Polymer-based actuators: *Back to the future*

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Polymer-based actuators play a key role in the area of smart materials and devices, and for such reason different polymer-based actuators have appeared in recent years being implemented in a broad-range of fields, including biomedical, optical or electronics, among others. Although being possible to find more types, they are mainly classified into two main groups according to their different working principles: electromechanical -with electrical to mechanical energy conversion- and magnetomechanical -with magnetic to mechanical energy conversion. The present work provides a comprehensive and critical review on the recent studies in this field. The operating principles, some representative designs, performance analyses and practical applications will be presented. The future development perspectives of this interesting field will be also discussed. Thus, the present work provides a comprehensive understanding of the effects reported in the past, introduces solutions to the present limitations and, *back to the future*, serves as a useful guidance for the design of new polymer-based actuators aiming to improve their output performances.

1. Introduction

Polymer actuators are a class of smart materials capable to modify their shape, as a response to changes in environmental conditions, such as temperature or pH, or applied signals, including electrical (Figure 1a) or magnetic inputs (Figure 1b)[1, 2]. In this way, those materials are capable of transduce specific energy variations into motion [1]. Due to this ability, actuators find a wide range of applications, including packing, food processing, factory automation, materials handling, microfabrication, microelectronics, robotics, lab-on-a-chip systems or machine tools, among others [1, 3, 4].

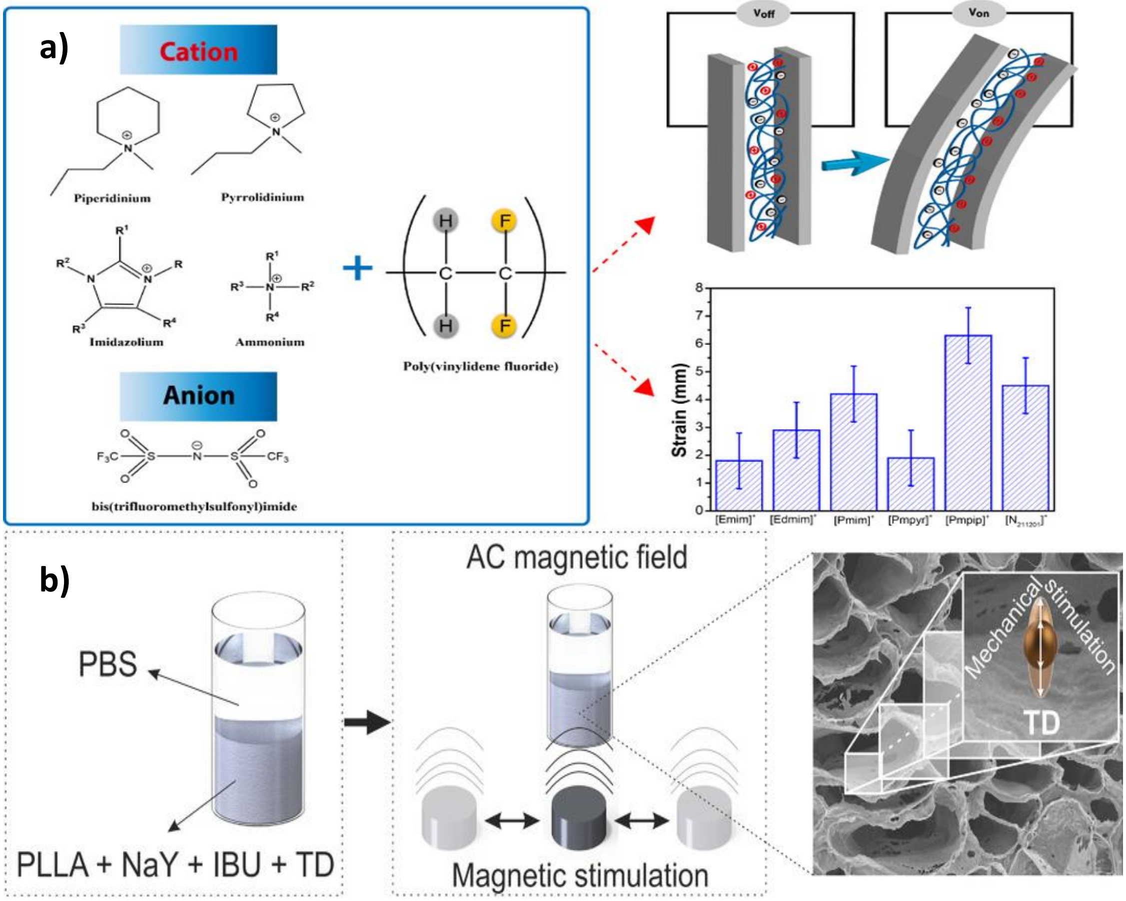

Figure 1. a) Electromechanical ionic liquid (IL)/poly(vinylidene fluoride) (PVDF) actuator: the applied voltage leads to a movement of the ions that, in turn, produces the bending motion of the material. Reproduced with permission from [5]. b) Schematic representation of a drug release mechanism under magnetic actuation from polymer-based membranes composed of poly-L-lactic acid (PLLA) and Terfenol-D (TD). Reproduced with permission from [6].

Since the first work reporting an aircraft electric actuator in 1944 [7], more than 150000 scientific journal papers have been dedicated to the study of actuators.

There are examples of actuators based on shape memory alloys, piezoelectric ceramics, electroactive polymer (EAP) actuators - including electronic actuators and ionic actuators-, magnetoactive, electrostatic and ferrofluids [8]. In recent years, polymer-based actuators are gaining increasing attention due to their tunnability: polymer materials can be soft and hard depending on their chemical and physical structure that allows the design of soft actuators for handling soft living tissues (for example) and hard actuators useful for handling heavier materials [1, 9]. Additionally, the large variety of different physical and chemical inputs providing mechanical variations, broad range of strain, stresses and conformation variations, simple processing in a large variety of forms and shapes, are advantages of polymers when compared to ceramic or metallic based actuators [10].

Polymer-based actuators are traditionally divided into several groups, namely: i) based on the elastic relaxation of shape after deformation; ii) based on the change of orientation of mesogen groups (liquid-crystalline actuators); iii) based on a reversible change in the volume and iv) the ones where the driving force is surface tension [1].

Additionally, most of the polymer-based actuators which are being reported in the literature actuate with one of five different methods: electric field, magnetic field, pneumatic, ionic and thermal [2], being electrical and magnetic actuators increasingly used (the research papers on electromechanical actuators being double than the ones of the magnetomechanical ones - Figure 2). Electromechanical actuator are characterized by the high energy conversion efficiency (up to 95%) and excellent duty cycle control [11]. Magnetomechanical ones allow to apply known and controllable forces over many types of elements and structures with no need of contact and no need of any kind of material medium [12].

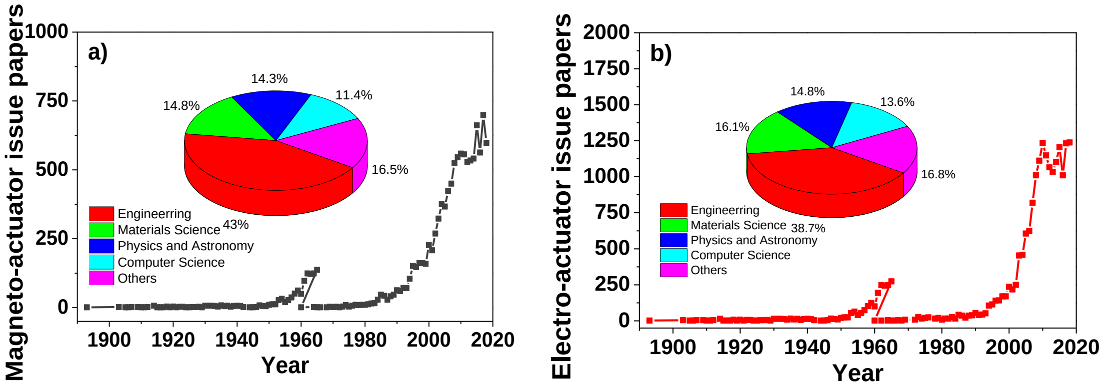

Figure 2. Published papers on SCOPUS database regarding the issue: a) Magneto-actuators and b) Electro-actuators. The insets reveal the percentage of the documents by subject area.

This perspective reports on the achievements in polymer-based electrical and magnetic actuators for technological applications and discusses the underlying physicochemical principles and actuator design strategies. Future research opportunities and implementation barriers are also presented in order to promote further research in this increasingly needed field.

2. Magnetomechanical actuation

2.1 Background

Magnetomechanical actuators show several advantages over other types of actuation methods which make them highly implemented in technological applications. Thus, they allow the production of relatively large forces over large distances (as opposed to the small operation distances of electrostatic actuators), are mechanically robust, show extended working range and fast response time[13], low field operation[13, 14], generate high mechanical forces [15], large strain[16], and allow highly controlled and localized actuation[17]. As a consequence of those advantages and the developments in soft magnetic materials, the advances in power electronics/digital control technology, and the continuous demand for higher performance motion control systems, magnetic actuators are currently intensively being investigated [18, 19]. The main principle of magnetic actuation is to drive materials with magnetic forces and/or torques. A low-frequency and quasi-static magnetic field is the most common approach to apply forces and torques directly to magnetic materials without the need for any tools or direct contact [20]. Magnetic forces and torques induced on a magnetized material are expressed as follows:

$$\overrightarrow{fm}= \int_{V_m} (\vec{m}.\Delta)\vec{B}\; dV_m \tag{1}$$

$$\overrightarrow{\tau m} = \int_{V_m} \vec{m}.\vec{B} dV_m \tag{2}$$

where V_m is the volume of the magnetized object, $\vec{B}$ is the flux density of the applied field in Tesla, and $\vec{m}$ is the magnetization of the object in Ampere/meter)[21, 22].

From equations 1 and 2, it is observed that both the magnetic force and magnetic torque are proportional to the gradient of the magnetic field. In this way, by changing materials and magnetic field gradients, different actuator types and topologies are emerging with varying operational characteristics, in terms of displacement, rotary or linear, speed of response, positional accuracy and duty cycle [19].

Additionally, the mechanical response of magnetomechanical actuators is conditioned by the intensity of the magnetic force, which in turns is proportional to the magnetic susceptibility (χ_m) of the material. For high values of magnetic field, the magnetization of the magnetic element becomes equal to the magnetization saturation (M_s). Usually, these actuators are excited with high enough intensity to reach the saturation level, and consequently, Ms takes a highlighted position against the magnetic susceptibility in the material selection. In addition, if a high frequency movement is required, χ_m will be an important selection parameter to achieve the desired mechanical response[23].

As the magnetic force is applied, a movement in response to that force will be observed in the magnetomechanical actuator. Such force is dependent on the stiffness of the material, its viscous coefficient and on the interaction with the surrounding environment, and will be a key aspect of the final mechanical response [23].

Another relevant characteristic of the actuator is its composition. Typically, most of the magnetomechanical devices are based on magnetostrictive materials such as Dysprosium (Dy), Terbium (Tb), Terfenol-D, NiFe, SiFe, CoFe, Galfenol, Alfenol, Fe-Ga, Fe-Al and Fe-Co, ferromagnetic materials (Co, Fe, Ni, $CoFe_2O_4$, Fe_3O_4) or piezomagnetic materials (MnF_2, CoF_2, $DyFeO_3$) [23, 24]. By combining a magnetostrictive actuator such as Terfenol-D with a piezoelectric element such as lead magnesium niobate-lead titanate (PMN-PT), magnetoelectric (ME) actuators such as the one represented on Figure 3 can be developed (an applied magnetic field induced strain is mechanically transferred to the piezoelectric phase, which experiences a change in electrical polarization). Among the different geometries exploited for magnetostrictive actuators, it is worth mentioning this ME class of magnetomechanical actuators once such coupling is very interesting to both material science and engineering viewpoints, due to the potential technological applications such as ME tools for biomedicine or ME motors [24].

