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When Cellulose Moves: Smart Sensors and Actuators

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Cellulose, the most abundant biopolymer on Earth, has rapidly gained attention as a key component in the development of smart materials capable of dynamic and adaptive behavior. This review highlights the unique motion and actuation capabilities of cellulose-based systems, focusing on their ability to convert external stimuli into mechanical movement. The recent progress in cellulose-derived soft actuators that respond to moisture, electric fields, thermal and light input, and magnetic forces is detailed – enabling programmable shape changes, bending, twisting, and locomotion. Alongside these, cellulose-based soft sensors that transduce environmental cues though magnetic, resistive and capacitive mechanisms are examined. Special emphasis is placed on the synergy between sensing and actuation in multifunctional devices that mimic natural movement and responsiveness. Applications in wearable devices, soft robotics, biomedical systems, energy harvesting, smart packaging, and bioinspired technologies are explored to demonstrate the practical potential of these materials. The challenges in achieving robust, reversible, and multi-stimuli-responsive motion are also addressed, and future directions for scaling up and integrating cellulose-based smart systems into real-world applications are proposed.

1. Introduction

1.1. Cellulose

The ever-growing concern for the environment has fueled a rising fascination with cellulose and cellulose-based materials. Numerous researchers have embraced the journey of isolating, characterizing, and discovering novel uses for cellulose. Cellulose results from the reaction of glucose molecules with the condensation of water. Cellulose is a linear-chain polymer that presents three hydroxy groups per anhydroglucose unit (AGU). To adjust the preferred bond angles of acetal oxygen bridges, every second AGU ring rotates 180° in the plane. The repetitive unit of cellulose is cellobiose.[1,2] The groundbreaking work of Hermann Staudinger in 1920 marked a pivotal moment in understanding cellulose's polymeric structure. Cellulose structure, as can be observed in **Figure 1**, is composed of a β-D-glucopyranose unit linked by (1 → 4) glycosidic bonds.

Cellulose is one of the most abundant low-cost and renewable materials on the planet. As a response to a growing environmental awareness, the interest in biobased materials, such as cellulose, is continuously increasing.[3,4] Cellulose and cellulose derivatives are used, among others, for coatings, laminates, optical films, pharmaceuticals, and textiles.[5–7] The development of bio-based materials with low environmental impact will pave the way for the development of high-performance smart materials. Cellulose-based smart materials are usually in the form of aggregates and gels, particles, fibers, membranes, and films.[5] In recent years, the interest in nanocellulose, including cellulose nanocrystals (CNCs), cellulose nanofibers (CNF), and bacterial cellulose (BNC), has surged due to its high aspect ratio, excellent mechanical properties, and large surface area.[8] They offer unique opportunities in advanced material design, particularly in the realm of functional flexible electronics and smart sensing.[9,10]

1.2. Smart Materials

A smart structure is a system that contains multifunctional parts that perform sensing, control, and actuation. Smart materials are used to build these smart structures that present actuation and sensing functions.[11] These materials present inherent sensing, controlling, actuation, or information processing capabilities in their structures. The "intelligence quotient" (I.Q.) of smart materials is correlated with their responsiveness to stimuli and their agility to respond.[11] The smart materials can be tailored, task-specific, or designed to provide enhancement of a specific property. Strain, stress, temperature, chemical composition (such as pH), electrical field, magnetic field, water content, radiation, among others are some of the stimuli that may act upon smart materials ability to receive, transmit, or process a stimulus.

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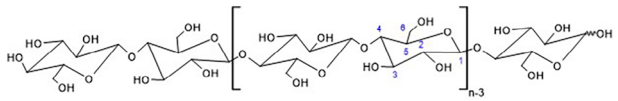

Figure 1. Molecular structure of cellulose (n = degree of polymerization). Cellobiose is the repetitive unit of cellulose, and the formation of hydrogen bonds is due to the existence of –OH.

Smart materials can be categorized as: i) sensors-that perform a non-mechanical response upon a mechanical stimulus and ii) actuators-materials that react to non-mechanical stimulus with a mechanical reply (see **Figure 2**).[12]

The advances in material science, structural engineering, and manufacturing significantly motivate the transformation of materials from their traditional, rigid forms with rectangular architectures to soft, flexible, stretchable forms with various architectures.[13] Motivated by the wonders of the Natural world and nature smart systems, considerable endeavors have been dedicated to converting external stimuli into desired mechanical motion using cellulose and cellulose-based materials. The pursuit of smart materials systems with functional-oriented structural design, with high-efficiency multi-responsive materials has become a hot topic in the research of soft-smart materials.

1.3. Cellulose-Based Soft Sensors

In this review, a broader definition of sensor will be considered and designed as a device that can detect and measure physical properties or changes in the environment, converts it into a signal, which can be read by an observer or an instrument. Sensors can detect several physical properties involved in many different systems, such as temperature (thermocouples and temperature detectors), pressure (piezoelectric), magnetism (Hall effect and magnetochemical sensors), motion and proximity (capacitive, inductive, and infrared motion sensors), gas (catalytic, metal-oxide, and electrochemical sensors). A sensor is usually made of several materials, and it can combine metals, semiconductors, ceramics, glass, and polymers. Most of these materials are derived from non-renewable resources. In the case of cellulose-based soft sensors, the materials involved in their structure are mostly based on cellulose and cellulose derivatives. Cellulose-based soft sensors, with their biodegradability and eco-friendly properties, are not only a sustainable choice but also a cost-effective one. They are pointed as less expensive and easier to produce than conventional sensors.[14–18] While they may not match the efficiency of conventional sensors, their affordability should reassure potential users about their practicality.[19–22]

1.3.1. Magnetic

Magnetic sensors typically consist of a magnetic component and a Hall sensor, the latter being responsible for converting external stimuli – such as mechanical forces or deformation – into changes in magnetic fields. The magnetic component is usually a permanent magnet or a soft composite containing magnetic particles. When subjected to external stimuli, these magnetic particles experience alterations in their magnetic field, which can be detected and quantified by the Hall sensor.[23]

Beyond mechanical sensing, magnetic fields have also been exploited to modulate the optical properties of materials, enabling multifunctional sensor applications.[24,25] One of the main advantages of using magnetic systems in actuators is their capability for contactless, remote sensing. Magnetic sensors can be based on semiconductor materials (e.g., Galliun Arsenide, Silicon) specially in Hall effect sensors, or on magnetic elements-based materials (e.g., Nickel, Iron) in the case of magnetoresistive sensors. Cellulose-based magnetic sensors emerge as a highly promising alternative due to their flexibility, biodegradability, lightweight nature, and compatibility with environmentally friendly fabrication processes. By incorporating magnetic nanoparticles into cellulose matrices, it is possible to create functional composites capable of responding to magnetic fields while retaining the mechanical adaptability of cellulose. This combination paves the way for the development of sustainable, flexible, and potentially disposable magnetic devices, particularly in applications such as biomedical monitoring, soft robotics, and smart packaging.

Hausmann et al.[26] recently reported use of cellulose nanocrystals (CNCs) as birefringent building blocks inside microparticles to manipulate light polarization. This manipulation is achieved utilizing a microfluidic platform to combine the microparticles with small concentrations of superparamagnetic iron oxide nanoparticles. Because of their combined properties it is possible to convert an input magnetic stimulus into an output optical signal with a well-defined frequency.[26] Yang et al.[27] stated a flexible and strong cellulose/iron oxide (Fe_3O_4) composite film based sensor where the inorganic particles are fixed in the cellulose matrix via a coordinate bond between Fe_3O_4 and the hydroxyl group of cellulose. The authors state that is possible to prepare a cellulose-based film with an excellent saturated magnetization as well as strong UV shielding properties using a simple methodology that could be used as magnetic and UV sensors.[27] Recently, bacterial cellulose was elected as the core component of a flexible magnetic sensor to develop an "intelligent jacket." The flexible, biodegradable, and self-powered electromagnetic sensor was prepared by in situ co-precipitation of $CoFe_2O_4$ nanoparticles onto the bacterial cellulose fibers paired with a post-process of hot-pressing and magnetizing. The magnetic film was sewed on the elbow of the jacket sleeve and allowed wireless monitor four typical human motions in different speeds. The combination of the flexible magnetic film with copper coils allowed an excellent performance of the intelligent jacket in the sensing, making the electromagnetic sensor developed by Chen et al. a promising device to be used in the wearable field.[28]

