

A Perspective on Cephalopods Mimicry and Bioinspired Technologies toward Proprioceptive Autonomous Soft Robots

Goffredo Giordano,* Marco Carlotti, and Barbara Mazzolai*

Octopus skin is an amazing source of inspiration for bioinspired sensors, actuators and control solutions in soft robotics. Soft organic materials, biomacromolecules and protein ingredients in octopus skin combined with a distributed intelligence, result in adaptive displays that can control emerging optical behavior, and 3D surface textures with rough geometries, with a remarkably high control speed (\approx ms). To be able to replicate deformable and compliant materials capable of translating mechanical perturbations in molecular and structural chromogenic outputs, could be a glorious achievement in materials science and in the technological field. Soft robots are suitable platforms for soft multi-responsive materials, which can provide them with improved mechanical proprioception and related smarter behaviors. Indeed, a system provided with a “learning and recognition” functions, and a constitutive “mechanical” and “material intelligence” can result in an improved morphological adaptation in multi-variate environments responding to external and internal stimuli. This review aims to explore challenges and opportunities related to smart and chromogenic responsive materials for adaptive displays, reconfigurable and programmable soft skin, proprioceptive sensing system, and synthetic nervous control units for data processing, toward autonomous soft robots able to communicate and interact with users in open-world scenarios.

1. Introduction

The light-manipulation properties and the 3D surface textures of the skin of biological species have always fascinated the

G. Giordano, Dr. M. Carlotti, Dr. B. Mazzolai
Bioinspired Soft Robotics Laboratory
Italian Institute of Technology
Viale Rinaldo Piaggio 34, Pontedera, PI 56025, Italy
E-mail: goffredo.giordano@iit.it; barbara.mazzolai@iit.it

G. Giordano
The BioRobotics Institute
Scuola Superiore Sant'Anna
Viale Rinaldo Piaggio 34, Pontedera, PI 56025, Italy

G. Giordano
Department of Excellence in Robotics & AI
Scuola Superiore Sant'Anna
Piazza Martiri della Libertà 33, Pisa, PI 56127, Italy

 The ORCID identification number(s) for the author(s) of this article can be found under <https://doi.org/10.1002/admt.202100437>.

© 2021 The Authors. Advanced Materials Technologies published by Wiley-VCH GmbH. This is an open access article under the terms of the Creative Commons Attribution License, which permits use, distribution and reproduction in any medium, provided the original work is properly cited.

DOI: 10.1002/admt.202100437

scientific community.^[1–7] Regardless, every time a living being reveals novel adaptive and dynamically reactive mimicry behavior, it inspires and fosters futuristic and unexpected technological outcomes.^[8–12] At the biological level, the visual crypsis is the ability of a species to resemble the surroundings by matching the coloration and the geometrical patterns of the habitat. In this sense, a living being can control its appearance optically (thanks to arranged and optimized structures at the mesoscopic scale, by means of pigmentation, or emissive elements) and morphologically (it can show wrinkles and textures on the body to evade detection or observation).^[13–18] Both these mechanisms are characterized by a time response that ranges from milliseconds to hundreds of seconds. In nature, several species take advantage of cryptic abilities, as, e.g., in cephalopods,^[7] crustaceans,^[19] reptilians,^[1,20,21] insects,^[22,23] birds,^[24,25] shells,^[26,27] plants,^[28,29] among many. The biotic color changing and body patterning relate to reproductive, communicative, and defensive and/or predatory strategies. Unfortunately, the nervous or central control-chain system that steers these behaviors in animals and plants, is still somehow fogged for scientists.^[7,30–32] A complete knowledge about their central information process systems, can open to astonishing developments in many scientific branches, from neurobiology^[33,34] to quantum biology.^[35] Undoubtedly, the most debated case of study in the natural world are cephalopods, which not only are highly evolved and specialized in fast adaptive color changing displays, but also are able to actuate their skin to generate 3D patterns, when exposed to specific mechanical, thermal, optical, or chemical stimuli. Soft muscular arrangements,^[36–38] spatially distributed and expandable absorbing components (namely chromatophores),^[39,40] iridescent elements (namely iridophores),^[41,42] and bright white scatterers (namely leucophores)^[43] are responsible for textural, postural and color changing. Moreover, octopuses are a remarkable intelligent species, able, for example, to open a jar or avoid predators in the order of seconds.^[44] Thus, cephalopods are usually regarded as a perfect example of embodied intelligence,^[45] thanks to the symbiosis between their body's mechanics and morphology, and the distributed sensory neuro-motor-control system. Their “learning”, “mechanical” and “material intelligence” will be our lodestars along this review, resulting in a fascinating connection between

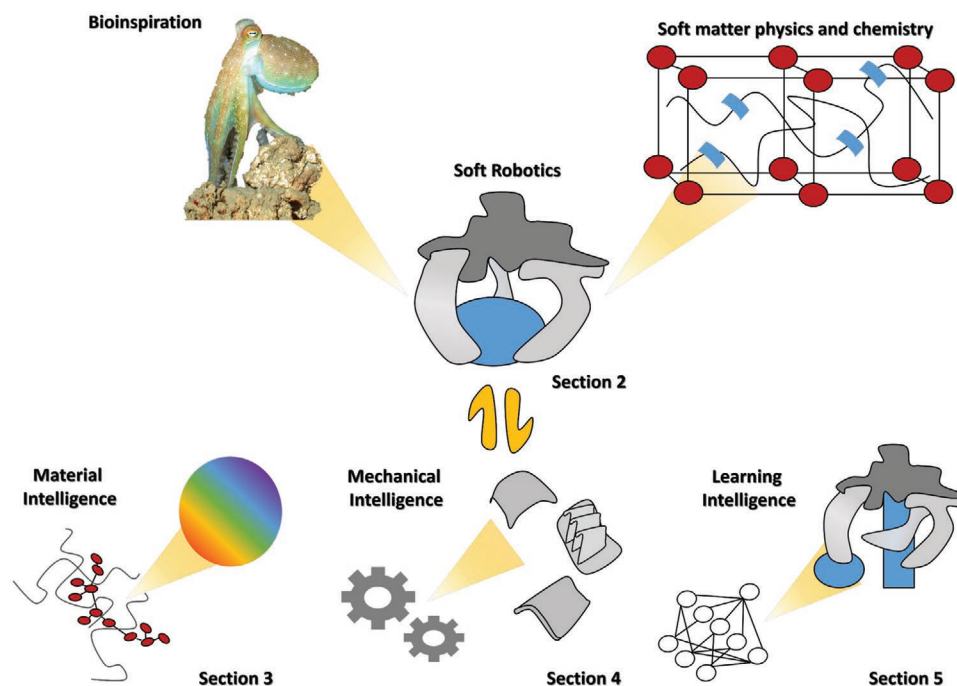


Figure 1. A schematic representation of fusion between bioengineering (robotics and biology) and material science (soft matter physics and chemistry) that can drive to develop bioinspired soft proprioceptive robots. Like biological organisms, a new era of machines is expected to act autonomously in open-world scenario. To achieve this challenge, they require a physical intelligence (i.e., mechanical and material intelligence) to adapt, conform, be resilient and independently behave due to internal and external perturbations, and a computational/ artificial neural intelligence to learn and integrate with collective systems. Interdisciplinarity between the soft robotics and the soft matter communities can open to astonishing adaptable autonomous and stimuli-responsive bioinspired machines. Biology is a source of inspiration for the bioengineering, and the latter is further trying to translate the adaptation principle in robotics. Thus, soft robotics, aimed at developing robots with high deformability, adaptation, gentle interaction with the environment, is growing fast, promoting the spreading of such autonomous machines in the open-world day-life. The “material” and “mechanical intelligence” of some species as, e.g., cephalopods are inspiring roboticists and scientists to develop adaptive chromogenic and texturing displays to effectively communicate with the external environment. At the same time, the responsiveness and ease of computation of the information by means of biological nervous system can be a source of inspiration for innovative processing data system, with the aim of innovative biologically inspired machine learning architectures. Fusing the “material”, “mechanical” and “learning intelligence” borrowed from biological platforms, in the proximal future we envisage new soft adaptable, autonomous and proprioceptive robots able to work in various scenarios. Picture of octopus by NURC/UNCW/ NOAA on Flickr, used in the public domain.

the biological world and bioinspired industrial and technological outcomes.

In the recent years, an interdisciplinary effort among the soft robotics^[46] and the soft matter communities,^[16,47–50] is laying the groundwork for innovative bioinspired sensors, actuators and control systems aiming at proprioceptive, multi-integrated, and autonomous robots (**Figure 1**). In particular, soft robots can be regarded to be prominent examples of a perfect multi-level platform. Innovative materials, mechanical design and learning control can provide fast, adaptive, and on-demand mechanical and optical displays to tackle various tasks in different scenarios.^[51] Taking inspiration from biology, by combining the basic principles of the cephalopods’ neuro-motor-control system and the secrets of their skin, roboticists and researchers can develop deformable and compliant materials capable of sensing the external light and hierarchically react changing the texture and coloration,^[52] thus obtaining dynamic learning control systems. Such adaptation is of paramount importance for a soft robot, which is required to reliably perform in different environments. To the best of our knowledge, there are no soft robots coexisting in open-world scenarios with humans or

other living beings, pointing to the fact that the latter is still an open challenge for robotics. Indeed, despite the development of many bioinspired soft robotic end-effectors as arms,^[53,54] graspers,^[55–57] or platforms as crawlers,^[8,58,59] coilers,^[60] multi-gait^[61] and growing robots,^[62,63] there is a large demand for innovative enabling technologies able to reduce mechanical and control complexities. A viable option could come from the “material”, “mechanical” and “learning intelligence” of the end-effector or robot, furthermore avoiding the presence of elements that hamper the intrinsic softness of the robot body (e.g., wirings, tubes, motors, bulky batteries, rigid skeletons, etc.).^[64] The concept of “mechanical intelligence” here will be entrusted as conformability, morphological adaptability, and the self-capability to real-time react while interacting with the environment. Along the perspective, the “material intelligence” will be intended mostly as conferring smart mechano-responsive properties to the soft constitutive materials. Nonetheless, more in general, it can regard multi-X capabilities (functional, modality, physics, etc.), self-X (adaptation, healing, powering, growing, etc.) and taxis (chemo-, pH-, photo-, magneto-, etc.) behavior, physical (re)programmability, modularity, and more in general

various stimuli-responsive features.^[65] These smart material properties have to report reliable and prompt information on the status of the end-effector, or on the interactive robotic body region, while actuated or while encountering environmental stimuli. Finally, a “learning intelligence” is required to pose robotic platforms able to measure, interpret and self-adapt to complex and real-world scenarios. The learning intelligence should formally lie in an artificial control brain that could be centralized or distributed. With the concept of learning, we gather together the complex system of a distributed network of neuromorphic interfaces along the robotic body or fully endowed at the end-effector tip, and the computation mechanisms, that should be able to acquire, analyze, recognize, and process data and patterns.^[66] Thus, the learning and recognition intelligence should act as an encoder of the physical intelligence^[67] (hereafter an interchangeable term to label the combination of mechanical and material intelligence). This challenging achievement should drive toward proprioceptive and embodied intelligence-based innovative machines that can mediate responses to external stimuli by sensorial capacities, or, e.g., optically and mechanically reconfigure to replicate camouflage as in the biotic counterpart.^[51] Thus, there is an increasing demand for miniaturized task-responsive sensor-actuator-controller solutions, directly embeddable in robotic platforms or manipulators, able to communicate and process data in efficient computational “artificial brain” programmed to learn fast and independently act in a various and unstructured scenarios, as, e.g., destroyed buildings due to catastrophes looking for survivors, or for cataloguing biodiversity or animal groups behavioral.^[68] Additional remarkable results could comprise: energy on-demand saving,^[47,69–72] effective communication in the working-task environment responding to the ambient stimuli,^[73] as, e.g., signaling a danger, a touch occurred, a forthcoming breach (for human-machine interaction),^[20,74–77] providing untethered actuations,^[78–80] and/or, more in general, infusing the soft robotic system with an effective mechanical proprioception.^[81] In particular, bioinspired smart chromogenic responsive materials and 3D shape changing soft actuators/platforms are conducive for a broad ensemble of applications ranging from behavioral life-science,^[82] up to industrial and practical use. The case for mimicry and cryptic behavior in soft robotics includes exploration, monitoring or even rescue in natural environments, where camouflaged robots may hide, and be protected from animal attacks, or even approach them for studying their behavioral and living world, and consistently validate scientific hypothesis.^[83] Of course, soft camouflaging robots may also support surveillance^[84] in harsh or military scenario.^[85] However, optically and mechanically adaptive soft platforms open up to camouflaging electronic displays^[86] and wearables,^[87] or at the same time can find application in the field of soft robotic facades with energy-saving feature,^[88] that can modulate energy harvesting or show heat adaptation shields.^[89] Further applications can result in soft robots for entertainment, fashion, and generic interaction with humans or living-beings.^[90,91]

In this review paper, we will focus on multifunctional and embeddable systems for soft robotics to manipulate income/outcome optical feedback for communicative purposes, or for perceiving the external stimuli in order to modify the shape

conformation to address a task, addressing efficient bioinspired learning approaches. We will do this referring to cephalopods, analyzing their biological structure and function, and comparing them to the state of the art of their man-made technological counterparts. Thus, in Section 2 we will firstly present some bioinspired soft robots, as platforms on which to embed the smart sensors, actuators, designs and control systems, to obtain environmentally responsive and adaptable machines in unstructured scenario. Section 3 will guide the reader through cephalopod’s bioinspired stimuli-responsive and electronics-free chromogenic optical materials, and dynamic photonic structures that can be fully integrated in soft embodiments and actuators to obtain a hierarchically light-matter responsive skin. In Section 4, we will analyze programmable and reconfigurable soft skin for 3D texture patterning, and generic shape-changing actuators for locomotion, grasping and environmentally-driven tasks. Actuation methodologies will be shown to obtain stimuli-responsive pre-engineered smart design. Beside the “material” and “mechanical intelligence”, we will present in Section 5 machine recognition approaches to perform advanced activities and boost robotic “learning intelligence” toward proprioceptive autonomous robots (actually soft robots mostly relies on open-loop systems). This section will provide the last approaches toward a new era of closed-loop proprioceptive soft robots, with biologically inspired machine learning approaches and neuromorphic elements that can profit from the integration of the aforementioned sensors and actuators. Indeed, while the latter are responsible for morphological computing reducing the controller complexity, for more complex tasks, learning will be needed to openly address the interactions between the effector and the environment.^[51,92,93] Again, solutions can arise from soft animals that incorporate a complex network of decentralized control and specialized sensing systems for collecting information from the environment.^[94] One of the biggest challenges in soft proprioceptive robots is multimodal sensing and the synthetic replacement of a nervous system to perceive multiple physical parameters and transfer the data to the de-/centralized control system. Along the section we will discuss how a distributed network of embedded soft sensors and neuromorphic interfaces can drive proprioceptive information to the artificial brain able to compute and process data. Physical intelligence should be respectfully integrated with software algorithms including neural networks, central pattern generators, behavior trees and modular architectures, optimized with reinforcement learning, evolutionary algorithms and imitation, to demonstrate innovative working strategies toward proprioceptive autonomous artificial neuro-motor-control systems.^[95–97] In this vision, we require new materials acting as smart bidirectional transducers that can provide physical intelligence, while at the same time integrating sensing, actuation and computational functions for learning and model-predictive closed-loop control.^[66] The new paradigm that we will analyze consists in the fusion of an artificial physical-intelligence with computational- and neural-intelligence (hereinafter terms to also refer “learning intelligence”). This is a most prominent frontier to develop proprioceptive and exteroceptive soft machines with capabilities of mimicking and changing optical and mechanical appearance. In the same section, we will summarize and comment on current challenges in the area of data-processing,

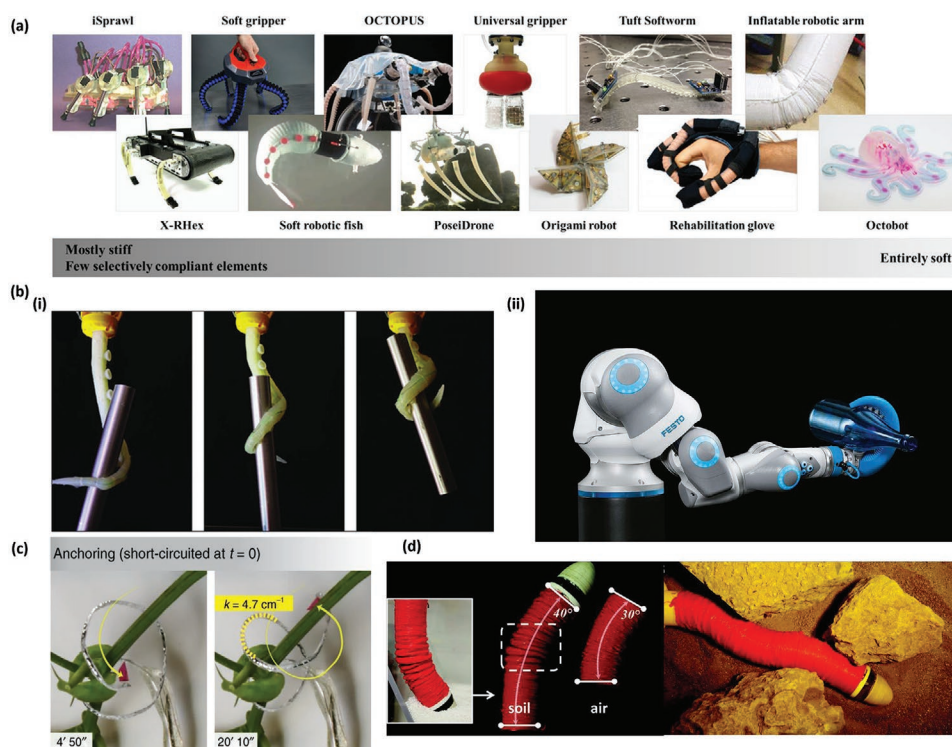


Figure 2. A selection of bioinspired robots and a representation of their evolution in terms of body compliancy and mechanical intelligence. a) The soft robotics evolution, where rigid skeleton and stiff components are going to be substituted to obtain an entirely compliant system. Reproduced with permission.^[46] Copyright 2016, American Association for the Advancement of Science. b-i) An octopus-inspired soft robotic arm developed to work in pipeline-like constrained and confined environment. Reproduced with permission.^[53] Copyright 2019, Wiley-VCH GmbH. ii) TentacleGripper based on the octopuses' tentacle for form-fitting grasping. Reproduced with permission. Copyright Festo SE & Co. KG, all rights reserved. c) A reversible and variable-stiffness tendril-like soft grasper to adaptively and gently coil around soft bodies. Reproduced with permission.^[60] Copyright 2019, Nature Publishing Group. d) Self-growing robot with embedded additive manufacturing system for locomotion in unstructured and complex environment with passive morphological adaptation. Reproduced with permission.^[129] Copyright 2019, Mary Ann Liebert Inc.

machine learning, and control to address various geometrical and optical conformation, while details about control in general for soft robotics^[98] will be mentioned by means of some representative work. In conclusion, along the perspective, the adaptation concept in soft robotics will be treated as a multi-level approach to “sense”, “conform” and “learn” toward the sought embodied intelligence for mechanical proprioception to address unstructured environments and tasks.

2. Bioinspired Soft Robots

Soft robotics offers innovative technological platforms able to adapt and react to external stimuli, providing safer and more robust interaction with the humans and the environment.^[98] These capabilities overcome the limitation posed by rigid materials and components, and open to applications impossible to achieve with conventional robotic systems.^[98,99] For example, the latter require high control complexity and do not allow passive adaptation. Contrary, for soft robotics the intrinsic intelligence of the constitutive materials (granular media,^[100] elastomeric and polymeric materials,^[101] hydrogels,^[102,103] showing a vast range of compliancy, i.e., from 10 kPa to 1 GPa^[104]), and the vast range of actuation, allow to adapt to the environment,^[105] gently interact with unstructured and complex objects,^[106] show large

deformation and shape- and size-changing,^[107] self-repairing from damages,^[108] embed multi-physics properties,^[109] and react with the external stimuli with (pre)-programmed or real-time output,^[110] in most of the cases even reducing the complexity of the control system (Figure 2a).^[69,46,111] In particular, soft matter can easily integrate smart multifunctional micro- and nanoscale systems, such as multi-responsive chemical embedded units, and disruptive transferring-data paradigms. From this perspective, robotics borrowed the concepts of “material” and “mechanical” intelligence from biology to replicate adaptation. Thus, robots with physical intelligence found several applications in biomedicine,^[112] different locomotion modalities^[113] as crawling on different terrains,^[114,115] overcoming obstacles,^[116] and self-healing.^[117] In this regard, several bioinspired solutions came from the study of the abilities and characteristics of cephalopods. In Figure 2b-i,ii we report some example of octopuses' bioinspired soft grippers,^[118,119] that exploit the flexibility of the soft material and the integration of suction cup elements^[120–124] to firmly grasp objects in confined, constrained,^[53] or open environments.^[125] In such examples, bioinspiration allows to solve some critical issues, such as coiling without damaging the organic or inorganic support^[60,126] (Figure 2c), not interfering with the surrounding ecological environment,^[127] or avoiding obstacles in simulated rescue situations^[128,129] (Figure 2d). The softness of their body structure, the theoretically infinite degree

of freedoms of soft manipulators, and their adaptability to different objects and terrain are a herald for the development of innovative technological machines. Nonetheless, at the present stage both soft manipulators and robots still require further refinement to succeed in an open-world scenario, with its infinite variabilities. In the following sections, we will examine multi-responsive soft matter technologies capable to provide soft robots with the ability to change their optical and mechanical appearance for communicative or user-machine interaction (“material” and “mechanical intelligence”). Finally, we will discuss different data-transfer and process approaches for open-world tasks (“learning intelligence”).