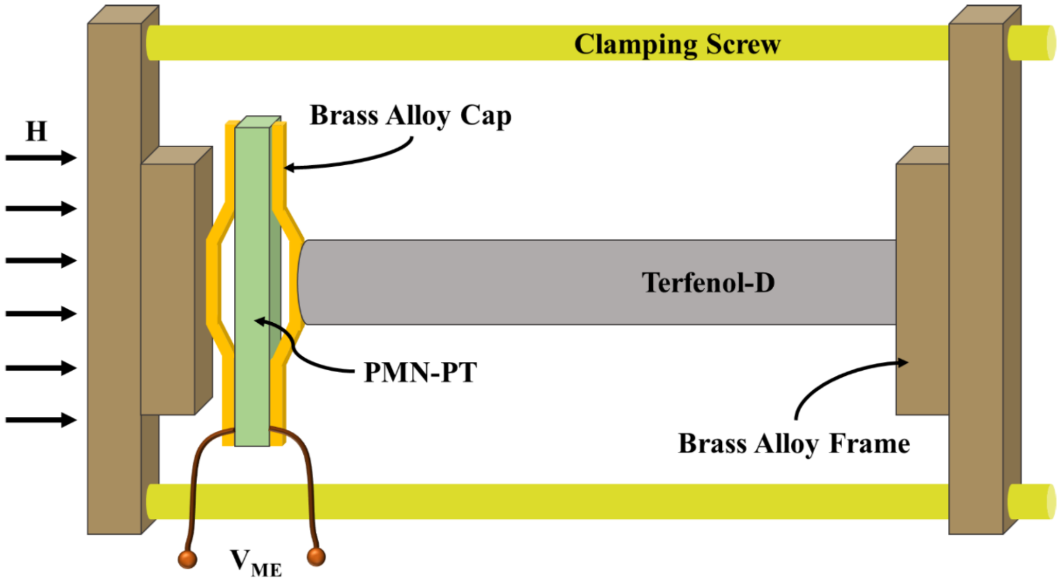

Figure 3. Schematic representation of a ME actuator based on Terfenol-D and a PMN-PT cymbal-like transducer proposed in [25]. Reproduced with permission from[24].

Incorporating the last advances in microfabrication and nanofabrication, nanocomposites have also been used for the fabrication of the magnetostrictive actuators. Magneto-mechanical nanocomposites are obtained by adding a magnetoactive component (magnetostrictive, ferromagnetic or piezomagnetic) in a host matrix. Based on the mechanical-chemical requirements, polymer matrices stand as the best option. In particular, elastomers (such as polydimethylsiloxane, PDMS) are the preferred choice because of their capacity for elastic deformation. Despite of this, some thermoplastics and thermosets (such as PMMA, PU, PS, or Epoxy) can be found in the literature acting as matrix for magneto-mechanical polymer composites[24].

2.2 Applications: developments, limitations and needs

Applications of polymer-based magnetic actuators include: aerospace (electromechanical and electro-hydraulic actuators for flight control surfaces); automotive (short-stroke actuators, for applications such as high-speed diesel fuel injector control valves; active and passive magnetic bearing actuation systems for electric vehicle flywheel energy storage units); industrial (long stroke linear motors for high-speed packaging and manufacturing applications); healthcare (linear actuators for resonant electro-mechanical systems, such as positive-displacement diaphragm air-compressors); informatics (multi-degree-of-freedom spherical actuators for force-feedback joysticks, manipulators)[19] and microfluidics (actively manipulation of fluids to induce chaotic advection and

increase the interfacial area of two fluids, thereby decreasing the length scale over which diffusion has to take place to mix the fluids).
As representative examples, a flexible and wearable thin form factor magnetic actuator (magnetic latching + flex PCB actuation coils + laterally shielded magnets) was developed for dynamic haptic feedback, overcoming some limitations of traditional soft actuators (coarse and non-localized stimuli) by placing several hard actuation cells by flexible printed electronics, in a soft 3D printed case [26]. Nevertheless, the Joule heating, led to a loss of robustness on long-term repetitive actuation at high frequency. The incorporation of thermal insulator layers, the increase of the air-flow inside the actuator and the introduction of printed shells are strategies to increase the long-term reliability of the magnetic actuator.
A closed-loop control system for an insertable robotic camera system for single incision laparoscopic surgery composed of permanent magnets (sEPMs) and the tail-end internal permanent magnets (tIPMs) was reported [27], allowing tilt motion and pan motion control. The magnetic actuator exhibits a unified control of anchoring, navigating, and rotating the camera without on-board electric motors (Figure 4a). In order to allow a convenient use by surgeons, the camera on-board electronics needs to be integrated. There is also a need of introducing an inertial sensor to provide the camera orientation feedback wirelessly, the dimensions and the weight need to be further reduced and the control cables removed. Additionally, the proposed system needs to be tested *in vivo*. Further, to solve the size and weight problems related to heavy and large electromagnetic coils, they can be changed by lightweight permanent magnets [28].
Also in the biomedical field two actuators with different Fe_3O_4 filler concentration, 10wt.% and 20wt.%, were evaluated for implantable applications [29]. It was discovered that increasing the magnetic filler concentration allows to increase the displacement of the actuator (5 mm) and material stiffness at a magnetic field of 0.23T. The reported polymer magnetic actuator demonstrated large motion, high-frequency operation and most importantly, wireless actuation without physical electrical connection or onboard electronics.
By using poly-l-lysine-coated Fe_3O_4 nanoparticles, the synthesis and characterization of magnetic actuators for neural guidance was reported [30]. Migration experiments under external magnetic fields confirmed that these polymer-based magnetic actuators can effectively actuate the cells, thus inducing measurable migration towards predefined directions, allowing to enhance or accelerate nerve regeneration and to provide guidance for regenerating axons. Nevertheless, the potential adverse effects of the magnetic actuators such as toxicity, poor water solubility and the nanometer size of the fillers require further considerations.

The fabrication of hydrogel-based magnetic actuators by the direct incorporation of carbon-coated cobalt nanoparticles into the backbone of a styrene maleic anhydride-based hydrogel was described, as well as its application as a flexible and strong magnetic actuator [31]. The reported chemical modification (cross-linking) of the particles prevents metal loss or leaching. Since metals have a far higher saturation magnetization and higher density than oxides, the resulting increased force/volume ratio allowed significantly stronger magnetic actuators with high mechanical stability (elongation of 1cm at 0.53 T), elasticity (stretched to 123% of their initial length), and shape memory effect. These properties may facilitate the use of soft actuators in medical implants, heart pump components or micro-pumps for controlled drug delivery and will eventually pave the way to the use and development of flexible machines.

On an innovative approach, a compliantly coupled silicone dielectric elastomer actuator was developed using magnetic repulsion [32], where two membranes are compliantly coupled using the repulsion of two permanent magnets (Figure 4b). The compliantly coupled mechanism was evaluated, demonstrating an adjustable phase shift between the two outputs that could not be achieved using previous rigidly coupled designs. This phase control scheme of two outputs from a single dielectric elastomer offers an alternative approach towards potential applications such as active vibration absorption and gait changes in biomimetic locomotion, while the active expansion generated from in-phase activation of the two membranes can potentially be exploited for active shape changes in applications such as suction cups. The performance of such magnetic actuator based on dielectric elastomers can be further optimized by using magnetic modulating mechanisms and by decreasing initial magnet distance within the stable range [33].

Regarding polymer-based magnetic composite actuators, a magnetic polydimethylsiloxane/NdFeB composite membrane was fabricated using a soft lithography process producing a maximum membrane deflection up to 12.87 µm [34]. The functionality test of the composite actuator for fluid pumping resulted in an extremely low sample injection flow rate of approximately 6.523 nL/min at a magnetic field of 0.63 mT. The performance on real environment requires further optimization at very high injection resolutions for high accuracy dosages, in particular in lab-on-chip and drug delivery systems. The deflection in this type of microfluidic polymer-based magnetic composite actuators can be maximized with the use of permanently magnetized magnetic fillers and with the combination of thicker and thinner layers of the actuator [35]. By using the same actuator composition, the application of the diamagnetic levitation of a small floater magnet for the accomplishment of a magnetically actuated membrane actuator in polymer composite technology was demonstrated [36] . The reported actuator exhibited a 14 nm/mA normalized displacement at an AC magnetic field of 1.2 mT and 200 Hz. Future work should be devoted for miniaturization of the actuator concept and the integration in a PDMS based microfluidic chip for fluid manipulation.

By using the same polydimethylsiloxane matrix and doping it with Fe, a 3D printed deformable magnetic micro-actuator was fabricated [37], capable to output up to 27 µN blocking force at a magnetic field of 160 mT. This micro-actuator can be used as a mixer in lap-on-chip applications and as the anchoring and propulsion legs of endoscopic capsule robots. Despite solving some problems of the existing methods for manufacturing polymer-based magnetic actuators, such as the high complexity and difficulty to use for the fabrication of micro-sized high-aspect-ratio materials, additional work is needed including experimental/theoretical studies on fluid dynamics and biocompatibility.
Still with respect to actuator membranes, a compact and simple electromagnetic micro-actuator structure was developed using PDMS with embedded NdFeB magnetic particles [38]. Experimental results revealed that a maximum deflection of 9.16 µm for an applied magnetic field of 0.98 mT was obtained within 40 seconds. Despite this polymer composite flexible membrane actuator being premeditated as part of a compact micropump for a fluidic injection system in a Lab-on-Chip device, it can also be for applied in robotic and biomedical actuators.

By mixing electroactive, magnetoactive and conducting polymer concepts, an electrolyte-free conducting polymer magnetic actuator was developed based on camphorsulfonic acid doped polyaniline gel [39] (Figure 4c). This actuator exhibits a large swinging speed (9000 swings/minute) that lies in the range of those of flies and bees (1000-15000 swings/min), and it is fatigue-resistant (> one million cycles) in the presence of an external magnetic field (250 mT) and significantly low driving voltage (< 0.5 V). Such features allow the development of efficient biomimetic actuators that mimic the performance of animal muscles.

In the area of robotics applications, the design and fabrication of miniature soft electromagnetic actuators has been described[40] that worked based on the Lorentz force principle. Such actuators were produced from silicone, liquid metal eutectic gallium indium (EGaIn) alloy and NdFeB magnetic powder (Figure 4d). Results proved that the polymer-based actuators were able to operate over a scalable range of voltages or currents (50 mA to >1 A, or 50 mV to 1 V respectively), yielding displacements of up to 1 mm, and retained most of their performance when stretched up to 100%, or bent to angles of 38°.

Finally, a soft spiral-shaped micro-swimmer was described composed of stimuli-responsive N-isopropylacrylamide-co-acrylic acid hydrogel encapsulating magnetic EMG 707-nanoparticles with propulsion force controlled by pitch change [41].

With a stimulation of an AC magnetic field of 20 mT at 5Hz, a shrinking ratio of ≈0.3 was reported suitable for applications such as microscale biochemical tools, including autonomous soft-robots and soft micro-probes.