1.3.2. Capacitive

Capacitive sensors function by converting external stimuli, such as deformation, pressure, or displacement, into measurable changes in capacitance. By employing various structural designs, these sensors can be tailored to detect a wide range of signals. The capacitance, C of a parallel-plate capacitator is defined by the equation:

$$C = \frac{\varepsilon_0 \varepsilon_r S}{s} \quad (1)$$

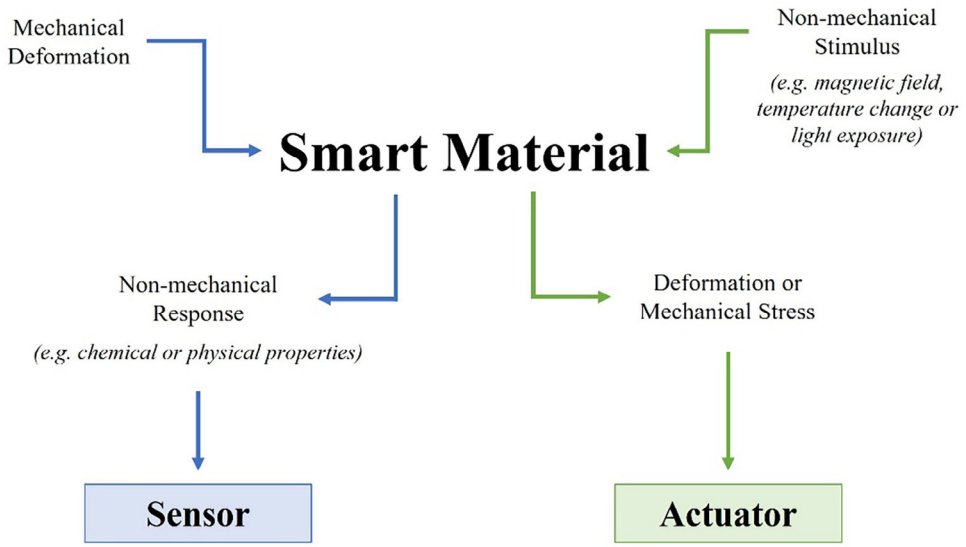

Figure 2. Categorizing Smart Materials: a sensor is a smart material that change its non-mechanical properties in response to a mechanical load and an actuator is a smart material will deform in response to a non-mechanical stimulus, adapted from.[12]

where ε_0 is the absolute dielectric constant, ε_r is the relative dielectric constant, s is the overlapping area between the two parallel plates, and d is the distance between the parallel plates. Variations in either s or d due to external forces result in a corresponding change in capacitance. This is the fundamental working principle of capacitive sensors: the external signal induces a mechanical or geometrical change that is transduced into an electrical signal through in capacitance.[23]

In the context of sustainable electronics, cellulose-based capacitive sensors have emerged as a promising alternative to conventional designs. In these systems, both the conductive plates and the dielectric layer can be derived from cellulose or cellulose-based composites, thereby eliminating the need for less environmentally friendly materials commonly used in traditional capacitors, such as copper (Cu), Nickel (Ni), indium tin oxide (ITO), synthetic polymers, and glass.[29,30]

Cellulose offers several advantages as a core material for soft-matter capacitators. Its intrinsic properties – biodegradability, mechanical flexibility, low weight, and availability from renewable sources – make it an attractive candidate for the next generation, eco-friendly electronic devices.[31–33] Furthermore, cellulose-based capacitive sensors exhibit high sensitivity and responsiveness, making them suitable for a range of applications including wearable electronics, pressure mapping, and flexible health-monitoring devices. The combination of environmental sustainability and functional performance positions cellulose capacitors as a key component in the advancement of green and flexible electronics.[22,34,35]

Mahadeva et al.[36] reported a flexible capacitive-type humidity and temperature cellulose-polypyrrole nanocomposite sensor. The nanocomposite was prepared by polymerization-induced adsorption of a polypyrrole layer onto a wet regenerated cellulose membrane. The dried cellulose-polypyrrole nanocomposite films thickness varies from 14.5 to 17.2 µm. The flexible cellulose-based sensor presents a reversible behavior with good response and recovery with capacitance increase dependent of humidity and temperature.[36]

Cheng et al. conducted a study demonstrating the use of cellulose acetate (CA)-coated screen-printed carbon electrodes (SPCEs) for capacitive sensing in conductive electrolytes. The CA layer functions as a semi-permeable dielectric film that modulates the electrode-electrolyte interface, enhancing capacitive signal stability and selectivity by restricting the diffusion of interfering species while allowing ion exchange relevant to microbial activity. The primary application of this sensor is to assess soil microbial indicators. Due to enzymatic activity and microbial respiration, these indicators serve as proxies for carbon balance and nutrient cycling, offering valuable insights into soil health. Monitoring soil health, particularly in stressed agricultural environments, is essential for promoting sustainable land management practices. Implementing eco-friendly technologies enhances the sustainability of these approaches.[37,38] In this study the 10-s dip-coating procedure for CA/SPCEs demonstrated impressive stability over a month, indicating their suitability for prolonged application in the observation of agricultural soil conditions.

The coated film CA coating undergoes progressive thinning or degradation though various mechanisms, including hydrolysis, enzymatic action, and microbial decomposition. These processes involve the sequential breakdown of cellulose into disaccharides and β-D-glucose, facilitated by cellulase and β-glucosidase, respectively. As degradation processes, capacitance increases, allowing the extent of material decomposition to be quantified based on capacitance variations.[39] Consequently, cellulose-based capacitor devices provide viable methods for evaluating soil health.

1.3.3. Resistive

Resistive sensors convert the change of the external signal into a change of resistance. The resistance R is defined by the formula

$R = \rho l/s$, where ρ is the electric resistivity, l is the length, and s is the cross-sectional area. The resistance can be changed if l or s is changed. The basic principle of resistive sensors is to convert the deformation or force information into the change of l or s, and as a consequence the resistance value. Resistive sensors are designed for different structures to measure different signals, such as stretching, normal force, and shear force.[23]

A cellulose/polyaniline sensor manufactured using a simple and scalable industrial process reported by Ragazzini et al.[40] The sensitivity of the sensor made of conductive cellulose fibres coated with polyaniline was 13%, a response time comparable to the commercial digital-output relative humidity and temperature sensor. The good recovery capacity and quick response to changes in the humidity highlight the feasibility of a low-cost sustainable humidity sensor. An innovative approach that uses directly laser-writing conductive electrodes onto TEMPO-oxidized cellulose paper to produce an all-cellulose humidity sensor was reported recently by Zhu et al.[41] This sensor demonstrates high sensitivity and linearity across a wide relative humidity range (11–98%). Its fast response to moisture and recovery times combined with its sensitivity makes it suitable for applications like monitoring plant transpiration and human perspiration. Conductive stretchable hydrogels fabricated with TEMPO-oxidized cellulose nanofibers combined with graphene with self-healing properties for the use of wearable monitoring human motion was reported in 2020.[42] TEMPO-oxidized cellulose nanofibers combined with carbon nanotubes and polyacrylamide matrix were used to produce a multifunctional wearable sensor that exhibited both high compressive (≈2.55 MPa at 60% strain) and tensile (≈0.15 MPa) strength. The cellulose-based conductive hydrogel also presented excellent intrinsic self-recovery properties and anti-fatigue capacity.[43] The use of nanocellulose in multifunctional hydrogel sensors was reported to enable Near Infrared (NIR) photo responsiveness and repeated use,[44] while the inclusion of carbon nanotube complexes has yielded systems with excellent self-recovery and mechanical-electrical stability.[43] Nanocellulose has played a pivotal role in the development of conductive hydrogels designed for flexible resistive sensors.[45] The integration of nanocellulose into hydrogel matrices to create highly responsive and durable sensor platform was recently reported by Zheng et al.[46] A double network hydrogel with nanocellulose was developed to provide self-adhesive, biocompatible, and environmentally resilient performance. A similar conductive fiber-based nanocellulose hydrogel fabricated via microfluidic spinning with high toughness and reliable strain sensitivity was reported in 2024 by Zhu et al.[47] More recently, a conductive nanocellulose-based hydrogel inspired by mussel adhesive strategies was reported by Ma et al. to be suitable for wearable human-motion monitoring.[48] The produced hydrogel presented key properties for continuous monitoring under deformation such as fatigue resistance, long term reliability and self-healing. Gong et al. reported the use of multifunctional regenerated silk fibers reinforced with conductive cellulose nanofibers to be used in flexible electronic devices, smart dressings, and underwater smart textiles.[49] The developed fibers can comprehensively monitor human motion and weak signals such as blinking and swallowing with high sensitivity (100 ms) and waterproof properties for 22 days. Furthermore, the fibers can be used as smart dressings in wound monitoring sensing humidity.

1.4. Cellulose-Based Soft Actuators

Cellulose-based soft actuators are an appealing class of materials and devices that take advantage of the well-known properties of cellulose in line with the demand for flexible eco-friendly materials. Cellulose based actuators are continuously advancing as a subject of ongoing research, focusing on enhancing their performance, response time, and fabrication methods.[50] Due to their composition, cellulose-based actuators are known to generate mechanical responses to different stimuli such as temperature, light, moisture, changes in electric field, and magnetic field.[51,52] This class of actuators has gained significant attention in recent years for their possible application in robotics, biomedical and wearable devices, sensors, and a variety of other fields.[52–54] In the next topics the actuators presented are categorized based on the driving signals.

