unit with limited possible directions and folding angle. For example, the crystalline arrangement in Figure 5.20 (a) can go in four directions and fold $\pm 90^\circ$. However, defining the Hamiltonian Path of an amorphous structure is more complex as the path taken to fill the shape ignores the lattice order. Accounting for the designer perspective where the main output is the shape, realising the Hamiltonian Path of an amorphous structure requires a string with a high degree of adaptability regarding the folding angles, length of segment and folding sequence, hence giving a high number of possibilities for the Hamiltonian Path to form the desired shape. In Figure 5.20, the paths illustrated are only in 2D, and therefore does not exhibit any entanglements seen in braids, knitting or knots e.g. overhand knot or figure 8 [176] [81]. Overall, this justifies the approach taken in this chapter where the folding sequence can be addressed by altering the design and structure of the hinges to create knots for the purpose of locking string-based structure similarly to stoppers on climbing ropes. Extending the concept of knots to crystalline structures, it is possible to take the method to translate continuous knots into discontinuous knots and apply it to create crystalline knots. Similar to stopper on climbing ropes, these knots would ensure the structure integrity, tightening and rigidifying the structure by increasing the inter-module friction, or bringing novel methods for anchoring robotic systems using the cow's hitch for example. Such concepts could be utilised for swarm of robotic worms to obtain specific functions or shapes.

As mentioned before, crystalline and semi-crystalline structures exhibit tight requirements such as a constant folding angle with a deviation as small as possible and a high controllability over the folding sequence to prevent collisions. In the case of amorphous structures, the requirements needs to be adjusted for the change of concept. The folding pattern and folding rate (in $^\circ/s$ remains important, however the folding accuracy mattered less. The main reasons lies in the topology of the knot. Once a similar topology has been achieved, folding further the hinges result only in a tighter knot. Taking the overhand knot as an example, once the loop

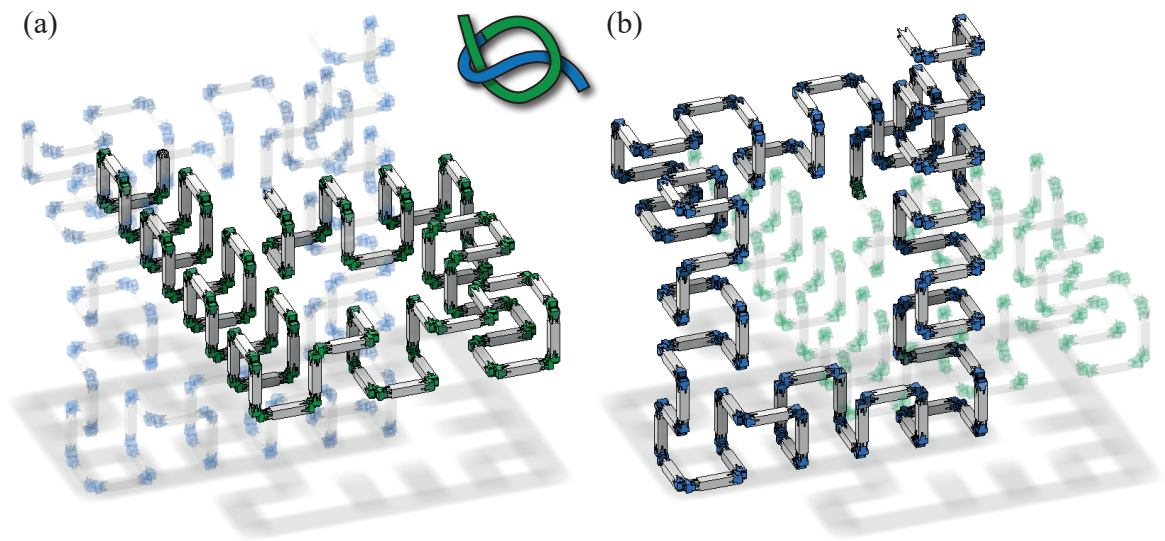


Fig. 5.20 Illustration of the crystalline overhand knot. (a) represents the loop in green of the knot and (b) the knot's helix in blue.

is formed and the helix started passing through it, folding further the helix will only tighten tighten the knot. Obtaining such a physical change on a crystalline structure would require to change the length of the segments of the string rather than altering the folding angle of the hinges. It is this discrepancy that illustrates the difference in nature of the two approaches to build up matter and result in stark differences between the folding requirements.