3. Smart and Chromogenic Responsive Materials for Adaptive Displays in Soft Robotics

In the recent years, several research groups fabricated exemplary soft robots comprising smart materials (“material intelligence”) that allowed them to change optical appearance in laboratory controlled environments.^[130–134] At the same time, electronics-free chromogenic mechanosensing have also attracted much attention, proposing new technological platforms comprising efficient mechanochemical systems and tunable dynamic photonic strategies directly embeddable in elastomeric host matrices. In the future, such technology could also allow machines to manage optical data on-demand, consequently implementing safe body patterns and texture, and providing visual feedback to interface and communicate with the external environment.

3.1. Cephalopods Camouflage Behavior as an Example of Income/Outcome Light Manipulation

The skin of cephalopods shows a hierarchical structure whose light manipulation strategy involves chromatophore, iridophore, and leucophore organs (Figure 3a-i,ii). Cephalopods’ abilities in this regard and the underlying mechanisms were described extensively.^[7,36–39] At the technological level, the camouflaging behavior is a perfect example of multifunctional devices for sensors, actuators and a fast/efficient control of the skin relying on light absorption, reflection, transmission and emission light^[135] (Figure 3b-i). A soft robot endowed with a synthetic light-manipulating skin could provide an optical feedback as a result of a mechanical perturbation, that can be used to communicate information about its status (e.g., high stretching, mechanical failure), or a completed task (e.g., grasping successfully occurred), etc. (Figure 3b-ii). In this section, we will pose the attention on the organs’ structure, and the state-of-the-art technological counterparts. Indeed, scientists developed devices to replicate cephalopod’s chromatophore organ behavior,^[136,137] or multilayered ordered and mechanically-tunable optical components as counterpart of iridophores,^[138,139] or self-assembled nanoparticles acting as the leucophores to widely scatter the light.^[140–142]

3.2. From Chromatophore as Absorbance Elements through Molecular Mechanochromic Devices

In the cephalopod skin, behind a semi-transparent epidermis layer, one can find the chromatophores, pigmented sacculi, that

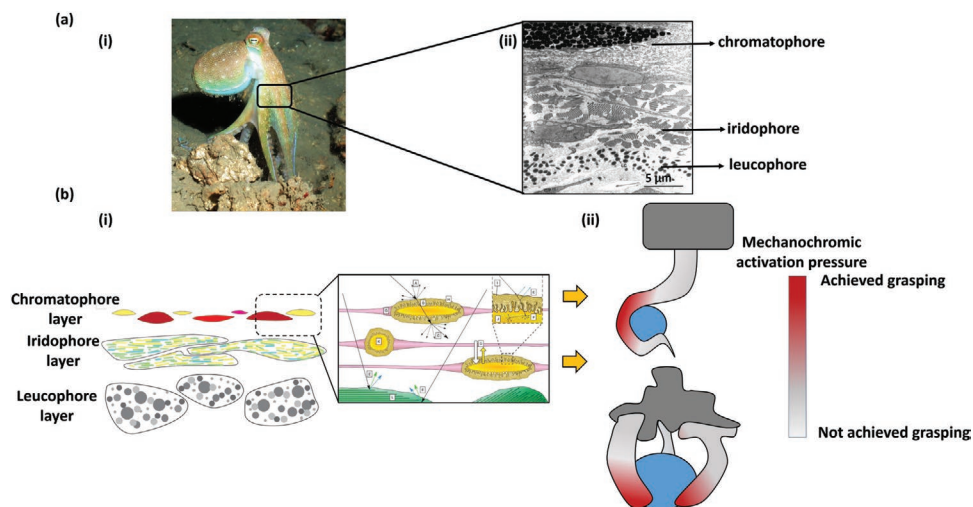


Figure 3. The octopuses’ skin hierarchical structure and the importance of colorimetric feedback to communicate with open world-scenario. a-i) Octopuses change coloration for communicative, reproductive, defensive or predatory strategies. Picture of octopus by NURC/UNCW/NOAA on Flickr, used in the public domain. ii) Hierarchical structure of the octopuses skin (cp = chromatophore, ip = iridophore, lc = leucophore). Reproduced with permission.^[40] Copyright 2001, Wiley-Blackwell. b-i) Left: pictorial reproduction of the optical behavior of the octopuses’ skin. Chromatophores act as a pigmentary absorptive elements, iridophores and leucophores as structural reflective component. Illustrated by Ariel Zych. Right: inset of the behavior of the yellowish chromatophore, with internal structural reflection of spherical and lamellar components. Reproduced with permission.^[135] Copyright 2019, Nature Publishing Group. ii) Pictorial representation of a soft robot communicating with the external environment whereas a grasping movement is applied. The sensible changing of coloration by mechanosensing can be decoded, e.g., by an applied pressure, or a by the approaching of the material failure threshold. The color legend is reported and represents a color-based visual feedback. Color-based outputs are fundamentals for alarming or communicating with color-sensitive living being or human-made detectors.

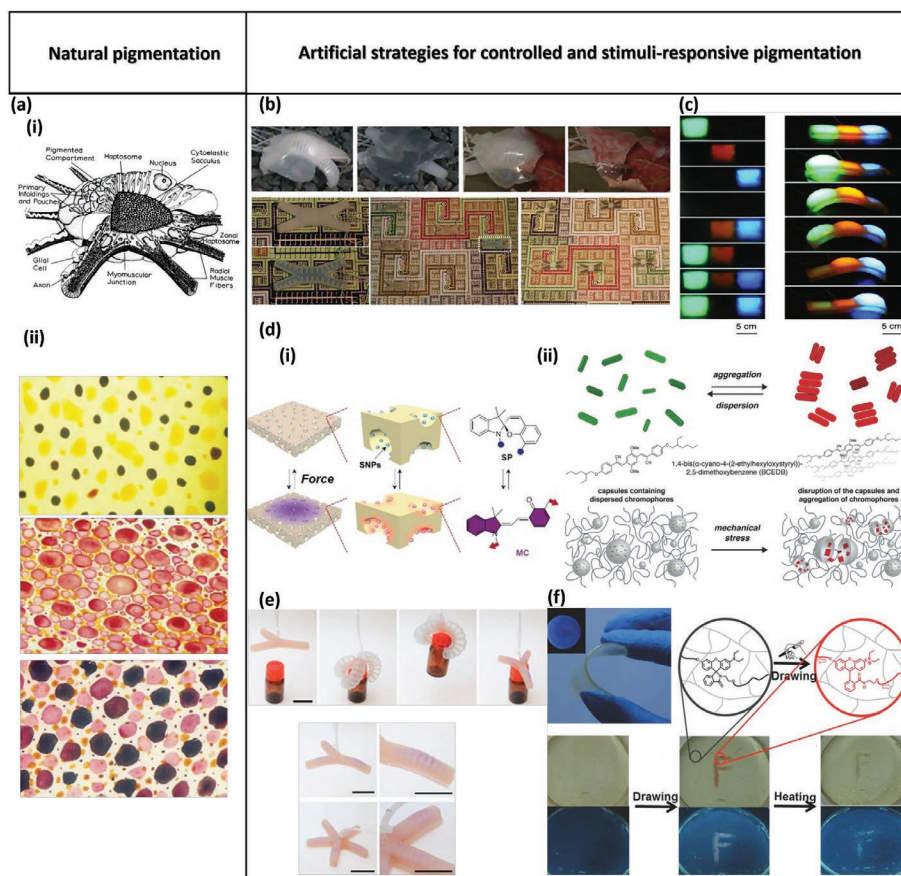


Figure 4. Octopus-inspired pigmentation and artificial strategies for controlled and stimuli-responsive colorimetric behavior. a-i) Internal structure of a chromatophore. Radial muscle fibers are responsible for the increasing or decreasing area of the optical filter element. Reproduced with permission.^[40] Copyright 2001, Wiley-Blackwell. ii) Typical palette in cephalopods' skins. Reproduced with permission.^[40] Copyright 2001, Wiley-Blackwell. b) Fluid-driven camouflaging soft robot. Reproduced with permission.^[8] Copyright 2012, American Association for the Advancement of Science. c) Optoelectronically-driven changing coloration for optical signaling. Reproduced with permission.^[145] Copyright 2016, American Association for the Advancement of Science. d-i) A polymeric soft skin with micropore confined mechanochromic nanoparticles. Reproduced with permission.^[161] Copyright 2019, Wiley-Blackwell. ii) An encapsulated mechanoluminescent excimer-forming dye blended in a polymeric matrix. Reproduced with permission.^[162] Copyright 2018, Wiley-Blackwell. e) SP-doped Ecoflex for a soft gripper robot. Mechanochromism is exploited for spatial-resolved grasping identification regions. Reproduced with permission.^[75] Copyright 2015, American Chemical Society. f) Rhodamine-doped PU for mechanochromism and photochromism. Reproduced with permission.^[178] Copyright 2015, Wiley-Blackwell.

can assume different colorations such as brown, red, black, orange or yellow (Figure 3a-ii).^[40] Each sacculus is surrounded by about 15–25 radial muscles.^[143] A dense and packed system of radial nerves is responsible for reconfigurable pigmented displays, that continuously change appearance expanding or retracting the pigmented area (Figure 4a-i,ii).^[7] Technologically, a synthetic soft and dense chain-like system can be reproduced by polymeric matrices, which show a large range of mechanical performances, from elastomeric to brittle. The optical appearance of a polymer can vary by dispersing dyes and pigments or by covalently binding them to the main chain.^[144] For example, Morin et al.,^[8] proposed the use of colored solutions in fluidic channels in a soft robot to control camouflage in predetermined backgrounds (Figure 4b). This classical approach requires external pigmented-fluids tank or an entangled network of fluidic channels to cover all the surface of the camouflaging body, failing an adaptive approach. Other researchers introduced optoelectronically active layers into elastomeric matrices to electronically control the optical outcomes (Figure 4c).^[145] The

presence of classical electronics (waiting for the advent of reliable stretchable electronics) still present issues such as the different compliancy between optoelectronic components and soft substrates, brittleness and opacity. However, color changing systems with optoelectronic elements showed remarkable results in a soft robotic finger for gesture applications, paving the way for the scaling up of this approach depending on the particular condition.^[146]

Hereinafter, we will analyze multifunctional and optically responsive solutions for soft robotics and manipulators aimed at reducing the use of standard electronics (not compliant with the soft body). An example of this comes from smart polymers and composites comprising functional units able to show a fast and reliable chromatic response to mechanical perturbations without hampering their mechanical performances. In the recent years, mechanochromism based on the variation of absorption^[147,148] or emission^[149–151] properties, lead off as emergent solutions for optical response in soft material like polymers,^[152] self-assembled liquid crystals,^[153,154] or gels,^[155,156]

that find applications in soft robotics.^[157] Mechanochromism is a macroscopic change in the optical properties of a material or device that relies on a selective chemical or structural transformation at the molecular level. In addition, phenomena related to resonance energy transfers (RET) and excimers formation, typical of aggregachromic systems, have also been reported to be responsive toward mechanical stimuli.^[158,159] At the technological level, the mechanochromic polymers are often regarded as mechanically induced on-off optical switch for structural failure or as visual indicator of strain and damage. Mechanochromic units can be covalently bonded to polymer chains^[160,161] or dispersed in the matrix^[150,162–164] (Figure 4d-i,ii), depending on their working principle. The latter, for instance, is typically the case of aggregachromic species where the mechanical stimulation can trigger the disruption of aggregates triggering a dramatic change in optical properties. Conversely, bistable molecular systems, i.e., species that can exist in different thermodynamically accessible states and that can be subjected to interconversion due to external stimuli (such as light, heat, solvent polarity, acoustic, electromagnetic radiation, etc.) can be incorporated directly in the polymeric chain. Extensive reports about the theory and application of such systems can be found in refs. [165–168]. One of the most investigated mechanochromic species is spiropyran (SP).^[75,169] These compounds can switch reversibly to the zwitterionic, blue/violet merocyanine (MC) form upon application of different stimuli. By functionalizing the core, SP derivatives can be covalently linked to several polymers such as polyacrylate (PMMA,^[160] PMA^[170]), polydimethylsiloxane (PDMS),^[169,171] Ecoflex,^[75] polyesters,^[172] polyurethane (PU),^[173] and hydrogels,^[155] showing reversible mechanochromic response over several cycles (Figure 4e). Innovative applications of SP-based systems include stretchable mechanochromic electronic skins,^[161] 3D printed mechanochromic materials,^[174] and visual-indicators for safe surgery.^[175] Next to SP, other mechanochromic compounds find applications as colorimetric strain sensors,^[176,177] touch sensors,^[178,179] chromogenic probes in the visible and near-infrared,^[159,180,181] and in devices for biological^[182] and medical use^[183] (Figure 4f). Similarly, mechanoresponsive luminescent materials, thanks to the high susceptibility of fluorescence emission to chemical and environmental variations, often find applications as precise probes for mechanical stress, heat, pH, and electromagnetic radiation.^[184] Aggregachromic systems, usually based on fluorescent molecules able to organize in precise supramolecular assemblies with defined optical properties (due to the formation of excimers or RET from a donor to an acceptor), are an interesting platform to obtain a finely-tuned and reversible response to several stimuli.^[185,186] Upon mechanical stress, the aggregates can break or modify their structure which affect the optical properties. The nature of supramolecular interactions, comprising a large number of weak bonds, allow a broad and precise transition between the optoelectronic properties of the aggregates and those of the isolated molecules, resulting in a high level of sensibility.^[159,162,186,187] Interestingly, many of these systems could act synergically with covalently-bound ones, yielding to more precise information about the mechanical history of the devices.^[188]

We summarized examples of the most common mechanoresponsive units found in the scientific literature in **Figure 5**,

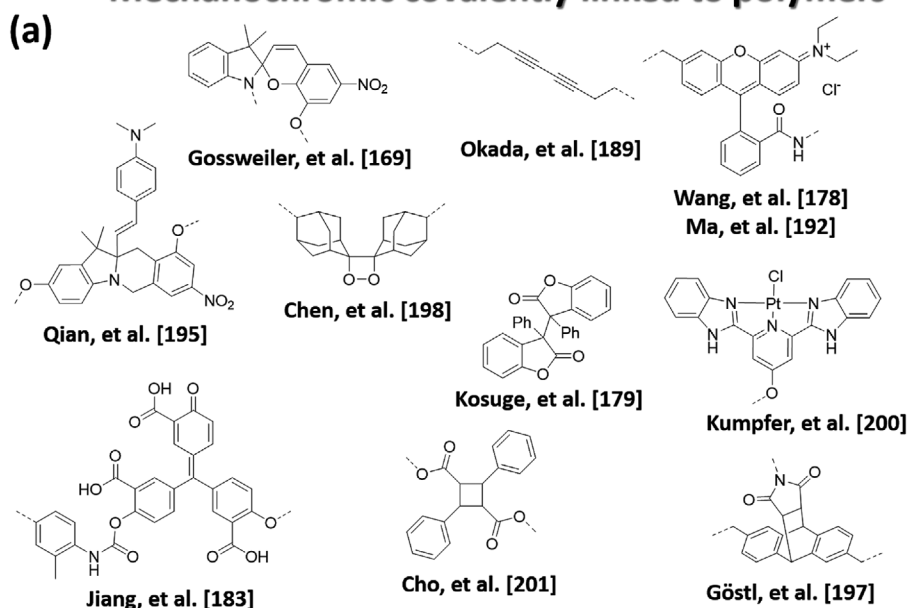
organizing them between those moieties that are covalently linked to polymers (Figure 5a)^[169,170,195–201,178,179,189–194] and those molecules that are employed as dispersions in polymeric matrices (Figure 5b).^[159,162,176,202–204] The list is not exhaustive and for mechanochromic materials we address the reader to the comprehensive reviews of Sagara et al.^[150] and Chen et al.^[205] Moreover, several innovative molecular systems which can display different colors upon change of conformation or crystal structures, could expand the scope of mechanochromic materials in the near future, making more colorations available and improving the performances of the materials in terms of sensibility and reliability.^[206–212] Applications such as sensors, displays, memories, optical waveguides for hydrostatic or flex measurements, and light-emitting fabrics,^[151,186,204,213–217] represent a solid foundation for the spreading of mechanoresponsivity in the robotic communities. The self-healing properties found in several of the aforementioned systems are also of particular interest for soft robotics. We will not discuss them here and we refer the reader to refs. [64,150,217]. The technical framework is ready to image a scaling up of the mechanochemistry in soft machines and grippers, adopting the colorimetric feedback and the manipulation of the income/outcome light, for communication for signaling a danger, damage, or touch occurred or even display some predetermined optical patterns or drawings.

3.3. Iridophore as Ordered Physical Nanostructures with Structural Reflective Behavior

Behind chromatophores, one may find the reflector cells responsible for structural colors ranging from pink, yellow, green, blue, silver and white (**Figure 6a-i**). The nanostructured multi-layered (iridophores) or spherical (leucophores) reflector structures, acting as photonic crystals, produce iridescent coloration by constructive interference or diffraction. Iridophores consist in lamellar stacks of spatially periodic membranes^[4] filled with reflectin proteins and cytoplasm^[42] (Figure 6a-ii,iii). The lamellar aggregation can vary from serpentine, tilted, and parallel assemblies respect to the surface of the skin.^[218] The nervous system controls the peripheral nerves that act on acetylcholine modifying the thickness and the refractive index (n) of the platelets, two physical key-parameters for photonic crystal iridescent response (i.e., photonic stop band, PSB).