An actuator system produces strength, torque, or displacement when energies from different forms are applied. This energy can be electric, magnetic, photonic, thermal in other words, actuators can transform energy from one form to another in the form of motion. These devices are applied in robotics, aerospace and defense systems, automobiles, medical and healthcare devices, and general electronic systems.[50,55,56] Soft matter actuators are promising for several applications from soft robotics and biomedicine to energy harvesting and aerospace technology. This includes artificial muscles and bio and microrobots, micromanipulators, moisture sensors, optical switches, adaptive surfaces and lightweight materials, smart textiles, and morphing structures for aerospace applications.[34,53]

1.4.1. Moisture Responsive

Moisture-responsive Cellulose-based soft actuators are an appealing class of materials and devices that take advantage of the well-known properties of cellulose in line with the demand for flexible eco-friendly materials. Cellulose-based actuators are continuously advancing as a subject of ongoing research, focusing on enhancing their performance, response time, and fabrication methods. Due to their composition, cellulose-based actuators are known to generate mechanical responses to different stimuli such as temperature, light, moisture, changes in electric field, and magnetic field.[19,20]

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Changes in moisture levels inside the material allow a controlled movement of the cellulosic structure. As cellulose is inherently hydrophilic it can absorb the moisture present in air making it possible to take advantage of the swelling and deswelling to change shape and create movement. Humidity sensors typically consist of a material that detects humidity and electrodes. Materials that sense humidity, including ceramics, semiconductors, and polymers, demonstrate a variation in their electrical signal due to the adsorption and desorption of water vapor.

Several compounds have been applied in moisture responsive soft materials. Silk fibers, cotton and wool yarns, carbon

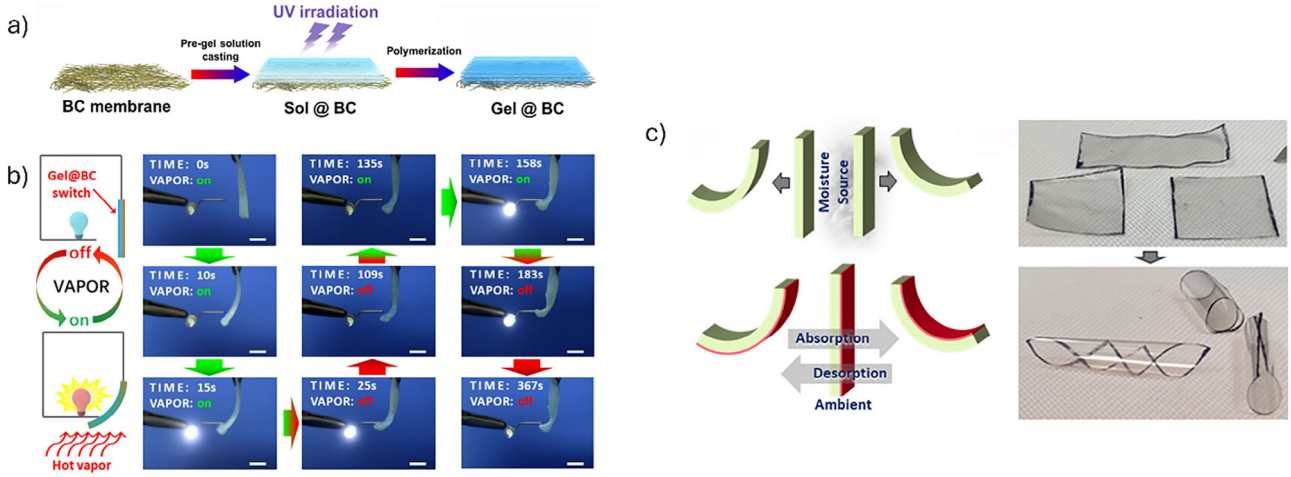

Figure 3. Moisture Responsive cellulose-based actuators: a) Schematic of the fabrication process of Gel@BC bilayer hydrogel, b) illustration of a vapor-triggered electrical switch based on Gel@BC as sensor and experimental images of Gel@BC bilayer hydrogel when used as a smart circuit switch to turn on- and -off LED lamps recurrently (scale bar is 5 mm).[59] c) Actuation of cellophane under moisture gradient,[62] reprinted under the Creative Commons 4.0 License.

nanotubes, and graphene, are examples of successful materials used in these devices.[57] In the case of cellulose, moisture responsive cellulose-based materials have been widely explored. Cellulose contains hydroxyl groups (-OH), that have affinity with water molecules, forming hydrogen bonds and causing expansion and swelling in humidity environments. Native and modified cellulose derivatives can be applied in such responsive displays, and the response in the presence of humidity can be increased with the crosslinking, surface modification, doped-ionic liquid or cellulose in sol-gel form.

Geng et al. produced a liquid crystal based cellulose motor induced bending mechanism.[58] A key characteristic of liquid crystal elastomers and networks is the connection between orientational order and mechanical deformation. Orientational order can be altered by numerous stimuli, such as the existence of moisture, giving rise to stress and strain and resulting of shape changes. The bending of the HPC due to moisture occurs due to the interaction between the orientation order of the rigid rod-like fragments in the network and mechanical stress. The change in the orientational order occurs due to the absorption of water molecules. Thus, it was built a motor that consists of a circular loop of cellulose film, which passes over two wheels. When the moisture is present near one of the wheels on one side of the film, rotation of the wheels occurs. With the wheel rotation, the humid film dries in a dry air atmosphere, returning to its initial form.

Hou et al. developed a fiber-reinforced bilayer hydrogel film, inspired by leaf structure, primarily designated as a rapid steam sensor.[59] The system comprises a non-swelled bacterial cellulose (BC) layer acting as a rigid scaffold, and a water-swellable PNIPAM (poly(n-isopropylacrylamide))/clay hydrogel serving as the active matrix (**Figure 3a**). The resulting Gel@BC bilayer hydrogel, exhibited remarkable mechanical properties, with the BC acting as a continuous reinforcement phase, mimicking the structural function of leaf veins. The integration of the PNIPAM/clay hydrogel into BC network resulted in a tough interface based on multiple hydrogen bonds, with interfacial toughness reaching $\approx 10^{-2}$ J m^{-2}. Notably, although the system was primarily developed for sensing proposes, it also demonstrated rapid and reversible bending behavior upon exposure to steam (Figure 3b), a core criterion to be considered as moisture-responsive material, within the scope of this review.

Moisture responsive sensors actuators, based on cellulose, can also be used as fog water collectors. A fog harvesting method based on the use of advanced fibers obtained through a single-step wet spinning of a mixture of hydrophobic polyvinylidene fluoride (PVDF) and hydrophilic cellulose fibers.[60] Xu *et. al*, used amphiphilic phase-change cellulose nanofibers (CNFs) with enhanced fog water harvesting properties. Polymer chains were selectively attached to the carboxyl groups (-COOH) of CNFs, achieving the balance of hydrophilicity and hydrophobicity, improving the fog capture efficiency and minimizing swelling and liquid film formation, two parameters that decreases the performance of conventional CNF.[61]

A cellulose film-based fast-morphing and motion-programmable soft actuator was developed by Wei and Ghosh.[62] This actuator, based on hygroscopic bending of cellophane (regenerated cellulose) coated and non-coated films, can generate caterpillar-like movement (Figure 3c). The uncoated cellophane film is capable of continuously moving on a moist substrate via self-directed bending–rolling–flipping (or oscillating) cycles. The anisotropic expansion, along with a reasonable moisture absorption and desorption rate, turns cellophane as an excellent option for an actuator activated by increasing of moisture.

1.4.2. Electrically Responsive

Electrically responsive materials can deform when exposed to an electric field, such as dielectric elastomers. Dielectric elastomers are generally composed of one dielectric layer sandwiched between two flexible electrode layers. When a high voltage is applied, the electric field between the two electrodes can generate Maxwell stress, which will squeeze the deformable dielectric

layer, causing a decreasing thickness and the expansion of the area. This mechanism is responsible for the conversion of electrical energy into mechanical energy.[23] The more common designs are based on the integration of conductive elements in the polymeric structure and use the piezoelectric effect or the ion migration effect as an actuating principle. Due to its inherent properties, cellulose can be used to fabricate electro-active actuators where its crystallinity and ion migration are the main influencers or the actuation performance.[52,63]

The work presented by Kim and coworkers[63] showcased both actuating principles. A tri-layered sandwiched structure reported to assemble a bacterial cellulose-based actuator. Cellulose is used as an ecofriendly alternative to most piezoelectric polymers with an enhanced ion migration effect. The middle layer contains freeze-dried bacterial cellulose embedded with an ionic liquid and the two outer layers are composed of a conducting polymer. Unexpected large bending deformation was observed and attributed to a synergy between the interfacial bonding of the bacterial cellulose and the conducting polymers used, without the occurrence of delamination, and the sponge-like porous structure that allowed the absorption of a large quantity of ionic liquid and consequent enhancement of the electrochemical properties.

Yang et al. reported a biocomposite cellulose ionic liquid actuator that presented an enhancement of the electrochemical and electromechanical properties when doped with multi-walled carbon nanotubes (MWCNT).[64] Based on the test results, the performance of the electrolyte layer post-doping treatment has significantly increased when compared to the pure electrolyte layer, and the electrolyte layer with a doping amount of 0.8 wt.%, with the best performance. The presence of MWCNT significantly altered the internal specific area of the electrolyte layer, leading to a rise in the internal charge capacity and specific capacitance. Moreover, the porosity of the cellulose ionic liquid electrolyte layer increased when MWCNT are added allowing to an increase the electron transfer efficiency between the electrolyte and the electrolyte layer contributing to increase the displacement of the actuator. The tensile strength and flexibility were improved. The tensile strength and flexibility were enhanced as well as the effectiveness of the biocompatible ion actuator.