Chapter 6

Conclusions and future work

This section concludes the work presented in this thesis by discussing the results and proposing future work directions.

6.1 Discussions

Reference	Type of string	Module Size	Module number	Torque	Medium	Mass in medium	Power Source	Folding in	Actuation Method	Assembly time	Remote folding	Untethered	Reversibility	Addressability	Reprogrammability	Sensing	Assembly and Deformation	Application field(s)
Yim et al. [167]	Chain	3x3x3 mm	999*	$3.2e^{-2}$ Nm [†]	Guide exit	36 g	Magnets	3D	Magnetic pull/push	108 s	Yes	/	Yes	/	/	/	Self-assembly and self-folding	Medical implants
Fass et al. [189]	Chain	20x20x10 mm	6	$2.5e^{-1}$ Nm [†]	Planar surface	$>>0^†$	Magnets	2D	Magnetic pull/push	0.2 s	Yes	Yes	Yes	/	/	/	Self-assembly and self-folding	Minimally invasive surgery
Griffith et al. [172]	Chain	25x25x3 mm	20*	$2.1e^{-1}$ Nm [†]	Air cushion	$\sim 0^†$	Magnets + Air	2D	Magnetic pull/push onto air cushion	30 s	Yes	/	Yes	/	/	/	Self-assembly and self-folding	Programmable matter
Han et al. [43]	Chain	14Øx9 mm	20*	$2.9e^{-2}$ Nm [†]	Agitated hot water	$\sim 0^†$	Magnets + Air + Heat	2D	Magnetic pull/push into bubbling water	10 s	Yes and No	Yes	Yes	/	/	Yes	Self-assembly and stretching	Programmable matter
Gu et al. [185]	Chain	15x5x5 mm	9*	$3.4e^{-3}$ Nm	Guide exit	$>0^†$	Magnets	3D	Magnetic pull/push	13 s	Yes	/	Yes	/	/	/	Self-assembly and self-folding	Minimally invasive surgery
Li et al. [7]	Ribbon	4x5x0.5 mm	5	$1.42e^{-2}$ Nm [†]	Planar surface	320 mg [†]	Magnets	2D	Magnetic push/pull	/	Yes	Yes	Yes	/	/	/	Self-folding	Robotics and programmable matter
Ijäs et al. [194]	Chain	3x2x2 nm	8064	/	Solution	~ 0	Atoms	3D	Atomic forces	/	Yes	Yes	/	/	/	Yes	Self-assembly and self-folding	Drug delivery
Douglas et al. [195]	Chain	3x2x2 nm	7308	/	Solution	~ 0	Atoms	3D	Atomic forces	12h	Yes	Yes	/	/	/	Yes	Self-assembly and self-folding	Drug delivery
Bai et al. [191]	Chain	3x2x2 nm	7249	/	Solution	~ 0	Atoms	3D	Atomic forces	12h	Yes	Yes	/	/	/	/	Self-assembly	Programmable matter
Cheung et al. [173]	Chain	77x38x38 mm	7*	1.5 Nm	Planar surface	>371 g	Electricity	3D	Servo Motors and Pivot	3 s	Yes	/	Yes	Yes	Yes	/	Torsion	Robotics and programmable matter
Knaian et al. [176]	Chain	1x1x1.5 cm	4*	$4.5e^{-4}$ Nm	Planar surface	14 g	Electricity	3D	Servo Motors and Pivot	5 s	Yes	/	Yes	Yes	Yes	/	Torsion	Robotics and programmable matter
Yim et al. [166]	Chain	1.3cm x cm x 0.1 mm	$>30^*$	/	Planar surface	$>0^†$	Electricity	3D	Servo Motors and Pivot	15 s	Yes	/	/	/	/	/	Pulling and Folding	Robotics and programmable matter
Sung et al. [175]	Chain	$>cm$ x cm x <0.5 mm	57	$4e^{-2}$ Nm	Hot water	$>0^†$	Heat and Magnetic field	3D	Smart Materials Magnetic steering	32 s	/	Yes	/	/	/	Yes	Self-folding	Robotics
Tibbitts et al. [168]	Chain	$>cm$ x 6.4Ø mm	63*	/	Hot water	$>0^†$	Heat	3D	Smart Materials	>40 mn	/	Yes	Yes	/	/	Yes	Self-bending	Programmable matter
Uchida et al. [28]	String	$>cm$ x Ø1 mm	/	/	Hot water	$\sim 0^†$	Heat	3D	Smart Materials	/	/	Yes	Yes	/	/	Yes	Self-bending	Medical
Wang et al. [128]	String	$>cm$ x $>Ø10$ mm	/	/	Hot air or water	$>0^†$	Heat	3D	Smart Materials	$>60s$	/	Yes	/	Yes	/	Yes	Self-bending and Torsion	Programmable matter
Chapter 3	Chain	30x5x5 mm	6*	$1.4e^{-5}$ Nm	Hot water	~ 0	Heat	3D	Smart Materials	4 s	/	Yes	Yes	/	Yes	Yes	Self-folding	Metamaterials and Programmable matter
Chapter 4	Ribbon	122x3x1 mm	7	/	Hot water	~ 0	Heat and Magnetic field	2D	Smart Materials magnetic steering	41 s	/	Yes	Yes and No	Yes	/	Yes	Self-bending	Robotics and programmable matter
Chapter 5	Ribbon	136x5x0.5 mm	6*	$2.8e^{-6}$ Nm	Planar surface	>0	Heat via Magnetic field	3D	Smart Materials induction heating	275 s	Yes	Yes	/	Yes	/	Yes	Self-folding	Programmable matter

