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Smart Bioinspired Material-Based Actuators: Current Challenges and Prospects

Alejandro Palacios, Léon Chiriatti, Simon Poppinga, Thomas Speck, and Vincent Le Houérou*

This research review discusses several examples of plant movements, either depending on the direction of the triggering stimuli (tropisms) or not (nastic responses), which have served as inspiration to develop smart biomimetic actuators. In addition, it presents an overview of the multiple approaches for the development of autonomous actuators based on synthetic materials, as well as of their advantages and disadvantages, applicability, and limitations. The classification is based on structural and conformational characteristics (mono-, bi-, or multimaterial assemblies, their orientation, chemical structures, and geometrical configurations). Additionally, this review presents an alternative formulation and extension of the pioneering Timoshenko's model, which provides an understanding of the underlying mechanical principle of bilayer bending actuation. Finally, upscaled applications of this actuation principle are described, focusing mainly on biomimetic architecture. Attention is given to previously reported real-life applications based on bio-based materials and material systems. Furthermore, this review discusses the multiple challenges for synthetic materials when an upscaling perspective is intended. In this sense, key aspects such as time responsiveness and mechanical amplification, in terms of speed, displacement, and load-bearing capability, are discussed.

1. Introduction

Some plants exhibit movements upon external stimulation, which are either dependent or independent of the direction of the respective stimulus, known as tropistic and nastic responses, respectively.^[1] For instance, the leaves of *Mimosa pudica*, commonly known as the sensitive plant, exhibit a particular case of a mechanically triggered nastic leaf movement for herbivore defense. An opening and closing of ion channels, leading to osmotic gradients between so-called motor cells and entailing water flows inside the commonly named pulvinus structure, enables the leaves to execute such sudden hydraulic movements (Figure 1a).^[2] Due to the fact that the hydraulic motion is morphologically determined and independent of the direction of the touch or injury stimulus, it is termed nastic. The Venus flytrap, a carnivorous plant, is another fascinating example of nastic movement. Its traps can perform a snapping motion in a matter of few hundred milliseconds^[3,4] to capture its mostly arthropod prey (Figure 1b). Here, in addition to hydraulics as already described above for the sensitive plant, the release of stored elastic energy plays an important role by speed-boosting the actuation. Due to the fact that the response is unalterable and independent to the (direction of the) stimulus (i.e., touch by prey), it is also termed nastic motion.

The flower of the bird-of-paradise (*Strelitzia reginae*) is an example of extrinsically actuated nastic motion. The body weight of pollinating birds causes a perch-like petal structure to bend and open due to torsional buckling, thereby exposing the stamens and facilitating the transfer of pollen to the bird's feet^[5] (Figure 1c).


In contrast to the nastic hydraulic and hydraulic-elastic motions of the sensitive plant and Venus flytrap, which involve physiological sensory and actuation processes, nastic hygroscopic motions do not require such physiological processes and are solely driven by changes in environmental humidity.^[6] Probably the most popular example of such motion is the humidity-dependent opening and closing of pine cones, i.e., the woody female seed-bearing organ of pines. The humidity-responsiveness of the individual scales on the cone enables seed release under favorable conditions for wind dispersal, i.e., when

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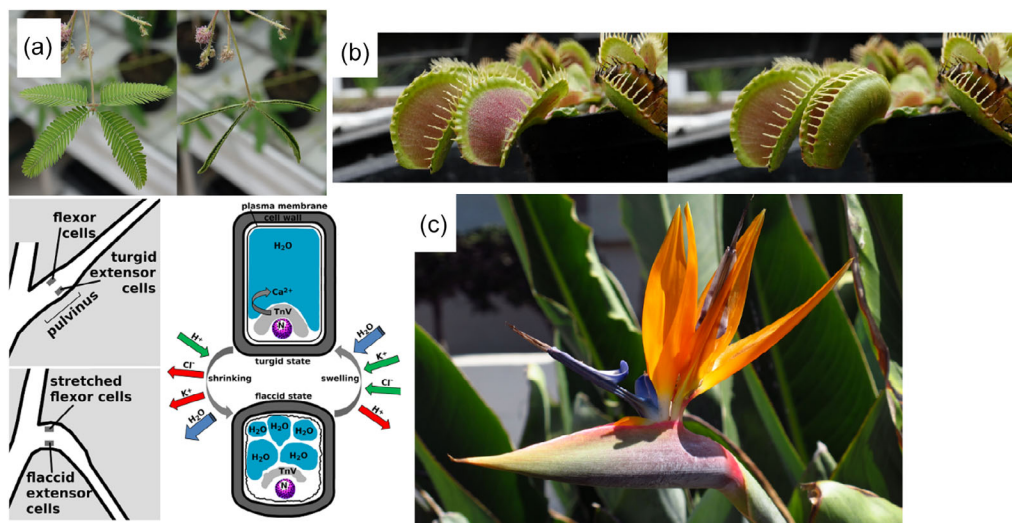


Figure 1. Examples of nastic movements in plants. a) The leaves of the sensitive plant (*Mimosa pudica*) react to mechanical stimuli (touch) with bending and folding motions, driven by metabolically costly hydraulics (i.e., changes of turgor pressure at the cellular level) (lower schematic images reproduced with permission.^[125] Copyright 2013, Wiley) b) The carnivorous Venus flytrap (*Dionaea muscipula*) is able to snap for prey in a matter of a few hundred milliseconds. The trap motion is driven by hydraulics (similar to the sensitive plant shown in (a) in combination with the release of stored elastic energy. c) The bird-of-paradise (*Strelitzia reginae*) develops a perch for pollinating birds, which, under the body weight of the animal visitor, bends downward and opens. This is an example of an extrinsically actuated motion.

it is dry, and is based on a mismatch in swelling of the different scale tissues.^[7–9]

Depending on the thickness of these tissues, hygroscopic actuation can be rather slow, taking, e.g., several minutes or even hours for a pine cone to open or close. The hygroscopic actuation of smaller structures like the involucre bracts of weather thistle flowers (*Carlina acaulis*) (Figure 2b) takes only a couple of minutes. Moreover, the bracts can undergo multiple

successive bending deformation processes, which is not fully understood yet.^[10]

In contrast to the nastic responses described above, tropisms depend on the direction of the respective stimulus.^[11] There are positive reactions, like positive phototropism also known as “sun tracking” in sunflowers^[12] (Figure 2c), and negative reactions, like negative phototropism also known as skototropism in English ivy shoots^[13] (Figure 2d). They are all based on

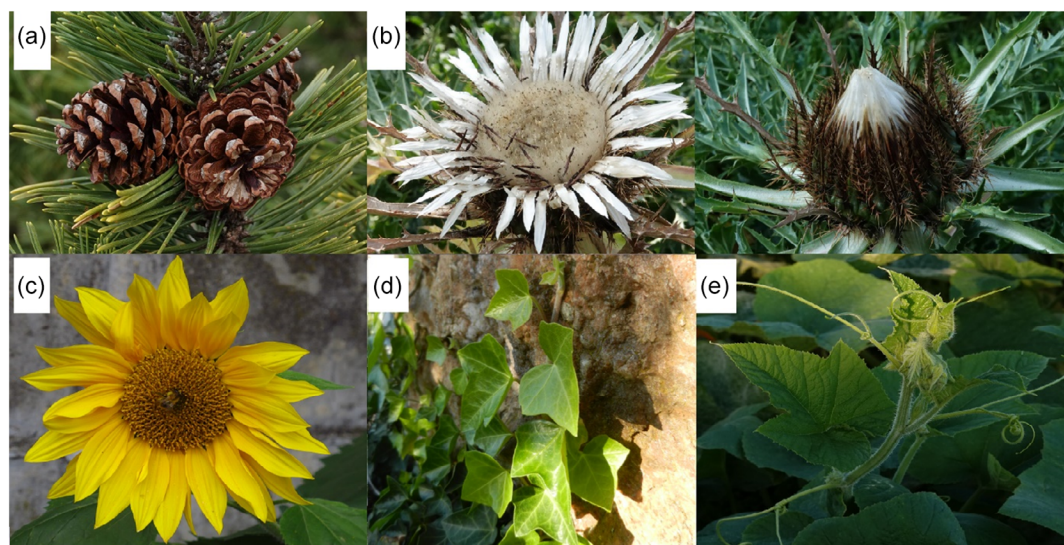


Figure 2. a) Open pine cones under low humidity, i.e., dry conditions. b) Open inflorescence of *Carlina acaulis* open at low relative humidity (RH) conditions (left image) and closed at high RH conditions (right image). c) Sunflowers (*Helianthus annuus*) exhibit positive phototropism. d) English ivy (*Hedera helix*) shoots grow actively away from light (negative phototropism). e) Autonomous movements of young tendrils in pumpkin (*Cucurbita pepo*).