Hygroexpansive Electrothermal Paper Actuators (HEPAs) based on cellulose, were reported by Hamedi et al.[65] These actuators were made from paper, conducting polymer, and adhesive tape. HEPAs can be used in several applications due to their low production cost, low weight, easy printability, and porosity. HEPAs crafted from paper, conducting polymer, and tape can be triggered by the hygroscopic properties of paper, managed electrically by heating. The key component is the conductive pathway (employed for electrothermal heating), created by covering the fibers in the paper (through the thickness of the sheet) with a conductive polymer (PEDOT:PSS), thereby forming a paper/polymer composite that preserves the paper porous and hydrophilic characteristics. When electrothermal heating occurs, the moisture level in the paper is reduced. When the electrothermal heating element is switched off, the paper/polymer composite takes in moisture from the surroundings (moisture content rises), causing it to expand. The authors highlighted four advantages of HEPAs: Lightweight, inexpensive, and biodegradable properties; easy to fabricate by using simple printing techniques; they operate if dented and scratched; they can actuate several times without degradation or performance decreasing.

1.4.3. Thermally and Photo Responsive

Thermally responsive actuators are triggered by changes in temperature to produce movement through specific mechanisms incorporated into their structure of composition. These mechanisms can be designed to take advantage of properties such as thermal expansion and contraction, different thermal conductivities between different materials, and thermal responsiveness. On the other hand, photoresponsive actuators are stimulated by interacting with light, using light sensitive elements or components incorporated into the cellulosic material such as photoactive molecules or polymers. When exposed to specific wavelengths or intensities of light, these additives undergo changes that result in controlled movement or changes in the shape of the actuator.

To an extent, both heat and light are closely related to each other as stimuli since both involve the movement and transfer of energy in the form of electromagnetic radiation, as the two are manifestations of the same phenomenon. As such, cellulose-based actuators using light or heat as stimuli are, often, the using both stimulus for the same effect. The molecular constitution of the thermal responsive materials is changed when these materials are heated, directly by joule heating or indirectly via ultraviolet waves, to a certain temperature resulting in the decline of their elastic modulus or contraction deformation. They can be designed for thermal expansion and contraction to integrate fillers with different thermal conductivities combining with thermoresponsive polymers. Polymeric materials are the most well-known thermally and photo-responsive soft-matter components used in such systems. The so-called smart polymers are polymers that change their properties according to the conditions in their environment. Such conditions mainly include temperature and pressure, providing the required mechanisms for being applied thermally and photo-responsively. Cellulose in nanocrystalline form (CNC) can have properties like those of these smart polymeric materials. The system's mechanical and optical properties bear stimuli-responsive surface modifications.

Zang et al. have investigated the effect of cellulose nanocrystals (CNC) on the mechanical and thermal properties of waterborne polyurethane (WPU), demonstrating that CNC incorporation enhances both characteristics (**Figure 4a**).[66] They proposed a simple filtration method to prepare flexible strain sensors, by combining CNC-reinforced WPU with carbon nanotubes (CNT). The sensor performance was found to depend on the CNT loading, with the CNT0.50-CNCs/WPU formulation (0.50 g of CNT per 1 mL of CNCs/WPU suspension) showing a balanced profile of thermal stability, tensile strength, and electrical signal responsiveness. Compared to CNT-only/WPU systems, the inclusion of CNCs improved CNTs dispersion by reducing nanotube bundling and enhancing the polymer matrix stiffness. Among various formulations CNCs/WPU with 1.00 wt.% CNC content exhibited the highest thermal stability and tensile strength, indicating an optimal reinforcement effect. These results suggest that CNCs not only serve as reinforcement agents but also contribute to improving the dispersion and interfacial compatibility

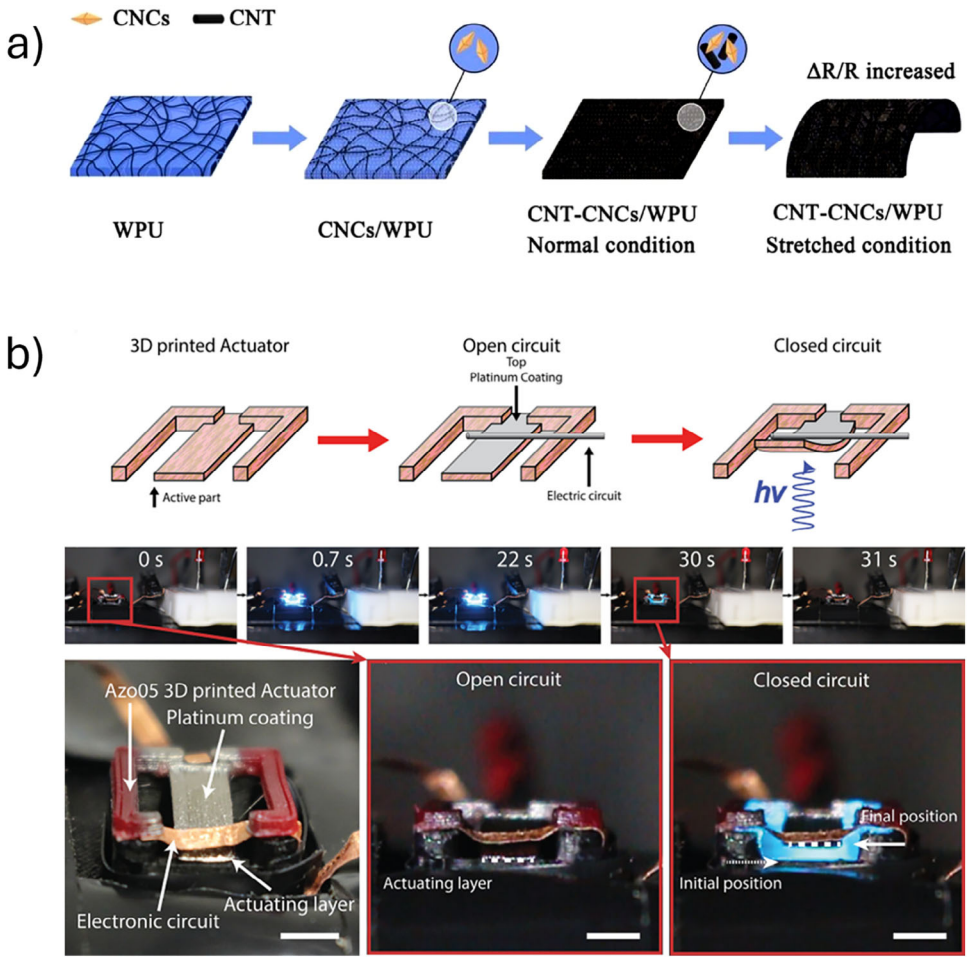

Figure 4. Thermally and photo-responsive cellulose-based flexible devices: a) illustration for the normal and stretched strain sensor produced using a waterborne polyurethane (WPU) enhanced by cellulose nanocrystals (CNC) and combined with carbon nanotubes (CNT).[66] b) 3D printed actuator assembly in an activation circuit: when illuminated it starts moving and after 22 s the red LED switches on, if the light is switched off the actuator comes back to its original position, and the circuit is open again. Scale bars correspond to 3 mm.[70]

of CNTs in WPU matrices, leading to enhanced sensor performance relative to non-CNC containing systems.

A bilayer TEMPO-oxidized nanofibers hydrogel with thermos-responsive, shape memory, and self-sensing assembled into a flexible sensor able to achieve real-time sensing signals driven by temperature changes was reported by Ma et al.[67] The nanofibers contributed as reinforcement to the mechanical strength and endowed the hydrogel actuator with strong interfacial bonding. Recently, a novel hydrogel actuator where TEMPO-oxidized cellulose nanofibers are embedded into a delignified wood scaffold. By combining the delignified wood scaffold with the possibility to tune the orientation of the cellulose nanofibers the authors achieved a strong interfacial bonding between the components. By controlling the orientation and ratio of isotropic/anisotropic layers, specific bending or curling motions were achieved under thermal stimulation.[68]

Wang et al. developed a smart hydrogel with programmable and reversible adhesion behavior under thermal stimuli by introducing a thermally responsive hydroxypropyl cellulose (HPC) polymer unit into the hydrogel network.[69] The wet hydrogel's surface mechanical characteristics, and optical transmission also respond to temperature stimuli, allowing them to be programmed and altered with temperature fluctuations. Utilizing its programmable and switchable adhesion capability, the hydrogel can act as a smart gripper for the precise capture and release of items both in air and underwater. Programmable control over dry and wet adhesion strength was also achieved. This work has improved the prospectives about the design of programmable adhesives, responsive materials, and smart devices.

Photoresponsive actuators are usually built by adding light-sensitive fillers to polymers. When exposed to light the photoresponsive actuators can reversibly deform by bending, contacting, or expanding. Changes in the wavelength, intensity, and irradiation time of light allow to program a specific response and achieve the desired motion. Photochemical reactions can also be used to provide energy to be converted to mechanical energy. Photoresponsive actuators are stimulated by interacting with light, using light sensitive elements or components incorporated into the cellulosic material such as photoactive molecules or polymers. When exposed to specific wavelengths or intensities of light, these additives undergo changes that result in controlled movement or changes in the shape of the actuator.