Fig. 6.1 Comparison of the literature on self-folding 1D structures and the characteristics of the 1D structure developed in this thesis. The asterisc in the module number column depicts the 1D structures' number of module demonstrated. The cross in the torque and mass in medium correspond to calculated values inferred from the information within the respective studies.

6.1.1 Challenges of robotics and 1D structures

In this subsection, the challenges of robotics and 1D structures that were presented in section 2 are discussed under the light of the contributions of the technical chapters of this thesis.

In the recent years, few scientific studies were published about self-folding 1D structures. Their technical challenges includes the difficulty of self-folding such structures and their lack of structural integrity once folded. From a physical point of view, these challenges are highlighted by the required self-folding torque which is exarcebated by the friction with the environment [193] and the 1D structure design and length [180]. The structural integrity is showcased by the stiffness of the hinges along with the structure self-load on its hinges and external load that may be applied. Due to these two technical challenges, 1D structures larger than a centimetre have received limited attention, therefore 1D structures have seen limited development, proposal for new design and applications. Regarding the range of designs of 1D structures, they can be organised with their similarities with crystalline, semi-crystalline and amorphous structures. Based on Figure 6.1 and due to their well-defined shape, crystalline structures are the most common approach, regardless of the folding technique or environment. To date, most crystalline structures are limited to cuboid and thus do not necessarily require complex algorithms, thus explaining their popularity. On the opposite, 1D structures based on semi-crystalline structures are less popular and have been reported in only three studies. One of the notable difference to crystalline structure is that semi-crystalline structures can output complex shapes more easily, but require complex algorithms to conceptualise most shapes. Finally, due to the complex and inter-twinning topologies, amorphous structures are not reported in the literature so far owing to their complex and empirical nature.

Owing to the development and usage of alleviating environments such as air cushion tables [172], water tanks [43] [169] or free fall methods [179], the self-folding of 1D structures was enabled. As seen in the work of Tibbits [169] and his lab, crystalline structures were self-folded using hydrogel actuators in a water tank, however the subsequent folding lasted for around 40 mn and, due to the 3D printed nature of the string, the structure could not be reprogrammed. Here, the contribution of chapter 3 is the design of reprogrammable chain able to utilise neutral buoyancy to alleviate and negate gravity to fold large structures. This development overcome the 2D self-folding limitation of cushion tables and flat water tanks [43], while providing a long lasting alleviation of required torques due to the slow cooling of the hot water within the tank. As a result, the folding of the 1D structure does not require fast motion to fold completely, thus enlarging the range of foldable structures.