The iridophores can be physically modelled as 1D photonic crystals, namely distributed Bragg reflectors (DBRs).^[219] In our day-life, a vast range of applications exploit the principles of photonic crystals,^[220] as antireflective and birefringent coatings,^[221] colorimetric solvent vapor sensors,^[222,223] tunable optical material for smart windows,^[224,225] photovoltaics,^[226] lasing,^[227] and telecommunications.^[228] The manipulated light in presence of 1D photonic crystals is highly angularly dependent (Lambertian like air mode emission profile).^[229] At the technological level, synthetic dielectric mirrors are fabricated by means of alternating layers of oxides or polymers. The former periodical structure (which, e.g., makes use of alternating high- n TiO₂, and low- n SiO₂) are usually prepared via several deposition techniques (e.g., sputtering, chemical vapor, etc.), following the Bragg's law (namely quarter-wave

Mechanochromic covalently linked to polymers



Aggregachromic dyes dispersed in polymer matrices

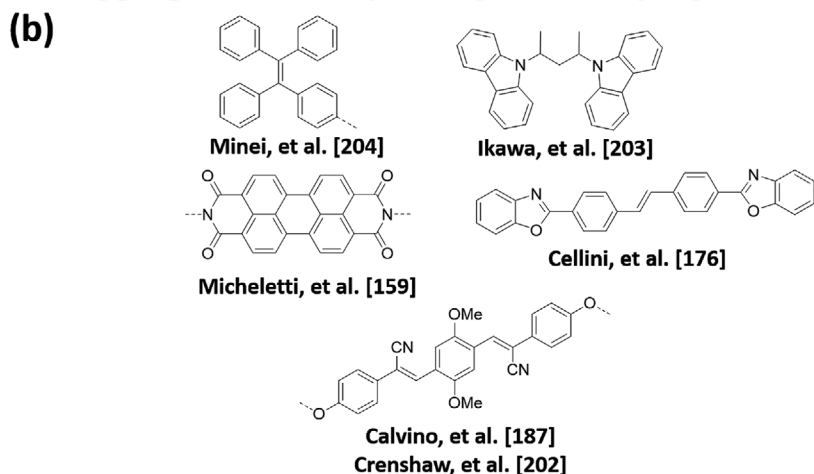


Figure 5. Functional molecular motifs employed in smart mechanochromic polymers. Here the adoption of mechanochromism is entrusted as material intelligence embodiment to replicate chromatophores and muscular-driven natural pigmentation in octopuses. a) Examples of mechanochromic dyes covalently linked to polymeric chains where the color change is related to breaking or rearrangements of bonds. b) Examples of aggregachromic pigments dispersed in polymeric matrices where the change in absorption and emission is due to the different optoelectronic properties of the aggregated and dispersed form of the molecules.

condition) to generate a broad PSB. Incident light is constructively reflected if the thickness of the periodical layers is comparable to the wavelength; whereas the n should be constant in the optical range that has to be reflected and show negligible absorption, in order to modulate the light deflection for constructive interference without losses. Although oxides provide a large dielectric contrast in the quarter-wave stacks ($n_{\text{TiO}_2} \approx 2.6$, $n_{\text{SiO}_2} \approx 1.46$, $\Delta n \approx 1.2$) showing large PSB, they are brittle and suffer the lack of a dynamically tunable PSB, prohibiting the adoption of the aforementioned technology in soft robotics. For optically dynamic solutions, mechanically stretchable 1D polymeric photonic crystals offer several advantages. During

the stretching, the thickness of the alternating polymer layers diminishes, affecting the PSB and allowing tunable structural mechanochromism.^[230] However, in the case of polymers, the n contrast reduces dramatically, i.e., $\Delta n \approx 0.1 - 0.3$ (Figure 6b-i). This narrows the PSB, requiring an increase in the number of the periodic couples to improve the performances (Figure 6b-ii). In Figure 6b-iii we theoretically compare the optical performances of an oxide-based DBR with 7 couple of $\text{TiO}_2/\text{SiO}_2$ and a dynamic strain-dependent polymeric-based DBR with 30 couple of polystyrene-polyisoprene (PSPI)/PDMS (Transfer Matrix Method). Moreover, fabrications issues such as wrinkling, corrugations, defects on surfaces can drastically

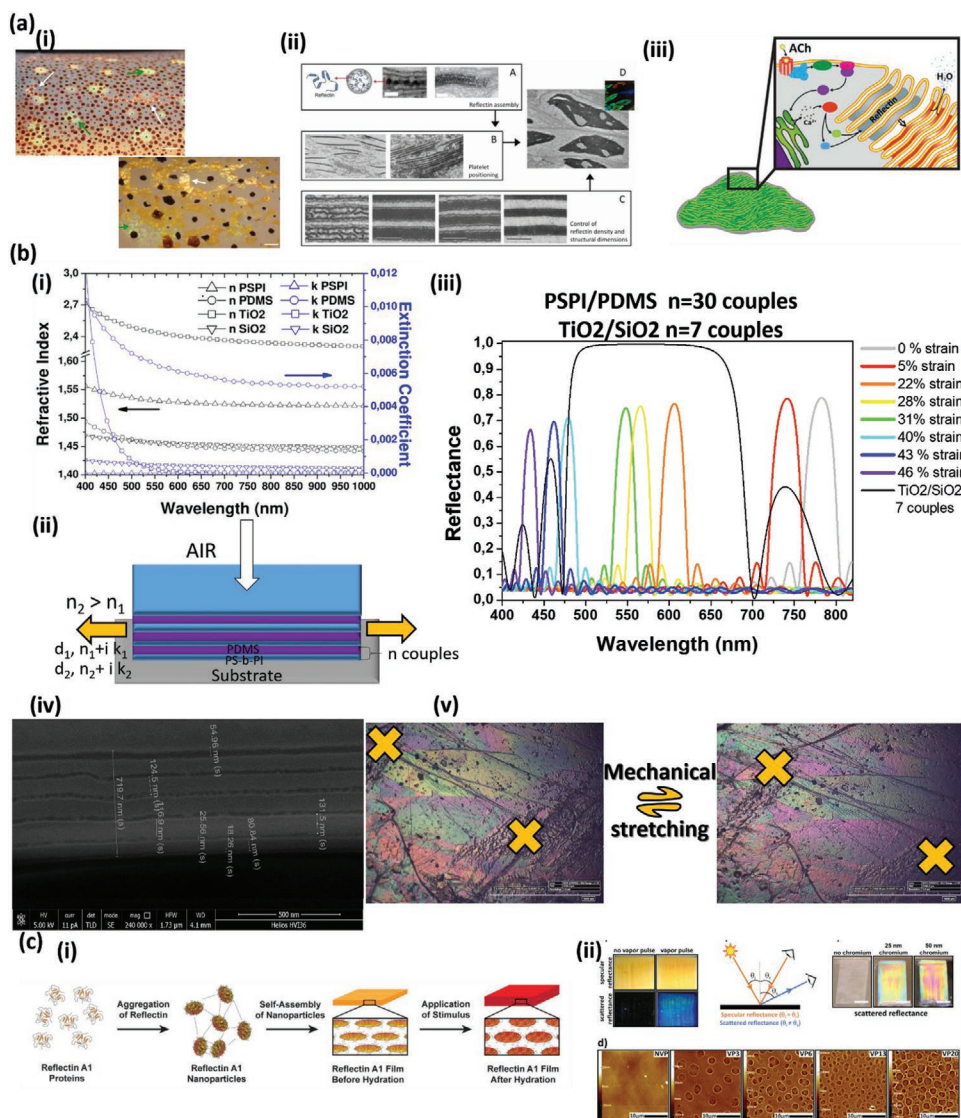


Figure 6. Cephalopod-inspired 1D photonic crystals and biohybrid iridescent systems. a-i) Identification of iridescent regions in cephalopods' skin. Reproduced with permission.^[135] Copyright 2019, Nature Publishing Group. ii) Biological assembly of iridophore in cephalopods. Reproduced with permission.^[13] Copyright 2019, IOP Publishing Ltd. iii) Tunable iridescence is due to the reversible introduction of water or reflectin flux, resulting in a dynamic control on thickness and refractive index of membrane invaginations. Reproduced with permission.^[42] Copyright 2013, National Academy of Sciences. b-i) Refractive indexes and extinction coefficients of oxide (TiO_2 , SiO_2) and polymeric (PSPI, PDMS) dielectrics determined by ellipsometric measurements. ii) Pictorial representation of the polymeric DBR (30 couples of PDMS, PSPI) mechanically tunable by stretching-controlled system with thickness derived by quarter-wave Bragg law. iii) Computation of the photonic bandgaps determined by transfer matrix method for a DBR of 30 couples of PDMS, PSPI under stretching, and a not mechanically tunable DBR of 7 couples of TiO_2 , SiO_2 . iv) SEM acquisition for 5 couples of PDMS, PSPI fabricated by spin coating technique. v) An optical microscope image of a corrugated and not uniform spin coated PDMS, PSPI polymeric DBR. The mechanical stretching underline the blue shifting of some macroregions due to the reduced thickness of the multilayered structure. Yellow cross aim is to help the reader in the identification of the regions where there is the structural mechanochromism. c-i) Self-assembled nanoparticles in reflectin-based solution that changes coloration due to hydration. Reproduced with permission.^[241] Copyright 2016, Wiley-Blackwell. ii) The presence of reflectin aggregation change the scattering characteristics of a reflectin-coated film. Reproduced with permission.^[246] Copyright 2017, American Institute of Physics.

affect the performances. The most adopted fabrication process is spin coating.^[231–234] As example, we report some SEM and optical microscope pictures extracted by a spin coated stretch-tunable polymeric DBR fabricated with PDMS, and PSPI (Figure 6b-iv,v). Further fabrication methodologies include photolithography,^[235] laser-beam diffraction pattern techniques,^[236]

co-extrusion,^[237] roll-to-roll doctor blade coating^[238] and bar coating.^[239,240] In the recent years, an innovative extent is to fabricate polymeric iridescent devices employing materials with a tunable n (i.e., with biotechnological solutions). Remarkably, several studies showed the possibility to exploit self-assembled nanoparticles in reflectin-based spin-coated solutions^[241–245]

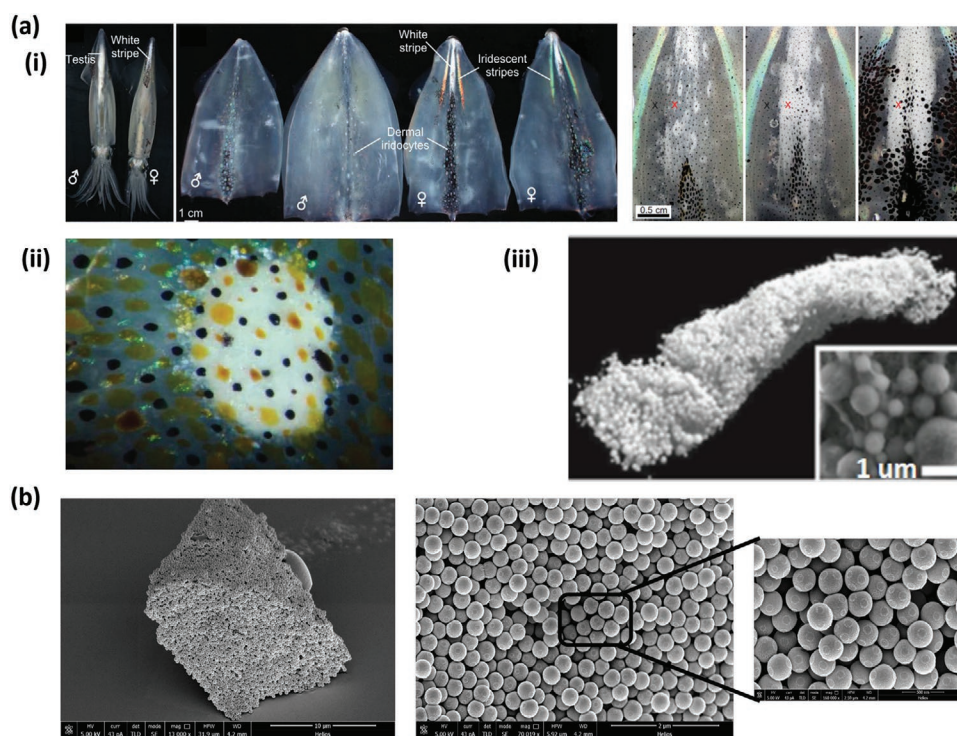


Figure 7. Cephalopod-inspired 3D photonic crystals and spherical self-assembled polymeric counterpart. a-i) Tunable leucophores and iridocytes for reproductive skills. Reproduced with permission.^[248] Copyright 2013, Company of Biologists Ltd. ii) Bright incoherent white scattering due to iii) spherical aggregation. Reproduced with permission.^[251] Copyright 2013, Wiley-VCH. b) From left to right PSPI SEM pictures showing self-assembly of spherical nanostructure as for the biological counterpart.

to obtain layers that react with the external environment (Figure 6c-i), and change their coloration^[246] (Figure 6c-ii). Bioelectronics and biochemistry can lead off to optical and mechanical transduction for dynamic devices (actually in vitro), controlling injection and extraction of protons altering thicknesses and refractive indexes of reflectin-based films.^[247] The latter can shed light on the biological significance about the neural control influx/efflux of ions and water that modify the geometry and spectroscopic evidence of lamellar structures. Despite these futuristic devices indicate new paths about arrangement at the nanoscale of self-assembled protein and their macroscopic optical behavior, further investigation for spatiotemporally control of ion-transportation in biomacromolecules and structural mechanochromism are required for the design and development of advanced octopuses-inspired technologies.

3.4. Leucophore as Incoherent Structural Scatterers

Behind the iridophores there are the leucophores. These broadband scatterers are responsible for white spot in cephalopods^[248] (Figure 7a-i,ii). The light is manipulated by a huge quantity of proteinaceous spheres with diameter ranging from 250 to 1250 nm^[7] acting as a 3D photonic crystal (cells with Mie-scattering spheres, containing reflectin protein).^[249] Leucophores are inherently soft and flat, and there are no nerves or muscles driving them^[250] (Figure 7a-iii). However, behavioral experiments suggest an optimal light tunability for adaptive

background display, modulating brightness and contrast with the environment, thus enforcing the necessity to investigate the neurobiological circuits driving these reflector cells.

At the technological level, synthetic incoherent scatterers were reproduced in various studies.^[140,251–254] The role of the scattered light is ubiquitous in the commercial and industrial markets where spectral coloration purity is required (e.g., smart display, fingerprint identification, energy consumption, fashion, mimetic fabrics, architecture and civil engineering). In general, bottom-up approaches such as self-assembly processes are favored to fabricate 3D photonic crystals or ordered microstructure. For example, di-block copolymers show photonic bandgap in the visible range thanks to self-assembled ordered nanostructures.^[255–259] In Figure 7b (from left to right), a self-assembled spherical structure of PSPI block copolymer dissolved in toluene is reported after a thermal solvent annealing (below 100°C). Cao et al.^[259] showed a promising way to fabricate large-sized nanostructures, investigating the influence of the spin coating parameters and solvent vapor annealing on the self-assembly of multi-geometrical ordered structure. Further, Jacucci et al.^[260] investigated the optimization of the morphology of the scatterer elements rather than the tunability of their refractive index, obtaining silica-based spheroid-cylindrical particles as colloids, thus showing a new class of pigment microspheres for ultra-bright coatings. A vast range of geometrically variable photonic crystals (with different inter-domain distances, sub-100 nm) can be achieved by di-block copolymers resulting from microphase separation (spheres, cylinders, lamellae).^[261] To scale up

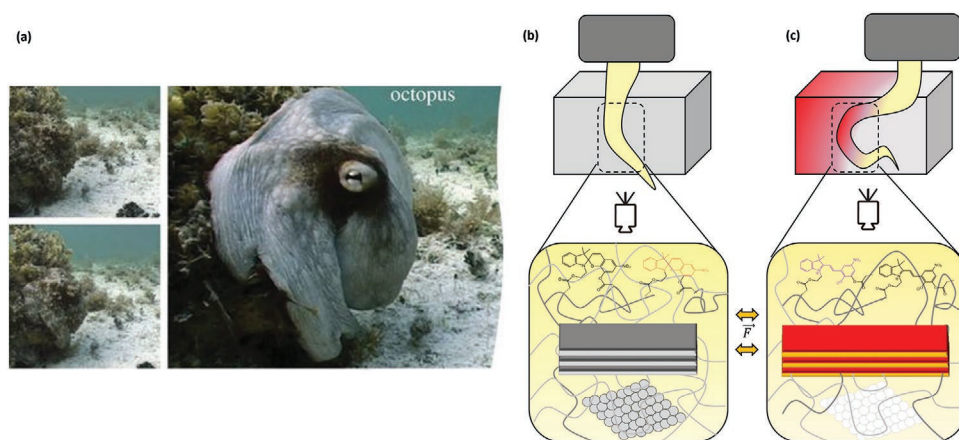


Figure 8. The fascinating bioinspired camouflage behavior can be replicated in a bionic artificial skin for communicative, energy saving, mimicry purposes. a) Cryptic behavior of a cephalopod that camouflages close to a coral reef. Different periods recreate the mechanical and optical camouflage behavior. Reproduced with permission.^[14] Copyright 2013, The Royal Society. b) A pictorial representation of a bioinspired soft robotic arm, whose skin is functionalized with a bottom-up combined and reversible molecular and structural mechanochromic strategy. The polymeric host matrix is endowed with mechanochromic molecules acting as chromatophore counterparts, and multilayered and spherical photonic crystals blended as iridophores and leucophores. c) A macroscopic movement can afford a mechanical perturbation showing a macroscopic changing coloration due to a changing chemical identity of the molecular mechanochromic elements, and different reflectance spectra from the structural mechanochromic modules.

large devices able to modulate photons in the visible and near-infrared light range (350–1500 nm), one needs to control some parameters such as the order-to-order or order-to-disorder temperature, the molecular weight, the volume fraction of block, the neutral solvent dispersion, the film thickness, and the necessity to overcome the smaller 150 nm limit of inter-domain distances (Bragg's diffraction law).^[262]

3.5. Combined Molecular and Structural Mechanochromism in a Soft Robot Inspired by Octopuses' Hierarchical Light Manipulation Strategies

Octopuses exploit mimicry and crypsis as an extent to protect their soft body from predators (Figure 8a). Their strategy fuses the 3D surface body texturing and the environmental adaptive light manipulation (also in benthic zones where light condition is poor)^[263] as we described in the previous sections.

By referring to the chromatic response mechanisms and technological platforms proposed so far, we may envisage a light modulating soft robot capable of enhanced mechanosensing (with continuous optical read-out), and with communicative and energy saving purposes.^[264] Smart chromogenic materials can be also endowed in soft grippers without requiring external power sources, to confer electronics-free reversible optical cues for mechanotransduction at a certain pressure threshold, or achieved grasping. Thanks to the aforementioned bioinspired technologies (counterparts of the cephalopods' organs), a polymeric hosting material could act as a mechanochromic skin, combining both molecular and structural mechanochromism, to achieve a broad spectrum of colorations and patterns. To date, there are very few examples of fully integrated camouflaging skin.^[10,11,265,266] A promising approach, was reported by Clough et al.^[267] In the latter, the authors combined chemical and physical mechanochromism to obtain high dynamic range

mechano-imaging. With a similar approach, we envisage in Figure 8b a hypothesis of octopus-inspired soft manipulator with a hierarchical soft and adaptive camouflage skin system. In Figure 8b,c in place of chromatophores, some mechanochromic molecules are cross-linked to soft elastomeric matrixes. Thus, the soft robotic skin, employing polymeric materials (e.g., polysiloxanes, polyacrylates, polyurethanes, etc.), can be doped with different mechanochromic compounds,^[268] in its entirety or in specific engineered regions, to show multi-color and tunable/adaptable displays upon mechanical stimulation. In achieving this, dopants that do not alter the mechanical properties of the host materials significantly should be preferred. Indeed, it is known that the addition of small amounts of SP do not affect the mechanical properties of bulk polysiloxanes;^[180] conversely, aggregachromic materials may generate colloidal grains that efficiently modifies the structural composition of the soft host matrix.^[186,269] Stretch-tunable polymeric DBRs can substitute organic iridophore. An inorganic counterpart cannot modulate n as effectively as a biological structure relying reflectin proteins or water filling. However, examples combining protein and polymeric films showed remarkable tunability (Section 3.3), opening to the possibility of the integration of biotechnological solutions for the addressed task. At the bottom of the synthetic camouflaging skin, one may engineer different block copolymers blends to increase or modulate the dominant environmental wavelengths to replicate leucophores activities. The possibility to integrate algorithms and control techniques to stretch-tune or mechanically-tune each chromogenic device fully immersed in the hosting material, can provide optical camouflaging features to the soft platforms or end-effectors. As introduced before, such camouflaging devices might be adopted for basic biological behavioral studies in animal communities, or as displays for on-demand energy saver in efficient machines, or for human-interactive devices, or in the field of surveillance or military assets.