Muller at al., reported a photoresponsive movement in 3D printed cellulose nanocomposites, introducing biocompatible and sustainable cellulose nanocrystals (CNC) into a soft liquid crystal elastomer (LCE), to facilitate the printing process by direct ink writing (DIW) (Figure 4b).[70] Through this technique, it is possible to took benefit from the anisotropic mechanical properties that arise from the alignment of these nanoparticles induced by extrusion, allowing for the direct printing of structures with customized geometries and mechanical characteristics. A doping process, that incorporates azobenzene photo-switches into the composite, grants photomechanical properties to the printed materials.

Zhou et al. studied the effect of modified cellulose nano crystals (MCNC) dosage on emulsion fluorinated polyacrylate, as a fabric finishing agent, and latex film properties, as self-healing materials for injuries.[71] The introduction of MCNC increases the mechanical property and thermal stability of latex films. For dosages over 0.4 wt.%, the latex film had good self-healing properties. Morphological analysis, by SEM-EDS analysis, revealed that the emulsion modified with photo-responsive cellulose nanocrystals was evenly applied to the fabric surface. When the dosage of MCNC was 1.0 wt.%, the surface damage of the finished fabric can be repaired, providing a new approach to obtaining stable and durable fabrics. Recently, a single-layered conductive gradient hydrogel that integrates TEMPO-oxidized cellulose nanofibers combines rapid programable motion with self-sensing capability was used to construct soft-grippers, artificial irises, and even a self-sensing tongue mimic.[72] The gradient structure is responsible for the fast deformation when exposed to thermal or near-infrared stimuli, presenting a bending speed of ≈21° s^{-1} at ≈50 °C. Its structure is also responsible for a conductive gradient enabling real-time monitoring via resistance changes.

1.4.4. Magnetically Responsive

Magnetically responsive materials are frequently composites that join magnetic particles with soft materials such as polymers. The magnetic field can be used to magnetize the particles generating regular magnetization curves, with the possibility to vary the field direction and amplitude. The magnetized particles can turn and align with the magnetic field, and the torque produced can shrink, elongate, or bend the entire material. By incorporating magnetic elements or materials within cellulose-based structures, it is possible to produce magnetically responsive actuators. The integration of magnetic particles or materials with the cellulosic matrix allows the induction of controlled motion in response to the changes in the magnetic field present. This can be done by adopting strategies such as aligning magnetic particles and taking advantage of the attraction or repulsion of magnetic poles and magnetostrictive effects.

Magnetic-responsive nanoparticles impregnated in cellulose are the most promising magnetically responsive actuators. The efforts to explore and enhance the magnetic properties of these systems have garnered significant attention across chemistry and physics research fields.[73,74] Ferrites, particularly Fe_2O_3 and Fe_3O_4, are the most well-known nanoparticles that impart unique characteristics to these devices due to their supermagnetic behavior, stability, and magnetic responsiveness.[75,76] These nanoparticles can be synthetized through wet-chemical routes such as co-precipitation, sol-gel, or hydrothermal methods, promoting the viability and sustainability of their production process.[77,78] Beyond the inherent biocompatibility, biodegradability, and sustainability of cellulose, the integration of magnetic nanoparticles adds considerable versatility. This makes them suitable for a wide range of applications, including drug delivery systems,[79] magnetic hyperthermia for cancer treatment,[80] and biomimetic soft robotics.[81]

Ferrofluid-Impregnated Paper Actuators were reported by Ding et al.[82] They present a cost-effective technique for creating small magnetic actuators by using paper infused with a light mineral-oil-based ferrofluid. Different types of papers (soft tissue paper, cleanroom paper, Whatman-1 filter paper, printer paper, and newspaper) were infused with oil-based ferrofluid, micromachined using a CO_2 laser, and coated with a parylene-C thin layer. The absorption ability of various papers was examined, with the soft tissue paper showing the greatest capacity, able to take in ferrofluid up to six times its initial weight.

Magnetic-responsive iron oxide (Fe_3O_4) nanoparticle-impregnated cellulose paper actuators were develop by Wang et al.[73] Chromatograph paper infiltrated with Fe_3O_4 nanoparticles was created by using an affordable blending technique, a low cost. After defining the fundamental material characteristics of the Fe_3O_4/paper nanocomposite, beam-, accordion-, and star-shaped actuators were created from the nanocomposite, and their actuation performance is examined through experiments. It was reported that when exposed to an external magnetic field, the paper-based actuators exhibit deformation, but reversibility and stability. The low magnetic hysteresis loop of the Fe_3O_4/paper nanocomposite allows stable and reversible performance in flexural actuation of the beam-like actuator. Tomás et al.[83] developed hybrids of cellulose nanocrystals decorated with magnetic nanoparticles, to both enhance and provide magnetic responsiveness to tendon-like hierarchical fibrous scaffolds, by creating a system that allows the remote stimulation of in vitro cells. With this work, it has been shown that the inherent physical signals of the tendon-like scaffolds can direct human adipose stem cells (hASCs) mechano-transduction processes, enhancing their tenogenic differentiation and positively influencing the expression patterns of anti-inflammatory/pro-healing and matrix remodeling gene markers.

1.4.5. Combined Stimuli

Combined stimuli actuators react to several external stimuli that can include temperature, pH, moisture, light, electric fields, and magnetic fields are key to developing adaptive materials for soft robotics, biomedical applications, and smart textiles. These actuators can exhibit more complex and programmable deformations, enhanced controllability, and increased versatility when multiple stimuli are combined. The designing of cellulose-based actuators to respond to multiple stimuli or combinations of stimuli is a growing area of interest in material science, by integrating various responsive mechanisms. This strategy allows intricate and precisely controlled movements.

Yang et al. have developed dual-response actuators with Janus structure films, by combining cellulose 3,5-di-tert-butyl-4

hydroxybenzoate (CBH) and water-swellable carboxymethyl cellulose (CMC), prepared by the solution casting method.[84] The obtained dual responsive film predicts the concentration of ethanol in aqueous solutions through the different deformation angles formed. The film is also inspired by nature, since it can mimic the petals of morning glory flowers, acquiring formation values with ethanol proportion variation.

Li et al.[85] developed a multi-stimuli responsive actuator with color- and morphing-change abilities made of cellulose nanocrystals-based cholesteric liquid crystal structure and its response behaviors to water and temperature. This structure affords high water absorption and expansion capacity. The material was inspired by alternate chemochrome present in some animal skins and butterfly wings. This study provides a method to simultaneously achieve multiple stimulus responses and mimic the vivid structural color showcasing a novel strategy to visualize the movement of flexible CNC-based actuators, with an emphasis on applications on flexible robotics, anti-counterfeiting, and information storage areas. Chen et al.[86] designed a dual-responsive, which can be driven by light or humidity, with programmable shape deformations. The bending rates and amplitudes of such deformations can be accurately controlled and programmed by adjusting the grayscale of patterns with computational assistance.

Responsive hydrogel cellulose-based actuators have also been developed in the last few years. Mo et al.[87] developed a rapid thermo-responsive hydrogel actuator by inducing a gradient distribution of renewable tunicate cellulose nanocrystals (TCNCs) in the poly(N-isopropyl acrylamide) (PNIPAM) matrix, induced by a DC electric field. The study offered a novel approach for creating hydrogel actuators that respond quickly by utilizing nanomaterials from renewable biomass nature. Park et al.[88] report a strategy that realizes a programmable thermo- and light-responsive hydrogel actuator with enhanced mechanical properties by exploiting the structural advantages of bacterial cellulose. They developed a hydrogel actuator system driven by temperature and light, featuring improved mechanical properties for accurate control of the response areas. Thus, it utilized PNIPAM as a temperature-sensitive polymer framework, gold nanoparticles (AuNPs) as a light-heat mediator converter, and nanofibrillar bacterial cellulose (BC) as a substrate for the nanoparticles and an interwoven polymer for the hydrogel. In the PNIPAM/BC hydrogel, BC plays a crucial role in forming physical cross-links via hydrogen bonding and entanglement with PNIPAM chains, which results in enhanced mechanical properties and quicker responses to temperature changes.

2. Applications

The unique properties and ability to respond to a wide range of stimuli grant cellulose-based actuators the potential for an extensive assortment of diverse applications. As stated before, cellulose and its derivatives come as more renewable, eco-friendly alternatives to the usual materials used in sensor technology. Even if they donot match the efficiency of their more conventional counterparts, cellulose-based sensors are not only a reliable alternative but also present a wide range of types, including magnetic, capacitive, resistive, colorimetric, and optimal waveguide sensors. In this section, examples of the applications of different types of cellulose-based smart materials will be presented, as presented in **Figure 5**.