irreversible growth processes. Finally, autonomous movements, such as movements of young tendrils or searcher twigs in climbing plants (Figure 2e), which “search” for a support structure for attachment, are not caused by external stimuli but are triggered autonomously by internal growth processes.

With such diversity in responses observed in plants, we present in the following section an exploration in the literature of bioinspired and especially plant-inspired actuators^[14] using synthetic materials with a special focus on the synthetic materials used and the actuation principles involved.

2. Elementary Actuators Based on Synthetic Materials

Similar to the tropistic and nastic movements found in nature, engineering materials can be tailored in different ways to exhibit a responsive behavior which yields a particular movement when exposed to an external stimulus. In the absence of any external (or extrinsic) actuation source except the one coming from the material itself, the approach is further referred to as material-based actuation: the motion emerges directly within the material. A first category comprises shape-memory materials (SMMs), which have the property of recovering a particular shape after being deformed under a load. This behavior is explained by the recoverable strain, damping capacity, or pseudo-elasticity of the material whose stiffness, position, deformation, or shape are affected by stimuli such as temperature, electricity, and magnetic fields.^[15,16] The three types of SMMs are metal–alloy-, ceramic- and polymer-based. In metal alloys, a reversible phase transition between two different lattice structures (martensite and austenite) occurs when changing the temperature, allowing the material to regain a specific shape when heated. In ceramics, the memory effect is achieved by blending mica minerals in a glassy matrix.^[15] The heating process under load generates a net strain as the amorphous phase expands. By cooling the material while applying the load, the structure remains deformed at low temperatures. When the material is heated again, the stress is released to recover the original shape. Additional types of ceramics include ferroelectric components to cause phase transitions when large electric fields are applied.^[15] Notably, this first type of SMMs is based on microscopic deformations of the material structure during fabrication.

Shape-memory polymers can be considered a separate category, given that they usually consist of a fixed phase and a reversible phase. For instance, block copolymers with soft and hard segments or cross-linked networks with fluid fillers can display reshaping when exposed to particular solvents, pH, ion concentration, or light wavelength variations.^[16]

Another elementary actuator type is directly inspired by plant movements based on turgor pressure changes due to osmotic alterations within so-called motor cells. In this respect, an analytical approach based on an idealized modeling of the osmotic processes occurring at the plant cellular level has been proposed^[17] and typical actuation times of few minutes and load capabilities of up to 20 N have been estimated. The significance of the relative size between the driving force structure (i.e., the osmotic membrane) and the energy storage structure (here, the bulging of an elastic membrane) has been highlighted. It is

interesting to note that this parameter appears somehow analogous to the surface-to-volume ratio, which is known to be a critical factor in the exchange between two media. The latter analytical approach was validated experimentally by manufacturing and testing a centimeter-scale model of the proposed osmotic actuator.^[18] This work, deliberately restricted to a single activation induced by a sodium chloride (NaCl) osmolyte, was further extended to reversible voltage-triggered actuation based on ions electrosorption.^[19]

The versatility of polymers has led to exploring another type of reaction to external stimuli that does not necessarily imply recovering a specific form. Instead, in recent years, new approaches have proposed the concept of self-evolving deformations. Here, the material contracts or expands in a predetermined direction to produce a specific movement under the effect of a particular stimulus. For instance, this notion has been applied to self-deforming materials that bend or stretch when submitted to light.^[20] More complex designs have used 3D printing to produce pre-calibrated hinge-like structures in the form of grids that induce hyperbolic surfaces.^[21] Here, actuation is not tuned at the microscopic level like in metal alloys or ceramics. Rather, the response is driven by the material properties and the type of stimulus.

Remarkably, another class of self-evolving actuators consists of material assemblies fabricated as films or strips that exhibit a characteristic bending when exposed to a certain stimulus. Bending actuating films can be produced in different ways and under particular conditions depending on the desired application, the source of the stimulus, or the way the actuation is generated. Hereunder, we explore numerous of the previously reported approaches of bending film actuators. In the following sections, we categorize the actuators as monomaterial, layered, and structured.

2.1. Monomaterial Actuators

Bettoti et al.^[22] studied monolayer materials capable of inducing autonomous bending by exposing a cellulose-based sample to a moisture gradient, in which one side of the film is exposed to humid air (above a water droplet), while the opposite side remains in contact with ambient air (Figure 3a). A gradient of water content within the film causes the curvature, which can be controlled by adjusting the water vapor pressure over the droplet by adding other compounds such as ethylene glycol or by changing the temperature of the droplet. Additionally, another example of monolayer materials based on cellulose was reported by Wei et al.^[23]

Fu et al.^[24] proposed another variant of actuating films made of a single material. The authors drop-casted liquid polydimethylsiloxane (PDMS) onto salt crystals attached to an adhesive tape, then solubilized the salt in water to remove it from the solid cross-linked PDMS film to obtain what they termed “Janus films” (one side completely flat and the other one is characterized by a high roughness). The difference in topography causes bending upon submersion in hexane since the highly porous side can take up solvent more easily than the smooth side. The advantage of Janus films over the monolayer system proposed in ref. [22] is that the film can be exposed to a gradient-free environment.

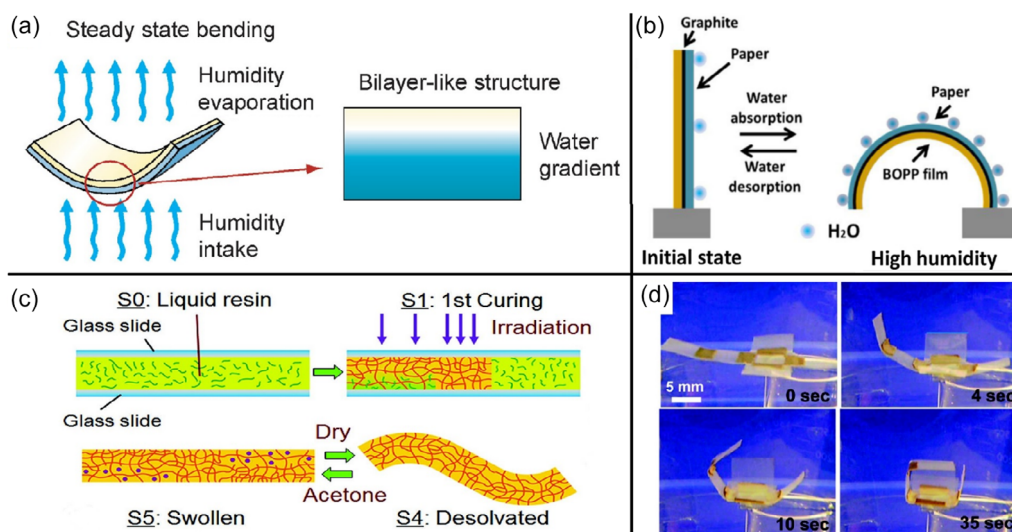


Figure 3. Possible approaches to fabricate self-sufficient actuators: a) Monolayer material with only one face exposed to the stimulus. Reproduced with permission.^[126] Copyright 2015, Wiley Online Library. b) Bilayer assembly with one layer that reacts actively to the stimulus. Reproduced with permission.^[43] Copyright 2021, Elsevier. c) Graded (gray-scale) actuator (cross-linking density variation along the thickness). Reproduced with permission.^[81] Copyright, 2016 Wiley Online Library. d) Hinge-like assembly that bends into a predetermined shape. Reproduced with permission.^[54] Copyright 2011, American Chemical Society. All images were taken from the original sources with no further modifications.