In regards to semi-crystalline structures, the current studies consist of the work of Yim et al. where a sliced structure was folded and actuated by pulling strings [167], the work

of Sung et al. where the folding of semi-crystalline structures was achieved using smart materials, and the work of Li et al. [7], who self-folded magnetic composites into halbach arrays. Of all these self-folding, none is shown to be reversible in the literature, hence the contribution of chapter 4 in which, hydrogel hinges enabled the structure to fold between straight and folded 1D structures. In addition, the embedment of the hydrogel onto a shape memory polymer allowed the structure to lock itself in the folded position, which resulted in a structure with variable stiffness, memory function and the firm prehension for robotic purposes. This behaviour is neither proper to hydrogels nor shape memory polymers, hence the novelty of the approach. This enables semi-crystalline structures for robotic applications that requires varying topology as depicted in other studies such as the work of Miyashita et al. [23] who demonstrated the relation between topology and functionality in origami robots.

Most of research regarding 1D structures and amorphous arrangement relates to knot theory [227], weaving/knitting topologies and enabled functionalities or smart sutures able to self-tighten but unable to self-knot. All these structures are assembled by hand and the self-knotting to lock the topology in place is not demonstrated in the literature by self-folding or self-bending. This showcases the contribution of chapter 5 where a methodology is proposed to design overhand knot and its self-folding is achieved. Consequently, the work presented here breaches the design limitation to obtain new topologies by using addressible self-folding which is shown in few studies using 1D structures based on smart materials.

6.1.2 Robustness and folding accuracy

Similar to Christmas lights, where one defective diode leads to all following diodes turning off due to circuit breakage; the main disadvantage of a self-folding string lies in its serial nature. If one of the hinges is defective, the entire structure is directly affected, causing it to be misformed, its structural properties to be altered, and potentially unemployable. The serial nature and propagation of errors in a crystalline structure is highlighted by the following equation but is conceptually translatable to both semi-crystalline and amorphous structures:

$$p_i = p_{i-1} + L_i \begin{bmatrix} \cos(\theta_{i-1}) + \cos(\alpha_{i-1}) \\ \sin(\theta_{i-1}) \\ \sin(\alpha_{i-1}) \end{bmatrix} \quad (6.1)$$

where i is the index indicating the hinge, i is its position, L_i is the length of the segment $i + 1, i$, and θ and α are the folding angles in the xy- and xz-planes, respectively. However, even under perfect conditions, the hinges are susceptible to the load applied and the overall torque they perceive. Regardless of the extent of the load, the hinges will always deform when loaded according to Equation 3.5 presented in Chapter 3. The resulting deformation