Lu et al.[94] developed a carboxymethyl cellulose (CMC) based, skin-inspired hydrogel-elastomer hybrid for mechanical-thermal multimode sensing with high biocompatibility, transmittance, and fatigue resistance. This sensor consisted of an ionic transport conductor material, lithium chloride (LiCl), which provided good mechanical, electrical, and sensing properties through a wide range of temperatures, a polymeric matrix, polyacrylamide (PAM), and an encapsulating outer-layer elastomer, polydimethylsiloxane (PDMS), for increased mechanical properties, waterproof properties, and long-term water retention. The CMC was used as a biocompatible nanofiller forming hydrogen bonds with PAM, increasing the toughness and fatigue resistance of the sensors, by transferring the load from the polymer chains to the nanocellulose. Furthermore, both the hydrogel and elastomer went through silane modification to increase the interface adhesion between both. Indeed, the authors developed a multimode mechanical-thermal sensor, with excellent temperature tolerance and waterproof properties, with the potential for underwater applications.

Han et al.[95] reported a polymer ionic-electronic conductor (MIEC) based multiparameter sensor device, with nanofibrillated cellulose (NFC) as a structural material, capable of independently measuring pressure, temperature, and humidity. For this, the authors freeze-dried a water dispersion with poly(3,4-ethylenedioxythiophene) (PEDOT), as the electrically conducting material that provides electronic thermovoltage, with polystyrene sulfonate (PSS) as the ionic conducting polymer that provides the ionic thermovoltage peak, with NFC, which form the mechanical structure the gels and increases the mechanical strength and with glycidoxypropyl trimethoxysilane (GOPS) as the crosslinker, which induces elasticity of the aerogel. The independent measurements of pressure, temperature, and humidity are assured by using a transduction protocol, based on mechanosensitive effect, electronic Seedbeck, and ionic Seebeck effects, that convert the device characteristics into a current-voltage-time coordinate system, from which it is possible to take all the necessary information.

2.1. Wearable Devices

There has been a shift from more conventional electronic devices to more high-performance devices, such as flexible electronics. Flexible electronics overcome the physical rigidity of conventional electronics, which are incompatible with applications, such as wearable devices, that require compatibility with nonplanar surfaces.[96]

However, the demand for flexible electronics requires the study and development of new materials that meet the requirements, such as high flexibility, biocompatibility, and low environmental impact. While materials such as polyimide, silicone rubber, and PDMS, exhibit the required mechanical properties, they lack the necessary biocompatibility. In this sense, flexible biomaterials, such as chitosan, pectin, and spider silk are promising materials for flexible electronics, presenting both the required mechanical properties and biocompatibility.

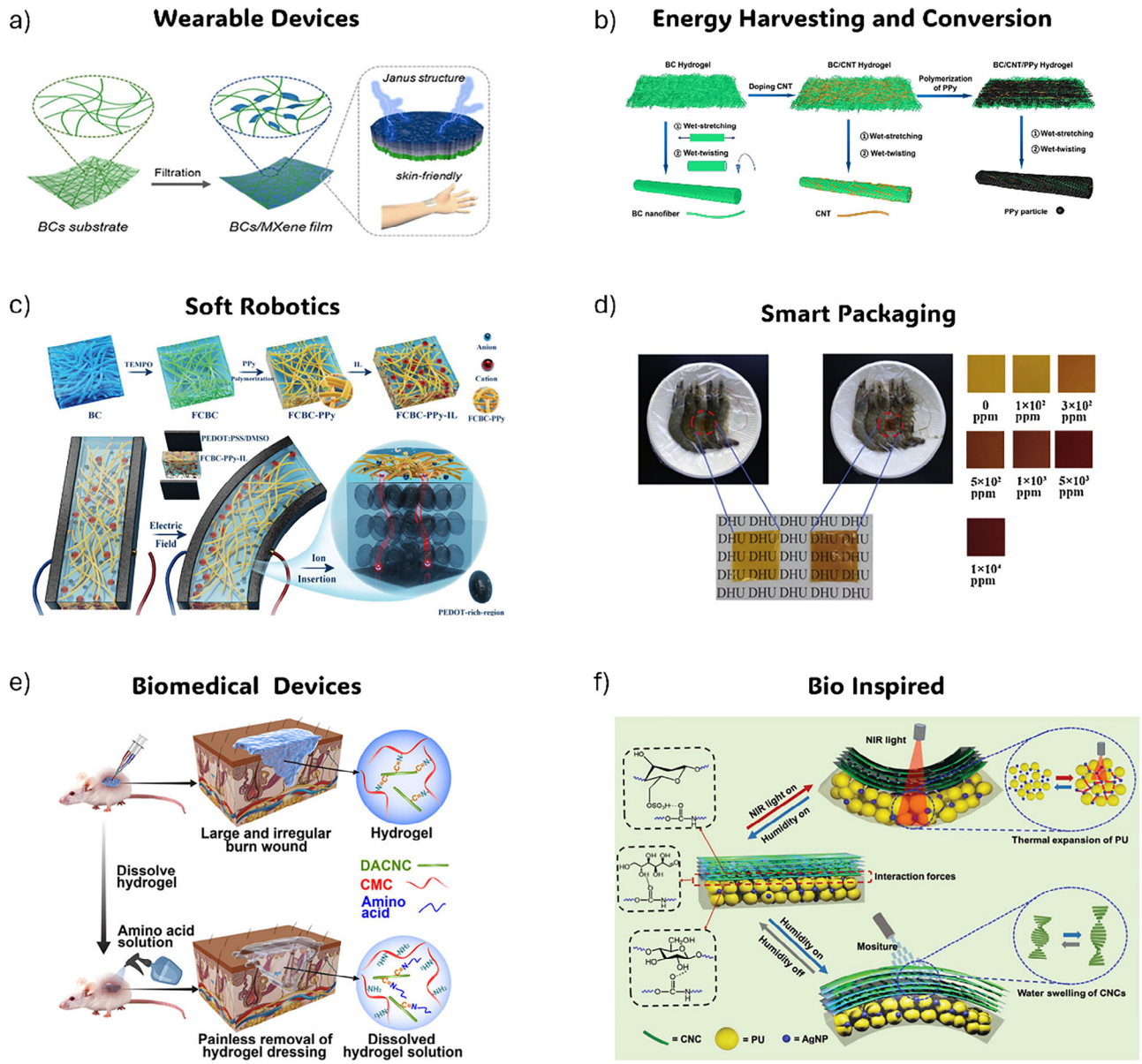

Figure 5. Cellulose-based smart materials: a) Schematic representation of a wearable device produced using bacterial cellulose,[89] b) Schematic illustration of the fibers used to develop a wearable fabric-based triboelectric nanogenerator energy harvesting and conversion,[90] c) schematic diagram of the production of the cellulose-based nanocomposite and actuation mechanism of the soft ionic robot,[91] d) Images of the cellulose-based film shrimp before and after shrimp spoilage,[92] e) schematic of the carboxymethyl chitosan (CMCS)/aldehyde-modified cellulose nanocrystal (DACNC) hydrogel self-healing properties usage,[93] and f) bio-inspired layer composite film based on cellulose nanocrystals development and its light- and humidity-responsive actuation.[85]

Cellulose is not only one of the planet's most abundant low-cost and renewable materials but also the most abundant natural polymer. Cellulose is a flexible material, meeting the requirements for flexible electronics. Furthermore, it can be transformed into conductive carbon materials via carbonization.[32] In this section, different examples of the application of cellulose in wearable devices will be presented.

Jing et al.[97] developed a polyvinyl alcohol (PVA)/cellulose nanofibril (CNF) dual crosslinked hydrogel, used as a capacitive ionic skin sensor for the detection of strain and pressure. The pressure sensors were produced by placing a dielectric layer between two hydrogel layers that acted as the conductive layers connected to two metallic electrodes, while for the capacitive strain sensors, a stretchable dielectric layer was used. The authors could produce hydrogels with properties such as high transparency, high stretchability, biocompatibility, and spontaneous self-healing ability, and their system achieved high sensitivity, to the point of detecting the pressure of the human breath. By introducing a secondary CNF network, not only the mechanical strength and viscoelasticity were improved, but also a high sensitivity of the capacitive sensor was achieved.