However, it does not work in contact with air, given that PDMS is a hydrophobic polymer that can only be activated using organic solvent vapor (like hexane). More recently, Lei et al.^[25] proposed a similar fabrication process to produce Janus films using chitosan.

Alternatively, Zhan et al.^[26] reported the fabrication of an actuator based on cellulose. They obtained cellulose stearyl esters with different degrees of substitution that showed a peculiar alternating bending behavior when exposed to a water vapor gradient over warm water. Interestingly, this behavior is analogous to the periodic one obtained by mechanical frustration in responsive monomaterial actuators with restrictive geometries.^[27]

Regarding the possible applications for this kind of actuators, although some exciting prototypes have been developed for use in soft robotics,^[24,28] none seems to appear suitable for meter-sized applications yet.

2.2. Layered Actuators

A typical approach to bending actuators consists of a bilayer strip made of two different materials bonded together: a passive layer, which does not react to the stimulus from the environment, or reacts negligibly, and an active layer that responds by expanding or contracting according to the stimulus acting on it. The mismatch in expansion behaviors causes the bending.^[29] One distinction between numerous actuators lies in the stimulus responsible for the bending, which could be temperature,^[30,31] relative humidity,^[32,33] solvent vapor pressure,^[34] electricity,^[35,36] light,^[37–40] pH,^[41] or salt concentration^[42] (the latter two apply to actuation in an aqueous solution). It is worth mentioning that an actuator can be triggered by more than one stimulus separately or simultaneously, depending on the fabrication and materialization of the strip.

Weng et al.^[43] explored a straightforward approach by assembling the actuator using conventional paper, depositing graphite on it with a standard pencil, and attaching a bioriented polypropylene adhesive tape (BOPP) on top (Figure 3b). Despite its simplicity, the actuator can be multiresponsive due to a mismatch of the coefficient of thermal (CTE) and hygroscopic expansion (CHE) between the BOPP layer and the cellulose present in the paper layer. Moreover, graphite can induce a photothermal effect when exposed to light of various wavelengths (from ultraviolet to infrared), and it is also electrically conductive, which can induce a rapid temperature rise when electricity is applied to the actuator. In addition, the BOPP layer acts as a passive layer when relative humidity changes, given that its CHE is supposedly zero.

Similarly, Amjadi et al.^[44] built an actuator made of BOPP and paper, with the difference that instead of graphite, the authors used silver nanowires attached to the paper by coating the sample with a mixture of poly(3,4-ethylenedioxythiophene) and polystyrene sulfonate (PEDOT:PSS). Thus, the actuator also responds to humidity, temperature, light, and electricity. Taccola et al.^[35] explored the use of PEDOT:PSS for an electrically driven actuator, given that this mixture can release humidity into the air when exposed to an electrical current and potentially take up water when the current is interrupted. They fabricated their actuator using PDMS as the passive layer, and were able to control the bending by changing the relative humidity and applied voltage. In addition, a similar actuator based on PEDOT:PSS was tested by Dingler et al.^[45]

The fundamental difference between an active/active and an active/passive bilayer system is that, in the first case, both layers can exhibit a non-negligible response to the stimulus (or stimuli) at the same time to maximize the curvature and responsive time of the actuator^[46,47] or to create a multibending system able to move in different directions.^[48]

One may understand the double bending actuation as the ability of a system to bend outward and inward with a unidirectional orientation (i.e., without twisting). For example, Wang et al.^[48] built an actuator that could bend in both directions by applying two different stimuli independently. They designed laminates made of pyrrole ferric chloride and an agar component (based on agarose polysaccharide), which can bend under multiple stimuli: agar hygroscopicity is responsible for the actuation triggered by humidity. Chlorinated pyrrole compounds can react to solvent vapors such as hydrogen chloride (HCl) and ammonia (NH₃) due to protonation and deprotonation reactions that cause water migration within the actuator toward and from the agar component. Additionally, thermal changes or exposure to infrared light can trigger the actuation due to CTE mismatch or photothermal effect in the pyrrole layer, respectively.

In contrast, researchers have extensively studied polymers with low critical solution temperature (LCST) to build smart materials,^[49] as they can behave as actuators displaying double bending under the same stimulus. In polymer science, LCST refers to the temperature below which a polymer is soluble in a solvent at any concentration; above LCST, the interactions between polymer chains and the solvent are not enthalpically or entropically favorable. Thus, the polymer collapses into coils and is no longer soluble. To obtain this property, the polymer chains need to have strong interactions with their solvent at low temperatures, such as hydrogen bonds that promote the dispersion of polymer molecules in the solvent; conversely, a temperature increase weakens such hydrogen bonds.^[50] The most studied polymer with an LCST is poly(N-isopropylacrylamide) (PNIPAAm),^[51,52] given that its LCST is about 31 °C in water. The copolymerization with hydrophilic monomers allows tuning this value.^[53] Many reported studies used PNIPAAm for actuation in aqueous solution^[54–56] including electrospinning^[57,58] and multilayering,^[59] while others showed its actuation abilities in contact with air.^[60,61]

Gao et al.^[62] proposed a different approach. The authors developed a copolymer with LCST by functionalizing polyvinyl alcohol (PVA) with N,N-diethylethylenediamine (DEEDA) and borax (also known as sodium borate). The actuator could bend in both directions in contact with air depending on relative humidity and temperature. This study reported the bending kinetics as a function of the two stimuli; here a significant effort was made to characterize the material properties (including the CTE and CHE). The authors also suggested using infrared light to trigger a photothermal actuation. Remarkably, the advantage of assembling an actuator such as in Gao et al.^[62] is that actuation relies on a chemical modification in the structure instead of a mere variation of physical conditions that can quickly deteriorate the response over multiple uses of the actuator.

Other authors have reported the fabrication of bilayer bending actuators using electrospinning,^[63,64] where they tuned the response with better precision through the inclusion of additives, variation of matrix composition, and filler crystallinity. Furthermore, Alexander et al.^[64] incorporated and discussed a relevant variable that affects the bending actuation and its response time, namely the diffusion of water in the active layer. However, to the best of our knowledge, most studies conducted on electrospinning have worked successfully exclusively in solution.

A significant number of studies have focused on the fabrication of the bilayer-actuators based on response to light, which can be classified into two categories. On the one hand, as mentioned previously, a light source can induce a photothermal effect to increase the temperature of the sample, eventually coupled with a hygroscopic shrinkage due to water evaporation. Tai et al.^[65] discussed the use of a polyvinylidene fluoride (PVDF) layer in combination with single-walled carbon nanotubes (SWCNTs). The authors chose this particular assembly to maximize the CTE and CHE difference between both materials. PVDF has a high CTE, is hydrophobic, and has a low CHE. In contrast, SWCNTs (with a high CHE) are known for having a rare negative CTE and having an outstanding thermal conductivity, which is very practical and efficient when using light as a heat source.