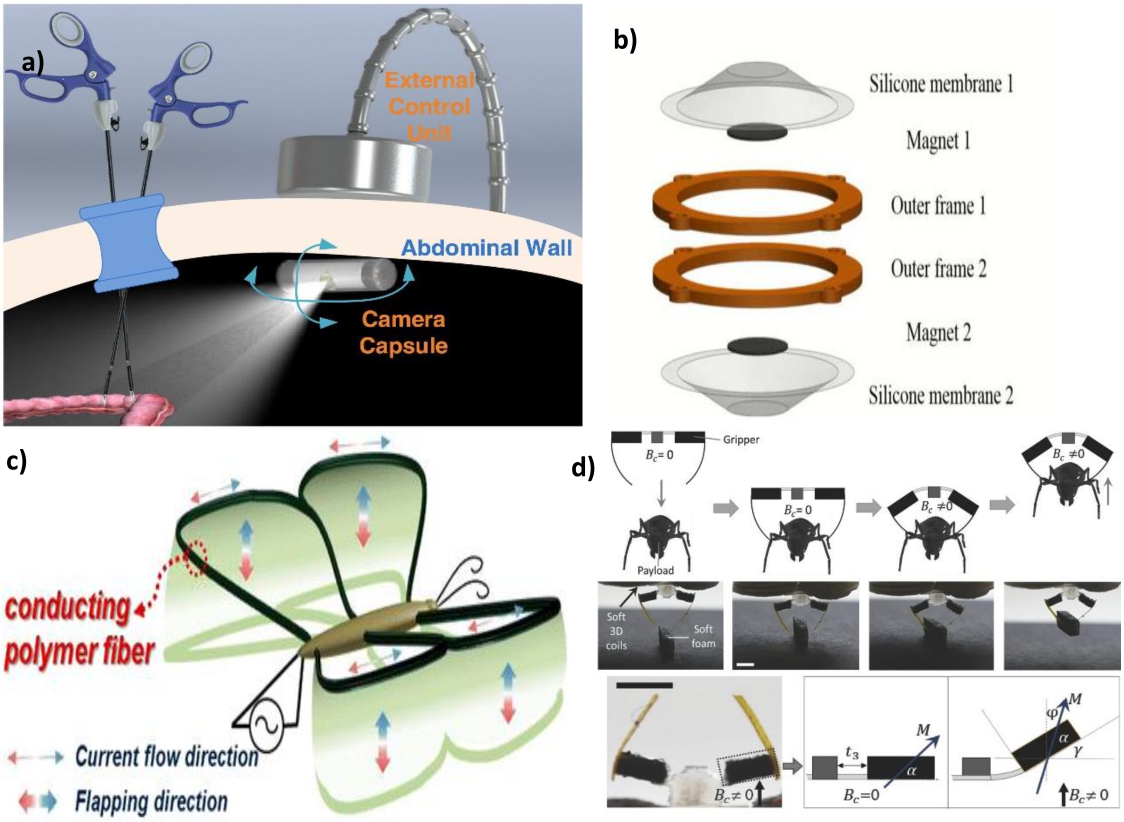

Figure 4. Some of the discussed applications of polymer-based magnetomechanical actuators: a) Insertable robotic camera system for single incision laparoscopic surgery [27]; b) Compliantly coupled dielectric elastomer actuator using magnetic repulsion [32]; c) Electrolyte-free conducting polymer-based actuator exhibiting flapping wing motions under applied magnetic fields[39] and; d) Miniature electromagnetic actuators for robotic applications[40]. Images reproduced with permission from [27], [32], [39] and [40], respectively.

In order to further improve the applicability of magnetomechanical polymer-based actuators such as the ones shown in Figure 4, long-term analytical and numerical analyses that consider creep, fatigue, and fracture need to be implemented [42].

3. Electromechanical actuation

3.1 Background

Electromechanical actuators are known by its ability to convert electrical signals into a mechanical force. In this field, important efforts have been performed to achieve the development of actuators with high speed responses at low voltages [43]. In this context, challenging steps involve the precise control of the actuation performance through the current-voltage, displacement-voltage and displacement frequency characteristics of the materials. The main challenge of this type of electromechanical transducers relies on the fabrication actuators with size reduction, important for the micro mechatronic systems, with wide performance range in terms of displacement, force and frequency response [44, 45]. Typically, the development of polymer-based actuators involves electroactive polymers (EAPs), conductive polymers and shape memory polymers [40]

Electroactive polymers (EAPs) defined as polymers with the ability to change the mechanical properties in response to an electrical stimulation, have emerged as an interesting class to the actuators due to their intrinsic properties, low cost, flexibility and lightweight [43, 46]. For electromechanical actuators based on EAP, the actuation mode is dependent on the EAP type: electronic or ionic EAP.

3.1.1 Electronic EAP-based actuators

Electronic electroactive materials are generally divided in dielectric elastomers (DE), electrostrictive and ferroelectric polymers and liquid crystal elastomers [47]. In dielectric elastomers the mechanism of actuation results from the electrostatic interactions between the electrodes or from a molecular re-structuration within the material. This type of actuation is observed in polymers with low elastic modulus [2]. In the case of dielectric elastomer actuators, the actuator consists of an elastic polymer with compliant electrodes coated in both sides (top and bottom) [43]. The mechanism of actuation involves the application of a voltage across the device. The applied voltage results in Coulombic attraction between both charged electrodes, leading to an electrostatic Maxwell stress on the polymer in the thickness direction. As a result, compressive strains are formed by the electrostatic interactions between the two surfaces of the elastomer. Due to the elastomer properties, namely its ability in maintain constant the volume, shrinkage in one direction leads to an expansion in another direction (**Figure 5a**).

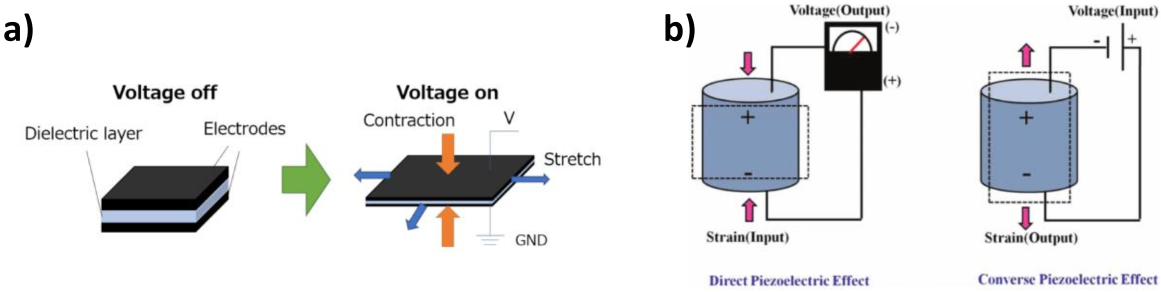

Figure 5. a) Schematic representation of the general mechanism of dielectric elastomer actuators [48]. b) Schematic representation of the direct and converse piezoelectric effects, the converse being the one used for mechanical actuator applications. Reprinted with permission from [49].

The effect is reversible and when the voltage is removed, the film returns to its original shape and size [2, 43]. These actuators can yield large strains (10% to 100%), promoting a high work per unit volume per cycle. Besides behind an interesting class of materials for actuator development, several limitations are associated with the electroactive dielectric elastomers, namely the application of large driving voltages to achieve large actuation strains, limiting their applicability [44].

In the case of electrostrictive relaxor ferroelectric actuators, the applied electric field induces the alignment of the poled domains within the polymer matrix, remaining the polarization permanent after the electric field removal. Liquid crystal elastomer-based actuators combine a polymer network and a liquid crystal mesogen. With increasing temperature, the liquid crystal mesogen transition from the nematic phase to the isotropic phase occurs, resulting in the deformation of the material [50, 51].

The stress and strain resulting from the bending motion can be quantitatively evaluated based on the theory of elasticity that is valid for all EAP-based actuators. The actuator deformation functional free energy associated to the elastic deformation ΔF_{el} and to the free energy associated to the isotropic expansion ΔF_{ex} as a resulting of the applied voltage can be expressed as follows [52]:

$$\Delta F = \Delta F_{el} + \Delta F_{ex} \tag{3}$$

In which the ΔF_{el} and ΔF_{ex} are determined using elastic constants (μ and K), the tensor strain (u_{ij}) and the stress resulting from the applied voltage, as described by [52]:

$$\Delta F_{el} = \int dr \left[\mu \left(u_{ij} - \frac{1}{3} \delta_{ij} u_{ij} \right)^2 + \frac{1}{2} K {u_{ll}}^2 \right] \tag{4}$$

$$\Delta F_{ex} = \int dr [-p u_{ll}]$$

Different elastomers have been used to develop electrically driven actuators, such as silicon-based elastomers modified with polar thioacetate groups [53], polydimethylsiloxane (PDMS), polyurethane (TPU) [54], acrylic (3M VHB) [55, 56] and modified acrylic elastomers [57].

3.1.2 Piezoelectric actuators

Piezoelectric polymers have been gained special attention in the development of EAP polymer-based actuators and their working principle is based on the reconfiguration of the electrical dipoles under and external stimuli (electric field or mechanical solicitations) [46]. The main advantage of this class of polymers relies on the fabrication of lightweight and cost-effective materials that enable electromechanical devices [44].

Piezoelectric materials are able to convert the electrical energy into a mechanical movement and, contrarily, the conversion of pressure or vibration into electrical energy [58]. The application of a tension and compression leads to the generation of voltages of opposite polarity, proportional to the applied force (direct effect). The development of actuators based on this type of materials allows to develop devices able to produce a small displacement when a voltage is applied (**Figure 5b**).

Within the wide variety of piezoelectric polymers derived from both synthetic, such as PVDF and its co-polymers, poly(L-lactic acid) (PLLA), and poly(hydroxybutyrate)

(PHB), and natural sources like cellulose, chitin, keratin, among others [59], PVDF and its co-polymers are the most commonly used for the development of actuators [60, 61].

3.1.3 Ionic EAP-based actuators

In ionic EAP actuators, the performance is mainly dependent on ions diffusion and mobility [44]. The mechanical response results from the mobility or diffusion of the ions into the polymer matrix when an electrical field is applied [43]. Contrarily to the electronic electroactive polymers, the ionic electroactive materials appear as a promising class of low driving voltage actuators (<3 V) when compared with electronic actuators [42]. Other advantages associated with ionic EAPs actuators are related with the lightweight, and capability of working in an aqueous medium [43]. Different polymer matrices and conductive fillers are combined to design an actuator. An interesting strategy relies on electromechanical actuators involving a highly ionic hydrated polymer-metal composite (IPMC). Typically, IPMC actuators are composed of an ion exchange membrane sandwiched between high surface area electrodes (e.g. gold nanoparticles or ionic polymer composites with carbon nanotubes, CNTs) [43]~~.~~

The most commonly used polymers for IPMC actuators development are cation exchange membranes based on perfluorinated polymers, such as Flemion and Nafion, the actuation mechanism resulting from hydraulic and electrostatic effects [62]. As schematized in **Figure 6a**, upon an applied voltage, the metal electrodes charge and the hydrated free counterions of Nafion migrate towards the oppositely charged electrode, which results in the bending of the actuator. The back relaxation due to the diffusion of the free water molecules, is observed upon the saturation of the oppositely charged electrode by the mobile ions, allowing the reverse of the device polarity [43] .