Ma et al.[89] developed Janus bacterial cellulose (BC)/$Ti_3C_2T_x$ Mxene film, through facile vacuum filtration, for skin-contactable wearable devices (Figure 5a). Mxenes are a family of 2D transition metal carbides/nitrides with large aspect ratios and metal conductivity with potential for wearable devices. The authors used the BC as the skeleton of the structure, trapping Mxene nanosheets. The composite showed properties, such as high flexibility, high transparency, high folding endurance, high air permeability, and high electromagnetic signal shielding effectiveness, required for skin-touchable smart electronics devices. Not only did the BC contribute to the mechanical properties of the films, but it was also used as structural material. Nanocellulose-based flexible electroluminescent (EL) devices, where a dielectric luminescent layer is sandwiched between two silver nanowires-cellulose nanocrystals with II crystalline allomorphs (CNC II) based electrodes, have been pointed out as favorable option for future use in the next generation of flexible green electronics.[98] Lu et al. reported the use of biomass-derived CNC II with high transmittance to be used as film-forming agent for transparent electrodes.[99] These electrodes when included in a sandwich-structured EL devices presented high stretchability, long-term stability, damage resistance, biocompatibility and could work normally under water and over a wide temperature range (≈20–70 °C).[100]

2.2. Energy Harvesting and Conversion

With the growth in wearable technology, new challenges arise to meet all the requirements, typically associated with standard rigid technology, such as the availability of energy. Typically, electronics run on rechargeable batteries, and, considering this factor, even when transitioning from rigid to flexible devices, this technology is limited by the rigidity and short lifetime of the batteries. In this sense, energy harvesting presents an ideal alternative to solve this issue, by using different sources, such as body heat, sunlight, or even by taking advantage of piezoelectricity, to generate energy.[101]

There are different considerations to consider when developing an energy harvesting device. For example, when using piezoelectric devices, although lead-based ceramics, such as lead zirconate titanate (PZT) present high piezoelectric coefficients, they are limited by their rigidity, brittleness, and toxicity. In this sense, polymers, such as cellulose and its derivates come as great piezopolymer alternatives, when looking for flexible, low-cost, and biocompatible devices.[102]

Pusty et al.[103] fabricated a piezoelectric nanogenerator (PNG) based on a cellulose/PDMS nanocomposite incorporated with silver nanoparticles (Au NPs). By producing a composite of cellulose and PDMS, the dielectric constant was increased when compared to only PDSM, and the improvement of the dielectric properties led to the enhancement of the piezoelectricity. The Au NPs were incorporated to increase the piezoelectric output voltage and enhance the current of the device. The authors were able to reach open circuit voltages of ≈6 V when using a periodic force of 3 N, and a short circuit current of ≈700 n, and the device had the capability of charging a 10 µF capacitor in 677 s.

Hu et al.[90] developed biodegradable, cellulose-based, conductive macro fibers, and designed a wearable fabric-based triboelectric nanogenerator (TENG) for energy harvesting and biomechanical motion monitoring, driven by Maxwell's displacement current (Figure 5b). The fibers were produced by wet-stretching and wet-twisting BC hydrogels incorporated with carbon nanotubes (CNTs) and polypyrrole (PPy). BC was used as a biodegradable and biocompatible structural material, conferring high tensile strength to the final device, reaching 449 MPa. Indeed, the authors created a device with an electrical conductivity that reached a value of 5.32 $S.cm^{-1}$, and with the capacity of powering commercial electronics, showing a maximum open-circuit voltage of 170 V, a short-circuit current of 0.8 µA, and output power at 352 µW, when used as an electrode.

2.3. Soft Robotics

Robotics allow human- or anime-like functions to be mimicked and performed by machines, with improved strength, speed, and accuracy. Furthermore, inside the field of robotics, two subfields can be found, the first of which is hard robotics. Hard robotics is based on the use of rigid materials and involves the use of electric actuators or pressurized fluids and power supplies. Despite the success of this field, it presents certain limitations, such as the lack of collaboration potential in most cases, due to the dangers associated with the large forces and rapid movements of hard robots and the use of non-compliant materials, which leads to a lack in adaptability to different tasks. In this sense, the field of soft robotics comes as a way to overcome some of the challenges associated with hard robotics.[104]

Soft robotics consists of a safer, lighter, and simpler option, which involves the use of softer materials, such as elastomeric materials, and can be adapted to different tasks, due to the ability to conform autonomously to different shapes. To develop soft robotics, varied materials can be employed, such as polyvinylidene fluoride (PVDF) and nafion, which are commonly used as ionic electrolyte membranes and can be used to develop ionic electroactive polymer (IEAP) actuators, which are an example of typically used artificial muscles. However, actuators based on PVDF and nafion present certain drawbacks, such as low bending strains, high cost, and lack of biocompatibility and biodegradability. In this sense, materials such as cellulose can come as an environmentally friendly, and biocompatible alternative.[91,104]

Wang et al.[91] developed ultralow voltage high-performance bioartificial muscles based on functional carboxylated bacterial cellulose (FCBC) and polypyrrole (PPy) (Figure 5c). FCBC was obtained by performing 2,2,6,6-tetramethylpiperidinel-oxyl radical (TEMPO) induced oxidation on BC, which allows the carboxylate groups to be selectively oxidized, obtaining a homogeneous dispersion of individual fibers. Following this, by taking advantage of the functional groups, the authors performed chemical oxidative polymerization of pyrrole (Py) on the surfaces of the FCBC nanofibers, resulting in a highly porous and conductive FCBC/PPy-based nanostructure. As an electroactive conducting polymer (ECP) PPy was used for its high conductivity, biocompatibility, biodegradability, high theoretical capacity, good energy density, inherent redox switching, and facile synthesis. Finally, the fast-drying casting of FCBC-PPy with an ionic liquid (IL) 1-Ethyl-3-methylimidazolium tetrafluoroborate, [EMIM][BF4] was performed to produce FCBC-PPy-IL nanobiocomposite

membrane. The actuator was obtained by depositing, on both sides of the FCBC-PPy-IL membrane, a flexible and conductive polymer, poly(3,4-ethylenedioxythiopene)-polystyrenesulfonate (PEDOT:PSS), treated with dimethyl sulfoxide (DMSO), which improves the ion migration and charge transport within electrodes on both sides. The designed FCBC-PPy-IL membrane showed a large ion-accessible surface area, excellent electron donor-acceptor interactions, high charge storage ability, and easy-fast ion movement within the membrane matrix. Furthermore, the bioartificial muscles presented astonishing actuation performances of the bioartificial muscle, which include a large bending strain, broad frequency bandwidth, long actuation durability under ultralow harmonic input of 0.5 V, and excellent biocompatibility.

Nasseri et al.[105] developed pH-responsive hydrogels for soft robotics with pre-determined microstructural anisotropy, shape-transformation, and self-healing, based on zwitterionic monomers and asymmetric cellulose nanocrystals. The hydrogels were obtained by copolymerization of 3-dimethyl (methacryloyloxyethyl) ammonium propanesulfonate (DMAPS) and methacrylic acid (MAA) in the presence of CNC nanoparticles. Not only were the zwitterionic monomers used to achieve self-healing properties and cytocompatibility, but also P(DMAPS-MAA) copolymers are known to be pH-sensitive, conferring the hydrogels their pH-sensitive behavior. The cellulose nanocrystals confer the hydrogels with swelling and mechanical properties, due to the sheer-induced alignment of the nanocrystals, enabling the programming of stimuli-responsive shape transformation. Furthermore, the self-healing properties allow the use of a cut-and-paste strategy, which combined with the structural anisotropy conferred by the cellulose nanocrystals, allows the programming of stimuli-responsive shape transformation, allowing the obtention of complex stimuli-triggered shape-morphing systems. Indeed, based on the system developed by the authors, it was possible to transport light cargo, using tethered and untethered soft grippers.

2.4. Smart Packaging

While conventional food packages simply have the passive role of creating a barrier that delays adverse effects of the environment on a certain product, with the key requirement of the used materials being inertness, modern food packages present an active role, interacting with the environment on a way that contributes to the protection of the product. Furthermore, modern packages can be separated into active packages, which serve the purpose of prolonging the shelf-life of a product by interacting with the environment, and intelligent packages, which serve the purpose of sensing and sharing information regarding the condition of the product. However, both types of packaging are not mutually exclusive and can be used together to develop smart packages.[106]

There are many ways in which active packages can improve the shelf-life of a product. They can present different types of properties, such as antimicrobial, oxygen scavenging, odor absorption, and moisture control properties. For example, pediocin or nisin can be fixed on cellulose casings and used to inhibit the growth of Listeria monocytogenes (which can be present in ham, turkey meat, and beef). Furthermore, intelligent packages can give different types of information, for instance, they can act as biosensors, gas sensors, indicators of freshness, and temperature control units, among others. By way of illustration, methyl red/cellulose membranes can be used as indicators of the release of volatile amines (released during fish spoilage) through changes in their color.[107]

Bandyopadhyay et al.[108] developed a polyvinylpyrrolidone (PVP)/CMC/BC film, as a bioadhesive for food packaging, and compared the properties with PVP/BC and neat BC films. BC is a material with different applications when it comes to food packaging technology, ranging from being used in edible food packaging, to being used in antimicrobial food packaging. The produced PVP/CMC/BC films showed the highest values of bioadhesiveness, highest elongation at break, and tensile strength, making these films appropriate for wrapping or packaging tape, where a better seal is necessary. Furthermore, the addition of the CMC to the films conferred some resistance to the transmission of light, since it made the films darker, which can reduce the photodegradation of the products by increasing their shelf-life.

Yordshahi et al.[109] developed BC films impregnated with *Lactobacillus plantarum* to be applied as an antimicrobial technology against *Listeria monocytogenes* for wrapping ground meat. BC was chosen since it exhibits good mechanical properties, as well as high porosity and water absorption capabilities, allowing it to be used as a carrier. Not only is the BC film appropriate for the carrying of *L. plantarum*, but it also allows a quick release, upon wrapping of the meat, due to the hydration of the film, which is advantageous when dealing with a product with a finite shelf-life. The technology developed by the authors allowed the doubling of the overall shelf-life.