Similarly, Guo et al.^[55] reported the elaboration of a hydrogel with dispersed silver particles able to respond in aqueous solution to infrared light, given that the presence of the metal in the matrix is responsible for a photothermal effect. Alternatively, Zhang et al.^[66] explored a more specific tunability of SWCNTs. First, the authors deposited the nanotubes on a polycarbonate matrix. Then, they used metal-enriched, chirality-enriched, high-pressured carbon monoxide (HiPCO)-synthesized SWCNTs to adjust the light wavelength that could be applied as a stimulus. As a result, wavelengths between 600 and 1250 nm could trigger the actuator. Additionally, another study^[39] showed the effect of light power on the radius of curvature.

Li et al.^[40] reported a different approach using light. They fabricated an actuator based on a PDMS matrix with dispersed chromium dioxide (CrO₂) particles and used a laser beam as a light source while keeping the sample exposed to a magnetic field. The demagnetization of CrO₂ causes the bending behavior when the temperature in the matrix increases upon exposure to the laser beam. This actuation is achievable thanks to the relative low Curie temperature of CrO₂ which is 124 °C.

On the other hand, another light-based approach consists of using liquid crystal elastomers (LCE). Several authors have extensively studied these materials to develop light-driven actuators.^[67] Notably, they have reported that azobenzene compounds can exhibit deformations due to reversible isomerization of the molecule when exposed to ultraviolet light,^[68] heat, and humidity.^[69,70] However, isomerization in fluorinated azobenzene molecules can be triggered directly by sunlight, as Kumar et al.^[71] reported the fabrication of a self-oscillating actuator. Furthermore, other types of bending actuators using azobenzene LCE in solution^[72] and in contact with air^[73] have shown the anisotropic effect of cutting uniaxially oriented film strips at different angles, which induces a particular response (bending or twisting) depending on the cutting angle. Moreover, Iamsaard et al.^[73] demonstrated how it is possible to tailor a twisting or bending actuation based on the type of cell (twisted or planar) that constitutes the crystals inside the film. Lastly, it is possible to tune azobenzene LCE to respond to longer wavelength light stimuli, either by chemically treating the material with acid^[74] (which produces a bending actuation with visible light) or by adding a dye capable of absorbing light with a specific wavelength, as reported by Wang et al.^[75] where the actuator is sensitive to both UV and infrared light. Various approaches for similar bilayer-like

structures include the use of natural materials such as flax fibers,^[76] microbial cells,^[77] proteins,^[78] as well as xerogels.^[79]

Complementarily, we may understand a multicomponent actuator as either a system with more than two layers or multiple materials in one single heterogeneous layer with discrete regions. In multilayer systems, each layer should add a specific feature to the actuator to tune the sensitivity under different stimuli or to protect the integrity of the actuator.^[80] Conversely, a system with multiple materials in a single layer can induce shape transformations beyond simple bending, given that each region within the sheet can respond independently to a particular stimulus.^[56] For example, Thérien-Aubin et al.^[56] explored the generation of actuators starting from single sheets of monomers exposed to UV radiation for photopolymerization, followed by submersion in water and a second selective photopolymerization using masks to delineate certain areas of what they called “binary gels.” Next, the authors carried out additional attempts using copolymerization to extend the heterogeneity of the sheets. Depending on the initial monomers, they reported 3D shaping of the actuator under changes in temperature, pH, ionic strength, and carbon dioxide (CO₂) concentration. Then, by patterning pairs of monomers in stripes, the actuator was built with four distinct zones that responded differently to each stimulus, achieving more complex 3D shapes.

Finally, it should be noted that the principle of layered actuation applies to graded materials. This might be obtained by gradually changing the active property within the material, which, for example, can be achieved by chemical functionalization. Zhao et al.^[81] proposed using a photocurable resin exposed to a light source (Figure 3c); in this case, the light intensity decreases with penetration into the resin, causing a gradient of crosslinking density in the structure. The authors used this practical approach to control the bending of thin sheets to induce origami shapes by acetone desolvation. Here, they controlled the shaping by restricting the exposed areas of the sheet according to the desired final shape. Similarly, Wu et al.^[82] studied the use of resin-based 3D printing to generate what they call a grayscale, which is the same principle applied by Zhao et al. but instead, they opted for UV radiation. The light intensity changes along with the penetration depth, allowing to obtain strips of material with different crosslinking degrees throughout its thickness. This functionalization emulates a bilayer system with different affinities for a particular solvent. Namely, the authors showed the bending behavior of the actuator by submerging it in acetone.

In another approach, Schäfer et al.^[83] used the photo-crosslinkable fluorescent copolymer P(DMAA-*co*-MABP-*co*-RhBMA) dissolved in either H₂O or IPA to achieve gradient modifications in paper sheets. Here, the single-layer paper sheets showed hygro-responsiveness (i.e., bending reactions due to changes in humidity), thus acting like functional bilayer materials. The developed humidity-responsive actuators out of a single sheet of paper have several advantages over multilayered structures, reducing the risk of delamination and simplifying the design and construction.

Also, direct applications of photoinduced actuators in soft robotics have been proposed. One example of this, is the helicopter-like actuator presented by Wang et al.^[84] The authors were able to fabricate a device with an ultrafast rotation in response to infrared light that could fly over short distances.

Similarly, Li et al.^[85] proposed a jumping and rolling actuator driven by light. They took advantage of the elasticity of a hydrogel based on poly(sodium acrylate) and the photothermal effect of iron oxide nanoparticles.

2.3. Structured Actuators

Other studies have focused on the fabrication process of complex assemblies yielding to programmed kinematics. For instance, the impact of anisotropy on actuation behavior has been investigated by some authors. Alexander et al.^[86] studied the effect of aligned PVA nanofibers and their orientation angle to induce a bending response along with a twisting effect in the actuator. The authors showed that the orientation also influences the effective diffusion of water within the active layer. Athanasopoulos and Siakavellas^[87] assembled a bilayer sample containing oriented polyethylene (PE) and evenly distributed aluminum strips with a 45° orientation. The difference between a low CTE in the aluminum strips and a high CTE in the PE layer results in twisting behavior upon heating or cooling. In addition, Song et al.^[88] reported the elaboration of anisotropic layers by applying a magnetic field to align the magnetic powder particles inside the actuator. Similarly, Sun et al.^[89] reported an actuator with Fe₃O₄ nanoparticles that reacted under application of a magnetic field.

Stoychev et al.^[90] fabricated a different type of anisotropic actuator with holes on specific sheet locations to govern how the bend (fold) should occur. The size and shape of the hole also determine the geometry obtained after folding, as complex 3D motions can be achieved. The authors showed that their approach also allows for ultrafast response as the actuator must overcome a buckling threshold to be able to fold. This introduces the buckling, or snap-through effect as a possible solution to deal with the deformation response speed of actuators.

Additionally, Le Duigou et al.^[91] reported the assembly of an actuator based on 3D-printed polylactic acid (PLA) with wood fibers. The authors evaluated the performance as a function of process conditions, namely, the printing width and postcompression, to assess the effect of porosity on total water uptake during submersion in water. Correa et al.^[92] fabricated an actuator by 3D-printing a cellulose-based filament bonded to a nylon layer. The authors detailed an elaborated design that considers different angular orientations of the printed pattern to generate curvatures at specific angles. Furthermore, they segmented the passive layers and placed them on defined sections on both sides of the 3D-printed wood layer to achieve multidirectional curvatures. Their actuator was successfully tested upon submersion and in contact with humid air.