The actuator response of IPMC actuators can be improved by the hydration level, ionic content, surface conductivity and electrode layers flexibility. Additionally, also carbon-based nanostructure materials, such as CNTs, fullerenes, graphene and graphene oxide, and carbon black have been employed as electrode materials in the fabrication of metal free actuators due to its interesting properties (relatively inexpensive and high electrical conductivity, large surface area and unique mechanical properties) improving the actuation tip displacement, speed response and force of the actuators. Further, the presence of a carbon-based electrode layer promotes a higher diffusion of the hydrated ions within the membrane, leading to an electric double layer formation [62].

3.1.4 Ionic liquid-polymer based actuators

Ionic liquid (IL)-polymer based materials have been gaining increasing attention in the development of ionic actuators. The combination of polymers with ILs, defined as salts composed by cations and anions, have been used to develop ionic electromechanical actuators. The main advantages of these electromechanical actuators are associated with their lightweight, high flexibility and low operation voltage. In this type of actuators, the applied voltage leads to the ions transport and redistribution near the electrodes, through the migration of the cations to the negative side and the anions to the positive side, resulting in a bending motion (**Figure 6b**) [63].

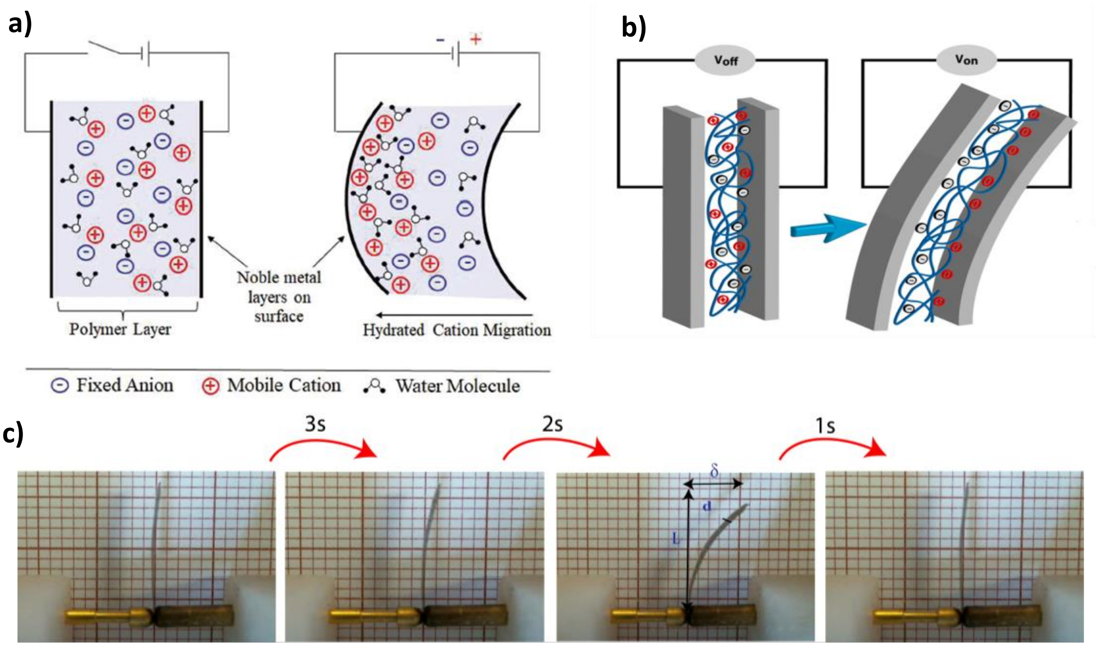

Figure 6. a) Schematic representation depicting the basic actuation mechanism of an IPMC. Hydrated cations move toward the anode under an applied voltage causing the membrane to swell on one side and contract on the other. b) Schematic representation of the bending mechanism actuation of ionic actuators. Reprinted with permission from [43] and [63], respectively. c) Bending motion as a function of time for [PVDF]/[Pmim][TFSI] composites at 5 V and 100 mHz . Reprinted with permission from [63].

The bending response of an electromechanical actuator is dependent of the electrode area and the strain of the sample. Despite this general interpretation, a deeper understanding of the bending motion mechanism is needed, in order to allow tailoring actuator response [52, 63].

The quantification of the electromechanical response in IL polymer based actuators upon the application of an applied field is performed through the analysis of the displacement of the material, being the bending response related to sample free length (L), thickness (d) and displacement (δ) as indicated in Equation 5 (**Figure 6c**):

$$\varepsilon = \frac{2d\delta}{L^2+\delta^2} \times 100 \tag{5}$$

Different studies concerning the development of IL-polymer based actuators have been performed, in which ILs such as N,N,N-trimethyl-N-(2-hydroxyethyl) ammonium bis(trifluoromethylsulfonyl)imide ([N_{1112}(OH)][TFSI]) and 1-ethyl-3-methylimidazolium ethylsulfate ([Emim][C_2SO_4])) [64], 1-ethyl, (1-hexyl and 1-decyl)-3-methylimidazolium bis(trifluoromethylsulfonyl)imide ([C_2mim][TFSI], [C_6mim][TFSI] and [C_{10}mim][TFSI] respectively) and 1-ethyl (1-hexyl)-3-methylimidazolium chloride ([C_2mim][Cl] and [C_6mim][Cl]) have been incorporated e.g. into a PVDF fluorinate matrix [61, 65].

3.1.5 Conductive polymer-based actuators

Electromechanical actuators also rely on conductive polymers, also known as conjugated polymers due to the presence of altering single or double bonds in the polymer chain. This type of EAPs are activated by ion transport and are usually chemically or electrochemically actuated, being necessary an electrolyte for their actuation [2].

It is to notice that only few conductive polymers have been explored for actuators design. Among the most commonly used polymer is polypyrrole (PPy), due to its ability to generate large actuation strain and stress. However, PPy based materials present some drawbacks associated with high rigidity, low conductivity and ion diffusion rate. Another conductive polymer with interesting properties (high, stable and tuneable electrical conductivity) is poly(3,4-ethylenedioxythiophene) (PEDOT). Carbon nanotubes are also an interesting approach for actuator development due their high conductivity and high mechanical strength [66].

The mechanism of actuation in conductive polymers is volume change driven, involving a dimensional change of the material through the incorporation of ions between polymers, as a result of the charge addition or removal from the polymer backbone. The ions flux from the electrolyte leads to a swelling or contraction of the material. Conductive polymer actuators can be classified as cationic or anionic driven, depending on the type of mobile ions introduced in the actuation process (**Figure 7**) [66, 67].

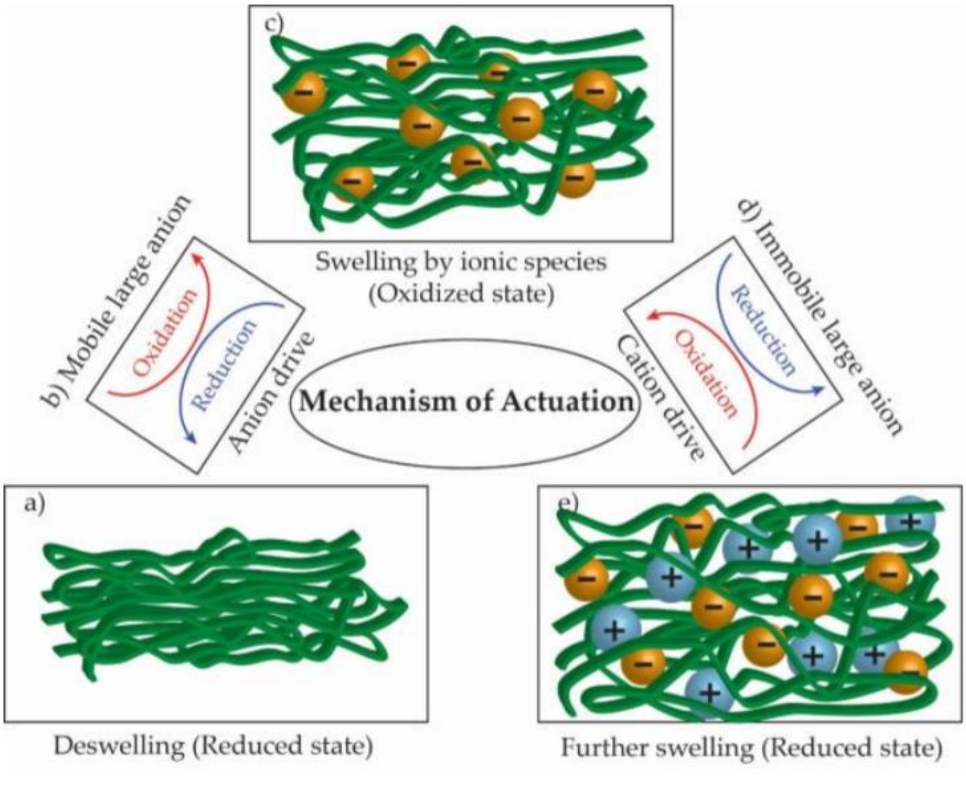

Figure 7. Mechanism of electro-chemo-mechanical actuation in conductive polymers. (a, c, e) Volume changes in conductive polymers via (b, d) two different redox pathways [67].

Actuators based on CNT materials demonstrated a similarity in the bending motion to IPMC actuators, involving in the actuation mechanism the ions mobility of a solvent within a polymer. With an applied electrical field, a swelling or contraction of the CNTs occurs depending on the ions position (entering or leaving the polymer regions). Due to the high electrical conductivity of the CNTs, the ions collected onto the CNTs surface induce a reconfiguration of the electrostatic charges with the potential variation, and in this sense into the CNT electronic structure, inducing dimensional changes in the material [66].

3.1.6 Shape memory- based actuators

Shape memory polymers have attracted significant attention in the development of electromechanical actuators due to its ability to be deformed into a temporary shape and then recover to its original shape through a specific external stimulus [68]. Several advantages are associated to this type of materials such as easy fabrication, high elastic deformation (most of the materials develop strains up to more than 200 %) and tailorable recovery [68, 69]. Several mechanisms of actuation have been reported for this type of actuators, including heat, light, electrical, magnetic field or solvent. The most commonly used shape memory polymers, such as shape memory polyurethane (SMPU) [70], present thermo-mechanical or electro-thermo-mechanical couplings with the ability to recover from high deformations, when compared with shape memory alloys. Particularly for electromechanical actuators, shape memory materials responds with a shape change to an applied voltage in a reversible manner [71].

With respect to electromechanical actuators, shape memory materials responds with a shape change to an applied voltage in a reversible manner [71]. For electro-activated SMP, as most of the SMP presents high electrical and thermal resistance, conductive fillers such as CNTs [72, 73] , carbon nanofibers [74] and metal particles [75] are usually incorporated into the SMP, leading to a Joule heating as a result by the current transfer through the conductive filler within the SMP. The deformation recovery is triggered once the temperature of the system reaches the transition temperature of the SMP [76] .