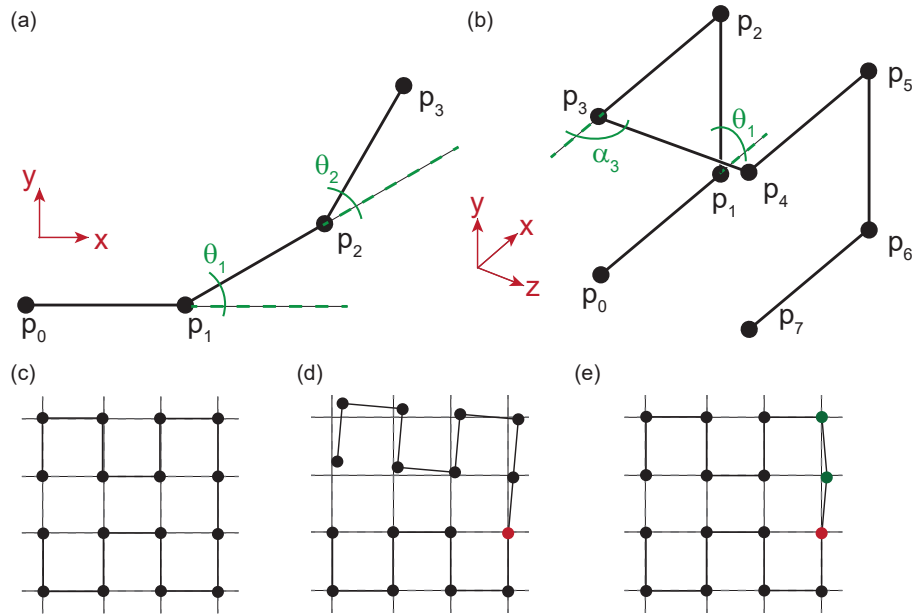


Fig. 6.2 Illustration of the position of the folding hinges of a string in 2D (a) and 3D (b). A cubic crystalline structure ideally arranged (c), with one hinge misfolding (d), and corrected (e).

affects the entire structure, as shown in Figure 3.15. This implies that the hinges of a self-folding string should be carefully designed to resist extensive loading or that the environment in which the string is placed should be well understood to have the structure perform as intended. Reversing the previous argument, the required stiffness for a hinge should be scaled with the size of the structure, meaning that a given hinge design is suitable for a certain size of the structure. Solutions can be found to improve the hinge performance and structure accuracy. The first solution could be to optimise the selection of the Hamiltonian path to reduce the number of hinges or the torque they receive. A second possibility is to search for Hamiltonian circuits instead of Hamiltonian paths. Where the Hamiltonian path has its start and end far from each other, the Hamiltonian circuit has neighbouring start and end. If connected together, the structure can resist two breakages before splitting into two distinct structures. In addition, upon connection of the start and end of the string before folding would allow the hinges to be partially controlled by their surrounding hinges, thus increasing the success rate. A third possibility could be to mimic more atomic and molecular interactions, where the string elements interact with each other chemically and mechanically. In my thesis, the elements only interacted mechanically, but implementing methods for the elements to bond together would improve the overall stiffness and resilience of the structure. Finally, provided the string could be controlled externally or the string was

capable of proprioception, a set of two hinges could compensate for the inaccuracy in a 2D or 3D structure (see Fig. 6.2 (d) and (e)), turning the disadvantage of the serial nature into a feature to correct position errors. However, the inaccuracy compensation would require each hinge to be addressable for independent control and a feedback loop to be implemented within the string, which would require on-board computation.

6.1.3 Scalability and size of the workspace

With an increasing number of hinges and string length, the following challenges are raised: folding torque, resistance of the hinges (or connections), folding sequence, collisions during self-folding, and size of the environment. In Chapter 3, I proposed a solution to the challenge of folding torque by allowing the string to experience micro-gravity using neutral buoyancy. Theoretically, in this way, an infinitely long string can self-fold when given an infinitesimal folding torque. However, this solution, similar to other self-folding strings, is limited by the size of the environment supplying the energy required to fold. This raises the challenge of the size of the environment compared with that of the string. In Chapter 5, the edge of the string received a limited amount of energy from the induction coil; however, the string could fold into a knot. One could draw a parallel between this challenge and spaghetti in a pan. Once immersed in boiling water, the spaghetti softens and the edge out of the pan enters the water. As the structure self-folds, the string agglomerates and it gradually enters the workspace where they can fold. The main condition for this to occur is to have the string anchored at the centre of the workspace, or to have the centre of mass of the string aligned on the centre of the workspace, so that the string self-folds around it. Thus, the self-folding of a string could be functional regardless of the global or local nature of the stimulus used to trigger the smart material, highlighting the versatility of the folding mechanism for self-folding strings.