In terms of fabrication methods, the use of 3D printing for actuator production has become more prevalent in recent years due to its practicality and easiness to control thickness and anisotropy. Lin et al.^[93] printed an actuator based on polyrotaxane and polydimethylacrylamide (PDMA) with a postmethylation process that allowed the actuator to bend with changes of temperature in water and even a change in transparency due to the reversible degree of crystallinity. Additionally, Tyagi et al.^[94] reported the fabrication via 3D printing of a microactuator based on two commercial hydrogels (Ebecryl 4491 and Hydromed D4). Due to the size of the pieces (a few hundred micrometers), their

deformation in water took a few seconds. They used the same actuator to create a hinge-like structure for a possible application in soft robotics. Moreover, Shin and So^[95] created a bilayer actuator based on PLA 3D-printed over a paper substrate that could be used for a temperature-sensitive electrical circuit. By varying the temperature, the bending of the actuator allowed the activation or deactivation of the circuit. The authors were able to tune the responsive time by changing the thickness. They additionally tested the performance of the actuator after multiple uses.

A step beyond conventional 3D-printing is 4D printing, a term used to describe composite materials that can deform in a pre-defined way.^[96] Some authors include them in the category of SMMs.^[97] Although this type of actuator is conventionally 3D-printed, filling fibers are generally integrated into the composite materials to allow for anisotropic deformations. In addition, the actuation can be customized by adjusting the size, orientation, shape, and spatial distribution of the fibers within the matrix. This method refers to the so-called “fiber architecture”.^[98] Notably, Yao et al.^[99] reported the fabrication of an actuator based on a lanthanide-ion-coordinated supramolecular hydrogel. Due to the interaction between europium (Eu) ions and nonconjugated chromophores, the material was able to change luminescence and opacity as a response to changes in humidity or hydration.

Interestingly, Nishiguchi et al.^[100] described the fabrication of an actuator based on 4D printing with a variable density. The authors were able to tune the architecture of the composite in the range of nanometers by using multiphoton lithography. Their actuator consisted on a matrix of PNIPAAm with dispersed gold nanorods (AuNRs), where through plasmonic heating of the AuNRs, a very fast deformation of the actuator was achieved.

Other approaches include the assembly of hinge-like structures, which use the same principle already explained for bilayer systems, with the difference that hinge actuators are direct applications for soft robotics and self-assembled memory materials (including origami-inspired designs). Notably, by building a hinge instead of a complete layer, the actuator can locally induce more significant deformations in a short time and favors kinematics in three dimensions. For instance, Zhang et al.^[54] fabricated a system using PNIPAAm as the main compound attached to hinges made of SWCNTs. The actuator was assembled as a flat sheet, and it could bend in water at different temperatures to display multiple shapes depending on the sheet form (cubes, stars, among others). The authors reported actuation durations (full motion) as fast as 35 s in water at 48 °C, significantly faster than conventional bilayer actuators with continuous layers (Figure 3d).

Furthermore, Yamamura and Iwase^[101] reported the fabrication of a hinge-like 4D printed actuator made of a thermoplastic polyurethane. The authors showed that it was possible to generate folding and unfolding deformations after the hinge reached a given curvature by applying an external force below the glass transition temperature.

2.4. Theoretical Aspects of Elementary Material-Based Actuation

The literature review presented in the previous section highlights a commonality among all synthetic material-based actuators,

namely, that the bending motion is produced by an in-plane strain gradient across the actuator thickness (i.e., the out-of-plane direction). In this regard, three idealized mechanisms (Figure 4) are likely to be identified to account for this fundamental driving force of bending. Interestingly, each of these mechanisms involves a mismatch that can be found either: 1) in the material expansion properties, when the actuator consists of a multilayered or graded assembly (i.e., internal mismatch: case I); 2) in the environmental conditions, when they differ on each side of the actuator (i.e., environmental-based gradient: case II); 3) in surface exchange differences, when the response is controlled by a surface modification on one face of a monomaterial-based actuator, such as roughness in the Janus film (see Section 1.1) for example (surface conditions mismatch: case III).

With reference to the numerous examples presented in Section 1, the actuators' bending capabilities could rely on either case I, II, III, or any combination.

Although these three approaches and their possible combinations open up a wide panel of actuator designs, the fundamental principle of bending actuation caused by a strain gradient across the actuator thickness might be analytically investigated using the basic bilayer assembly (case I) as the elementary building brick of material-based actuation. Timoshenko's pioneering framework of a bilayer beam curvature caused by thermal expansion^[29] is the obvious starting point. Since then, many generalized formalisms involving various strain origins have been reported in literature.^[45,46,102–104] In particular, models for multilayered assemblies have been developed to achieve zero stress at interfaces subject to delamination^[103] and gradients across the film thickness have been invoked to correctly capture the curvature behavior of some gold/polypyrrole-based composites.^[104]

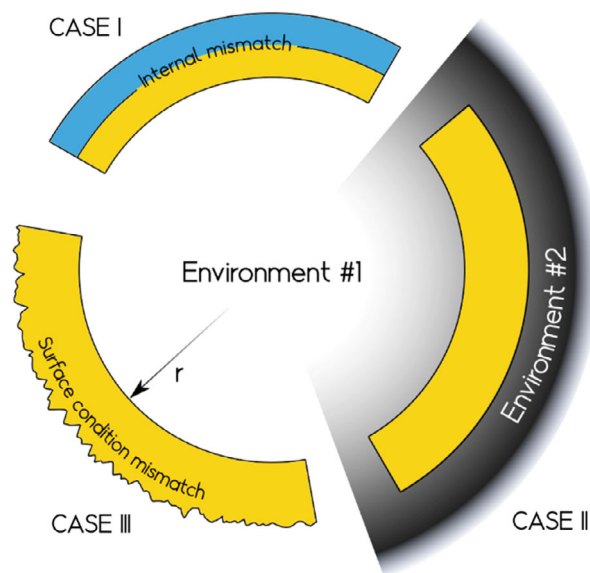


Figure 4. Idealized cases related to material-based bending actuation. r indicates the radius of curvature. Case I: the actuator generates an internal mismatch that results in a given curvature. Case II: the response is driven by the roughness of the surface of one of the phases. Case III: the actuator is exposed to two different environments or conditions that trigger the response.

Here, the objective is to reveal the underlying mechanical principle that governs the actuation and to identify the essential parameters and their quantitative role. For this purpose, Equation (1) presents an extension of the original Timoshenko's equation for bimetal thermostats to multiple stimuli. This equation, which gives the radius of curvature r of a bilayer actuator when subjected to homogeneous environmental stimuli, intends to serve here as an entry point to obtain a mathematical insight into the key parameters leading to the most efficient response.

$$\frac{1}{r} = \chi \frac{\overrightarrow{\Delta C} \cdot \overrightarrow{\Delta S}}{h} \quad (1)$$

$$\text{with } \overrightarrow{\Delta C} = \begin{pmatrix} \Delta CTE \\ \Delta CHE \\ \vdots \end{pmatrix} \text{ and } \overrightarrow{\Delta S} = \begin{pmatrix} \Delta T \\ \Delta RH \\ \vdots \end{pmatrix}.$$

In this form, Equation (1) shows that the efficiency of an autonomous bending actuator in terms of curvature is essentially driven by the following aspects (see Figure 5): 1) The magnitude of each component of $\overrightarrow{\Delta C}$, which gathers the mismatches in expansion coefficients between layer 1 and layer 2 (thermal, hygroscopic etc.). 2) The amplitude of each component of $\overrightarrow{\Delta S}$, which gathers the stimuli variations (temperature, relative humidity, etc.) between two states of equilibrium. 3) The total thickness $h = a_1 + a_2$ of the actuator. 4) The actuator design described through the agglomerating parameter χ (Equation (2)). Noteworthy, Equation (2) shows that, for the bilayer-like elementary actuator of interest, two coupled components, namely, mechanical (through n , i.e., Young's moduli ratio) and geometrical (through m , i.e., the thickness of layer 1 (a_1) over the total thickness ($h = a_1 + a_2$)), are jointly involved in the assembly efficiency.