3.2 Applications: developments, limitations and needs

3.2.1. Electronic EAP actuators

Electromechanical actuators are commonly applied in several areas, such as robotics, biomedical applications as artificial muscles, or haptic applications, among others.
Electronic EAP based actuators based on dielectric elastomers, typically silicon or acrylic based polymers, have been used as artificial muscles and robotic applications [77, 78]. Besides its interesting properties, some disadvantages are associated to this type o polymer-based actuators, mainly due to the high operation voltages, typically above 1 kV [53]. Thus, significant efforts have been developed to design new types of dielectric elastomer actuators [79]. Silicon-based elastomers modified with polar thioacetate groups have been developed to achieve an actuator that responds to low voltages [53]. The distinct characteristics of these actuators relies on the increased dielectric permittivity and rather low elastic moduli, which allows the development of an actuator able to respond to low electric fields. Further, also the thickness of the films influences the actuator

performance. The actuators show mechanical losses that increase with increasing frequency (**Figure 8a and b**) and a actuators performance that increases with films thickness reduction with 5.6% lateral actuation strain at 650 V (**Figure 9**), lower than the traditional voltage applied to dielectric elastomers actuation [53]. Additionally, as shown in **Figure 8c**, thin films (B) containing 10% of silica particles ($B_{10\%}$) allow actuation at low voltages. Another interesting study relies on the development of low voltage phase transition-controlled force actuators based on graphene/Ag/ silicone rubber able to operate at 6 V and grip objects of different weights [80]. The magnitude of the feedback force can be adjusted through the application of a square wave signal and a minimal change in feedback force of 0.02 N was achieved. This devices can be applied in soft robots, bionic devices design, and medical field [80].

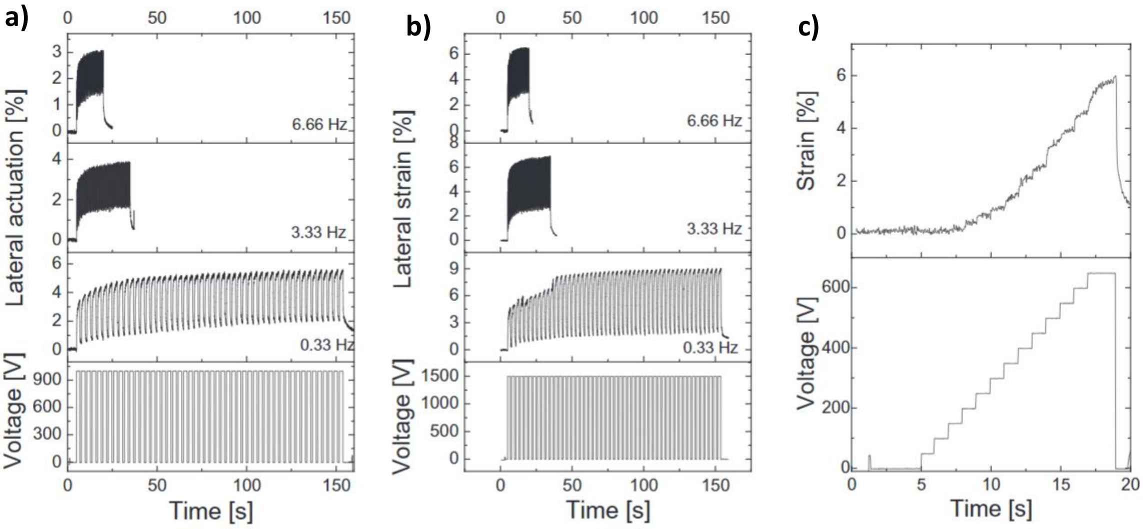

Figure 8. Cyclic actuation at 0.33 Hz 50 cycles, at 3.33 Hz 100 cycles and at 6.66 Hz and 100 cycles and 1000 V of $B_{10\%}$ (a) and 1500 V of $B_{5\%}$ (b). The thicknesses of the actuators were 62 mm and 87 mm for $B_{10\%}$ and $B_{5\%}$, respectively. Lateral actuation strain of a 24 mm thick film of $B_{10\%}$ tested using a voltage ramp of 50 V per 2 seconds up to 650 V (c). Reprinted with permission from [53].

PDMS allows to improve the electromechanical actuation of thermoplastic polymers like TPU [54]. TPU/PDMS blend films containing different PDMS loadings fabricated through a solution-assisted casting method exhibit a maximum areal strain of 2.3% under an electric field of 40 V μm^{-1}, being 60 times higher than for pristine TPU [54]. Dielectric elastomers with enhanced dielectric constant and high dielectric strength have been developed by molecular grafting of azobenzenes to silicon rubber-PDMS [81]. The grafting with 4.0% wt% of azobenzene induced a breakdown strength of azo-g-PDMS up to 89.4 V μm^{-1} (higher than that of pristine silicon rubber) and enhanced the electric field induced deformation, reaching a maximum area strain of 17% with 7.1% wt. of azobenzene, showing also a short time response of ~0.5 s. Interestingly, printable dielectric electroactive polymer actuators have been produced based on TPU [82], the incorporation of azo chromophores into the crosslinked network leading to an improvement of the strain at the break (550% for 10.3 wt% azo-dipole loading) (**Figure 9a**). Further, the actuation strain as a function of the applied electric field also reveals an increase by the introduction of the chromophores (**Figure 9b**), leading to an area expansion of 4.73% uniformly in all directions [82]. PDMS/SWCNTs composites are capable of a bending angle over 540° under a 12 V drive voltage [83].

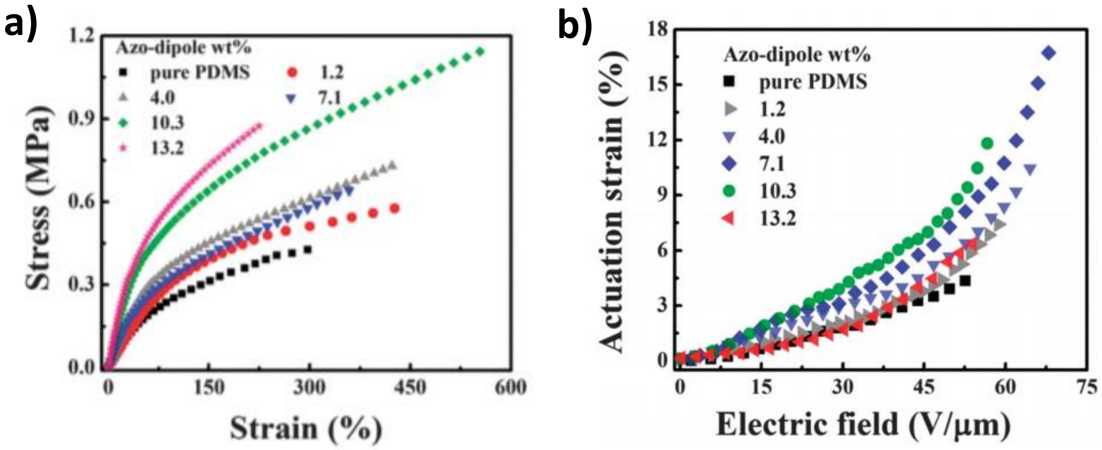

Figure 9. Dependence of the area strain of azo-g-PDMS films with different azo dipole mass fractions on the applied electric field. Reprinted with permission from [81].

Electrically driven soft robots have been also developed through elastic composite materials based on a silicon rubber that combines a strain up to 900% and a high stress (up to 1.3 MPa) with low density. The efficiency of the actuator and its operation is highly dependent on heating and cooling rate. Higher current or a higher distributing heat networks, during the resistive heating, leads to a faster material expansion. During the cooling, the process is facilitated by an optimized design of the actuator geometry and surface area [84].

Devices based on piezoelectric polymers are less expensive (low material cost and processing) that their ceramic counterparts, and do not require advanced microfabrication facilities. Also natural polymers such as silk and collagen are piezoelectric [85]. PVDF actuators fabricated by electric poling assisted additive manufacturing have been evaluated based on the converse piezoelectric effect under the application of an electric field of 1.2 kV. A displacement of approximately 3 μm and 10 μm displacements were obtained in the positive and negative polarity conditions, respectively [86].

Different applications are associated to this type of materials, such as tissue engineering [59]. Additionally, also a piezoelectricity wireless stimulation of a piezoelectric material leads to cells differentiation [85]. Piezoelectric actuators based in PVDF and its co-polymers have been also used in haptic applications, being developed a novel skinny button with multimodal audio and haptic feedback aiming the enhancement of a touch user interface. The application of a normal pressure by the fingertip on the polymer film, leads to the activation of an electric field, producing tactile-feedback vibrations, resulting in a multimodal audio-tactile skinny button [87].

3.2.1. Ionic EAP based actuators

IMPC actuators based on conductive carbon electrode nafion-based actuators were developed by the physical deposition of vulcan carbon (V)/multiwall carbon nanotubes (MWCNT)(V/M) ink on both faces of nafion membranes [62]. It was observed that V/M electrodes improved the ionic conduction and capacitive behavior of the nafion-based actuators. The maximum tip displacement of the actuators was evaluated immersing the actuator in a saturated solution of lithium chloride (LiCl). As a result, in response to a dc applied voltage, the migration of Li ions through the nafion membrane resulted in the bending actuation. A tip displacement of 44.5 mm was obtained in actuators comprising 20 wt% MWCNTs with higher ionic conductivity (26.9 mS cm^{-1}) and capacitive characteristic (45.2 μFcm^{-2}) compared to the Pt-based actuators (**Figure 10**). The actuators also present higher durability in open air [62]. However, the variation in the tip displacement of the actuators over time decayed gradually over time from a maximum value to zero [62]. In an another study, [88] the electromechanical performance of metal-free nafion-based electromechanical actuators was evaluated using poly(3,4-ethylene dioxythiophene):poly(styrene sulfonate) (PEDOT: PSS) and polypyrrole (PPy) as electrode materials.

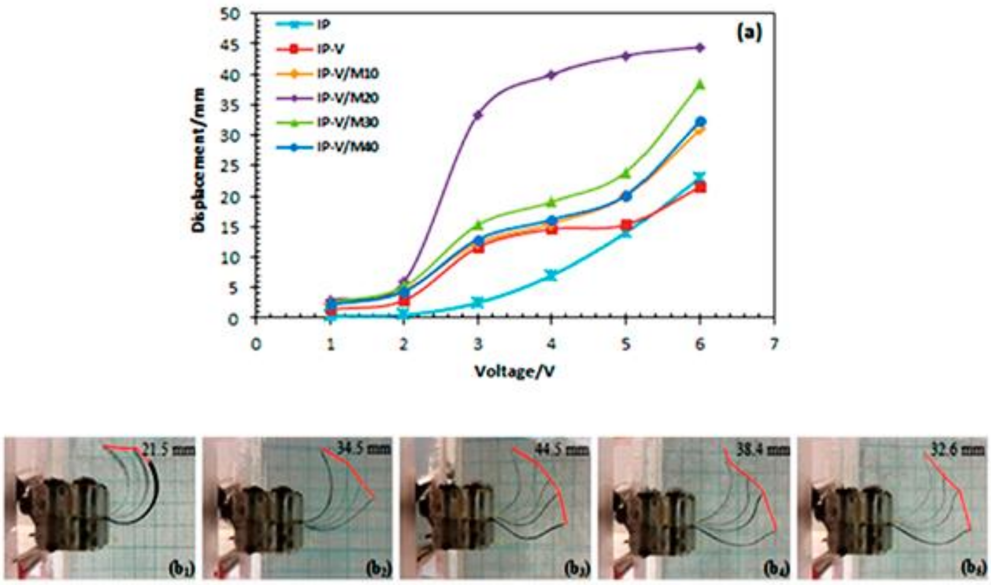

Figure 10. (a) Voltage dependence of the tip displacement of the IP-V/Mx actuators (x denotes the applied weight percentage of MWCNT) and (b) corresponding overlaid digital images captured at the starting point and end point for actuators (b_1) IP-V, (b_2) IP-V/M10, (b_3) IP-V/M20, (b_4) IP-V/M30, and (b_5) IP-V/M40 in response to DC voltage of 6 V in amplitude. IP corresponds to IMPC actuator, and in IP-V/Mx, x denotes the applied weight percentage of MWCNT. Reprinted with permission from [62].