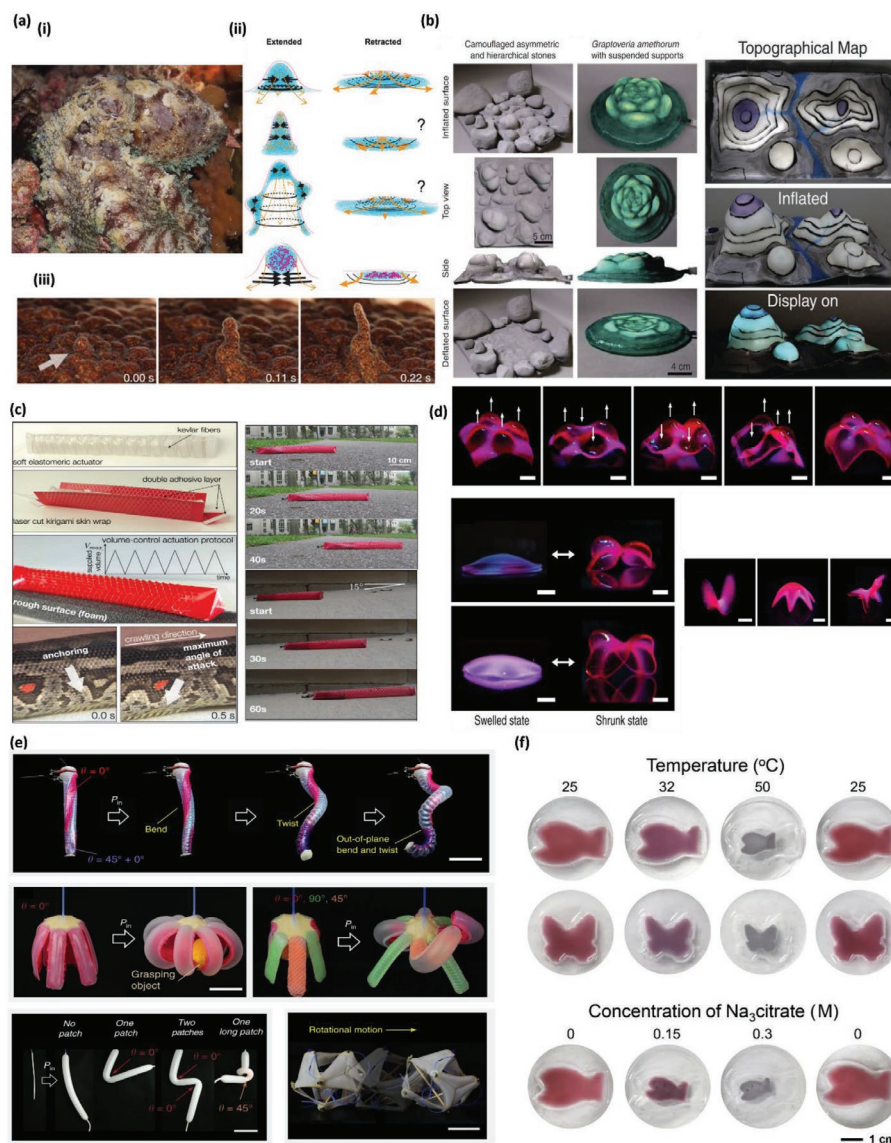


Figure 9. Taking inspiration from cephalopods' skin texturing and cryptic abilities, mechanically intelligent soft robots able to conform, adapt morphologically, and real-time react while interacting with obstacles or environmental constraints, can be developed. a-i) Picture of an octopus 3D texture patterning shaping as coral reef for defensive or predator strategy. Photo by Elias Levy on Flickr, reproduced under CC BY 2.0. ii) Schematic analysis of expression or depression of skin papillae, and iii) periods for the expression in the real octopuses' skin. ii) Reproduced with permission.^[270] Copyright 2014, John Wiley and Sons Inc. iii) Reproduced with permission.^[10] Copyright 2017, American Association for the Advancement of Science. b) 3D texture patterning of complex structure fluidic-driven for synthetic camouflaging skin. Reproduced with permission.^[10] Copyright 2017, American Association for the Advancement of Science. c) A kirigami skin with 3D patterns allowing a soft robot to crawl. Reproduced with permission.^[272] Copyright 2018, American Association for the Advancement of Science. d) Multimodular 3D shape configuration with and without preprogrammed control on the movements. Reproduced with permission.^[110] Copyright 2018, Nature Publishing Group. e) Reconfigurable soft skin for various trajectories and complex movements in soft robotics. Reproduced with permission.^[275] Copyright 2019, Nature Publishing Group. f) Volume and color change of hydrogel architectures by means of temperature and after immersion in deionized water and Na_3 citrate aqueous solution. Reproduced with permission.^[280] Copyright 2016, Nature Publishing Group.

4. Soft Actuators for 3D Surface Skin Textures and Shape-Changing Effectors

Cephalopods are astonishing soft camouflaging machines even thanks to their dynamic 3D physical texturing of the skin. Their hydrostat muscular architecture allow unparalleled cryptic patterns, e.g., the extension of their papillae ranges from flat on the skin (dynamically invisible), to rough geometry

when fully extended (\approx cm order)^[270,271] (Figure 9a-i,ii,iii). Monocular lateral vision (and not tactile) feedback is fundamental for the coordination of the 3D fine texture papillae expression.^[36] Further, the flexibility of the body, the dynamic control of the flexible arms, and the absence of a skeleton permits different body shapes and configurations, highlighting the role of cephalopods as unique living beings in the animal kingdom.

4.1. Reconfigurable Morphing Soft Skins and Programmable Optical Displays

Conformability – the ease of changing shape with programmable design – is a skill typical of living beings. To translate it to “mechanical intelligence” in machines and end-effectors, the designers require expertise in mathematical models for 3D structures in space, soft materials, predictive algorithms, and control systems to analyze external stimuli and respond accordingly. An important example of a synthetic tissue that obtains 3D target shapes starting from a 2D steady state is reported by Pikul et al.^[10] (Figure 9b). They developed a mechanism of synthetic tissue groupings consisted of elastomeric membranes embedded with inextensible textile mesh, actuated pneumatically. The system is independent from the material, thus encouraging the investigation of other soft material structures, which could tackle different issues of operating in open-world conditions. Origami or kirigami techniques are adopted for 3D programmed texturing and patterning design. Rafsanjani et al.,^[272] taking inspiration from snake skin, showed a new methodology based on kirigami, to replicate crawling movements, unlike adopting modular actuation for locomotion (Figure 9c). The mechanical instability induced a transformation from planar structure to a 3D texture pattern that emerged as a promising tool to obtain morphing and highly stretching skin structure. The main actuation methodologies for reconfigurable morphing skins are pneumatically- or fluidic-driven, because they are able to sustain high frequency and to provide fast movements. Nonetheless, programmable smart design approaches for 3D skin texturing can be also applied to soft robot and manipulators that are actuated with dielectric elastomers, liquid crystal elastomers, shape memory alloys and polymers, and swelling hydrogels (Figure 9d,e).^[110,273–276]

At the actuator level, one may find materials responsive to external stimuli, which show complementarity “material” and “mechanical intelligence”. Their use in soft skin layers may result in both a reconfigurable and pre-programmed texturing and patterning, and a time-resolved variable optical appearance at the same time.^[277,278] For example, Kim et al. patterned on a micro mirrors array, a pH- responsive polyelectrolyte hydrogel which acted as an artificial muscle, reversibly bending and switching from an opaque to transparent state.^[279] Temperature variations can modify the volume and color of gold nanoparticles colloids in a poly(N-isopropylacrylamide-co-acrylamide) hydrogel matrix, resembling the formation of predetermined hydrogel architecture (Figure 9f).^[280] Electric fields can be employed for the actuation of a dynamic plasmonic nanostructured cell comprising highly ordered gold/silver nanodomains to real-time manipulate the light and match environmental background, thus working as an active camouflaging display.^[281] Electrothermal actuation can be provided integrating transparent and flexible Ag nanowire percolation networks for a transparent walking robot and gripper with directional movement selectivity.^[282] Electrohydraulic transducer can show muscle mimetic properties with a fast response speed and high positional control, further showing optical transparency in liquid dielectric and the ability to self-sense the deformation state.^[283] Moreover, also by providing light to liquid crystal elastomers is possible to change their surface morphology^[284] while

changing the apparent coloration as well.^[285,286] In conclusion, the “material” and “mechanical intelligence” is exploited in a vast range of demonstrators. However, to obtain a complete camouflaging behavior (both in its mechanical and optical features), especially in a robotic platform, a scaling-up is required, which must also encompass the integration of a “learning intelligence”.

4.2. Programmable and Shape-Changing Actuators for Various Environment and Task

Nowadays, programmable-design and shape-changing actuators are considered particularly appealing to obtain soft robotic platforms capable of expand their competencies, attain multiple manipulations, smartly interact with the environment, and changing conformation in function of complex variables, other than replicate soft reconfigurable morphing skins. For example, taking inspiration from nature – in particular from soft underwater organisms –, researcher studied shape-changing mechanism of actuation able to replicate movement patterns for harsh and complex environment.^[287,288] One may find a vast literature about actuation systems for changing conformation and appearance to perform various tasks, and we recommend further references to the interested readers.^[289–292] One of the first approaches to investigate programmability and structural changes involved the study of small and tiny untethered actuators, whose reversible behaviors were driven by external stimuli such as humidity, temperature, light, etc.^[293–297] However, here we focus on various examples of shape changing soft robots and grippers, and how the latter can improve and foster autonomy and adaptation on a proprioceptive robot while facing a real-world scenario. These studies have shown highly desirable technological advancement for adaptation on different terrains,^[107,298] navigation in confined areas,^[299] lifting or twining around tiny object for precision tasks,^[300] and manipulation in remote areas^[301] or at the microscale.^[302] Thus, the “mechanical intelligence” of a soft effector can allow, e.g., to replicate the wide deformation of the tongue of a reptile, or the winding of vines around objects, opening to bionic results in flexible, accurate, and precise operations up to now impossible.^[300] In this regard, the changing of shape, conformability and self-reaction with the environment is obtained by ultrathin electrothermal actuators, whose large deformation movements are associate to thermal responsible soft silicone thin film, working at temperature below 70°C, showing ability to piecewise control (thanks to multiple electrode-system), and with a small power input.^[300] Nonetheless, a programmable and transformable pneumatic balloon-type soft robot, whose actuation can be tuned by means of a shell-structure, can walk, grab and move like an octopus showing its adaptability to different and variable environment.^[299] One of the main advantages of the programmable actuator with variable bending direction was the lack of necessity to refabricate the structure, that although the shell must be repositioned, opens to future real-time metamorphosis. The morphing adaptation and conformability of end-effectors, and not only as reported ahead for programmable soft skins, are crucial for future advances in hardware (physical intelligence) and software (learning intelligence) embodiments

to obtain shape-changing robots that can address environmental-specific shapes, gait, manipulation, or human-machine interaction. The necessity to integrate the physical intelligence of the effector with the intelligence of the artificial brain will be discussed in the following section, giving to the reader the idea of proprioceptive synthetic system able to decouple deformation and strain-dependent performance, target morphology, optimization of the shape changing or obstacles encountered. As a matter of fact, it is yet unclear how and when the changing of the shape should occur due to the lack of efficient and reliable neuromorphic and nervous synthetic systems that can transfer the proprioception to de-/centralized control system. However, it is widely recognized that programmable and shape-changing capabilities can increase the efficacy of the soft robotic platform and solve control problems.^[107] These challenges open to evolutionary and autonomous soft robotics that with the ease of multi-materials, and optimized control and algorithms, will spontaneously evolve as, for example, reported by Corucci et al.^[303] showing the possibility to change the behavior of a robot from an amphibious to a swimming one.

5. Proprioception and Control System for Soft Robots

Cephalopods' sensorimotor system is neural refined and hierarchically organized so that they can change their total body appearance (both coloration and texture) in hundreds milliseconds.^[304] In the brain, the optic lobes, the lateral basal lobes, and the anterior/posterior chromatophore lobes steer the skin patterns (by means of the neural control of the chromatophores, iridophores, leucophores and papillae).^[305] While the central nervous system controls the neural cells connected to chromatophores, the peripheral nervous system dynamically drives the papillae responsible for skin texture and the iridescent organs (Figure 10a-i). In particular, a peripheral control of the central nervous system allows nerves to dip the papillae, while other nerves are responsible for their expression.^[270] More detail on neural control of the skin papillae can be found elsewhere.^[306] Undoubtedly, while research on cephalopods' brain is still ongoing, technology can move astray, without the need of consistently replicate living beings. Interestingly, there are great expectations on the studies related to the learning approaches of cephalopods, due to their advanced cognitive abilities, and their large nervous system that rivals that of many mammals. Recent discoveries reported that, despite these animals show poor proprioceptive senses, the central nervous system uses peripheral information about the arm motion as well as tactile input to accomplish learning tasks that entail directed control of movement.^[34] The biological studies can be adopted also as benchmark for innovative proprioceptive strategies in robotics.^[307] We envisage that the acquisition and transmission of data inferred by the sensory-motor system ("mechanical" and "material intelligence"), can be provided by stretch-receptive sensor network,^[308] or unravel soft sensors network,^[309] decentralized control,^[94] neuromorphic systems,^[310] toward innovative data processing software-based approaches^[97] (Figure 10a-ii).

As previously stated, soft robots rely on open-loop control strategies, due to the lack of a distributed mechanical proprioception.^[98]

A relevant part of the research in soft robotics concerns control techniques. The topic, however, falls outside of the scope of this perspective and we address the reader to refs. [311–317] to access details about the actual control-technologies adopted in soft robotics and soft manipulators, and the possibilities that those studies can provide fostering, favoring and addressing the solutions discussed along the perspective toward efficient autonomous proprioceptive soft robots. A distributed mechanical proprioception is a fundamental prerogative to achieve autonomous systems, and could be conceived through the interplay between various "synthetic senses" and "synthetic data process systems". For example, Truby et al.^[318] employed a set of sensors for different motions (curvature, inflation, and contact) to measure the spatially distributed state of the soft robot and the object to be grasped.

Soft robots and grippers can benefit from the aforementioned income/outcome light manipulating sensors (as electronics-free counterpart of "vision"), both for communicative skill with the external environment (interventionists/users) and to acquire and process data from the working environment to adaptively conform or generate motion by means of a synthetic neuromorphic system. At the same time, mechanochromic elements could act as smart "touch" sensor if combined with an external vision system or an integrated optical decoder. In this section, we would like to provide the reader with a brief summary of proprioceptive sensors able to monitor changes in shape, motion, and/or touch, and that are also able to send reliable data to an artificial control system, thus acting as the synthetic counterpart of biological synapses and nerves. This issue was debated for a long time in the soft robotic community; however, the fusion between disruptive technologies for sensing and actuation, and a reliable control and process system is still largely missing. First, we will discuss examples of sensing systems for proprioception, to then proceed with neuromorphic elements and computational learning methodologies. An example of the former is offered by stretchable photonic elements integrated in a soft robotic body. In these prototypes, a waveguide allows for an integrated sensor system that does not alter the soft platform or manipulator mechanical structure, while at the same time providing reliable strain-dependent data. The most studied optical strain gauges are fiber Bragg sensor systems (FBG), which act as strain-resolving optical fibers for the shape sensing.^[319–324] However, the use of silica-based materials and the cost of the interrogation systems limits the scope of this technology. Further investigation on polymeric FBGs may allow the spreading of these systems in soft robotics by allowing fine tunability of the properties, ease of fabrication, and mechanical compliance with the host materials. Currently, many researchers have posed attention on novel technologies based on elastomeric soft optical fibers (Figure 10b-i).^[325–329] These latter, can act as pressure, strain, and/or touch sensors^[330,331] by measuring light losses,^[332–337] or exploiting the variation of the optical properties of an embedded fluorescent dye due to the mechanical perturbation.^[338–342] In addition, flexible and stretchable electronics-based systems for e-skin,^[81,343,344] based on resistive^[345,346] and capacitive sensors,^[347–349] are promising platforms for such applications, but they still lack in precision, reproducibility, and signal-to-noise ratio (e.g., in harsh environment electromagnetic immunity

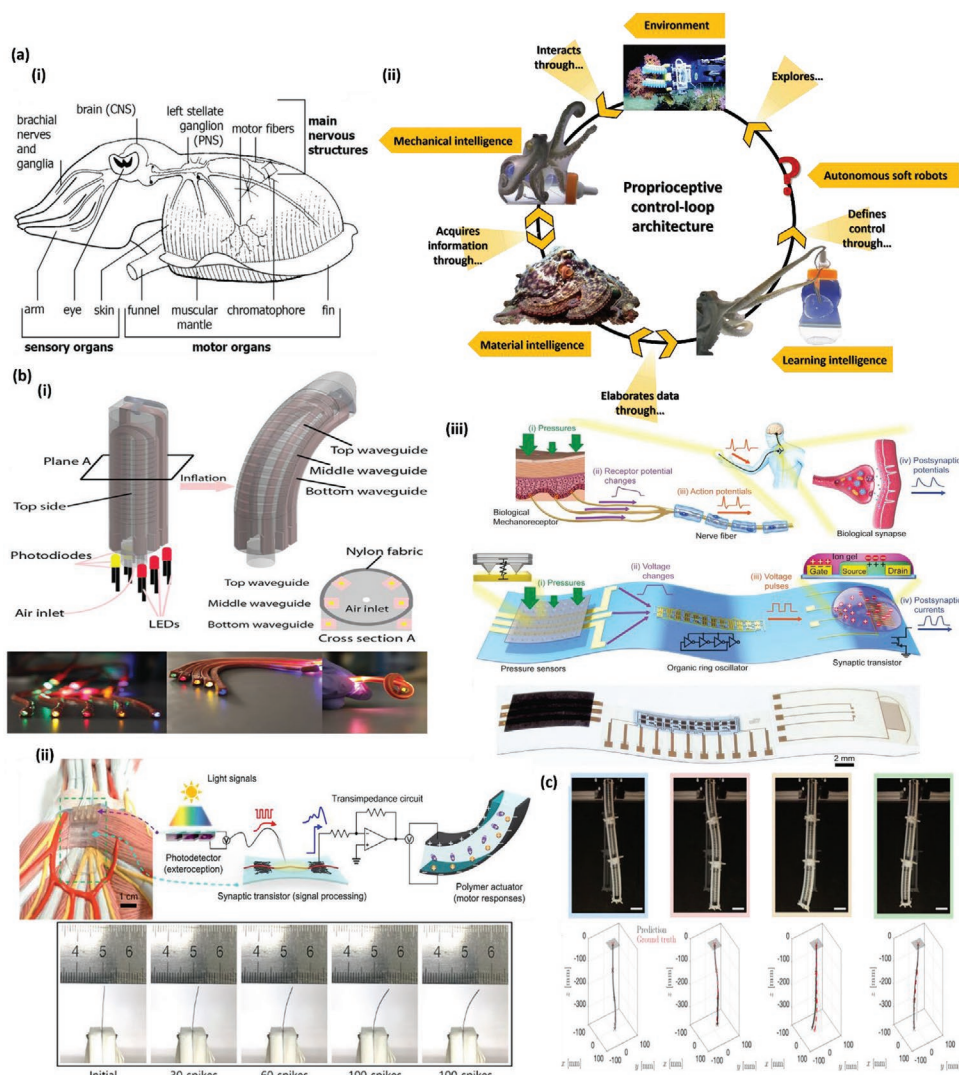


Figure 10. The network distribution of sensing elements that do not interfere with the mechanical compliancy of the bionic end-effector/robot, with neuromorphic strategies are the basement for artificial neural learning and computation intelligence toward mechanical proprioception and embodied intelligence. a-i) Pictorial representation of the main nervous structures (central nervous system, CNS, and peripheral nervous system, PNS, and sensory organs) and of the principal motor organs. Reproduced with permission.^[307] Copyright 2020, John Wiley and Sons Inc. ii) A schematic of the proprioception closed control-loop architecture. The environment acts as a complex scenario of physical interactions, disturbances, and inputs. Thus, a complex machine endowed with “mechanical” and “material intelligence” can transduce these interactions, acquiring and elaborating data by means of a de-/centralized synthetic nervous system, adopting stretch-receptive soft sensor network, neuromorphic systems, etc. Those data are processed to recognize the stimuli and consistently react, and explore for the environmental task, adopting innovative data processing software-based approaches, obtaining in future a soft autonomous proprioceptive robot. Mechanical, material and learning intelligence are bidirectional elements for sensory-motor feedback, and their outputs/ data are mediated by means of a synthetic nervous system as acting in biological neuro-sensory-motor counterparts. Reproduced with permission.^[57] Copyright 2016, Mary Ann Liebert, Inc. b-i) Proprioceptive sensors based on elastomeric optical fibers for shape, touch and pressure recognition on a soft robotic hand. The material of the sensors is fully integrated and compatible with the host matrix, resulting in an electronics-free soft sensor embodiment. Reproduced with permission.^[326] Copyright 2016, American Association for the Advancement of Science. ii) Representation of a light-sensitive sensor system, translating, by means of an artificial nerve, a motor response (exploiting the “material intelligence”). The polymeric actuator drives the optical spike in a strain-dependent motion. Reproduced with permission.^[365] Copyright 2018, American Association for the Advancement of Science. iii) Biological representation of the working functioning of mechanoreceptors. A pressure is mediated by receptor potential changes in multiple nerve fibers that combines synapses actions and contributes to information processing. A neuromorphic electronic system replicate the biological processing information chain, helping both in prosthetic and in robotics toward mechanical proprioception. Reproduced with permission.^[359] Copyright 2018, American Association for the Advancement of Science. c) Photographs and prediction outcomes of a soft robotic arm poses during different actuations. The neural network works properly in the reproduction of predicted configurations. Reproduced with permission.^[378] Copyright 2020, Institute of Electrical and Electronics Engineers.

should be preferred). They also further require a compliant integration with the soft host material to real-time feed the sensation in a feedback control loop for soft robotics.^[350] However,

although we considered proprioceptive sensors to provide feedback of the body and skin shape variations, soft robotics still lack about reliable proprioception and exteroception. This

results in machines that actually are not aware about their body during movements or whether are completing a complex task.^[309] Thus, the sense of the body, as regard providing feedback on both the end-effector (e.g., mechanosensing, temperature sensing, etc.), and along the soft body (e.g., position, movement, vision, etc.), is still a remarkable achievement to obtain for autonomous systems that control their bodies, adapt while growing, access dark and harsh environment, etc. We report some representative works in literature addressing some of these open issues.^[81,145,331,351–356]