Depending on the structure type, the volume of the structure and the length of the string scales differently. In the case of crystalline arrangement, the length of the string L_{tot} scales linearly with the volume of the target structure V_{tot} providing the crystal lattice is not missing a crystal. In the case of semi-crystalline arrangement, the length of the string $L_{sm_{tot}}$ is split into two sections: $Lp_{i-1,i}$ the length of a slice and $Lh_{i-1,i}$ the length of a hinge. In addition, the height between two of the n slices in the structure is noted h . The total length can therefore be expressed as $L_{sm_{tot}} = nLp_{i-1,i} + (n-1)Lh_{i-1,i}$. In the case of amorphous structure, if one considers the string has zero volume and the ends of the string are free, the total volume of the knot Va_{tot} is function of the tightening of the knots of the string. Consequently, the ratio $L_{a_{tot}}/Va_{tot}$ could be as high as the length of the string $L_{a_{tot}}$ allows or as low as the thickness and rigidity of the string allows. Therefore, compared to other structure configurations, amorphous structure has the potential to be the

most compact, which increases its resilience owing to the friction within the knot as the knot is tightened/compacted. This relates to scalability of the string, as a longer string could be folded if it is for an amorphous arrangement rather than a crystalline or semi-crystalline arrangement.

6.1.4 Scalability and complexity

In addition to the workspace size, folding inaccuracies and increase in the folding torque required, scaling up the number of hinges increases the string complexity. Scaling up highlights one of the main challenges of self-folding objects: collisions. In the case of a crystalline structure with cubic voxels, the string possesses two degrees of freedom (one rotation along two Cartesian axes); however, in the case of an amorphous structure, the string can have as many degrees-of-freedom as there are hinges. Instinctively, collisions within a crystalline structure can be predicted, or at least the potential location of collisions can be predicted. An intuitive example of this statement is that any hinge folding towards an already formed unit cell potentially generates a collision. In the case of an amorphous structure, the problem is much more difficult to imagine and one should be aided by CAD software. However, even if the complexity of the string increases with the number of hinges, there is a stark difference between crystalline and amorphous structures. Where crystalline structures are highly ordered and the position of the voxels is crucial, amorphous structures do not require as much folding accuracy, but rather in the folding sequence. Providing the helix and loop are folded to their minimum folding angle to be formed, further folding should only tighten and secure the knot. Thus, the requirement on the hinges differs from one structure type to another. In contrast to the amorphous and crystalline structures, the semi-crystalline structures do not exhibit any changes in complexity, as the hinges have only one degree-of-freedom and the layering within is not prone to collisions due to the parallel stacking. Consequently, the complexity remains constant regardless of the scale. Overall, one can draw a general rule linking the scaling and complexity of a self-folding string based on the total number of degrees-of-freedom of the string.

6.1.5 Physical properties and Hamiltonian path

The resistance of a material can be understood as the bonding force between molecules and crystals, in addition to the friction between them. This is reflected by the Hamiltonian path within a string-based structure through unit cell/unit-cell interactions. However, Hamiltonian paths are NP-complete problems, meaning that one must manually search for paths. To obtain the target structural characteristics, manual investigation would be inefficient, which

would justify the development of an algorithm searching for the Hamiltonian path that fulfils the following requirements:

- Propose a valid path,
- Avoid collisions while self-folding,
- Correct output physical properties,
- Improve structural strength and robustness to breakage.

Coupled with the compactness of the final structure, several strings could be self-folded and manually assembled or self-assembled to obtain a structure with reinforced physical properties. This could be achieved with structures with similar or different configurations. For example, a crystalline structure should serve as a base for an amorphous or semi-crystalline configuration to branch, entangle and nest within. Interlocking configurations could yield structures with reinforced robustness owing to unit-cell interactions. Thus, the Hamiltonian path can be optimised to enhance the characteristics of the final structure. In contrast to crystalline networks, semi-crystalline and amorphous networks exhibit only a single path outputting the desired shape or knot, rendering the problem of the Hamiltonian path specific to crystalline networks. However, for amorphous networks, allowing an algorithm to freely investigate Hamiltonian paths for knots can lead to new knots or classes of knots, yielding increased stability, instability, or functions. Alternatively, the implementation of knot aspects and theories into crystalline structures could produce crystalline knots. This could eventually be implemented to allow the string within the structure to lock by protecting its ends from environmental interactions.

6.2 Conclusion

In my thesis, I have presented different approaches for self-folding thermo-responsive chains and ribbons to replicate the crystalline, semi-crystalline and amorphous structures seen in matter.