The largest tip displacement (25 mm) was observed for the PPy-electrode actuators to a DC potential of 6 V mainly due to the higher ion and water permeability. The electro-mechanical energy efficiency of the developed actuators revealed a higher electro-mechanical energy efficiency for the PPy-electrode actuators (46.42%) when compared with the PEDOT electrode actuators [62]. Similarly to the previous study, the displacement decayed gradually over the time.

Electromechanical actuators based on covalently bridged black phosphorous/CNTs presents a strong applicability as soft actuators for bioinspired applications (e.g. wings-vibrating, artificial-claw and hand actuators) due to its large maximum strain (1.67%), low power consumption/ strain (0.04 W cm^{-2} $\%^{-1}$), frequency response (0.1–20 Hz), fast strain and stress rates (11.57% s^{-1}; 28.48 MPa s^{-1}), energy (8.48 kJ m^{-3}) density, high power (29.11 kW m^{-3}) and, moreover, excellent cycling stability (500 000 cycles) [89].

In the case of ionic actuators based on polymers and ILs, an interesting approach relies on the use of ILs as the conductive filler into a polymer matrix [90]. The inclusion of ILs into a polymer matrix allows the development of high conductive ionic EAP actuators due to the ions (cations and anions) mobility and diffusion into the polymer matrix and to the plasticizing effect of the IL, which results in a decrease of the T_g of the polymer and an increase of the electrical conductivity. The performance of the actuator can be tuned though the size of the ions, cation and anion type, concentration and IL chain length [44].

A strategy to improve the amount and rate of ion migration aiming the development of high performance actuators [90] relied in introducing hydrophilic polymer-polyvinyl pyrrolidone (PVP) into a PVDF polymer matrix to increase the chemical compatibility between the PVDF and the IL 1-ethyl-3-methylimidazolium tetrafluoroborate ($[Emim][BF_4]$). The IL was used to obtain a polar electrolyte film of PVDF/PVP with enhanced inner channels to promotes the adsorption of polar water and IL to develop either water- or IL-driven IPMC actuator. Both water-driven and IL-driven PVDF-based IPMCs exhibit high ion migration rates, improving the actuation frequency and higher levels of actuation force and displacement, being a promising strategy in the design of artificial muscles with tunable electromechanical performance for flexible actuators or displacement/vibration sensors [90].

Several studies have been also developed aiming the design of IL EAP based actuators for haptic technologies such as braille displays and biomedical applications [64, 80]. PVDF electroactive actuators based on IL/PVDF composites $[N_{1112}(OH)][TFSI]$ and $[Emim][C_2SO_4]$ have been developed [64]. A highest strain bending response of 10.5 mm was observed for the $[N_{1112}(OH)][TFSI]$/PVDF at 5 V demonstrating its potential as a non-cytotoxic material for biomedical applications [64]. Different ILs have been introduced into a PVDF polymer matrix to reach as bending responses of 0.53 % [61] or 0.3 % [65] at applied voltages of 10 V square signal, demonstrating the need to make efforts to improve the performance of the ionic EAP actuators. The influence of the IL cation type also reveals low bending responses (**Figure 11**) [63].

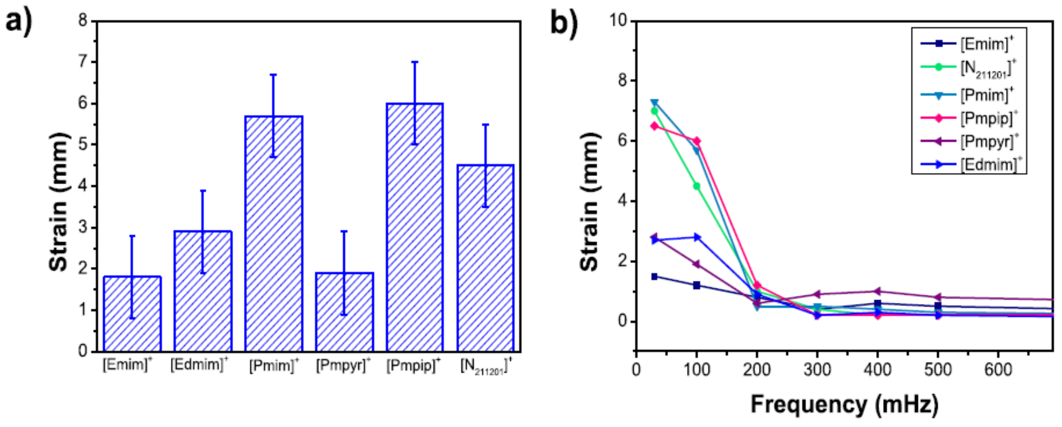

Figure 11. Displacement of the PVDF/IL composites with different IL types for an applied voltage of 5 V and a frequency of 100 mHz (a) and as a function of frequency (b). Reprinted with permission from [63].

High performance actuators based on cellulose nanofibers (CNFs) with single-walled carbon nanotubes and ILs (CNFs/SWCNTs/ILs) and poly(vinylidene fluoride-hexafluoropropylene) (PVDF-HFP)/IL were developed [91]. The effect of CNF and PVDF-HFP and CNF/IL as electrolyte species on the electrochemical and electromechanical properties of the actuators has been evaluated. The PVDF-HFP/IL electrolyte actuators exhibited a higher strain than the CNF/IL electrolyte actuators, both the electrodes being stable in air for at least 10 000 cycles. The frequency dependence of the CNF/SWCNT/IL electrode actuator displacement response showed the potential of the CNF/SWCNT/IL electrodes and the PVDF-HFP)/IL electrolyte actuators for application as electrochemical materials such as wearable and energy-conversion devices [91]. A three-layer material based on PVDF-HFP/CNTs and ILs able to develop a displacement of 1 mm when subjected to an applied voltage of 3V was evaluated as a thin braille display. However, besides the interesting effort, several problems are associated with the actuator, namely its durability, reliability and properties fluctuation [92].

ILs ([EMIM][BF_4] and 1-ethyl-3-methylimidazolium triflate ([EMIM][CF_3SO_3]) have been also combined with cellulose nanofiber/poly(3,4-ethylenedioxythiophene):poly(4-styrenesulfonic acid) (PEDOT:PSS) [93]. The developed flexible and robust films demonstrated potential as actuator materials (maximum strains ranging from 0.64 to 1.04%) for wearable energy conversion devices[93]. The ionic interpenetrating nanofibrillar network formed at the interface of the nanofibrillar thermoplastic polyurethane nanofibrillar matrix with the IL 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide [Emim][TFSI] allows the development of an ionic polymer artificial mechanotransducer (i-PAM) for tactile sensors with the ability to yields simultaneously an efficient wide bandwidth and a wide bandwidth. The developed actuator presented a stable operation up to 200 Hz at low voltage. These actuators can be also applied as soft gripper for surgical robots and an alphabet tactile rendering system [94].

Interesting efforts have been made to develop natural and sustainable soft actuators. A renewable biocompatible nanocomposite soft actuator based on three-dimensional porous electrodes by highly conductive polyaniline (PANI) doped with reduced graphene oxide/ carbon nano-sheets and natural biopolymer cellulose-based electrolyte dissolved in ionic liquid 1-butyl-3-methylimidazolium chloride ([Bmim][Cl]) was reported [95]. These green and environmental friendly actuators reveal large static deformation (up to 13.53 mm) and bending force (up to 12 mN) at the 5 V DC voltage without apparent back-relaxation, quick response speeds (approximately 10 s), excellent flexibility, water retention, as well as superior electrochemical and electromechanical properties compared with conventional ionic actuators (**Figure 12**). Due to all the briefly mentioned results and due to the biocompatibility and biodegradability, the developed actuators seem suitable for medical equipment, wearable devices and soft robots [95].

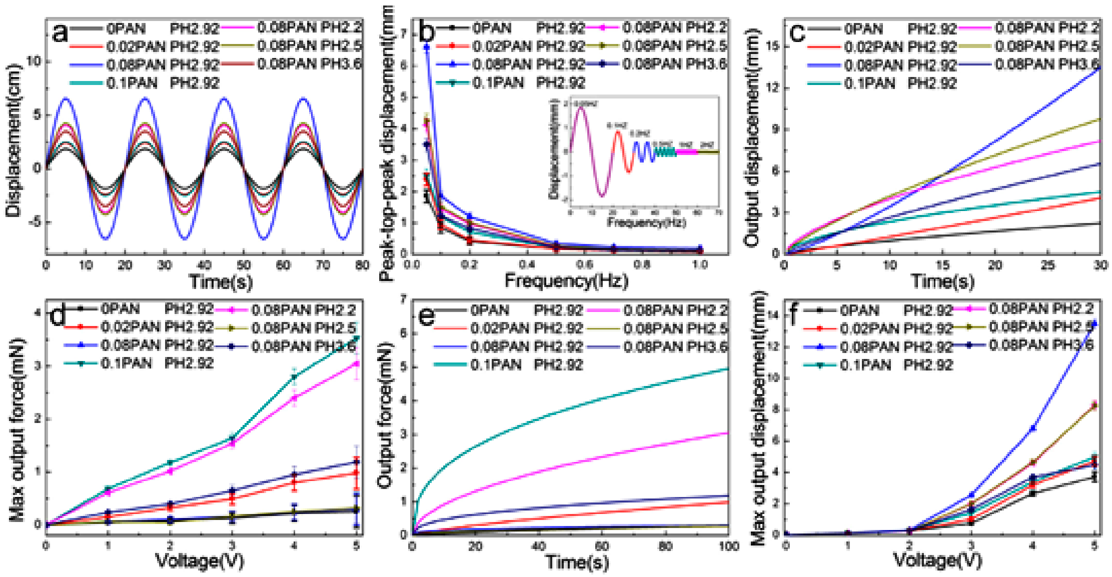

Figure 12. Electromechanical experiments of actuators with different PANI content and dissolving pH value: (a) deflection displacements at 5 V0.05 Hz sine wave voltage, (b) maximum displacement at 5 V sine wave voltage and different frequencies, (c) deflection displacement at 5 V DC voltage, (d) maximum output force of at 5 V DC voltage. Reprinted with permission from [95].

Natural PHB/1-buthyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide ([Bmim][TFSI]) soft actuators, water and moisture resistant, with a reversible macroscopic bending for applied potentials from 0.1 to 7 V and an excellent operation durability up to 105 cycles at 2 V were developed [96]. These actuators display interesting mechanical properties. The actuators demonstrated to be responsive for low voltages being observed a maximum tip displacement of 0.2 mm at 0.1 V, increasing the actuation with the applied voltage. The main advantage of this type of actuators relies on the development of a bioinspired actuator that not exhibit a hygroscopic behavior.