In vision to develop “synthetic senses” and “actuators” with the aim to replicate biological nerves and motor/control units in an autonomous system, the proprioceptive elements should require flexibility, mechanical compliancy, low-power consumption, high-density integration, and biocompatibility.^[315] The scientific debate to design new synthetic learning paths is growing fast in the last decades, thanks to more efficient computational machines (that are moving toward “understanding”) and innovative discoveries in the biological data processing functioning.^[69,314] Neuromorphic electronics and different sensing/motor elements, such as photo-,^[357] pressure-,^[358,359] and chemical-sensors^[360] are going toward matching the performances of biological synapses and nerves in perceiving and reacting to various events in real-world scenarios.^[361] The most promising are light-sensitive synthetic data processing systems based on 2D emerging materials,^[362] perovskite,^[363] biomaterials and biotechnological solutions.^[364] However, to the best of our knowledge, there are only a limited number of examples of applications in soft robotics, manipulators, or prosthetics (Figure 10b–ii,iii).^[365–367] The synthetic synapses to detect strain, touch and pressure can be helpful to avoid injuries or damages, or for human-machine interactions and communication.^[368] For example, piezoelectric nanogenerators can link spatiotemporal strain information with artificial intelligence to detect perception,^[369] but require flexible data storage, with large memory capacity, fast processing speed, complex data computation,^[370] and ultra-low energy consumption.^[371] All these features are still far from the realization in current soft robots. Unfortunately, it is yet unclear how to embed stretch-tunable network of neuromorphic systems to measure and interpret the actuator state, the shape of the effector and the environmental conditions acting along the body, but future advances in hardware and algorithms can solve these challenges.^[107]

As one may imagine, the “synthetic senses” and “actuators” introduced so far by means of “material” and “mechanical intelligence” require a recognition and “learning intelligence” to perform advanced activities and to acquire and process data toward an embodied intelligence. Digital intelligence, or as well-known artificial intelligence, has been playing an important role in the development of different aspects in the robotic fields enabling more robust, effective, and autonomous robots. For example, Cully et al.^[96] demonstrated an intelligent trial-and-error algorithm that enabled the robot, once deployed in its environment, to create a detailed map of the space and adopt this learning intelligence to adapt, and consequently perform its tasks even if damage occurred. The learning algorithm used, comprising a pre-computed space of thousands of different behavior-performance, can effectively estimate the behavior that a hexapod robot has to perform while walking, and quickly

discover a way to compensate for the occurred damage. This is an example of learning by training, adopting a Bayesian optimization procedure and the data acquired during the robot performances by its sensory-motor system. Thus, the machine learning technique searches for the maximum approximation of the performance function and selects the corresponding behavior. In this sense, the presented predictive algorithm represents an important step toward the tight coupling between an agent’s body and artificial brain. In terms of recognition and learning, the different training of sophisticated algorithm architectures can provide a 3D adaptive display, which can confer the soft robots and manipulators a conformational and task-driven movement in a various scenario. With reliable proprioception sensors, neural networks and deep learning methodologies could be used to confer the soft robot with mechanical proprioception. Machine learning progresses and studies are reported in refs. [311,372–375] for ease of consultation. Remarkably, one can find several examples of recurrent neural networks capable of predicting several scenarios such as the lateral and twist motion in a soft robotic finger,^[376] the tip position and contact force of a single actuator,^[377] and a 3D soft robot configuration for a trunk-like arm by overcoming the hysteresis issues related to piezoresistive sensorized skin (Figure 10c).^[378] Wall et al.^[376] investigated a dual solution to decrease the size network of sensing elements to reconstruct the shape of their effector, while minimizing the prediction error for 3D specific actuator deformation as flexional, lateral and twist. A redundant sensor layout for the selected deformations, a set of training data and a supervised learning algorithm to map the deformation and minimize the prediction error, followed by a recursive feature elimination automation to exclude the least relevant sensor during each iteration, are at the basis of the “learning intelligence” of their pneumatic actuated soft hand. Thuruthel et al.^[377] drew inspiration from the human perceptive system to build a soft actuator with a redundant sensor topology, a vision-based motion capture detector, and a recurrent neural network algorithm to model the kinematics of the robotic platform in real time. A long short-term memory network was adopted for the time series mapping of the spatial coordinate of the actuator tip and the forces applied during the training. Indeed, the system learned models on the external force applied predicting the contact, e.g., with the hand, using the stress–strain relationship of the soft body. Actually, redundancy in the sensory system is exploited to address robustness on multimodal and multi-physics parameters, highlighting its necessity to replicate synthetic nervous systems and connect the agent’s body and the behavior. However, the adoption of machine learning techniques, recursive analyses and learning-based approach are also useful for modeling the system as, e.g., on the design, placement, minimization, and fabrication step of the distributed network of embedded sensors, other than time-resolving the behavioral perception/prediction. Further, a model-based controller for a complicated non-linear dynamical system, as a non-linear actuator with a deep neural network approach was reported.^[379] Gillespie et al.^[379] achieved a position control precision of 2° for a one degree of freedom joint angle, adopting a deep neural network model predictive control, with constraints about severe hysteresis and unknown state interactions. Starting from the definition of the state variables and the physical constraints

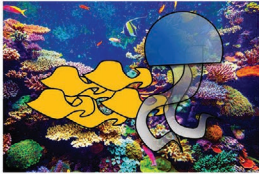
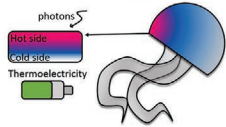

Tasks	Features	Potential applications
<p>Behavioral collective animal studies</p> 	<ul style="list-style-type: none"> • Transparent elements; • Compliant and fully embedded chromogenic materials; • Reconfigurable and camouflaging skins; • Ecological appearance; • Integrated and multifunctional intelligence. 	<ul style="list-style-type: none"> • Safe-interaction and mimicry for collective studies; • Possibility to reach human dangerous regions; • Imperceptible machines for lone animals.
<p>On demand energy saving</p> 	<ul style="list-style-type: none"> • Wrinkled and reconfigurable surfaces; • Thermochromic or electrochromic embodiments; • Embedded storage device; • Income light control depending on the light irradiation. 	<ul style="list-style-type: none"> • Solar thermal devices; • Artificial photosynthesis; • Camouflage solar panel; • Energetic efficient buildings.
<p>Human-machine interaction</p> 	<ul style="list-style-type: none"> • Compliant and fully embedded chromogenic materials; • Reconfigurable embodiments for unstructured scenario; • Intelligence for multivariable environment; • Energetic autonomy. 	<ul style="list-style-type: none"> • Possibility to reach human dangerous regions; • Safe-interaction with human; • On-demand communication for dangerous task or environmental perturbations; • Adaptability on unstructured scenario.

Figure 11. Outlook of soft robots with changing coloration and reconfigurable skins as tools to advance the proprioception in soft platforms. The mechanical proprioception in soft robots could come by means of chromogenic devices (material intelligence as, e.g., structural and molecular mechanochromism), integrated with the inherent mechanical intelligence (e.g., morphing texturing or task-oriented adaptation) of the end-effectors. Taking inspiration from the natural optical materials, mechanical design, proprioceptive senses and decision-making neural systems these futuristic machines can address issues such as: 1) safe-interact with collective animal groups and analyze their behavior, 2) exploit the multifunctional skin to save energy and transform incoming light in electric source, 3) safe-interact with humans with most effective communicative skills.

as inputs, the training of the nonlinear neural network (three hidden layers each with 200 nodes) fed with a random series of steps in both position, velocity, and stiffness at each joint angle, can produce a high-performing model-based controller. Estimation and kinetics feedback control are emerging also in regards to growing locomotion robotic systems able to perceive external stimuli to better address the task-driven movement,^[380] or energetically optimize the robotic system.^[381]

The fusion between smart materials (“material intelligence”), variable actuation systems (“mechanical intelligence”) and complex artificial brain architecture (“learning intelligence”), should provide in the proximal future a theoretical embodied intelligence in soft robotics and manipulators, aiming at solving, e.g., morphological adaptation to unpredictable environments. Soft robots are a prominent platform and the innovative technological developments described hereby, can be a disruptive technology for safe machine-human interaction, respectful ecological inspections, and for tasks that can result dangerous for humans.^[382]

6. Perspective and Conclusion

In the natural world, optical materials, mechanical designs, proprioceptive senses and decision-making neural systems, evolved in millions of years as living beings adapted to effectively survive in different environments. The scientific endeavor of trying to develop comparable technological systems and merge them together in a single machine brought together diverse scientific disciplines such as biology, material science, chemistry, physics, bioinformatics, mechanical design, fabrication, and computer science. However, despite the tremendous and unstoppable

growth that characterized these research fields, there still are grand challenges yet to solve to equal the natural adaptability in soft robotics and other man-made technological platforms.^[383] In the future, one should design soft robots and manipulators so that they are capable of accomplishing several functions, programmable to learn and coexist with the biotic open-world, and to relieve humans from dangerous, difficult, expensive, and complicated tasks (**Figure 11**). To achieve a better-responsive biomimicry behavior, we still require further research in functional materials, structural mechanics, integrated energy sources, multifunctional solutions, and software architectures.^[288,384] This endeavor will require a highly dynamic cooperation among these research fields, so that the optimization of single components and designs does not go astray from connectivity between them.

At the material level, recent developments in nanotechnology, computation, and characterization, can help to improve our understanding of hierarchical biological structures at different length-scales and their effect on the observed macroscopic properties and behavior. However, the replication of many biological architectures still prove to be an open technological challenge. Although polymeric matrices and soft materials can mimic many properties of biological skin and tissues, their dynamic interaction with an artificial brain and on-demand response are still out of our reach. The human-made devices and functionalities developed up to now, are well far from the specialization of the molecular components of a biological system.^[385,386] However, synthetic materials show complexities and variabilities (i.e., metastructures, blending of different elements, mechanical properties on demand, etc.), that can be engineered ad hoc for environments where living being cannot survive thanks to their biological building blocks, such as high

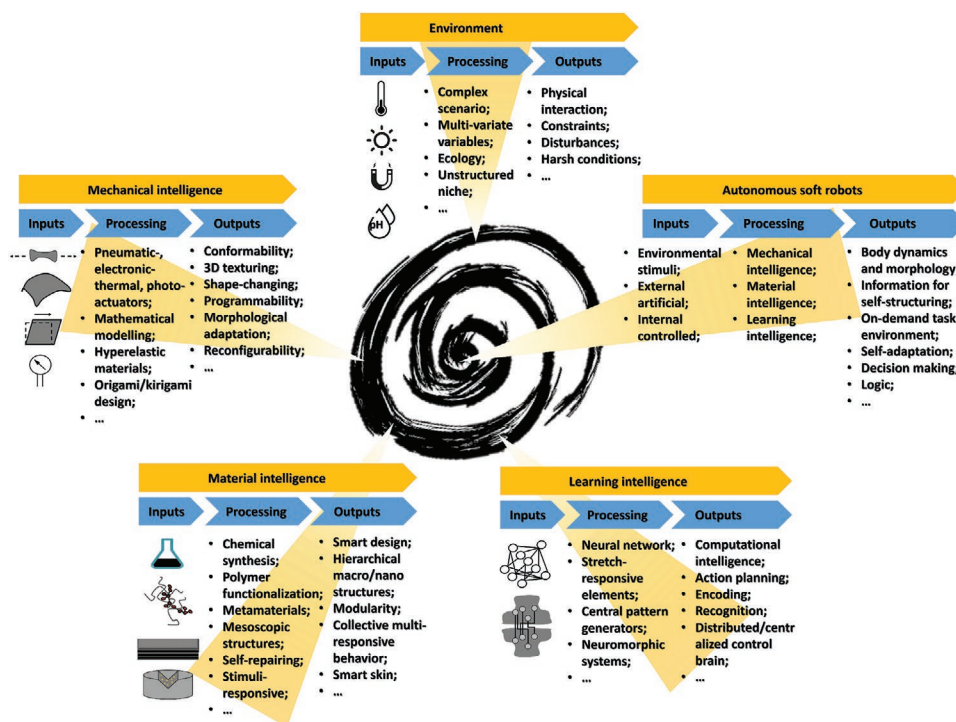


Figure 12. Conclusive diagram toward autonomous soft robots for real-world scenario.

temperatures, high pressure, lack of oxygen, venom environment (or even if they can survive they are extremely specialized for those environments).

At the mechanical level, mathematical and innovative computational techniques to reliably predict the behavior of soft and hyper-elastic materials are under development.^[387] Mathematically modelling how a macroscopic actuation affect a chain-like and multi-units complex system, i.e., the multifunctional skin, is computationally demanding, especially when molecular and atomistic details are needed, such as the case of chromophores bonded to polymer chains or the properties of the material in the interphase surrounding aggregates.^[186,388]

Finally, at the learning level, a soft robot requires a distributed proprioception enabled by a soft, sensorized, and morphing skin capable of communicate with the artificial control system and demand for 3D configurations or various advanced activities. However, machine learning approaches demand for a distributed network of sensors with high sensitivity, coupled with reliable readout electronics at small dynamics or motions. Whether the former is a challenge for the “material” and “mechanical intelligence” distributed in the soft robotic body, the latter can be a bottleneck for the closing of the control loop system in soft robotics. Thus, to discriminate different sensory feedback, one could envision innovative machine learning approaches relying on different domains (e.g., time and frequency).^[389]

In conclusion, multifunctional elements (“material intelligence”), mechanical properties for various geometrical conformations (“mechanical intelligence”), and fast learning and responsive processes (“learning intelligence”), combined with ecofriendly characteristics,^[72,390] are the basement to endow the

future soft robotic platforms with an embodied intelligence able to adapt and coexist with ecological environments and human users (Figure 12).

Acknowledgements

The author would like to thank S. Tirelli for the ellipsometric measurements in Figure 6b,C. Filippeschi for the SEM pictures in Figures 6b and 7b. M.C. gratefully acknowledges partial support from the European Union’s Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie grant agreement MP3 – no. 885881.

Conflict of Interest

The authors declare no conflict of interest.

Keywords

adaptive display, cephalopod camouflage, mechanochromism, neuromorphic approaches, physical and learning intelligence, reconfigurable smart skin, soft robotics

Received: April 13, 2021
Revised: June 17, 2021
Published online: August 5, 2021

[1] J. Teyssier, S. V. Saenko, D. Van Der Marel, M. C. Milinkovitch, *Nat. Commun.* **2015**, *6*, 6368.

- [2] V. E. Johansen, L. Catón, R. Hamidjaja, E. Oosterink, B. D. Wilts, T. S. Rasmussen, M. M. Sherlock, C. J. Ingham, S. Vignolini, *Proc. Natl. Acad. Sci. USA* **2018**, *115*, 2652.
- [3] D. D. Deheyn, N. G. Wilson, *Proc. R. Soc. B* **2011**, *278*, 2112.
- [4] R. Levenson, C. Bracken, N. Bush, D. E. Morse, *J. Biol. Chem.* **2016**, *291*, 4058.
- [5] L. M. Mähger, E. J. Denton, N. J. Marshall, R. T. Hanlon, *J. R. Soc., Interface* **2009**, *6*, S149.
- [6] J. J. Allen, D. Akkaynak, A. U. Sugden, R. T. Hanlon, *Biol. J. Linn. Soc.* **2015**, *116*, 377.
- [7] R. T. Hanlon, J. B. Messenger, *Cephalopod Behaviour*, Cambridge University Press, Cambridge **2018**.
- [8] S. A. Morin, R. F. Shepherd, S. W. Kwok, A. A. Stokes, A. Nemiroski, G. M. Whitesides, *Science* **2012**, *337*, 828.
- [9] H. H. Chou, A. Nguyen, A. Chortos, J. W. F. To, C. Lu, J. Mei, T. Kurosawa, W. G. Bae, J. B. H. Tok, Z. Bao, *Nat. Commun.* **2015**, *6*, 8011.
- [10] J. H. Pikul, S. Li, H. Bai, R. T. Hanlon, I. Cohen, R. F. Shepherd, *Science* **2017**, *358*, 210.
- [11] C. Yu, Y. Li, X. Zhang, X. Huang, V. Malyarchuk, S. Wang, Y. Shi, L. Gao, Y. Su, Y. Zhang, J. A. Rogers, *Proc. Natl. Acad. Sci. USA* **2014**, *111*, 12998.
- [12] P. Won, K. K. Kim, H. Kim, J. J. Park, I. Ha, J. Shin, J. Jung, H. Cho, J. Kwon, H. Lee, S. H. Ko, *Adv. Mater.* **2020**, *30*, 2002397.
- [13] A. McDougal, B. Miller, M. Singh, M. Kolle, *J. Opt.* **2019**, *21*, 073001.
- [14] E. Kreit, L. M. Mähger, R. T. Hanlon, P. B. Dennis, R. R. Naik, E. Forsythe, J. Heikenfeld, *J. R. Soc., Interface* **2013**, *10*, 20120601.
- [15] S. Tadepalli, J. M. Slocik, M. K. Gupta, R. R. Naik, S. Singamaneni, *Chem. Rev.* **2017**, *117*, 12705.
- [16] M. Kolle, S. Lee, *Adv. Mater.* **2018**, *30*, 1702669.
- [17] L. Wu, J. He, W. Shang, T. Deng, J. Gu, H. Su, Q. Liu, W. Zhang, D. Zhang, *Adv. Opt. Mater.* **2016**, *4*, 195.
- [18] G. Isapour, M. Lattuada, *Adv. Mater.* **2018**, *30*, 1707069.
- [19] D. Gur, B. Leshem, V. Farstey, D. Oron, L. Addadi, S. Weiner, *Adv. Funct. Mater.* **2016**, *26*, 1393.
- [20] M. Vatankehah-Varnosfaderani, A. N. Keith, Y. Cong, H. Liang, M. Rosenthal, M. Sztucki, C. Clair, S. Magonov, D. A. Ivanov, A. V. Dobrynin, S. S. Sheiko, *Science* **2018**, *359*, 1509.
- [21] C. GANS, D. BAIC, *Science* **1977**, *195*, 1348.
- [22] M. Burresti, L. Cortese, L. Pattelli, M. Kolle, P. Vukusic, D. S. Wiersma, U. Steiner, S. Vignolini, *Sci. Rep.* **2014**, *4*, 6075.
- [23] K. Chung, S. Yu, C. Heo, J. W. Shim, S. Yang, M. G. Han, H. Lee, Y. Jin, S. Y. Lee, N. Park, *Adv. Mater.* **2012**, *24*, 2375.
- [24] H. Yin, L. Shi, J. Sha, Y. Li, Y. Qin, B. Dong, S. Meyer, X. Liu, L. Zhao, J. Zi, *Phys. Rev. E* **2006**, *74*, 51916.
- [25] J. Wang, Y. Zhang, S. Wang, Y. Song, L. Jiang, *Acc. Chem. Res.* **2011**, *44*, 405.
- [26] A. Finomore, P. Cunha, T. Shean, S. Vignolini, S. Guldin, M. Oyen, U. Steiner, *Nat. Commun.* **2012**, *3*, 966.
- [27] L. Li, S. Kolle, J. C. Weaver, C. Ortiz, J. Aizenberg, M. Kolle, *Nat. Commun.* **2015**, *6*, 6322.
- [28] S. Vignolini, T. Gregory, M. Kolle, A. Lethbridge, E. Moyroud, U. Steiner, B. J. Glover, P. Vukusic, P. J. Rudall, *J. R. Soc., Interface* **2016**, *13*, 20160645.
- [29] S. Vignolini, E. Moyroud, T. Hingant, H. Banks, P. J. Rudall, U. Steiner, B. J. Glover, *New Phytol.* **2015**, *205*, 97.
- [30] T. J. Wardill, P. T. Gonzalez-Bellido, R. J. Crook, R. T. Hanlon, *Proc. R. Soc. B* **2012**, *279*, 4243.
- [31] P. Imperadore, S. B. Shah, H. P. Makarenkova, G. Fiorito, *Sci. Rep.* **2017**, *7*, 46564.
- [32] B. Geiger, J. P. Spatz, A. D. Bershadsky, *Nat. Rev. Mol. Cell Biol.* **2009**, *10*, 21.
- [33] B. Hochner, *Brain Behav. Evol.* **2013**, *82*, 19.
- [34] T. Gutnick, L. Zullo, B. Hochner, M. J. Kuba, *Curr. Biol.* **2020**, *30*, 4322.
- [35] J. McFadden, J. Al-Khalili, *Life on the Edge: The Coming of Age of Quantum Biology*, Broadway Books, New York **2016**.
- [36] D. Panetta, K. Buresch, R. T. Hanlon, *Biol. Lett.* **2017**, *13*, 20170070.
- [37] P. Graziadei, *Proc. R. Soc. London, Ser. B* **1965**, *161*, 392.
- [38] L. M. Mähger, R. T. Hanlon, *Cell Tissue Res.* **2007**, *329*, 179.
- [39] R. L. Sutherland, L. M. Mähger, R. T. Hanlon, A. M. Urbas, M. O. Stone, *J. Opt. Soc. Am. A* **2008**, *25*, 588.
- [40] J. B. Messenger, *Biol. Rev. Cambridge Philos. Soc.* **2001**, *76*, 473.
- [41] M. Izumi, A. M. Sweeney, D. DeMartini, J. C. Weaver, M. L. Powers, A. Tao, T. V. Silvas, R. M. Kramer, W. J. Crookes-Goodson, L. M. Mähger, et al., *J. R. Soc., Interface* **2010**, *7*, 549.
- [42] D. G. DeMartini, D. V. Krogstad, D. E. Morse, *Proc. Natl. Acad. Sci. USA* **2013**, *110*, 2552.
- [43] D. Froesch, J. B. Messenger, *J. Zool.* **1978**, *186*, 163.
- [44] A. K. Schnell, P. Amodio, M. Boeckle, N. S. Clayton, *Biol. Rev.* **2021**, *96*, 162.
- [45] A. Cangelosi, J. Bongard, M. H. Fischer, S. Nolfi, in *Embodied Intelligence* (Eds: J. Kacprzyk, W. Pedrycz), Springer Berlin Heidelberg, Berlin, Heidelberg **2015**, pp. 697–714.
- [46] C. Laschi, B. Mazzolai, M. Cianchetti, *Sci. Rob.* **2016**, *1*, eaah3690.
- [47] H. Zhou, J. Xu, X. Liu, H. Zhang, D. Wang, Z. Chen, D. Zhang, T. Fan, *Adv. Funct. Mater.* **2018**, *28*, 1705309.
- [48] V. Vallem, Y. Sargolzaeiaval, M. Ozturk, Y. - C. Lai, M. D. Dickey, *Adv. Mater.* **2021**, *33*, 2004832.
- [49] P. Boyraz, G. Runge, A. Raatz, *Actuators* **2018**, *7*, 48.
- [50] E. Sachyani Keneth, A. Kamyshny, M. Totaro, L. Beccai, S. Magdassi, *Adv. Mater.* **2020**, *33*, 2003387.
- [51] D. Howard, A. E. Eiben, D. F. Kennedy, J.-B. Mouret, P. Valencia, D. Winkler, *Nat. Mach. Intell.* **2019**, *1*, 12.
- [52] Y. Wang, S. E. Naleway, B. Wang, *Bioact. Mater.* **2020**, *5*, 745.
- [53] B. Mazzolai, A. Mondini, F. Tramacere, G. Riccomi, A. Sadeghi, G. Giordano, E. Del Dottore, M. Scaccia, M. Zampato, S. Carminati, *Adv. Intell. Syst.* **2019**, *1*, 1900041.
- [54] T. T. Hoang, P. T. Phan, M. T. Thai, N. H. Lovell, T. N. Do, *Adv. Mater. Technol.* **2020**, *5*, 2000724.
- [55] A. Yin, H.-C. Lin, J. Thelen, B. Mahner, T. Ranzani, *Adv. Intell. Syst.* **2019**, *1*, 1900089.
- [56] T. Ranzani, G. Gerboni, M. Cianchetti, A. Menciassi, *Bioinspiration Biomimetics* **2015**, *10*, 035008.
- [57] K. C. Galloway, K. P. Becker, B. Phillips, J. Kirby, S. Licht, D. Tchernov, R. J. Wood, D. F. Gruber, *Soft Rob.* **2016**, *3*, 23.
- [58] K. H. Petersen, R. F. Shepherd, *Fluid-Driven Intrinsically Soft Robots*, Elsevier Ltd, New York **2019**.
- [59] S. P. M. Babu, F. Visentin, A. Sadeghi, A. Mondini, B. Mazzolai, *IEEE Rob. Autom. Lett.* **2020**, *5*, 4055.
- [60] I. Must, E. Sinibaldi, B. Mazzolai, *Nat. Commun.* **2019**, *10*, 344.
- [61] R. F. Shepherd, F. Ilievski, W. Choi, S. A. Morin, A. A. Stokes, A. D. Mazzeo, X. Chen, M. Wang, G. M. Whitesides, *Proc. Natl. Acad. Sci. USA* **2011**, *108*, 20400.
- [62] A. Sadeghi, A. Mondini, B. Mazzolai, *Soft Rob.* **2017**, *4*, 211.
- [63] J. D. Greer, T. K. Morimoto, A. M. Okamura, E. W. Hawkes, *Soft Rob.* **2018**, *6*, 95.
- [64] S. Bauer, S. Bauer-Gogonea, I. Graz, M. Kaltenbrunner, C. Keplinger, R. Schwödau, *Adv. Mater.* **2014**, *26*, 149.
- [65] M. Sitti, *Extreme Mech. Lett.* **2021**, *46*, 101340.
- [66] Y. Mengüç, N. Correll, R. Kramer, J. Paik, *Sci. Rob.* **2017**, *2*, eaar4527.
- [67] A. Miriyev, M. Kovač, *Nat. Mach. Intell.* **2020**, *2*, 658.
- [68] C. Majidi, *Soft Rob.* **2014**, *1*, 5.
- [69] G. M. Whitesides, *Angew. Chem., Int. Ed.* **2018**, *57*, 4258.
- [70] R. Pfeifer, M. Lungarella, F. Iida, *Commun. ACM* **2012**, *55*, 76.
- [71] J. Yang, X. Zhang, X. Zhang, L. Wang, W. Feng, Q. Li, *Adv. Mater.* **2021**, *33*, 2004754.
- [72] F. Hartmann, M. Baumgartner, M. Kaltenbrunner, *Adv. Mater.* **2021**, *33*, 2004413.