Notably, each expansion coefficient ΔC_i (ΔCTE , ΔCHE ,...) of vector $\overrightarrow{\Delta C}$ is paired with its corresponding stimulus component counterpart ΔS_i (ΔT , ΔRH ,...) of vector $\overrightarrow{\Delta S}$:

$$\chi = \frac{6}{3 + \frac{(1+mn)^2}{(1+m)^2} (m^2 + \frac{1}{mn})} \quad (2)$$

$$\text{with } m = \frac{a_1}{a_2} \text{ and } n = \frac{E_1}{E_2}.$$

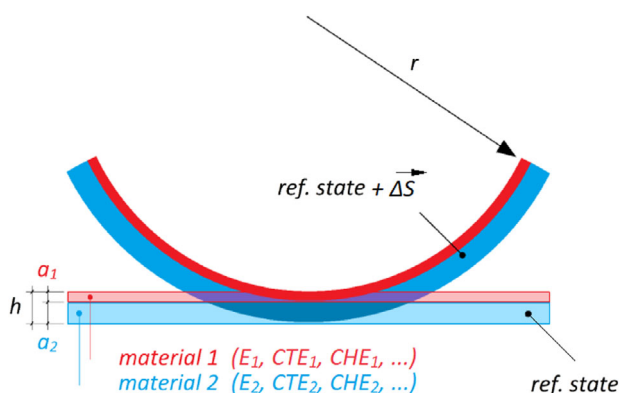


Figure 5. Bilinear-like model of an elementary autonomous bending actuator.

It should be emphasized that Equation (1) restricted to a given single stimulus leads to Equation (3), which duly meets Timoshenko's equation^[20] if temperature is considered.

$$\frac{1}{r} = \chi \frac{\Delta CTE \times \Delta T}{h} = \chi \frac{(\alpha_2 - \alpha_1) \times \Delta T}{h} \quad (3)$$

with α_1 and α_2 , the linear thermal expansions of both materials, considering $\alpha_2 > \alpha_1$. The graphical representation of Equation (2) in Figure 6 shows that it is possible to find an optimal region where χ reaches its maximum value ($\chi = 1.5$) and, therefore, the curvature of the bilayer actuator is maximized regarding the actuator design, independently from the other key parameters $\overrightarrow{\Delta S}$, $\overrightarrow{\Delta C}$, and h . The optimal region where χ equals 1.5 corresponds to an assembly design that satisfies the condition $E_1 a_1^2 = E_2 a_2^2$. Finally, although this condition is achieved (i.e., $\chi = 1.5$), Equation (1) points out that the magnitude of the resulting movement remains also governed by the mismatches in the expansion coefficients: the absence of a property gradient in the material assembly would entirely cancel out the corresponding stimulus effect, thus leading to no curvature contribution. In terms of material considerations, this insight straightforwardly draws the limitations of curvature efficiency, as a change in geometric ratio m involves an important adjustment of Young's moduli ratio to achieve a maximum actuating effect. This considerably constrains the possible choices in the design of efficient autonomous actuators.

The challenges to cope regarding the upscaling of autonomous bending actuators and biomimetic approaches that have been implemented on large scale are discussed in the next section through the prism of this mathematical description of an elementary bilayer actuator.

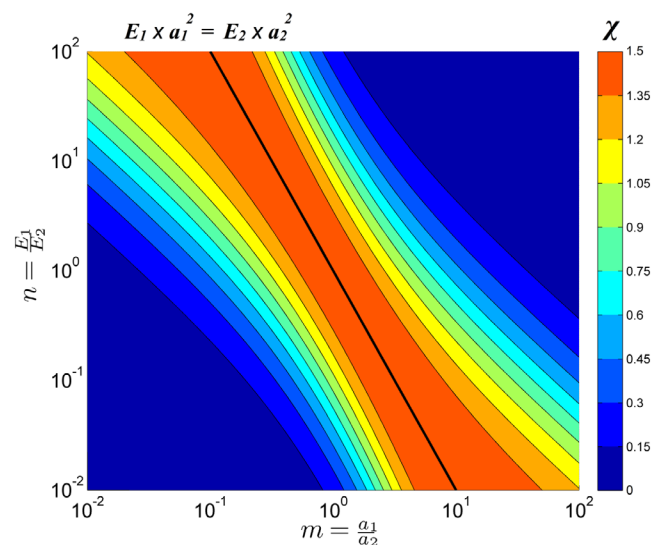


Figure 6. Value of the agglomerating parameter χ as a function of the layers' relative thickness and Young's modulus. The solid curve highlights the maximum curvature achieved for $\chi = 1.5$ when the assembly design satisfies the condition $E_1 a_1^2 = E_2 a_2^2$.

3. Upscaling Material-Based Actuators

3.1. Biomimetics and Examples of Applications in Architecture

In the past, actuators inspired by nature have been successfully developed for applications in engineering and architecture. One example is the Flectofin,^[5,105] a compliant system featuring a rigid backbone and a thin lamina. By bending the backbone thanks to an external mechanical actuation, the lamina deforms at a certain angle and creates a flat surface. Thus, the combination of several of these elements in parallel generates a hingeless shading system. The authors developed the prototype by taking bioinspiration from the bird-of-paradise flower (*Strelitzia reginae*), as mentioned above (Figure 1c).

Similarly, multibending actuation in nature may serve as a source of inspiration for exciting applications, where the required energy does not come from any electrical or mechanical source but rather from the environment itself. One may think of such applications when energy losses need to be minimized, or when the environment is the only actual energy source.^[106] This premise aligns with the current needs and could be a powerful approach toward environmental sustainability.^[107] Remarkably, this type of application appears advantageous to minimize long-term costs, given that the mechanism does not require highly elaborated systems with costly maintenance routines.

In modern architecture, designers have taken inspiration from nature to create innovative constructions that are either too complex to design or too costly to produce using conventional approaches. Although architects and engineers have neglected double curvature structures during the last century, they have incorporated these structures more frequently in building constructions worldwide in recent years.^[108,109]

Biomimetic wood-based prototypes have gained great importance for real-life applications in architecture. Indeed, several studies have explored bioinspired structures made of wood, given that this natural composite can be used as a self-forming material to generate double curvature shapes^[110,111] and interlocking pieces to form folded plates.^[112] It is also possible to obtain bilayer wood sheets that bend with temperature or humidity changes due to their anisotropic nature.^[113,114] As a natural composite, wood offers multiple advantages thanks to its microfibril structure and highly anisotropic behavior. In particular, the swelling pressure exerted on the cellular walls of the microfibrils can reach theoretically 158 MPa,^[115] which makes this material unrivalled in water responsiveness at a large scale. As a result, a variety of innovative applications have taken advantage of this material. For instance, it has been demonstrated that timber can be used to produce complex double curvature structures that are initially fabricated as flat sheets,^[116] up to the large structures in architectural scales.^[117] Computational ply design is the key factor to control the orientation and respective bending response; the parts are fabricated at low relative humidity, and they bend to reach the desired shape at high humidity levels.