Ding et al.[92] developed a pH-sensitive device, based on regenerated cellulose (RC) modified with an acidochromic die, incorporated into PVA, for real-time monitoring of food spoilage (Figure 5d). This device was applied to detect shrimp spoilage, by detecting the changes in pH caused by the production of ammonia. Not only did the devices show great sensitivity to pH, being capable of detecting ammonia at 100 ppm or above, but they also exhibited great reversibility. The tensile strength and elongation at break of the films were greatly improved with the addition of the modified RC. Furthermore, by using RC and modifying it with the acidochromic die it was possible to overcome the leakage problems that usually affect sensors produced by directly blending the pH-responsive product with a matrix.

2.5. Biomedical Devices

With the development of technology throughout the years, different materials have been studied for biomedical applications, such as gelatine, chitosan, and silk fibroin, due to their bio-renewable properties. Cellulose, being the most abundant natural polymer, is of great interest for biomedical applications. Furthermore, due to characteristics such as mechanical and rheological properties, biocompatibility, antibacterial properties, barrier properties, and ease of transformation into various derivates, cellulose presents great potential for different applications, for instance, tissue engineering, drug delivery, wound healing, and controlled release.[110]

Farzamfar et al.[111] synthesized cellulose acetate/gelatin-based 3D porous scaffolds loaded with gabapentin (GBP), an anticonvulsant drug, for neural tissue regeneration. The scaffolds were produced by wet-electrospinning, which was performed into GBP-containing baths. GBP was used since it has already shown potential for nerve regeneration applications. Gelatin was used due to its biocompatibility, biodegradability, and low cost. However, gelatin presents poor mechanical properties, so, in this sense, cellulose acetate was used, since it presents good mechanical properties, and degradability, and is compatible with wet electrospinning. Cell culture studies were performed and the scaffolds showed compatibility with primary rat Schwann cells (SCs). Furthermore, the scaffolds improved tissue regeneration when tested in adult male Wistar rats, on which a segment of the sciatic nerve was removed.

Huang et al.[93] produced a carboxymethyl chitosan (CMCS)/aldehyde-modified cellulose nanocrystal (DACNC) hydrogel, with self-healing properties, for deep partial thickness burn wound healing (Figure 5e). CMCS is a material that presents high solubility under physiological conditions, unlike chitosan. Furthermore, it is a biocompatible material with good water retention capabilities. Due to its high mechanical strength and stiffness, cellulose nanocrystal (CNC) was used as a reinforcement biocompatible material. Not only that but it was also used since it could easily be modified into DACNC, acting as a crosslinking agent. The obtained hydrogels presented good mechanical and self-healing properties, on-demand dissolving capabilities by using an amino acid solution, and the potential to support cell growth. Finally, in vivo tests showed that hydrogels could treat deep partial-thickness burn wounds. Indeed, the authors were able to produce a cellulose-based self-healing injectable hydrogel, with the capability to keep a moist environment, absorb wound exudate, and heal wounds.

Shahzadi et al.[112] developed hydrogels from CNC grafted with polyacrylic acid (CNC-g-PAA) and doped with magnesium oxide (MgO) for bactericidal applications and the controlled release of doxorubicin (DOX) a standard drug used in cancer therapy. The grafting with PAA increases the hydrogels' absorption capacity, while also conferring them pH-sensitive properties. Doping with MgO increases the mechanical properties of the hydrogels and confers antibacterial, antioxidant, and anticancer properties. The MgO/CNC-g-PAA hydrogels showed bactericidal efficiency against gram-positive bacteria and were able to entrap DOX and allow a stable and controlled release at an acidic pH of 5.8. Furthermore, the authors observed that the effect of DOX and MgO/CNC-g-PAA might trigger apoptosis in breast cancer cells, showing the potential of these hydrogels for targeted and controlled drug release for cancer treatment.

2.6. Bio Inspired

For many years, researchers have drawn inspiration from the structures and hierarchical organization found in nature to create new materials and devices with properties such as color-changing abilities, high mechanical strength, and self-healing capabilities. In this context, two terms are often used, which are biomimicry, and bioinspiration. Biomimicry refers to the field of research focused on recreating the structures and functions of complex biological. Bioinspiration, on the other hand, involves developing innovative materials, structures, and devices based on insights from biological systems, regarding their properties, structures, and functions.[113] Indeed, by taking inspiration from nature's evolutionary process that led to the development of multi-layer hierarchically structured multifunctional materials and their properties, it is possible to develop new materials with improved properties.[114]

Inspired by the environmentally sensitive properties of animal skin and the dynamic structural coloration of butterfly wings', Li et al.[85] developed multi-stimuli responsive bilayer CNC-based actuators, exhibiting both color-changing and light/humidity-responsive shape-morphing behavior (Figure 5f). These actuators were fabricated by assembling a CNC cholesteric film onto a polyurethane (PU) elastomer substrate. The addition of glucose during the casting process served as a thickener, facilitating the formation of a uniform cholesteric liquid crystalline structure, that provided stable optical properties and long-term actuation performance. Furthermore, to achieve excellent photothermal conversion efficiency and thermal conductivity, silver nanoparticles (AuNPs) were dispersed into the PU matrix. Indeed, by taking advantage of the high-water absorption, expansion capacity, extremely low coefficient of thermal expansion, and linear birefringence of the CNC-based cholesteric liquid crystal structure, the authors could develop a multi-stimuli responsive actuator, with an excellent variable chiral optical property and humidity/NIR light bidirectional reversible actuating performance. Thus, beyond structural color modulation, this work demonstrates a synergistic integration of optical and mechanical responses, making it a compelling example of bioinspired actuators.

Taking inspiration from the porous architecture of the *Thalia dealbata* stem Wang et al.[115] developed a biomimetic CNF-based aerogel with excellent mechanical, flame-retardant, and thermal-insulating properties. The anisotropic architecture with lamellae interlinked bridges of the *Thalia dealbata* stem contributes to its lightweight and robustness against strong winds. In this sense, CNF aerogels with this interconnected structure come as a biomimetic alternative to achieve ultralow thermal conductivity and high mechanical stiffness. However, due to the CNF's inherent flammability, the authors incorporated hierarchical alpha-zirconium phosphate (α-ZrP)/reduced graphene oxide (RPO) nanosheets, obtained through a space confinement strategy to grow two-dimension ZrP within multilayer graphene, to increase the flame resistance and thermal insulation of the CNF aerogel. While graphene works as a 2D barrier to restrain heat and mass presence, due to the sp^2-bonded carbon sheets, α-ZrP as a strong solid acid contributes to the catalytic carbonization of the polymer. By using a unidirectional freeze-casting technique to assemble the CNF and the α-ZrP/RPO into a biomimetic aerogel through the usage of lamellar ice crystals as templates, the authors were able to achieve excellent thermal insulation, mechanical stiffness and flame resistance perpendicular to the lamellar alignments.

3. Conclusion, Challenges, and Future Outcomes

Over the last years, remarkable advances have been made in the field of cellulose-based smart materials capable of both sensing

and actuation. Research in this domain has undergone notable growth, driven by the need to use more sustainable, biocompatible, and smart materials. The sophisticated mechanical transformations and adaptive response exhibited by cellulose-based materials make them particularly attractive for a wide range of applications. Cellulose, in its various forms, such as nanocrystals, nanofiber, and its derivatives, has been strategically engineered to serve as a foundation for responsive systems.

These efforts have led to the development of moisture-driven bending and twisting actuators, light- and heat-responsive films exploring anisotropic expansion, magnetically guided cellulose composites, and capacitive sensing platforms. Recent studies have demonstrated the integration of cellulose into wearable devices, where it functionates as a flexible, breathable, and biocompatible component for sensors capable of monitoring physiological signals with high performance. In the field of energy harvesting, the dynamic motion of cellulose-based systems has been extensively explored, establishing a foundation for sustainable, self-powered devices. Additionally, actuators driven from cellulose that mimic natural movements have been widely studied for the development of soft robotic systems. The inherent biocompatibility of cellulose, combined with the possibility to tailor its structural and surface properties, has further allowed its incorporation into smart packaging, particularly as part of sensors.

Despite this impressive progress, several challenges continue to impede the broader development and industrial translation of cellulose-based dynamic systems. Variability in cellulose sources, including differences in molecular weight and crystallinity, can lead to significant challenges in reproducibility and device performance. Mechanical resilience under prolonged or repeated actuation also remains a challenge to tackle, as many cellulose-based actuators and sensors display fatigue or hysteresis under repeated use. Furthermore, while individual responsiveness has been convincingly demonstrated, there remains a pressing need to develop robust cellulose-based platforms that can integrate multi-responsiveness systems.

The last few years have firmly established cellulose as a versatile, ecological sound material capable of underpinning soft responsive systems, which embody motion and responsiveness across multiple length scales. Looking forward, bridging cellulose chemistry, materials science, and advanced design strategies will be essential to overcome these existing challenges and unlock new paradigms in soft sensing and actuation. Such interdisciplinary efforts promise to transform cellulose from its traditional role as a structural biopolymer into a central enabler of intelligent, sustainable technologies poised to address the needs of the future.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

actuator, cellulose, sensor, smart material, stimuli

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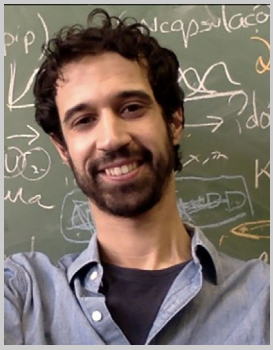

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