- [73] M. Ding, L. Jing, H. Yang, C. E. Machnicki, X. Fu, K. Li, I. Y. Wong, P.-Y. Chen, *Mater. Today Adv.* **2020**, *8*, 100088.
- [74] Q. Wang, G. R. Gossweiler, S. L. Craig, X. Zhao, *Nat. Commun.* **2014**, *5*, 4899.
- [75] G. R. Gossweiler, C. L. Brown, G. B. Hewage, E. Sapiro-Gheiler, W. J. Trautman, G. W. Welshofer, S. L. Craig, *ACS Appl. Mater. Interfaces* **2015**, *7*, 22431.
- [76] J. Hughes, S. Li, D. Rus, in *2020 IEEE Int. Conf. on Robotics and Automation*, IEEE, Piscataway, NJ **2020**, pp. 6913–6919.
- [77] J. Zhong, Y. Ma, Y. Song, Q. Zhong, Y. Chu, I. Karakurt, D. B. Bogy, L. Lin, *ACS Nano* **2019**, *13*, 7107.
- [78] S. Palagi, A. G. Mark, S. Y. Reigh, K. Melde, T. Qiu, H. Zeng, C. Parmeggiani, D. Martella, A. Sanchez-Castillo, N. Kapernaum, F. Giesselmann, D. S. Wiersma, E. Lauga, P. Fischer, *Nat. Mater.* **2016**, *15*, 647.
- [79] X.-Q. Wang, K. H. Chan, Y. Cheng, T. Ding, T. Li, S. Achavananthadith, S. Ahmet, J. S. Ho, G. W. Ho, *Adv. Mater.* **2020**, *32*, 2000351.
- [80] M. Rogó , H. Zeng, C. Xuan, D. S. Wiersma, P. Wasylczyk, *Adv. Opt. Mater.* **2016**, *4*, 1689.
- [81] H. Wang, M. Totaro, L. Beccai, *Adv. Sci.* **2018**, *5*, 1800541.
- [82] G.-Z. Yang, J. Bellingham, H. Choset, P. Dario, P. Fischer, T. Fukuda, N. Jacobstein, B. Nelson, M. Veloso, J. Berg, *Sci. Rob.* **2016**, *1*, eaal2099.
- [83] C. Laschi, *Science* **2017**, *358*, 169.
- [84] Y. Qiao, Z. Meng, P. Wang, D. Yan, *Front. Mater.* **2021**, *8*, 79.
- [85] J. Lee, H. Sul, Y. Jung, H. Kim, S. Han, J. Choi, J. Shin, D. Kim, J. Jung, S. Hong, S. H. Ko, *Adv. Funct. Mater.* **2020**, *30*, 2003328.
- [86] Y. Wang, H. Nie, J. Han, Y. An, Y.-M. Zhang, S. X.-A. Zhang, *Light: Sci. Appl.* **2021**, *10*, 33.
- [87] S. Hong, S. Shin, R. Chen, *Adv. Funct. Mater.* **2020**, *30*, 1909788.
- [88] B. Svetozarevic, M. Begle, P. Jayathissa, S. Caranovic, R. F. Shepherd, Z. Nagy, I. Hischer, J. Hofer, A. Schlueter, *Nat. Energy* **2019**, *4*, 671.
- [89] O. Graydon, *Nat. Photonics* **2018**, *12*, 445.
- [90] R. R. Ruckdashel, D. Venkataraman, J. H. Park, *J. Appl. Phys.* **2021**, *129*, 130903.
- [91] M. Han, S. Ahn, *Adv. Mater.* **2017**, *29*, 1606580.
- [92] A. Pal, V. Restrepo, D. Goswami, R. V. Martinez, *Adv. Mater.* **2021**, *33*, 2006939.
- [93] A. E. Eiben, N. Bredeche, M. Hoogendoorn, J. Stradner, J. Timmis, A. M. Tyrrell, A. Winfield, in *Artificial Life Conf. Proc. 13*, MIT Press, Cambridge **2013**, pp. 1056–1063.
- [94] T. Umedachi, T. Kano, A. Ishiguro, B. A. Trimmer, *R. Soc. Open Sci.* **2016**, *3*, 160766.
- [95] F. Silva, M. Duarte, L. Correia, S. M. Oliveira, A. L. Christensen, *Evol. Comput.* **2016**, *24*, 205.
- [96] A. Cully, J. Clune, D. Tapapore, J.-B. Mouret, *Nature* **2015**, *521*, 503.
- [97] B. Shih, D. Shah, J. Li, T. G. Thuruthel, Y.-L. Park, F. Iida, Z. Bao, R. Kramer-Bottiglio, M. T. Tolley, *Sci. Rob.* **2020**, *5*, eaaz9239.
- [98] D. Rus, M. T. Tolley, *Nature* **2015**, *521*, 467.
- [99] M. Wehner, R. L. Truby, D. J. Fitzgerald, B. Mosadegh, G. M. Whitesides, J. A. Lewis, R. J. Wood, *Nature* **2016**, *536*, 451.
- [100] E. Brown, N. Rodenberg, J. Amend, A. Mozeika, E. Steltz, M. R. Zakin, H. Lipson, H. M. Jaeger, *Proc. Natl. Acad. Sci. USA* **2010**, *107*, 18809.
- [101] K. Suzumori, *Rob. Auton. Syst.* **1996**, *18*, 135.
- [102] L. Ionov, *Adv. Funct. Mater.* **2013**, *23*, 4555.
- [103] P. Polygerinos, N. Correll, S. A. Morin, B. Mosadegh, C. D. Onal, K. Petersen, M. Cianchetti, M. T. Tolley, R. F. Shepherd, *Adv. Eng. Mater.* **2017**, *19*, 1700016.
- [104] T. J. Wallin, J. Pikul, R. F. Shepherd, *Nat. Rev. Mater.* **2018**, *3*, 84.
- [105] C. Majidi, *Adv. Mater. Technol.* **2019**, *4*, 1800477.
- [106] Z. Shen, H. Zhong, E. Xu, R. Zhang, K. C. Yip, L. L. Chan, L. L. Chan, J. Pan, W. Wang, Z. Wang, *Soft Rob.* **2020**, *7*, 535.
- [107] D. S. Shah, J. P. Powers, L. G. Tilton, S. Kriegman, J. Bongard, R. Kramer-Bottiglio, *Nat. Mach. Intell.* **2021**, *3*, 51.
- [108] R. F. Shepherd, A. A. Stokes, R. M. D. Nunes, G. M. Whitesides, *Adv. Mater.* **2013**, *25*, 6709.
- [109] A. Lendlein, R. S. Trask, *Multifunct. Mater.* **2018**, *1*, 010201.
- [110] A. Nojoomi, H. Arslan, K. Lee, K. Yum, *Nat. Commun.* **2018**, *9*, 3705.
- [111] R. Z. Gao, C. L. Ren, *Biomicrofluidics* **2021**, *15*, 011302.
- [112] M. Cianchetti, C. Laschi, A. Menciassi, P. Dario, *Nat. Rev. Mater.* **2018**, *3*, 143.
- [113] M. Calisti, G. Picardi, C. Laschi, *J. R. Soc., Interface* **2017**, *14*, 20170101.
- [114] L. Qin, X. Liang, H. Huang, C. K. Chui, R. C. H. Yeow, J. Zhu, *Soft Rob.* **2019**, *6*, 455.
- [115] M. Yu, W. Yang, Y. Yu, X. Cheng, Z. Jiao, *Actuators* **2020**, *9*, 26.
- [116] D. S. Shah, M. C. Yuen, L. G. Tilton, E. J. Yang, R. Kramer-Bottiglio, *IEEE Rob. Autom. Lett.* **2019**, *4*, 2204.
- [117] R. A. Bilodeau, R. K. Kramer, *Front. Robot. AI* **2017**, *4*, 48.
- [118] B. Mazzolai, L. Margheri, M. Cianchetti, P. Dario, C. Laschi, *Bioinspir. Biomim.* **2012**, *7*, 025005.
- [119] J. Fras, Y. Noh, M. Macias, H. Wurdemann, K. Althoefer, in *2018 IEEE Int. Conf. on Robotics and Automation*, IEEE, Piscataway, NJ **2018**, pp. 1583–1588.
- [120] G. Zhong, Y. Hou, W. Dou, *Int. J. Mech. Sci.* **2019**, *153–154*, 445.
- [121] S. Baik, D. W. Kim, Y. Park, T. J. Lee, S. Ho Bhang, C. Pang, *Nature* **2017**, *546*, 396.
- [122] S. Sareh, K. Althoefer, M. Li, Y. Noh, F. Tramacere, P. Sareh, B. Mazzolai, M. Kovac, *J. R. Soc., Interface* **2017**, *14*, 20170395.
- [123] G. Meloni, O. Tricinci, A. Degl’Innocenti, B. Mazzolai, *Sci. Rep.* **2020**, *10*, 15480.
- [124] F. Tramacere, M. Follador, N. M. Pugno, B. Mazzolai, *Bioinspir. Biomim.* **2015**, *10*, 035004.
- [125] M. Cianchetti, M. Calisti, L. Margheri, M. Kuba, C. Laschi, *Bioinspir. Biomim.* **2015**, *10*, 035003.
- [126] M. Yang, L. P. Cooper, N. Liu, X. Wang, M. P. Fok, *Opt. Express* **2020**, *28*, 35158.
- [127] G. Picardi, M. Chellapurath, S. Iacoponi, S. Stefanni, C. Laschi, M. Calisti, *Sci. Rob.* **2020**, *5*, eaaz1012.
- [128] E. W. Hawkes, L. H. Blumenschein, J. D. Greer, A. M. Okamura, *Sci. Rob.* **2017**, *2*, eaan3028.
- [129] A. Sadeghi, E. Del Dottore, A. Mondini, B. Mazzolai, *Soft Rob.* **2019**, *7*, 85.
- [130] C. Christianson, N. N. Goldberg, D. D. Deheyne, S. Cai, M. T. Tolley, *Sci. Rob.* **2018**, *3*, eaat1893.
- [131] H. Yuk, S. Lin, C. Ma, M. Takaffoli, N. X. Fang, X. Zhao, *Nat. Commun.* **2017**, *8*, 14230.
- [132] S. Banisadr, J. Chen, *Sci. Rep.* **2017**, *7*, 17521.
- [133] T. Li, G. Li, Y. Liang, T. Cheng, J. Dai, X. Yang, B. Liu, Z. Zeng, Z. Huang, Y. Luo, T. Xie, W. Yang, *Sci. Adv.* **2017**, *3*, 1602045.
- [134] P. Li, Y. Wang, U. Gupta, J. Liu, L. Zhang, D. Du, C. C. Foo, J. Ouyang, J. Zhu, *Adv. Funct. Mater.* **2019**, *29*, 1901908.
- [135] T. L. Williams, S. L. Senft, J. Yeo, F. J. Martin-Martinez, A. M. Kuzirian, C. A. Martin, C. W. DiBona, C.-T. Chen, S. R. Dinneen, H. T. Nguyen, C. M. Gomes, J. J. C. Rosenthal, M. D. MacManes, F. Chu, M. J. Buehler, R. T. Hanlon, L. F. Deravi, *Nat. Commun.* **2019**, *10*, 1004.
- [136] S. Zeng, D. Zhang, W. Huang, Z. Wang, S. G. Freire, X. Yu, A. T. Smith, E. Y. Huang, H. Nguon, L. Sun, *Nat. Commun.* **2016**, *7*, 11802.
- [137] J. Rossiter, B. Yap, A. Conn, *Bioinspiration Biomimetics* **2012**, *7*, 036009.
- [138] R. M. Kramer, W. J. Crookes-Goodson, R. R. Naik, *Nat. Mater.* **2007**, *6*, 533.
- [139] P. Kim, Y. Hu, J. Alvarenga, M. Kolle, Z. Suo, J. Aizenberg, *Adv. Opt. Mater.* **2013**, *1*, 381.