Similarly, two original ventilation systems have been proposed under the names HygroSkin^[118] and HygroScope.^[119] The first is a sandwich-like structure that closes and opens at high and low relative humidity, respectively. The assembly consists of two plywood sheets on the exterior and one internal extruded polystyrene (XPS) layer. The prototype was designed and fabricated

in Stuttgart and later installed in the so-called HygroSkin pavilion in Orléans, France. The second variant is a bilayer structure of wood and a mixture of epoxy resins and glass fibers. The actuator was carefully tuned by setting conditions during the fabrication to calibrate the response to a specific relative humidity. The prototype was then assembled on a large scale, showing significant bending to relative humidity changes (Figure 7a,b). These two prototypes are one-of-a-kind adaptations where the actuators were directly assembled at the macroscopic scale with impressive design and precise fabrication steps.

Barrett et al.^[80] designed a multicomponent bending system intended as a shading and ventilation structure for building applications. Changes in ambient temperature can trigger the actuator, called Thermadapt, which consists of four different layers: the outermost layer is a graphite-epoxy compound with a low CTE, attached to a composite layer, followed by an aluminum layer with a high CTE. The innermost layer is made of a foam of XPS, very lightweight and with a high CTE. The air cells inside the foam also contribute to significant bending with small temperature changes. After installation, the actuator bends outward to allow airflow inside the structure in hot conditions. Conversely, in cold weather, the trapped air inside the XPS contracts the whole assembly, creating a sealing effect to achieve thermal insulation inside the structure (XPS is a typical insulator in construction). Interestingly, the authors managed to fabricate relatively thick actuators (above 1 cm) by dividing the XPS layer into small elements glued together parallel to each other along the length of the piece. They have shown that this configuration increases lateral stiffness to obtain a unidirectional bending. It is worth noticing that XPS has a very low density, which allows gravitational effects to be neglected even in large-sized actuators, along with a low Young's modulus (≈ 25 MPa^[80]) that allows large deformations under low stresses.

One of the most challenging issues during the fabrication is the upscaling of the actuators to realistic thicknesses (in the centimeter range) while keeping a relatively fast response to changes in the environmental conditions.^[102,109] Material costs and fabrication complexity also play an essential role for potential large-scale applications, as many reported studies have explored assemblies with expensive compounds or highly elaborated production processes that are not easily transposable to an industrial scale. However, a successful application that overcame these scalability problems was reported by Rüggeberg and Burgert.^[102] They directly took different varieties of wood, fabricated an actuator capable of bending at multiple ambient conditions, and evaluated long-term performance under actual weather conditions (exposure to rain, dew, and wind) (Figure 7c,d).

Noteworthy, while wood is a very versatile and unique material, it can degrade over time. As a consequence, the responsiveness of wooden actuators may decline with multiple uses, or it may not be possible to guarantee long-term durability (based on the behavior displayed by the actuator fabricated by Rüggeberg and Burgert^[102]). This limitation is the reason why wood-based assemblies for autonomous movements are often considered with caution for engineering applications. It is therefore important to explore the viability of synthetic materials to produce actuators that can exhibit similar responsiveness while providing greater stability and a longer service life.



Figure 7. Wood-based actuators at large scale. a) Hygroscope prototype (Reproduced with permission.^[119] Copyright 2015, Elsevier) at 40%RH (fully open) and b) at 80%RH (fully closed). c) Curvature response of a bilayer wood actuator at 88%RH and d) 55%RH with attached load under real weather conditions.^[102] Taken from the original sources without further modifications.

It is important to emphasize that the design of these actuators, specifically aimed at shading systems and smart facades, must meet certain parameters to be considered attractive and feasible for commercialization on the market. Interestingly, Attia et al.^[120] have summarized pivotal considerations that might define the development of this field (commonly referred to as adaptive facades) in the future. The authors consulted several experts and concluded that while energy saving is a decisive factor in design, end-user satisfaction and minimal environmental impact will play an essential role. Remarkably, they argue that the design will need to consider elements such as human-oriented and service-oriented solutions, as well as smart building operability, circularity and resource efficiency. In addition, while technologies applied to chromogenic and solar active facades offer promising potential, dynamic shading systems and active ventilated facades (which include biomimetic approaches) are technologies with wide application scope and considerable commercial potential.

3.2. Challenges in Scaling Up Material-Based Actuators

The fabrication of real-life actuators is undoubtedly challenging and this field has been tentatively explored.^[109] For the sake of simplicity, the following discussion will be focused to the framework of temperature and humidity stimuli, since many actuators

in engineering intend to react to these two environmental stimuli. Nevertheless, one should keep in mind that other stimuli such as light, swelling, etc. could be considered and discussed in the same way to design an effective elementary actuator brick as described in part 2. For instance, light and wind are as important as temperature and relative humidity for shading systems applications in architecture.

When considering broader engineering applications (civil and mechanical engineering, soft robotics,...), the vast majority of authors opt for a bottom-up approach, i.e., starting from a given actuator design to then evaluate its efficiency. Conversely, with respect to Section 2, the present work aims at drawing the underlying scientific drawbacks related to the upscaling purposes as a starting point in a top-down methodology. Following this approach, three major axes for an effective upscaled autonomous actuation have been identified: i. serviceability; ii. response time; and iii. mechanical amplification.

3.2.1. Serviceability

A determining factor for the fabrication of actuators at a large scale consists of evaluating their stability in the long term. In this respect, the actuators must certainly fulfil minimal stability and resistance requirements. Thus, the design phase has to consider aging effects to guarantee the actuator's durability over time.

Furthermore, the phase lag observed with cycling stimulus must be addressed during the material selection and respective fabrication to mitigate material/assembly degradation leading to long-term actuation weakening. Additionally, in the absence of careful characterization, the evoked unwanted side phenomena may result problematic if a reliable actuator is to be designed. As it will be discussed along the following sections, a sensible choice should consider a light, reliable material, whose deformation can be easily controlled without losing its integrity when exposed to rain or other external factors. In addition, its sensitivity should be easily quantifiable with minor changes in the environmental stimulus.

The design of an actuator will certainly be discarded for industrial production if its applicability cannot go beyond the meso-scale. Unfortunately, this is where most actuators proposed in the literature to date fail to contribute. It is well established that specific actuators can be easily adapted to stimuli in solution and applied to fields such as soft robotics.^[121] However, the number of studies focused on tackling the persistent adaptability problems is scarce. The crucial point relates to fabrication factors. At the initial stage of selection, the materials' processability needs to be addressed. Regarding this aspect is perhaps a chemical functionalization the least reasonable option. The control of fabrication variables becomes more demanding as the batch size increases. For this reason, it may be advantageous to choose a physical process where all parameters are more easily controlled, no matter the production size. Naturally, cost (including raw materials and manufacturing) and processability must be crucial considerations. Ideally, components should be easy to obtain and process. The more materials and fabrication steps involved, the more complex it is to successfully produce an actuator at a large scale. Finally, it may be worth considering sacrificing the purity of expensive raw materials and dealing with the presence of contamination when designing manufacturing steps.

3.2.2. Response Time

Another challenging factor in upscaling a thin-film actuator is undeniably its time responsiveness. In a general framework, the actuator response time has to remain consistent with the characteristic time of the stimulus (i.e., the change in the environmental conditions in the context of interest). On the one hand, several applications could require an actuation response that tends to temporally match the environmental stimulus trend. In this case, a fast and complete movement must be achieved. For instance, if relative humidity increases in a ventilation system, the autonomous actuator will be useless if it cannot reach its maximal opening or closing before the relative humidity decreases. On the other hand, one may target an actuation kinetic allowing for the damping of the environmental variations. In this case, a finely tuned response in terms of both speed and amplitude may be sought.