Other naturally based electromechanically actuators were developed based on chitosan–polyaniline composite systems [97]. The actuator performance of the chitosan-polyaniline samples on attainment of the equilibrium swelling in hydrochloric acid (HCl) and sodium chloride (NaCl) aqueous solutions, revealed a reversible deformation with a bending in the direction of the positively charged electrode. Nevertheless, ~50% of deformation is developed within the first 60–90 s once the electric field is switched on. After that, the rate of sample bending drops and the bend value flattens out [97]. Another interesting type of naturally derived actuator with strong potential as a electroactive polymer actuator is based on silk fibroin containing different ILs (1-butyl-3-methylimidazolium tricyanomethanide ([Bmim][C(CN$_3$)]) and choline dihydrogen phosphate ([Ch][DHP])). Both composites developed suitable bending responses for applied voltages as low as 3 V, being the highest bending response ~ 0.5 at applied voltages of 4 and 5V for both [Ch][DHP]/SF and [Bmim][C(CN$_3$)]/SF composites, respectively [98]. Based on the interest in the natural origin of the polymer matrix, efforts must be carried out to improve actuation displacements [99].

Shape memory polymer-based actuators are also commonly employed in the development of electromechanical actuators. Due to its ability to recover shapes after a deformation process through an external stimulus, they have been used in medical devices, smart structural repair and actuators, among others. However, some disadvantages are associated to this type of materials, based on their lower elastic moduli and recover stresses [70]. Shape memory polymers show actuation forces in the range of 1-100 N and actuation speeds of 0.5-10 s/cycle, not allowing a rapid response performance, when compared with other smart materials (**Figure 13**) [100].

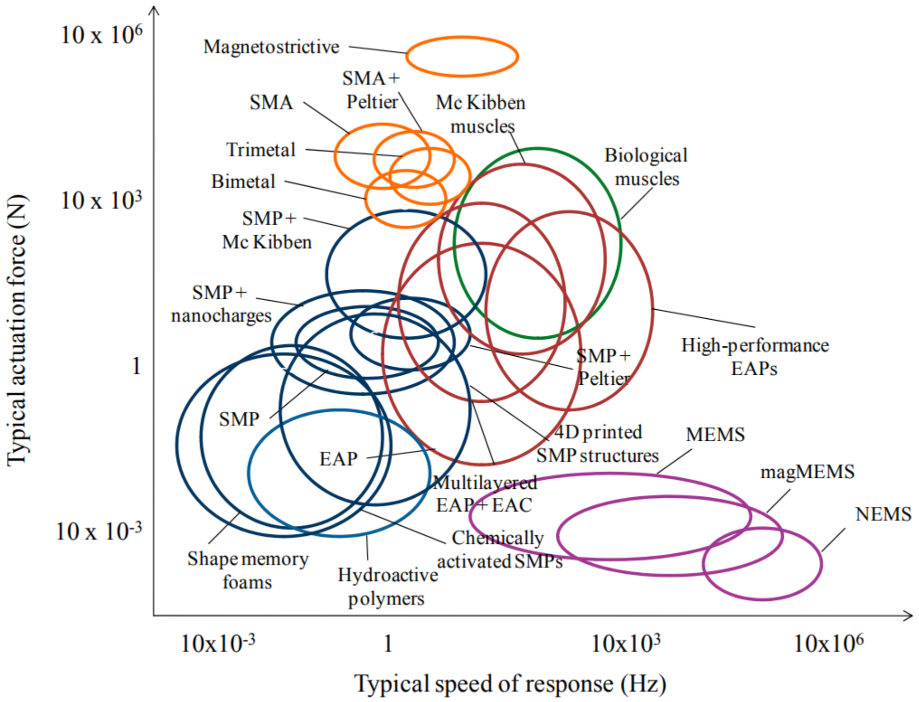

Figure 13. Typical actuation force vs. speed of response for actuators based on “smart” materials. Abbreviations: EAC, electroactive (normally piezoelectric) ceramics; EAP, electroactive polymers; SMP, shape-memory polymers; SMA, shape-memory alloys; MEMS, microelectromechanical systems; magMEMS, Magnetic microelectromechanical systems; NEMS, nanoelectromechanical systems. [100]

To overcome these limitations, shape memory-based composites incorporating different fillers, such as conductive fillers have been developed. On the case of a conductive shape memory composite, an electrically driven-actuator can be designed. The combination of piezoelectricity effect and shape memory effect was achieved through the incorporation of zirconate titanate (PZT) particles into SMPU films [70]. An enhancement of 133% in the maximum recovery stresses after the particles incorporation in the SMPU was observed. Due to the combination of the two effects, the actuators can generate higher displacements. The authors reported displacements of the “U” type actuator and the “Z” type actuator of 18 nm (about 4.7 times displacement of the corresponding film actuator) at 840 V and 75 nm (about 13.3 times displacement of the corresponding film actuator) at 600 V, respectively (**Figure 14**) [70]. Besides the high potential of the developed actuators to be integrated into nanomachine devices, efforts are still needed to improve the piezoelectric effect and to develop novel applications based on this composites [70].

Shape memory polymers can be also manufactured through printing technologies, including 3D or 4D printing [101].

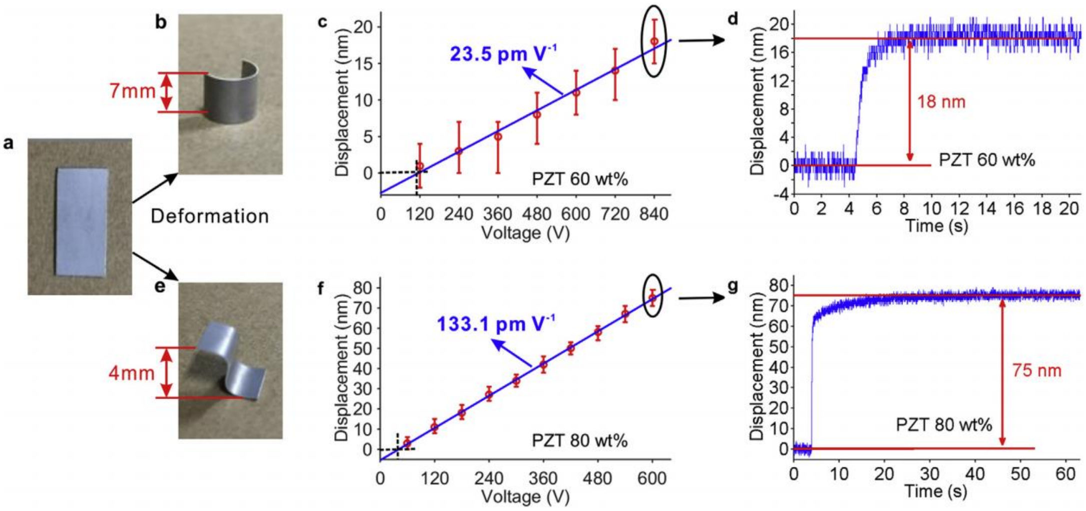

Figure 14. PZT/SMPU composite actuators and their nanoscale displacements. (a) Film actuator. (b) "U" type actuator with PZT 60 wt%. (c) Displacements of the "U" type actuator vs. applied voltages with a linear fitting line. (d) Displacement response of the "U" type actuator corresponding to a step voltage of 840 V. (e) "Z" type actuator with PZT 80 wt%. (f) Displacements of the "Z" type actuator vs. applied voltages with a linear fitting line. (g) Displacement response of the "Z" type actuator corresponding to a step voltage of 600 V. Reprinted with permission from [70].

An overview of representative applications of different types of electromechanical actuators are summarized in **Figure 15**.

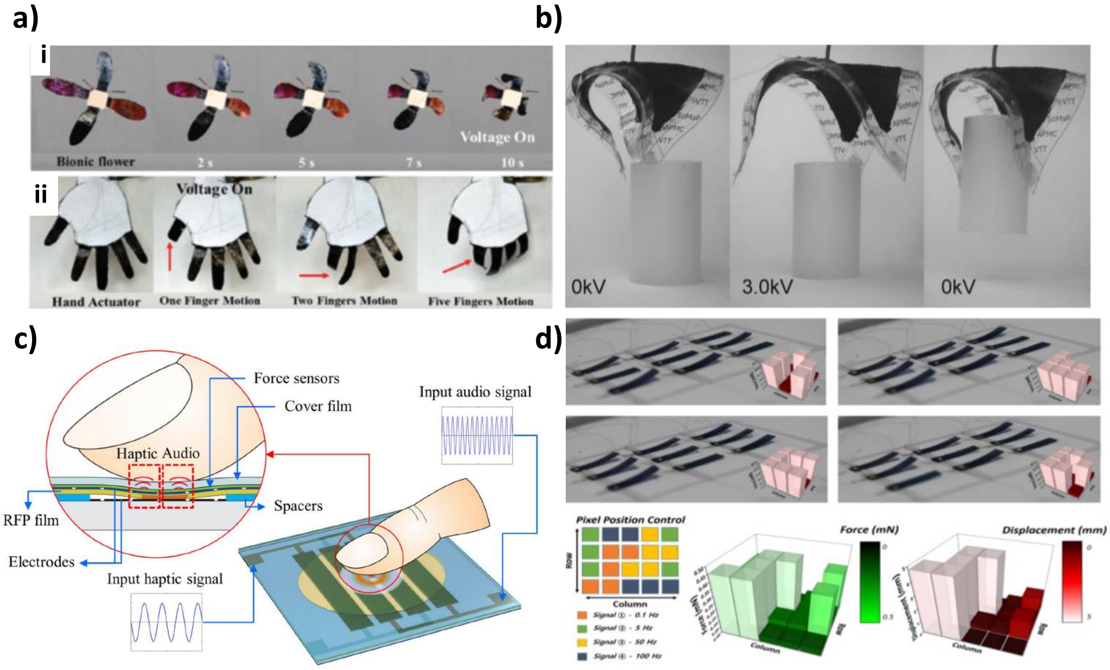

Figure 15 a) Biomimetic applications of electrochemical actuators [89]: i-bionic flower to mimic folding and blooming processes, and ii-hand actuator to mimic finger motions. b) Minimum energy structure provides gripper action. It opens when a voltage of 3.0 kV is applied, enough to grasp objects when the voltage is removed [78].

c) Design and functional mechanism of the audio-tactile skinny button: Conceptual design and functional mechanism of the flexible audio-tactile skinny button, in which aural and haptic sensations are produced simultaneously via the fretting vibration phenomenon [87]. d) Photographs showing effective i-PAM (3×3) tactile rendering of the alphabet by stimulating DC input of 2 V. Inset graph shows blocking force mapping according to the i-PAM conditions and rendering map of blocking force and displacement of the i-PAM (4 × 5) tactile array according to various frequency conditions [94]. Reprinted with permission from [89], [78], [87] and [94], respectively.

4. Final considerations and outlook

The present report provides and overview on the exciting and growing field of polymer-based actuators. Despite many exciting developments have been reported over the last decades (Table 1), the literature shows that polymer-based actuators have been only partially explored with respect to their best features, such as optimal mechanical force and the highest density of stored energy that allow compact and powerful devices.

Table 1: Summary of representative magnetomechanical (MM) and electromechanical (EM) actuators and their main characteristics discussed in this review.