- [140] A. Kumar, R. M. Osgood, S. R. Dinneen, B. D. Koker, R. Pang, L. F. Deravi, *Adv. Opt. Mater.* **2018**, 6, 1701369.
- [141] J. F. Galisteo-López, M. Ibisate, R. Sapienza, L. S. Froufe-Pérez, Ú. Blanco, C. López, *Adv. Mater.* **2011**, 23, 30.
- [142] J. Ge, Y. Yin, *Angew. Chem., Int. Ed.* **2011**, 50, 1492.
- [143] R. A. Cloney, E. Florey, *Z. Zellforsch. Mikrosk. Anat.* **1968**, 89, 250.
- [144] C. Fleischmann, M. Lievenbrück, H. Ritter, *Polymers* **2015**, 7, 717.
- [145] C. Larson, B. Peele, S. Li, S. Robinson, M. Totaro, L. Beccai, B. Mazzolai, R. Shepherd, *Science* **2016**, 351, 1071.
- [146] T. Bu, T. Xiao, Z. Yang, G. Liu, X. Fu, J. Nie, T. Guo, Y. Pang, J. Zhao, F. Xi, C. Zhang, Z. L. Wang, *Adv. Mater.* **2018**, 30, 1800066.
- [147] C. Calvino, L. Neumann, C. Weder, S. Schrettl, *J. Polym. Sci., Part A: Polym. Chem.* **2017**, 55, 640.
- [148] J. Li, C. Nagamani, J. S. Moore, *Acc. Chem. Res.* **2015**, 48, 2181.
- [149] Y. Sagara, S. Yamane, M. Mitani, C. Weder, T. Kato, *Adv. Mater.* **2016**, 28, 1073.
- [150] M. M. Caruso, D. A. Davis, Q. Shen, S. A. Odom, N. R. Sottos, S. R. White, J. S. Moore, *Chem. Rev.* **2009**, 109, 5755.
- [151] Q. Zhu, K. Van Vliet, N. Holten-Andersen, A. Miserez, *Adv. Funct. Mater.* **2019**, 29, 1808191.
- [152] F. Ciardelli, G. Ruggeri, A. Pucci, *Chem. Soc. Rev.* **2013**, 42, 857.
- [153] T. Kato, *Science* **2002**, 295, 2414.
- [154] Y. Sagara, T. Kato, *Angew. Chem.* **2011**, 123, 9294.
- [155] Y. Zhang, B. Ren, F. Yang, Y. Cai, H. Chen, T. Wang, Z. Feng, J. Tang, J. Xu, J. Zheng, *J. Mater. Chem. C* **2018**, 6, 11536.
- [156] H. Chen, F. Yang, Q. Chen, J. Zheng, *Adv. Mater.* **2017**, 29, 1606900.
- [157] H. Koshima, *Mechanically Responsive Materials for Soft Robotics*, John Wiley & Sons, New York **2019**.
- [158] R. Klajn, *Chem. Soc. Rev.* **2014**, 43, 148.
- [159] C. Micheletti, P. Minei, M. Carlotti, V. Mattoli, F. Muniz-Miranda, A. Perfetto, I. Ciofini, C. Adamo, G. Ruggeri, A. Pucci, *Macromol. Rapid Commun.* **2020**, 42, 2000426.
- [160] D. A. Davis, A. Hamilton, J. Yang, L. D. Cremer, D. Van Gough, S. L. Potisek, M. T. Ong, P. V. Braun, T. J. Martínez, S. R. White, J. S. Moore, N. R. Sottos, *Nature* **2009**, 459, 68.
- [161] J. Park, Y. Lee, M. H. Barbee, S. Cho, S. Cho, R. Shanker, J. Kim, J. Myoung, M. P. Kim, C. Baig, S. L. Craig, H. Ko, *Adv. Mater.* **2019**, 31, 1808148.
- [162] C. Calvino, A. Guha, C. Weder, S. Schrettl, *Adv. Mater.* **2018**, 30, 1704603.
- [163] A. Lavrenova, J. Farkas, C. Weder, Y. C. Simon, *ACS Appl. Mater. Interfaces* **2015**, 7, 21828.
- [164] A. Lavrenova, D. W. R. Balkenende, Y. Sagara, S. Schrettl, Y. C. Simon, C. Weder, *J. Am. Chem. Soc.* **2017**, 139, 4302.
- [165] V. I. Minkin, *Chem. Rev.* **2004**, 104, 2751.
- [166] J. T. C. Wojtyk, A. Wasey, N.-N. Xiao, P. M. Kazmaier, S. Hoz, C. Yu, R. P. Lemieux, E. Buncel, *J. Phys. Chem. A* **2007**, 111, 2511.
- [167] M. Sommer, *Macromol. Rapid Commun.* **2021**, 42, 2000597.
- [168] Y. Fang, W. Dehaen, *Coord. Chem. Rev.* **2021**, 427, 213524.
- [169] G. R. Gossweiler, G. B. Hewage, G. Soriano, Q. Wang, G. W. Welshofer, X. Zhao, S. L. Craig, *ACS Macro Lett.* **2014**, 3, 216.
- [170] S. L. Potisek, D. A. Davis, N. R. Sottos, S. R. White, J. S. Moore, *J. Am. Chem. Soc.* **2007**, 129, 13808.
- [171] M. H. Barbee, K. Mondal, J. Z. Deng, V. Bharambe, T. V. Neumann, J. J. Adams, N. Boechler, M. D. Dickey, S. L. Craig, *ACS Appl. Mater. Interfaces* **2018**, 10, 29918.
- [172] G. O'Bryan, B. M. Wong, J. R. McElhanon, *ACS Appl. Mater. Interfaces* **2010**, 2, 1594.
- [173] C. K. Lee, D. A. Davis, S. R. White, J. S. Moore, N. R. Sottos, P. V. Braun, *J. Am. Chem. Soc.* **2010**, 132, 16107.
- [174] G. I. Peterson, M. B. Larsen, M. A. Ganter, D. W. Storti, A. J. Boydston, *ACS Appl. Mater. Interfaces* **2015**, 7, 577.
- [175] G. Giordano, M. Gagliardi, Y. Huan, M. Carlotti, A. Mariani, A. Menciasci, E. Sinibaldi, B. Mazzolai, *Adv. Sci.* **2021**, <https://doi.org/10.1002/advs.202100418>.
- [176] F. Cellini, S. Khapli, S. D. Peterson, M. Porfiri, *Appl. Phys. Lett.* **2014**, 105, 061907.
- [177] K. Seshimo, H. Sakai, T. Watabe, D. Aoki, H. Sugita, K. Mikami, Y. Mao, A. Ishigami, S. Nishitsuji, T. Kurose, H. Ito, H. Otsuka, *Angew. Chem. Int. Ed.* **2021**, 60, 8406.
- [178] Z. Wang, Z. Ma, Y. Wang, Z. Xu, Y. Luo, Y. Wei, X. Jia, *Adv. Mater.* **2015**, 27, 6469.
- [179] T. Kosuge, K. Imato, R. Goseki, H. Otsuka, *Macromolecules* **2016**, 49, 5903.
- [180] W. Qiu, P. A. Gurr, G. Da Silva, G. G. Qiao, *Polym. Chem.* **2019**, 10, 1650.
- [181] G. R. Gossweiler, T. B. Kouznetsova, S. L. Craig, *J. Am. Chem. Soc.* **2015**, 137, 6148.
- [182] N. Willis-Fox, E. Rognin, T. A. Aljohani, R. Daly, *Chem* **2018**, 4, 2499.
- [183] Y. Jiang, *Mater. Sci. Eng., C* **2015**, 45, 682.
- [184] J. Zhang, Q. Zou, H. Tian, *Adv. Mater.* **2013**, 25, 378.
- [185] D. Görl, X. Zhang, F. Würthner, *Angew. Chem., Int. Ed.* **2012**, 51, 6328.
- [186] M. Carlotti, G. Gullo, A. Battisti, F. Martini, S. Borsacchi, M. Geppi, G. Ruggeri, A. Pucci, *Polym. Chem.* **2015**, 6, 4003.
- [187] C. Calvino, Y. Sagara, V. Buclin, A. P. Haehnel, A. del Prado, C. Aeby, Y. C. Simon, S. Schrettl, C. Weder, *Macromol. Rapid Commun.* **2019**, 40, 1800705.
- [188] K. Ishizuki, D. Aoki, R. Goseki, H. Otsuka, *ACS Macro Lett.* **2018**, 7, 556.
- [189] S. Okada, S. Peng, W. Spevak, D. Charych, *Acc. Chem. Res.* **1998**, 31, 229.
- [190] K. L. Berkowski, S. L. Potisek, C. R. Hickenboth, J. S. Moore, *Macromolecules* **2005**, 38, 8975.
- [191] J. P. Lee, H. Hwang, S. Chae, J.-M. Kim, *Chem. Commun.* **2019**, 55, 9395.
- [192] Z. Ma, Y. Ji, Y. Lan, G.-C. Kuang, X. Jia, *J. Mater. Chem. C* **2018**, 6, 2270.
- [193] Y. Lin, T. B. Kouznetsova, S. L. Craig, *J. Am. Chem. Soc.* **2019**, 142, 99.
- [194] D. Sakamaki, S. Ghosh, S. Seki, *Mater. Chem. Front.* **2019**, 3, 2270.
- [195] H. Qian, N. S. Purwanto, D. G. Ivanoff, A. J. Halmes, N. R. Sottos, J. S. Moore, *Chem* **2021**, 7, 1080.
- [196] Y. Sagara, M. Karman, E. Verde-Sesto, K. Matsuo, Y. Kim, N. Tamaoki, C. Weder, *J. Am. Chem. Soc.* **2018**, 140, 1584.
- [197] R. Göstl, R. P. Sijbesma, *Chem. Sci.* **2016**, 7, 370.
- [198] Y. Chen, A. J. H. Spiering, S. Karthikeyan, G. W. M. Peters, E. W. Meijer, R. P. Sijbesma, *Nat. Chem.* **2012**, 4, 559.
- [199] N. Jiang, S.-H. Ruan, X.-M. Liu, D. Zhu, B. Li, M. R. Bryce, *Chem. Mater.* **2020**, 32, 5776.
- [200] J. R. Kumpfer, S. D. Taylor, W. B. Connick, S. J. Rowan, *J. Mater. Chem.* **2012**, 22, 14196.
- [201] S.-Y. Cho, J.-G. Kim, C.-M. Chung, *Sens. Actuators, B* **2008**, 134, 822.
- [202] B. R. Crenshaw, C. Weder, *Chem. Mater.* **2003**, 15, 4717.
- [203] T. Ikawa, T. Shiga, A. Okada, *J. Appl. Polym. Sci.* **1997**, 66, 1569.
- [204] P. Minei, G. Iasilli, G. Ruggeri, V. Mattoli, A. Pucci, *Chemosensors* **2021**, 9, 3.
- [205] Y. Chen, G. Mellot, D. van Luijk, C. Creton, R. P. Sijbesma, *Chem. Soc. Rev.* **2021**, 50, 4100.
- [206] T. Suzuki, H. Okada, T. Nakagawa, K. Komatsu, C. Fujimoto, H. Kagi, Y. Matsuo, *Chem. Sci.* **2018**, 9, 475.
- [207] L. Wang, L. Liu, B. Xu, W. Tian, *Chem. Res. Chin. Univ.* **2021**, 37, 100.
- [208] J.-J. Liu, S.-B. Xia, Q.-T. Que, H. Suo, J. Liu, X. Shen, F.-X. Cheng, *Dalton Trans.* **2020**, 49, 3174.
- [209] D. Peng, L.-H. He, P. Ju, J.-L. Chen, H.-Y. Ye, J.-Y. Wang, S.-J. Liu, H.-R. Wen, *Inorg. Chem.* **2020**, 59, 17213.
- [210] Y. Tani, M. Komura, T. Ogawa, *Chem. Commun.* **2020**, 56, 6810.
- [211] S. Tan, Y. Yin, W. Chen, Z. Chen, W. Tian, S. Pu, *Dyes Pigm.* **2020**, 177, 108302.

- [212] T. Ishi-i, H. Tanaka, R. Youfu, N. Aizawa, T. Yasuda, S. Kato, T. Matsumoto, *New J. Chem.* **2019**, 43, 4998.
- [213] Y. Sagara, T. Kato, *Nat. Chem.* **2009**, 1, 605.
- [214] Z. Li, R. Toivola, F. Ding, J. Yang, P. N. Lai, T. Howie, G. Georgeson, S. H. Jang, X. Li, B. D. Flinn, A. K.-Y. Jen, *Adv. Mater.* **2016**, 28, 6592.
- [215] Y. Li, Z. Ma, A. Li, W. Xu, Y. Wang, H. Jiang, K. Wang, Y. Zhao, X. Jia, *ACS Appl. Mater. Interfaces* **2017**, 9, 8910.
- [216] M. J. Robb, W. Li, R. C. R. Gergely, C. C. Matthews, S. R. White, N. R. Sottos, J. S. Moore, *ACS Cent. Sci.* **2016**, 2, 598.
- [217] K. Imato, J. C. Natterodt, J. Sapkota, R. Goseki, C. Weder, A. Takahara, H. Otsuka, *Polym. Chem.* **2017**, 8, 2115.
- [218] W. J. Crookes, L. L. Ding, Q. L. Huang, J. R. Kimbell, J. Horwitz, M. J. HcFall-Ngai, *Science* **2004**, 303, 235.
- [219] P. Lova, G. Manfredi, D. Comoretto, *Adv. Opt. Mater.* **2018**, 6, 1800730.
- [220] D. Comoretto, *Organic and Hybrid Photonic Crystals*, Switzerland: Springer International Publishing, **2015**.
- [221] M. F. Weber, C. A. Stover, L. R. Gilbert, T. J. Nevitt, A. J. Ouder Kirk, *Science* **2000**, 287, 2451.
- [222] M. Malekovic, M. Urann, U. Steiner, B. D. Wilts, M. Kolle, *Adv. Opt. Mater.* **2020**, 8, 2000165.
- [223] S. Daqiqeh Rezaei, J. Ho, A. Naderi, M. Tavakkoli Yarak, T. Wang, Z. Dong, S. Ramakrishna, J. K. W. Yang, *Adv. Opt. Mater.* **2019**, 7, 1900735.
- [224] E. Lee, M. Zhang, Y. Cho, Y. Cui, J. Van Der Spiegel, N. Engheta, S. Yang, *Adv. Mater.* **2014**, 26, 4127.
- [225] S. Liu, Y. Long, C. Liu, Z. Chen, K. Song, *Adv. Opt. Mater.* **2017**, 5, 1601043.
- [226] W. Yu, X. Jia, Y. Long, L. Shen, Y. Liu, W. Guo, S. Ruan, *ACS Appl. Mater. Interfaces* **2015**, 7, 9920.
- [227] G. M. A. V. Kavokin, J. J. Baumberg, F. P. Laussy, *Microcavities*, Oxford Science Publications, Oxford **2007**.
- [228] A. Genco, G. Giordano, S. Carallo, G. Accorsi, Y. Duan, S. Gambino, M. Mazzeo, *Org. Electron.* **2018**, 62, 174.
- [229] X. Fu, Y. Mehta, Y.-A. Chen, L. Lei, L. Zhu, N. Barange, Q. Dong, S. Yin, J. Mendes, S. He, R. Gogusetti, C.-H. Chang, F. So, *Adv. Mater.* **2021**, 33, 2006801.
- [230] S. Kinoshita, S. Yoshioka, J. Miyazaki, *Rep. Prog. Phys.* **2008**, 71, 076401.
- [231] J. Bailey, J. S. Sharp, *Eur. Phys. J. E* **2010**, 33, 41.
- [232] J. Bailey, J. S. Sharp, *J. Polym. Sci., Part B: Polym. Phys.* **2011**, 49, 732.
- [233] N. Gibbons, M. Kolle, B. Zheng, J. J. Baumberg, U. Steiner, *Opt. InfoBase Conf. Pap.* **2010**, 18, 575.
- [234] Z. Wang, C. Wang, H. Hou, W. Liu, R. Nian, H. Sun, X. Chen, G. Cui, *Appl. Surf. Sci.* **2019**, 479, 1014.
- [235] Z. Wang, J. Zhang, J. Xie, Z. Wang, Y. Yin, J. Li, Y. Li, S. Liang, L. Zhang, L. Cui, H. Zhang, B. Yang, *J. Mater. Chem.* **2012**, 22, 7887.
- [236] S. Noda, *MRS Bulletin* **2001**, 26, 618.
- [237] T. Kazmierczak, H. Song, A. Hiltner, E. Baer, *Macromol. Rapid Commun.* **2007**, 28, 2210.
- [238] Y.-L. Ko, H.-P. Tsai, K.-Y. Lin, Y.-C. Chen, H. Yang, *J. Colloid Interface Sci.* **2017**, 487, 360.
- [239] D. Khim, H. Han, K. Baeg, J. Kim, S. Kwak, D. Kim, Y. Noh, *Adv. Mater.* **2013**, 25, 4302.
- [240] S. G. Bucella, A. Luzzio, E. Gann, L. Thomsen, C. R. McNeill, G. Pace, A. Perinot, Z. Chen, A. Facchetti, M. Caironi, *Nat. Commun.* **2015**, 6, 8394.
- [241] K. L. Naughton, L. Phan, E. M. Leung, R. Kautz, Q. Lin, Y. Van Dyke, B. Marmiroli, B. Sartori, A. Arvai, S. Li, M. E. Pique, M. Naeim, J. P. Kerr, M. J. Aquino, V. A. Roberts, E. D. Getzoff, C. Zhu, S. Bernstorff, A. A. Gorodetsky, *Adv. Mater.* **2016**, 28, 8405.
- [242] D. D. Ordinario, E. M. Leung, L. Phan, R. Kautz, W. K. Lee, M. Naeim, J. P. Kerr, M. J. Aquino, P. E. Sheehan, A. A. Gorodetsky, *Adv. Opt. Mater.* **2017**, 5, 1600751.
- [243] L. Phan, W. G. Walkup IV, D. D. Ordinario, E. Karshalev, J. M. Jocson, A. M. Burke, A. A. Gorodetsky, *Adv. Mater.* **2013**, 25, 5621.
- [244] L. Phan, D. D. Ordinario, E. Karshalev, W. G. Walkup IV, M. A. Shenk, A. A. Gorodetsky, *J. Mater. Chem. C* **2015**, 3, 6493.
- [245] T. Cai, K. Han, P. Yang, Z. Zhu, M. Jiang, Y. Huang, C. Xie, *Sci. Rep.* **2019**, 9, 5201.
- [246] P. B. Dennis, K. M. Singh, M. C. Vasudev, R. R. Naik, W. J. Crookes-Goodson, *APL Mater.* **2017**, 5, 120701.
- [247] D. T. Simon, E. O. Gabrielsson, K. Tybrandt, M. Berggren, *Chem. Rev.* **2016**, 116, 13009.
- [248] D. G. DeMartini, A. Ghoshal, E. Pandolfi, A. T. Weaver, M. Baum, D. E. Morse, *J. Exp. Biol.* **2013**, 216, 3733.
- [249] R. T. Hanlon, L. M. Mähger, G. R. R. Bell, A. M. Kuzirian, S. L. Senft, *Bioinspir. Biomim.* **2018**, 13, 035002.
- [250] R. A. CLONEY, S. L. BROCCO, *Am. Zool.* **1983**, 23, 581.
- [251] L. M. Mähger, S. L. Senft, M. Gao, S. Karaveli, G. R. R. Bell, R. Zia, A. M. Kuzirian, P. B. Dennis, W. J. Crookes-Goodson, R. R. Naik, et al., *Adv. Funct. Mater.* **2013**, 23, 3980.
- [252] G. R. R. Bell, L. M. Mähger, M. Gao, S. L. Senft, A. M. Kuzirian, G. W. Kattawar, R. T. Hanlon, *Adv. Mater.* **2014**, 26, 4352.
- [253] M. D. Weber, L. Niklaus, M. Pröschel, P. B. Coto, U. Sonnewald, R. D. Costa, *Adv. Mater.* **2015**, 27, 5493.
- [254] J. J. Walsh, Y. Kang, R. A. Mickiewicz, E. L. Thomas, *Adv. Mater.* **2009**, 21, 3078.
- [255] N. Sakamoto, T. Hashimoto, C. D. Han, D. Kim, N. Y. Vaidya, *Macromolecules* **1997**, 30, 5321.
- [256] N. Sakamoto, T. Hashimoto, *Macromolecules* **1998**, 31, 8493.
- [257] A. P. Tomsia, M. Rühle, D. R. Clarke, *Annu. Rev. Mater. Res.* **2008**, 38, 1.
- [258] R. Nagarajan, K. Ganesh, *J. Chem. Phys.* **1989**, 90, 5843.
- [259] W. Cao, S. Xia, M. Appold, N. Saxena, L. Bießmann, S. Grott, N. Li, M. Gallei, S. Bernstorff, P. Müller-Buschbaum, *Sci. Rep.* **2019**, 9, 18269.
- [260] G. Jacucci, B. W. Longbottom, C. Parkins, S. A. F. Bon, S. Vignolini, *J. Mater. Chem. C* **2021**, 9, 2695.
- [261] I. W. Hamley, *Prog. Polym. Sci.* **2009**, 34, 1161.
- [262] P. Mokarian-Tabari, R. Senthamarakannan, C. Glynn, T. W. Collins, C. Cummins, D. Nugent, C. O'Dwyer, M. A. Morris, *Nano Lett.* **2017**, 17, 2973.
- [263] M. Stevens, S. Merilaita, *Animal Camouflage: Mechanisms and Function*, Cambridge University Press, Cambridge **2011**.
- [264] J. M. Clough, C. Weder, S. Schrettl, *Macromol. Rapid Commun.* **2021**, 42, 2000528.
- [265] H. Yuk, T. Zhang, G. A. Parada, X. Liu, X. Zhao, *Nat. Commun.* **2016**, 7, 12028.
- [266] C. Xu, M. C. Escobar, A. A. Gorodetsky, *Adv. Mater.* **2020**, 30, 1905717.
- [267] J. M. Clough, J. van der Gucht, T. E. Kodger, J. Sprakel, *Adv. Funct. Mater.* **2020**, 30, 2002716.
- [268] T. Wang, N. Zhang, J. Dai, Z. Li, W. Bai, R. Bai, *ACS Appl. Mater. Interfaces* **2017**, 9, 11874.
- [269] S. C. Baxter, B. J. Burrows, B. S. Fralick, *Probabilistic Eng. Mech.* **2016**, 44, 35.
- [270] J. J. Allen, G. R. R. Bell, A. M. Kuzirian, S. S. Velankar, R. T. Hanlon, *J. Morphol.* **2014**, 275, 371.
- [271] J. J. Allen, G. R. R. Bell, A. M. Kuzirian, R. T. Hanlon, *J. Morphol.* **2013**, 274, 645.
- [272] A. Rafsanjani, Y. Zhang, B. Liu, S. M. Rubinstein, K. Bertoldi, *Sci. Rob.* **2018**, 3, eaar7555.
- [273] E. Siéfert, E. Reyssat, J. Bico, B. Roman, *Nat. Mater.* **2019**, 18, 24.
- [274] E. Hajiesmaili, D. R. Clarke, *Nat. Commun.* **2019**, 10, 183.
- [275] S. Y. Kim, R. Baines, J. Booth, N. Vasios, K. Bertoldi, R. Kramer-Bottiglio, *Nat. Commun.* **2019**, 10, 3464.
- [276] B. A. Kowalski, T. C. Guin, A. D. Augustine, N. P. Godman, T. J. White, *ACS Macro Lett.* **2017**, 6, 436.