The second point about time response when upscaling a thin-film assembly consists in the various time scales inherent to its response to each stimulus. Indeed, a temperature stimulus has the unique advantage of not depending on mass transfer phenomena compared to relative humidity, as temperature

homogeneity could be reached almost instantly if the assembly is designed with considering its thermal conductivity. The design of RH-driven actuators is undoubtedly more challenging as it requires dealing with a specific aspect, namely diffusion. In conventional materials (unstructured), the response is subject to the molecular structure and its water diffusivity. Consequently, the diffusion coefficient and the actuator thickness become the relevant variables. According to Fickian's law applied to plane sheets,^[122] the time required to reach water content equilibrium is directly proportional to h^2/D (squared thickness divided by diffusion coefficient). However, in accordance with assumptions related to Equation (1) and (3), the complete bending of the actuator is only achieved when the stimulus has reached its innermost layer, which necessitate in fact a certain time to process. Thus, a thickness increase will drastically affect the actuation time.

One possible solution is to modify the diffusion coefficient of the active layer. However, this endeavor cannot be easily carried out unless the molecular conformation is directly addressed. Nonetheless, chemical manipulation of the material structure could only partially raise the water diffusion in an extent that could not easily overcome the actuation constraints generated by increasing the system thickness to the centimeter range. As a consequence, a groundbreaking approach that takes advantage of relative humidity changes in the environment while minimizing stabilization times may not be feasible only through chemical functionalization of the material. Although the production scheme would be inexpensive and easy to implement, this route may not be viable as a stand-alone solution according to upscaling criteria.

Another way to a faster response would be to tremendously increase the surface to volume ratio in the material to accelerate the temperature/species transfer. The purpose is to overcome inherent problems associated with the material, typically to tackle the diffusion slowness. Both surface modification and feature of the bulk can be envisaged. Here, we can think of different approaches such as porosity, as proposed by Tahouni et al.^[11] surface patterning, or anisotropy. Increasing the surface-to-volume ratio by using microstructured material at various scales could participate to overcome diffusion limitations at relatively large thicknesses. Fruleux et al.^[123] applied 3D printing to include microchannels into hydromorph biocomposites. With the achieved biomimetic capillary water distribution within the printed hydraulic actuators, an eightfold improvement in the speed of shape-change transformation without compromising any other functional performance was achieved.

Finally, a bioinspired solution was pointed out by Poppinga et al.^[106] discussing that hydraulic actuators could potentially be designed in such a way that small subunits would be triggered in synchrony so that the overall global response of the actuator would be much faster in comparison to common actuators relying on diffusive processes. The main challenge here is to create materials with a similar microscopic organization. Unfortunately, still to date, the fabrication techniques reported to obtain such microstructuring of synthetic hygroscopic materials are challenging and expensive, in the perspective to generate large-scale actuators based on this approach.^[11]

3.2.3. Mechanical Amplification

The decisive factors that make the difference between a feasible upscaling and an unviable application lie in the actuator ability to follow a defined trajectory and to bear a given load. Hence, upscaling may be apprehended in terms of displacement and load-bearing capabilities, which could be referred to as mechanical amplification features. As far as material-based actuators are concerned, this field of research is still almost pristine to the best of our knowledge.

In terms of single movements, large displacements are usually targeted. Among the possible approaches, the scheme made of individual cells yielding to a macroscopic movement described by Poppinga et al.^[106] summarizes an interesting approach to reach finite displacements. Another solution lies in hinge-like structures, which are easy to implement to obtain an enhanced response in actuation. To program a movement into a material-based actuator, a coupling effect can be used by generating responses to simultaneous changes in relative humidity and temperature. The main issue here is the considerable difference in the responsiveness between both stimuli for most materials. This approach would be beneficial when the range of magnitude of the CHE and CTE are comparable or when the expected relative humidity variation can create a similar response to typical temperature changes (or vice versa). Additionally, in real-life applications, it may not be practical to pursue coupling effects since there is no real control over the environmental conditions. In this case, it is relevant to consider whether the benefit is significantly greater than triggering an isolated response. Noteworthy, this remark should be limited to simple applications where unidirectional bending is expected. Finally, it might be worth exploring self-responsive actuators based on the release of prior stored elastic energy (comparable to the behavior of the Venus flytrap described in the introduction) which, instead of relying on a mechanical activation, could use specific environmental conditions to trigger a sudden and large movement. Interestingly, buckling could successfully match both objectives of low actuation time and ample displacement. However, the overall thickness in the system would be increased which might cause problems in robustness and long-term stability. Here, a robust design of the materials systems is necessary to tune this buckling carefully.

In the case of complex movements, ambitious innovations like those envisaged by Poppinga et al.^[106] where the actuator should produce Gaussian curvatures with integrated tropistic and nastic responses, would require a vastly more complex design and calibration of a multivariable system. In a complex 3D trajectory, the kinematics could be handled by rigorous computation to control the actuation response. Remarkably, complex multistage kinematics might be achieved thanks to the coupling of distinct time response zones within the structure. The study from Tahouni et al.^[10] gives an interesting insight into this approach to achieve a complex and programmable motion. From a biomimetics point of view, it should be noted that natural structures such as those observed in pine cones or silver thistle bracts would not be conceivable without the anisotropic microstructuring at different scales. Consequently, it is improbable that a complex controlled response could be attainable in an artificial assembly without

turning to anisotropic structuring. However, the concept of anisotropy should not be simply regarded as a preferential orientation in each layer (for instance, fibers in a composite material) that alters particular material features such as in-plane and out-of-plane responses. Instead, using aspect ratio and characteristic size of anisotropic features as tuneable variables could lead to a complex response in both space and time domains.

When it comes to large-scale applications, the reaction time capability of the structure's actuation function remains a major challenge. Another crucial point lies in the bearing capability of the resulting structure. In fact, besides the force to be transmitted by the actuator in the application, another important challenge is the gravitational effect on larger structures, as they may collapse when the actuator reaches a certain upper mass limit. Therefore, it is essential to include gravity as a relevant variable, regardless of the approach used to design the actuator in general, which is a premise for upscaling. Furthermore, getting larger actuator requires also to cope with the inherent issues associated with diffusion and thickness as previously mentioned. Possible solutions can be envisaged by turning to honeycomb structures like those used in aeronautics. In this respect, Barrett and Barrett^[124] suggested the fabrication of a high-pressure adaptive actuator for commercial aircraft wings. Such structures display outstanding performance in terms of maximum strain versus blocked stress compared to conventional hydraulic and pneumatic actuators. Nonetheless, this particular application is highly dependent on an external pneumatic stimulus. Optimally, the ideal synthetic material should combine for instance a charge-proof honeycomb-like structure with an autonomous response to environmental stimuli.

4. Conclusion

Motile plant structures have proven to be suitable concept generators in numerous biomimetic approaches, as their versatile, fail-safe, and efficient motion sequences can be abstracted in many cases and transferred to novel material systems. This applies in particular to systems that react autonomously and self-sufficiently to changes in environmental conditions (e.g., humidity and/or temperature). Nastic movements, which are based on the anatomy and morphology of the respective motile structure, have been realized in a variety of ways, as summarized and discussed in this article. However, far less attention has been paid to tropisms, i.e., movement responses depending on the direction of the triggering stimulus. They represent the "holy grail" of plant movement biomimetics and could dramatically change the field of actuator development if versatile and reliable methods for their technical realization are developed in the future. In addition, self-sufficient biomimetic compliant systems, which autonomously react to environmental changes, usually lack user controllability. New developments in this area could dramatically increase the acceptance and use of such new and "smart" actuator systems.

In summary, the present research review provides an overview of the most important advances in the development of autonomous mesoscale actuators based on synthetic materials. Nevertheless, up to date, all actuators based on synthetic materials are not easily scalable, which hinders the potential large-scale

applications in, for example, architecture and civil engineering. Furthermore, the most relevant factors for the design of material actuators that be subjected to various stimuli at the macroscale have been highlighted and may be considered for future innovative developments and proposals that can successfully overcome the size and applicability limitations shown in the studies published so far.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

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