Type	Material	Stimulation	Actuation	Force	Application	Year	Ref.
MM	NaAlg + magnetic nanoparticle (Ferro Tech, EMG707) + p(NIPAN-co-AAC	20mT 5Hz	1.638 mm	not defined	micro-probes and soft robotics	2019	**[41]**
	Not revealed	0.05 T	75º tilt, 20-50 mm	50 N	Robotic camera	2018	**[27]**
	Silicone rubber + Fe_3O_4 (Ø65mm and 2mm)	0.23T	5 mm	not defined	Implantable devices	2017	[29]
	Poly-l-lysine/Fe_3O_4	1.25T	Not revealed	Not revealed	Neural guidance	2012	[30]
	carbon-coated Co /styrene maleic anhydride hydrogel	0.53 T	1cm	Not revealed	Robotic Muscle	2009	[31]
	silicone elastomer	0.5 T	14% stroke	500 mN	bioinspired locomotion	2019	[32]
	Dielectric elastomer	1.2 T	37 mm	2 N	soft robots and haptic feedback	2018	[33]
	polydimethylsiloxane/NdFeB (6x6 mm)	0.63 mT	12.87 µm deflection	Not revealed	drug delivery	2017	[34]
	PDMS/NdFeB (3×2 cm)	1.2 mT	14 µm	Not revealed	Fluid manipulation	2013	[36]

	PDMS/Fe (5x1mm)	160 mT	5mm deflection	27 µN	endoscopic capsule robots	2015	[37]
	PDMS+NDFeB (6x6x0.310mm)	0.98mT	0.0916 mm	Not revealed	Micropump system for Lab-on-Chip	2016	[38]
	camphorsulfonic/PANI	250 mT	6 mm	Not revealed	Flapping Wing	2015	[39]
	Silicon/ EGaIn)/ NdFeB	1.3 T	1 mm, 38º bent angle	0.15 N	stretchable electronics and soft robotics	2018	[40]
EM	PDMS modified with polar thioacetate groups	650V	5.6% lateral actuation strain	Not defined	dielectric in actuators.	2020	[53]
	VHB4910 membrane material	9kV	30.54% maximum axial strain	not defined	micro fexible actuator	2020	[79]
	Graphene/Ag (10 × 1c0 x2mm)	6V 2.83A	4 mm	60g	soft gripper, soft robots	2019	[80]
	TPU/PDMS	40 Vμm^{-1}	areal strain of 2.3%	not defined	not defined	2017	[54]
	Azobenzen-g-PDMS	3kV	area strain of 17%	not defined	not defined	2015	[81]
	TPU (Diabase X60 Ultra Flexible Filament)	5.1 and 8.2 kV	concentric circle paths: area expansion of 4.73% line paths: area expansion of 5.71% and. 4.91%	not defined	not defined	2019	[82]
	SWCNT+PDMS (thickness 20 µm)	12V	Radial 560°	not defined	not defined	2019	[83]
	Soft elastomer membranes+ethanol micro-bubbles (2g)	15V 1A	100% volumetric expansion	30 N	robotic applications	2017	[84]
	Vulcan carbon/MWCNT	6 V	44.5 mm	not defined	not defined	2018	[62]
	Liquid crystal mesogen RM257	1-3V	40%	3.92 N (s 0.312 MPa)	soft gripper, soft robots and artificial muscle	2019	[77]
	PVDF (50x50x0.3mm)	1.2KV	3 µm and 10 µm (positive and negative polarity conditions)	not defined	not defined	2019	[86]
	Black phosphorous(BP)/ CNTs	±2.5 V	21.4 mm	not defined	not defined	2019	[89]

	Lithium/PVDF [Emim][BF$_4$]/PVDF	10 V	12.3 mm 11.1 mm	33.33 mN 7.62 mN	artificial muscles	2018	[90]
	([N1112(OH)][TFSI])/PVDF [C$_2$mim][C$_2$SO$_4$]/PVDF	5V	10.5 mm	not defined	biomedical applications	2016	[64]
	[Pmim][TFSI]/PVDF [Pmpip][TFSI] c	5V	5.7 mm 6.0 mm	not defined	not defined	2019	[63].
	[C$_6$mim][TFSI])/PVDF [C$_6$mim][Cl])/PVDF	10V	4.4 mm 3.4 mm	not defined	not defined	2016	[61]
	[C$_2$mim][Cl])/PVDF [C$_6$mim][Cl]/PVDF [C$_2$mim][TFSI]/PVDF [C$_{10}$mim][TFSI])/PVDF	10V	0.42 mm 2.5 mm 3.73 mm 0.88 mm	not defined	not defined	2016	[65]
	[EMI][BF$_4$]/PVDF	10-20V	maximum strain 0.44–0.76%	not defined	not defined	2019	[91]
	PVDF(HFP)+Ionic liquid+ CNT 5x5mm)	3V	Radial 90°	3 gf	braille display	2016	[92]
	i-PAM/PEDOT:PSS	±2V	5.1 mm	0.4 mN	t haptic interface	2019	[94].
	PANI/MCN/RGO	5V	13.53 mm	12 mN	wearable devices and soft robots	2019	[95]
	Cellulose in IL	6Vac 0.05Hz	17.44 mm	5.93-79.7 mN	not defined	2019	[95]
	[Bmim][TFSI]/PHB	1V 3V 5V 7V	1 mm 5 mm 11mm 17 mm	not defined	not defined	2019	[96]
	Chitosan–PANI	3V	bending angle of 34°	not defined	not defined	2018	[97]
	[Ch]-[DHP]/SF [Bmim][C(CN$_3$)]/SF	4V 5V	0.5%	not defined	not defined	2019	[98]
	CNFs by hardwood bleached kraft pulp (LB): [EMI][BF$_4$]/ CNF/PEDOT:PSS [EMI][CF$_3$SO$_3$]/ CNF/PEDOT:PSS CNFs by bleached kraft pulp (BB): [EMI][BF$_4$]/ CNF/PEDOT:PSS [EMI][CF$_3$SO$_3$]/ CNF/PEDOT:PSS	2V	0.74% 0.64% 1.04% 0.78%	not defined	not defined	2018	[93]

Despite EAPs offer attractive characteristics including resilience, damage tolerance, and ability to induce large actuation strains when compared to the other actuation mechanisms (stretching, contracting or bending) some aspects need to be further optimized such as increase their actuation force, electromechanical conversion efficiency, lifetime and response speed as well as decrease the activation voltage. For that the processes of synthesizing, fabrication, electroding, shaping and handling need to be refined to maximize the actuation capability and robustness in an effort that join materials engineers, chemists, electromechanics, computer engineers, physicists and electronic engineers.

Regarding conductive polymer actuators the future progresses should focus on refining the synthetic methods and deriving novel assembly processes for better control of the size, composition, structure, and interface that will allow to increase their actuation capability. Conducting polymer actuators filled with nanotubes and nanofibers are particularly interesting in such scenario.

Such refining in the experimental studies should also be promoted on shape memory-based actuators in order to better understand the structure–property-relationships that will allow to optimize the tuning of the switching temperature to room-temperature (for biomedical applications) or to high temperatures (aerospace applications). The successful production of such novel polymer-based structures will be not only of interest to achieve the tailored switching temperature capability, but it will also be of great interest to obtain multifunctional materials suitable for harsh temperature conditions ready for application/commercialization/implementation.

Notwithstanding the biomedical field be one of the application areas in which the magnetomechanical materials will be able to find the most fruitful implementation arena, it is urgent the development of materials that simultaneously have high magnetization, biocompatibility, biodegradability and low size (µm level).

Considering mechanical force and displacement as main representative parameters, it is possible to identify some of the areas (Figure 17) where these materials may be successfully implemented, making it evident that the field of applicability is wider for actuators that can produce higher forces/displacements.

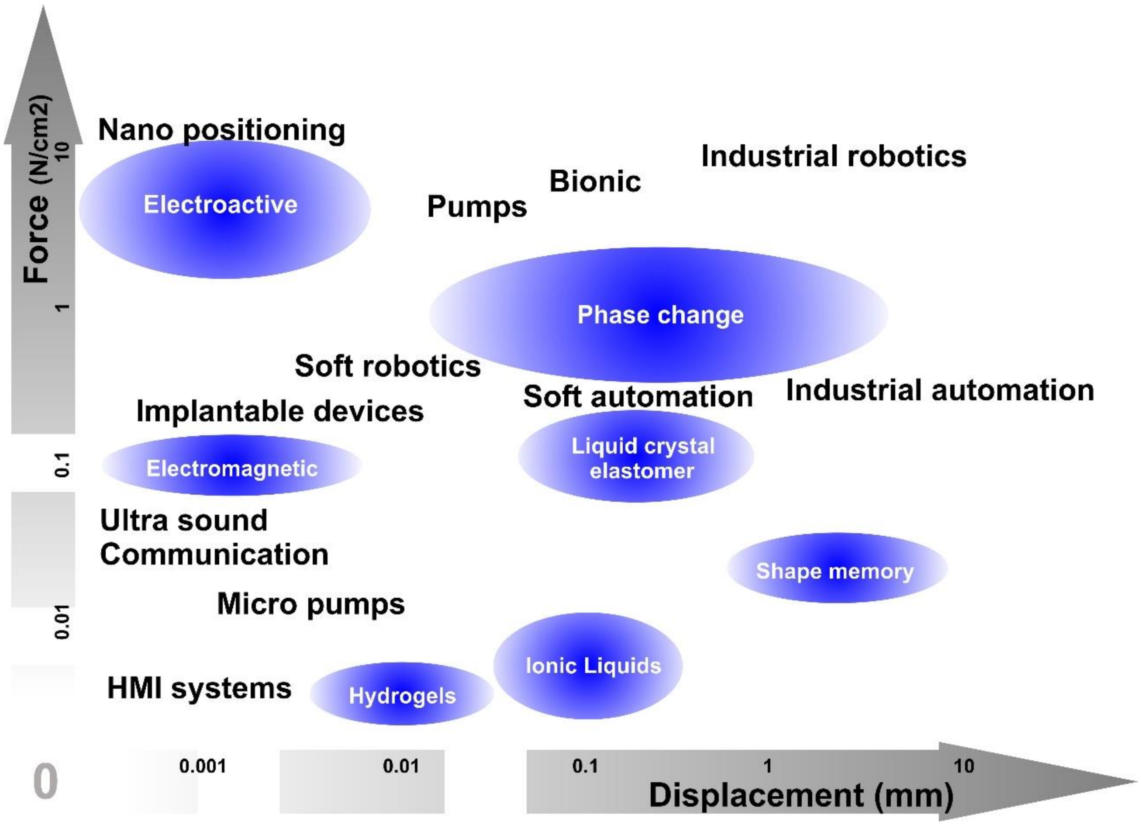

Figure 17. Graphical representation of the distribution of actuator categories (blue) considering force (divided by the area of material so that different types of materials can be compared), displacement and application areas.

Polymeric actuators for ultrasound communication systems is one of the few examples of applicability that is already widespread in the industry. Other polymer-based systems need to be further optimized, being the first and most basic level of research the tailoring of materials response for specific applications. This is especially important in the 'smart materials' research field, driven by the high demand for materials that will help to materialize the fourth industrial revolution closely connected to the Internet-of-Things concept. Further, the development of novel actuation principles can also see the light of the day, opening new interaction and application possibilities. After materials development, other level will emerge: the fabrication and integration of the actuators. At that level, several problems need to be addressed, mainly the ones related to the scalability of the production process, the reproducibility of the devices and their stability over time and under real conditions. This second level brings us to the third, the need to use theoretical tools such as evolutionary computing in order to find novel designs and design methodologies that can improve the existing polymer-based actuator technologies. We have also verified that the most successful polymer-based actuators are targeting applications that require small forces or small mechanical displacements, this can change induce future developments of advanced materials formulations.

At this point, it is impossible to predict how much polymer-based actuators technology will match or even beat nature soon. Such future has not been written yet, not even in this perspective, and for such reason we can safely cite Doc Brown from the «Back to the future» movie: *"Your future hasn't been written yet. No one's has. Your future is whatever you make it. So, make it a good one."*

So, let´s together, give rise to a good future for polymer-based actuators!

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