- [277] D. Rhee, S. Deng, T. W. Odom, *Nanoscale* **2020**, *12*, 23920.
- [278] M. del Pozo, C. Delaney, C. W. M. Bastiaansen, D. Diamond, A. P. H. J. Schenning, L. Florea, *ACS Nano* **2020**, *14*, 9832.
- [279] P. Kim, L. D. Zarzar, M. Khan, M. Aizenberg, J. Aizenberg, in *Proc. SPIE*, Vol. 7927, SPIE, Bellingham, WA **2011**, p. 792705.
- [280] J. E. Song, E. C. Cho, *Sci. Rep.* **2016**, *6*, 34622.
- [281] G. Wang, X. Chen, S. Liu, C. Wong, S. Chu, *ACS Nano* **2016**, *10*, 1788.
- [282] H. Lee, H. Kim, I. Ha, J. Jung, P. Won, H. Cho, J. Yeo, S. Hong, S. Han, J. Kwon, K. J. Cho, S. H. Ko, *Soft Ro.* **2019**, *6*, 760.
- [283] N. Kellaris, V. Gopaluni Venkata, G. M. Smith, S. K. Mitchell, C. Keplinger, *Sci. Rob.* **2018**, *3*, eaar3276.
- [284] A. Agrawal, P. Luchette, P. Palffy-Muhoray, S. L. Biswal, W. G. Chapman, R. Verduzco, *Soft Matter* **2012**, *8*, 7138.
- [285] Y. Shi, C. Zhu, J. Li, J. Wei, J. Guo, *New J. Chem.* **2016**, *40*, 7311.
- [286] X. Li, J. Liu, D. Li, S. Huang, K. Huang, X. Zhang, *Adv. Sci.* **2021**, <https://doi.org/10.1002/advs.202101295>.
- [287] M. Hermes, M. Ishida, M. Luhar, M. T. Tolley, in *Bioinspired Sensing, Actuation, and Control in Underwater Soft Robotic Systems*, Springer, Cham **2021**, p. 7.
- [288] C. A. Aubin, S. Choudhury, R. Jerch, L. A. Archer, J. H. Pikul, R. F. Shepherd, *Nature* **2019**, *571*, 51.
- [289] R. V. Martinez, J. L. Branch, C. R. Fish, L. Jin, R. F. Shepherd, R. M. D. Nunes, Z. Suo, G. M. Whitesides, *Adv. Mater.* **2013**, *25*, 205.
- [290] Q. He, Z. Wang, Y. Wang, A. Minori, M. T. Tolley, S. Cai, *Sci. Adv.* **2019**, *5*, eaax5746.
- [291] B. Mosadegh, P. Polygerinos, C. Keplinger, S. Wennstedt, R. F. Shepherd, U. Gupta, J. Shim, K. Bertoldi, C. J. Walsh, G. M. Whitesides, *Adv. Funct. Mater.* **2014**, *24*, 2163.
- [292] M. Guttag, M. C. Boyce, *Adv. Funct. Mater.* **2015**, *25*, 3641.
- [293] Y. Dong, J. Wang, X. Guo, S. Yang, M. O. Ozen, P. Chen, X. Liu, W. Du, F. Xiao, U. Demirci, B. F. Liu, *Nat. Commun.* **2019**, *10*, 4087.
- [294] H. Kim, S. Ahn, D. M. Mackie, J. Kwon, S. H. Kim, C. Choi, Y. H. Moon, H. B. Lee, S. H. Ko, *Mater. Today* **2020**, *41*, 243.
- [295] B. Xu, X. Han, Y. Hu, Y. Luo, C.-H. Chen, Z. Chen, P. Shi, *Small* **2019**, *15*, 1900006.
- [296] M. Sitti, H. Ceylan, W. Hu, J. Giltinan, M. Turan, S. Yim, E. Diller, *Proc. IEEE* **2015**, *103*, 205.
- [297] D. Lunni, M. Cianchetti, C. Filippeschi, E. Sinibaldi, B. Mazzolai, *Adv. Mater. Interfaces* **2020**, *7*, 1901310.
- [298] D.-Y. Lee, S.-R. Kim, J.-S. Kim, J.-J. Park, K.-J. Cho, *Soft Rob.* **2017**, *4*, 163.
- [299] T. Nakajima, T. Yamaguchi, S. Wakabayashi, T. Arie, S. Akita, K. Takei, *Adv. Mater. Technol.* **2020**, *5*, 2000201.
- [300] D. Li, S. Wang, J. He, H. Zeng, K. Yao, Z. Gao, M. Wu, Y. Liu, L. Wang, Z. Xie, X. Yu, *Adv. Mater. Technol.* **2021**, *6*, 2001095.
- [301] S. Chen, Y. Pang, Y. Cao, X. Tan, C. Cao, *Adv. Mater. Technol.* **2021**, *6*, 1906244.
- [302] Y. F. Zhang, C. J. X. Ng, Z. Chen, W. Zhang, S. Panjwani, K. Kowsari, H. Y. Yang, Q. Ge, *Adv. Mater. Technol.* **2019**, *4*, 1900427.
- [303] F. Corucci, N. Cheney, F. Giorgio-Serchi, J. Bongard, C. Laschi, *Soft Rob.* **2018**, *5*, 475.
- [304] A. T. Scaros, A. Andouche, S. Baratte, R. P. Croll, *J. Comp. Neurol.* **2020**, *528*, 1095.
- [305] C.-C. Chiao, R. Hanlon, *Oxford Research Encyclopedia of Neuroscience*, Oxford University Press, Oxford **2019**.
- [306] P. T. Gonzalez-Bellido, A. T. Scaros, R. T. Hanlon, T. J. Wardill, *Iscience* **2018**, *1*, 24.
- [307] R. Pfeifer, M. Lungarella, F. Iida, *Science* **2007**, *318*, 1088.
- [308] J. Tapia, E. Knoop, M. Mutný, M. A. Otaduy, M. Bächer, *Soft Rob.* **2020**, *7*, 332.
- [309] G. Soter, A. Conn, H. Hauser, J. Rossiter, in *2018 IEEE Int. Conf. on Robotics and Automation*, IEEE, Piscataway, NJ **2018**, pp. 2448–2453.
- [310] Y. Lee, T.-W. Lee, *Acc. Chem. Res.* **2019**, *52*, 964.
- [311] K. Chin, T. Hellebrekers, C. Majidi, *Adv. Intell. Syst.* **2020**, *2*, 1900171.
- [312] A. Hess, X. Tan, T. Gao, *Bioinspir. Biomim.* **2020**, *15*, 035004.
- [313] C. D. Santina, M. Bianchi, G. Grioli, F. Angelini, M. Catalano, M. Garabini, A. Bicchi, *IEEE Rob. Autom. Mag.* **2017**, *24*, 75.
- [314] K. Man, A. Damasio, *Nat. Mach. Intell.* **2019**, *1*, 446.
- [315] C. Laschi, M. Cianchetti, *Front. Bioeng. Biotechnol.* **2014**, *2*, 3.
- [316] K. Nakajima, H. Hauser, T. Li, R. Pfeifer, *Soft Robot.* **2018**, *5*, 339.
- [317] T. George Thuruthel, Y. Ansari, E. Falotico, C. Laschi, *Soft Rob.* **2018**, *5*, 149.
- [318] R. L. Truby, M. Wehner, A. K. Grosskopf, D. M. Vogt, S. G. M. Uzel, R. J. Wood, J. A. Lewis, *Adv. Mater.* **2018**, *30*, 1706383.
- [319] W. Zhuang, G. Sun, H. Li, X. Lou, M. Dong, L. Zhu, *Optik* **2018**, *165*, 7.
- [320] L. Xu, N. Liu, J. Ge, X. Wang, M. P. Fok, *Opt. Lett.* **2018**, *43*, 2503.
- [321] E. Al-Fakih, N. A. A. Osman, F. R. M. Adikan, *Sensors (Switzerland)* **2012**, *12*, 12890.
- [322] S. C. Ryu, P. E. Dupont, in *2018 IEEE Int. Conf. on Robotics and Automation*, IEEE, Piscataway, NJ **2018**, p. 3531.
- [323] S. Liu, Y. Ji, J. Yang, W. Sun, H. Li, *Sens. Actuators, A* **2018**, *269*, 313.
- [324] H. Wang, R. Zhang, W. Chen, X. Liang, R. Pfeifer, *IEEE/ASME Trans. Mechatronics* **2016**, *21*, 2977.
- [325] C. K. Harnett, H. Zhao, R. F. Shepherd, *Adv. Mater. Technol.* **2017**, *2*, 1700087.
- [326] H. Zhao, K. O'Brien, S. Li, R. F. Shepherd, *Sci. Rob.* **2016**, *1*, eaai7529.
- [327] N. Jiang, R. Ahmed, A. A. Rifat, J. Guo, Y. Yin, Y. Montelongo, H. Butt, A. K. Yetisen, *Adv. Opt. Mater.* **2018**, *6*, 1701118.
- [328] J. Guo, M. Niu, C. Yang, *Optica* **2017**, *4*, 1285.
- [329] J. D. Sandt, M. Moudio, J. K. Clark, J. Hardin, C. Argenti, M. Carty, J. A. Lewis, M. Kolle, *Adv. Healthcare Mater.* **2018**, *7*, 1800293.
- [330] S. Yun, S. Park, B. Park, Y. Kim, S. K. Park, S. Nam, K. Kyung, *Adv. Mater.* **2014**, *26*, 4474.
- [331] M. Totaro, A. Mondini, A. Bellacicca, P. Milani, L. Beccai, *Soft Rob.* **2017**, *4*, 400.
- [332] S. Sareh, Y. Noh, M. Li, T. Ranzani, H. Liu, K. Althoefer, *Smart Mater. Struct.* **2015**, *24*, 125024.
- [333] D. Lunni, G. Giordano, E. Sinibaldi, M. Cianchetti, B. Mazzolai, in *2018 IEEE Int. Conf. Soft Robotics RoboSoft 2018*, Institute Of Electrical And Electronics Engineers Inc., Piscataway, NJ **2018**, pp. 541–546.
- [334] L. Birolo, N. Alberto, J. L. Pinto, R. Nogueira, *Sensors (Switzerland)* **2012**, *12*, 12184.
- [335] J. Liu, Y. Hou, H. Zhang, P. Jia, S. Su, G. Fang, W. Liu, J. Xiong, *Sensors* **2017**, *17*, 196.
- [336] S. Song, Z. Li, H. Yu, H. Ren, *Mechatronics* **2015**, *29*, 28.
- [337] D. Shah, B. Yang, S. Kriegman, M. Levin, J. Bongard, R. Kramer-Bottiglio, *Adv. Mater.* **2020**, *33*, 2002882.
- [338] M. Choi, M. Humar, S. Kim, S. H. Yun, *Adv. Mater.* **2015**, *27*, 4081.
- [339] J. Guo, B. Zhou, C. Yang, Q. Dai, L. Kong, *Adv. Funct. Mater.* **2019**, *29*, 1902898.
- [340] J. Guo, X. Liu, N. Jiang, A. K. Yetisen, H. Yuk, C. Yang, A. Khademhosseini, X. Zhao, S. H. Yun, *Adv. Mater.* **2016**, *28*, 10244.
- [341] Z. Liu, D. Qi, G. Hu, H. Wang, Y. Jiang, G. Chen, Y. Luo, X. J. Loh, B. Liedberg, X. Chen, *Adv. Mater.* **2018**, *30*, 1704229.
- [342] A. Leber, B. Cholst, J. Sandt, N. Vogel, M. Kolle, *Adv. Funct. Mater.* **2019**, *29*, 1802629.
- [343] N. Lu, D.-H. Kim, *Soft Rob.* **2013**, *1*, 53.
- [344] J. A. Rogers, T. Someya, Y. Huang, *Science* **2010**, *327*, 1603.
- [345] Y. L. Park, C. Majidi, R. Kramer, P. Brard, R. J. Wood, *J. Micromech. Microeng.* **2010**, *20*, 125029.
- [346] C. Majidi, R. Kramer, R. J. Wood, *Smart Mater. Struct.* **2011**, *20*, 105017.
- [347] S. S. Robinson, K. W. O'Brien, H. Zhao, B. N. Peele, C. M. Larson, B. C. Mac Murray, I. M. Van Meerbeek, S. N. Dunham, R. F. Shepherd, *Extreme Mech. Lett.* **2015**, *5*, 47.

- [348] D. J. Lipomi, M. Vosgueritchian, B. C. K. Tee, S. L. Hellstrom, J. A. Lee, C. H. Fox, Z. Bao, *Nat. Nanotechnol.* **2011**, *6*, 788.
- [349] P. Roberts, D. D. Damian, W. Shan, T. Lu, C. Majidi, in *Proc. – IEEE Int. Conf. on Robotics and Automation*, IEEE, Piscataway, NJ **2013**, p. 3529.
- [350] S. Li, H. Zhao, R. F. Shepherd, *MRS Bull.* **2017**, *42*, 138.
- [351] M. D. Dickey, *Adv. Mater.* **2017**, *29*, 1606425.
- [352] J. Guo, C. Xiang, J. Rossiter, *Mater. Des.* **2018**, *156*, 586.
- [353] E. L. White, J. C. Case, R. K. Kramer, *Sens. Actuators, A* **2017**, *253*, 188.
- [354] S. Russo, T. Ranzani, H. Liu, S. Nefti-Meziani, K. Althoefer, A. Menciassi, *Soft Rob.* **2015**, *2*, 146.
- [355] Y. Yang, Y. Chen, *IEEE Rob. Autom. Lett.* **2018**, *3*, 656.
- [356] M. Luo, E. H. Skorina, W. Tao, F. Chen, S. Ozel, Y. Sun, C. D. Onal, *Soft Rob.* **2017**, *4*, 117.
- [357] L. Qian, Y. Sun, M. Wu, C. Li, D. Xie, L. Ding, G. Shi, *Nanoscale* **2018**, *10*, 6837.
- [358] J. Ge, S. Zhang, Z. Liu, Z. Xie, S. Pan, *Nanoscale* **2019**, *11*, 6591.
- [359] Y. Kim, A. Chortos, W. Xu, Y. Liu, J. Y. Oh, D. Son, J. Kang, A. M. Foudah, C. Zhu, Y. Lee, *Science* **2018**, *360*, 998.
- [360] M. Giordani, M. Berto, M. Di Lauro, C. A. Bortolotti, M. Zoli, F. Biscarini, *ACS Sens.* **2017**, *2*, 1756.
- [361] H. Park, Y. Lee, N. Kim, D. Seo, G. Go, T. Lee, *Adv. Mater.* **2020**, *32*, 1903558.
- [362] J. Rivnay, S. Inal, A. Salleo, R. M. Owens, M. Berggren, G. G. Malliaras, *Nat. Rev. Mater.* **2018**, *3*, 17086.
- [363] Y. Wang, Z. Lv, Q. Liao, H. Shan, J. Chen, Y. Zhou, L. Zhou, X. Chen, V. A. L. Roy, Z. Wang, *Adv. Mater.* **2018**, *30*, 1800327.
- [364] Z. Lv, Y. Wang, Z. Chen, L. Sun, J. Wang, M. Chen, Z. Xu, Q. Liao, L. Zhou, X. Chen, *Adv. Sci.* **2018**, *5*, 1800714.
- [365] Y. Lee, J. Y. Oh, W. Xu, O. Kim, T. R. Kim, J. Kang, Y. Kim, D. Son, J. B.-H. Tok, M. J. Park, *Sci. Adv.* **2018**, *4*, eaat7387.
- [366] H. Shim, K. Sim, F. Ershad, P. Yang, A. Thukral, Z. Rao, H.-J. Kim, Y. Liu, X. Wang, G. Gu, L. Gao, X. Wang, Y. Chai, C. Yu, *Sci. Adv.* **2019**, *5*, eaax4961.
- [367] M. Karbalaee Akbari, S. Zhuyikov, *Nat. Commun.* **2019**, *10*, 3873.
- [368] M. Zhu, T. He, C. Lee, *Appl. Phys. Rev.* **2020**, *7*, 031305.
- [369] Y. Chen, G. Gao, J. Zhao, H. Zhang, J. Yu, X. Yang, Q. Zhang, W. Zhang, S. Xu, J. Sun, Y. Meng, Q. Sun, *Adv. Funct. Mater.* **2019**, *29*, 1900959.
- [370] D. Kim, I.-J. Kim, J.-S. Lee, *Adv. Intell. Syst.* **2020**, *3*, 2000206.
- [371] W. Huang, P. Hang, Y. Wang, K. Wang, S. Han, Z. Chen, W. Peng, Y. Zhu, M. Xu, Y. Zhang, Y. Fang, X. Yu, D. Yang, X. Pi, *Nano Energy* **2020**, *73*, 104790.
- [372] M. Hofer, C. Sferrazza, R. D'Andrea, *arXiv Prepr. arXiv2012.06413* **2020**.
- [373] P. Hyatt, D. Wingate, M. D. Killpack, *Front. Rob. AI* **2019**, *6*, 22.
- [374] J. Hughes, A. Spielberg, M. Chounlakone, G. Chang, W. Matusik, D. Rus, *Adv. Intell. Syst.* **2020**, *2*, 2000002.
- [375] I. M. Van Meerbeek, C. M. De Sa, R. F. Shepherd, *Sci. Rob.* **2018**, *3*, eaau2489.
- [376] V. Wall, G. Zöllner, O. Brock, in *2017 IEEE Int. Conf. on Robotics and Automation*, IEEE, Piscataway, NJ **2017**, pp. 4965–4970.
- [377] T. G. Thuruthel, B. Shih, C. Laschi, M. T. Tolley, *Sci. Rob.* **2019**, *4*, eaav1488.
- [378] R. L. Truby, C. Della Santina, D. Rus, *IEEE Rob. Autom. Lett.* **2020**, *5*, 3299.
- [379] M. T. Gillespie, C. M. Best, E. C. Townsend, D. Wingate, M. D. Killpack, in *2018 IEEE Int. Conf. on Soft Robotics*, IEEE, Piscataway, NJ **2018**, pp. 39–45.
- [380] E. Del Dottore, A. Mondini, A. Sadeghi, B. Mazzolai, *Appl. Sci.* **2018**, *8*, 47.
- [381] F. Tedone, E. Del Dottore, M. Palladino, B. Mazzolai, P. Marcati, *Bioinspir. Biomim.* **2020**, *15*, 056006.
- [382] S. Coyle, C. Majidi, P. LeDuc, K. J. Hsia, *Extreme Mech. Lett.* **2018**, *22*, 51.
- [383] G.-Z. Yang, J. Bellingham, P. E. Dupont, P. Fischer, L. Floridi, R. Full, N. Jacobstein, V. Kumar, M. McNutt, R. Merrifield, B. J. Nelson, B. Scassellati, M. Taddeo, R. Taylor, M. Veloso, Z. L. Wang, R. Wood, *Sci. Rob.* **2018**, *3*, eaar7650.
- [384] B. R. Napolitano, W. Reinhart, J. P. Gevaudan, **2021**, *37*, 1200.
- [385] L. Persano, A. Camposeo, D. Pisignano, *J. Mater. Chem. C* **2013**, *1*, 7663.
- [386] J. Xie, H. Ping, T. Tan, L. Lei, H. Xie, X.-Y. Yang, Z. Fu, *Prog. Mater. Sci.* **2019**, *105*, 100571.
- [387] G. A. Naselli, B. Mazzolai, *Int. J. Rob. Res.* **2019**, *40*, 197.
- [388] C. I. Idumah, C. M. Obele, *Surf. Interfaces* **2021**, *22*, 100879.
- [389] D. Kim, S.-H. Kim, T. Kim, B. B. Kang, M. Lee, W. Park, S. Ku, D. Kim, J. Kwon, H. Lee, *PLoS One* **2021**, *16*, 0246102.
- [390] M. Schwartz, J. P. F. Lagerwall, *arXiv Prepr. arXiv2103.11005* **2021**.



Goffredo Giordano received his M.Sc. in condensed matter physics, and nanotechnologies for biomedicine and the environment in 2016 from the University of Salento, Italy. He is currently a Ph.D. student in Biorobotics at the Scuola Superiore Sant'Anna (Pisa), and affiliated scholarship student at the Istituto Italiano di Tecnologia (IIT) in the Bioinspired Soft Robotics group under the supervision of Dr. Barbara Mazzolai. He focuses on the development of mechanochromic biomimetic systems, stretch-tunable optical materials, fiber optic-based sensing systems, opto-electronics, and soft robotics.



Marco Carlotti received his M.Sc. (cum laude) in inorganic chemistry in 2014 from the University of Pisa. In 2019, he obtained a Ph.D. in Molecular Electronics at the University of Groningen under the supervision of Prof. R. C. Chiechi. He currently holds a Postdoc position as MSCA Fellow at the Center for Materials Interfaces of the Istituto Italiano di Tecnologia where he focuses on the development of new conjugated polymers and functional materials for organic electronics and microfabrication.



Barbara Mazzolai is an associate director for Robotics and Director of the Bioinspired Soft Robotics Laboratory at Istituto Italiano di Tecnologia (IIT). From 2011 to 2021 she was Director of IIT Center for Micro-BioRobotics. She graduated (M.Sc.) in Biology (with Honours) at University of Pisa, and received the Ph.D. in Microsystem Engineering from University of Rome TorVergata. She was Coordinator of EU FET-Open PLANTOID project, and she is Coordinator of EU FET-Proactive GrowBot and of EU FET-Proactive I-SEED projects. She is author and co-author of more than 270 papers in international journals, books, and conference proceedings in bioinspired and soft robotics.