Along the thesis, several solutions have been proposed to address the challenges of self-folding structures. In Chapter 3, I developed a chain that self-folds into cubic crystals for crystalline structures. The reprogrammability of the modular chain was achieved owing to the new hinge design and methodology that enabled an SMP-actuated hinge to be reset and the chain to be reconfigured. I also proposed an environment with reduced gravity, thus decreasing the required folding torque, to increase the scale of self-folded chain structures.

In Chapter 4, I developed a chain that self-folds into a cubic lamellae for semi-crystalline structures. I proposed a fabrication process to self-fold reversibly and permanently turn off the reversibility of the chain. In Chapter 5, I developed a ribbon that self-folds into an overhand knot for amorphous structures. I proposed to remotely self-fold the ribbon using magnetic induction and address the hinges by tuning the geometry of the magnetic receivers.

6.3 Future work

Even though my approaches relate to some of the main challenges of self-folding structures, only the compilation of the various features of each of my approaches will result in an actual solution: a modular reprogrammable string reversibly and remotely self-folding into any 3D shape. Steps have already been made in this direction, but as of today, they have achieved limited success. The next step would be to introduce a technique or method to remotely self-fold underwater while maintaining addressability in 3D.

Strings have been widely applied across scales and fields. At the molecular level, life originates from the entanglement of carbonated chains forming DNA. On a centimetre to metre scale, strings are weaved together to form plants, clothes, and boat sails. Beyond hundreds of metre, strings are used to sustain bridges and connect homes with data and power.

A first step could scale up the number of hinges within the string, while scaling down the size of the elements of the string. Thus, the intricacy of the string would be increased while refining the resolution, and size of the structures that can be self-folded.

As the scaling would be covered, attraction and repulsion between the string's elements could be implemented to replicate molecular interactions. This would increase the robustness of the final structure but would also guide the self-folding and allow the string to be encoded. In this way, the folding of the string could replicate the formation of polymer chains that exhibit a plethora of properties owing to their shape and atoms. This would help chemists understand and explain the behaviour and formation of materials such as polymers or DNA in a visible manner. Ultimately, this could allow chemists to develop new materials owing to a new understanding of chain interactions.

Future work could also consist of several independent strings self-folding simultaneously or sequentially around each other, thus mixing self-assembly and self-folding to entangle and form more intricate and complex structures. Thus far, the study of self-folding strings has focused on single strands; however, in reality, several strands form a string and several strings form a rope. The conglomeration of strands resemble the inner structure of plants. Depending on the arrangement of the fibers, the plants obtain stiff or flexible properties by Bourmaud et al. [228], or even motion in the case of sunflowers (heliotropism) or mimosa pudica. On the artificial side, through a weaving process, soft strings are widely used for clothing and rigid strings are used to create fences. Thus, large 2D and 3D structures with specific properties can be fabricated using thin strings.

With an increase in the number of unit-cells and the functionalisation of self-folding strings, the development of an algorithm for predicting the mechanical properties of the final structure is required. In addition to the prevention of collisions and reinforcement of the

structure to breakage, this algorithm could find patterns in the path to soften or rigidify the structure. Applied to knots, this algorithm could find new types of knots and, perhaps, new functionalities.

Currently, the string does not possess any feedback (other than the change in buoyancy or temperature) to indicate the state of folding; however, be it the position of the hinges or the extent of folding of the hinges, sensing would grant the string proprioception and exteroception. Coupled with the addressability, this could be used to rectify the misfolding of the hinges and ensure that the intended mechanical properties are provided by the structure, as well as to understand how the structure reacts to external stimuli.

Such strings could potentially be applied to biomedical robotics to perform *in vivo* operations. Owing to the presence of smart materials, *in vivo* strings could form stents after detecting a certain biomarker, a change in pH, or a change in temperature. Another possibility could be to use robotic strings that are able to perform *in vivo* surgeries, such as self-suturing, which should be achieved by piercing the tissue, tying knots, and self-tightening to close wounds.

If applied in space, the main limitations of the required folding torque to actuate long strings would be cancelled due to micro gravity. As a result, long strings could be compacted to be shipped to space, where they would fold themselves into frames to help build space stations, and reduce the time astronauts would spend on space walks to assemble such structures.

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