

# Electroactive polymer artificial muscles: an overview

F. Carpi & D. De Rossi

*Interdepartmental Research Centre 'E. Piaggio', School of Engineering,  
University of Pisa, Italy*

## Abstract

Electroactive polymers as smart actuation materials with muscle-like properties represent an emerging scientific field, bridging material science, mechanical and electrical engineering, and medical disciplines. EAP materials are commonly classified into two major families: ionic EAPs, activated by an electrically-induced transport of ions and/or solvent, and electronic EAPs, activated by electrostatic forces. This paper provides a brief overview on the field, highlighting fundamental features of the most relevant electroactive polymer technologies currently available.

*Keywords: actuator, artificial muscle, electroactive, polymer, transducer.*

## 1 Introduction

Electroactive Polymers (EAPs) represent a broad family of so-called ‘smart materials’ that are capable of transducing energy from the electrical to the mechanical form, and vice versa. As such, they are used for electro-mechanical actuation and mechano-electrical sensing, as well as mechanical energy harvesting to generate electricity [1-6].

EAPs are significantly studied as actuation materials for ‘artificial muscles’ requiring large mechanical compliance, effective down-scalability, high power-to-weight and power-to-volume ratios and high efficiency, typically precluded to conventional actuation technologies (namely electrostatic, electromagnetic, hydraulic, pneumatic and thermo-chemical motors).

EAPs consist of materials capable of changing dimensions and/or shape in response to opportune electrical stimuli. EAP based actuators show useful properties, such as sizable active strains and/or stresses in response to electrical



stimuli, high mechanical flexibility (compliance), light-weight, structural simplicity and versatility, ease of material processing, scalability, no acoustic noise, no generation of heat, and, in most cases, low costs [1-6]. These properties are being used for several kinds of applications. Usage spans from the micro- to the macro-scale in different sectors, such as medical and haptic devices, consumer electronics, and automation and robotic systems. Reported examples include micro-pumps and micro-valves for micro-fluidic systems (e.g. for lab-on-a-chip devices), controlled release of active compounds for medical therapeutic devices (e.g. insulin release in blood stream), miniaturized surgical tools for medical interventional systems (e.g. steerable catheters), miniaturized implantable actuators as components of artificial organs (e.g. mechanical stimulators of cardiac tissue), robotic systems (including medical and industrial robots), variable-stiffness devices (e.g. safe actuators for robots interacting with humans and vibration dampers for vehicles), active orthoses for rehabilitation systems (e.g. hand and limb orthoses), pumps and valves for macro-fluidic systems (e.g. for low-pressure hydraulics and pneumatics), tunable lenses for adaptive optics (e.g. for camera phones) and tactile displays for haptic systems (e.g. refreshable displays for Braille readers or displays with tactile feedback for control panels of consumer electronics) [1-6].

The rapid expansion of the EAP field has stimulated in Europe the creation of the 'European Scientific Network for Artificial Muscles' (ESNAM), which gathers the most active research institutes and industrial developers and end users [7].

EAPs are classified in two main categories: ionic EAPs (whose actuation is based on diffusions of ions and solvents) and electronic EAPs (whose actuation is based on electronic charging of the material). Each class presents the following sub-division in specific groups:

- ionic EAPs: polyelectrolyte gels (such as modified poly(acrylonitrile)), ionic polymer metal composites (IPMC) (such as Nafion/Pt), conducting polymers (such as polypyrrole (PPy) and polyaniline (PANi)) and carbon nanotubes (currently classified as EAPs even though they are non-polymeric macromolecular materials);
- electronic EAPs: piezoelectric polymers (such as poly(vinylidene fluoride) (PVDF)), electrostrictive polymers (such as copolymers based on PVDF), dielectric elastomers (such as silicone) and flexoelectric polymers (such as liquid crystal elastomers).

The following sections provide brief descriptions of the basic features of the most studied EAP families.

## 2 Polyelectrolyte gels

A polymer gel consists of an elastic cross-linked polymer network and a fluid filling its interstitial space. Gels are wet and soft and look like a solid polymer material, but are capable of undergoing large deformations through swelling and de-swelling. Polymer gels can be easily deformed by external stimuli and generate force or execute work externally. If such responses can be translated

from the microscopic level to a macroscopic scale, a conversion of chemical free energy into exploitable mechanical work is achieved [1-5].

As early as the end of the forties, studies about water-swollen polymer gels converting chemical energy into mechanical work were reported. Reversible contractions and dilatations, due to reversible ionizations of suitable groups (for example polycarboxylic (-COOH) groups), are obtained by alternating addition of alkalis and acids. Katchalsky denoted such transformations as “mechano-chemical reactions”. More generally, gels can undergo reversible order-disorder transitions, induced by changes either in temperature, irradiation, electric fields, pH (by chemical or electrochemical activation) or solvent properties. Fig. 1 lists different types of stimuli which are able to induce a mechanical response of polyelectrolyte gels.

Water swollen hydrogels are generally amorphous without any particularly ordered structure at molecular level. Since many years, polymer gels are being studied for the development of low-voltage soft actuators. They can also show shape memory effects. Concerning solvent-controlled activations, the structure of a gel can shift to a disordered state by means of an immersion in ethanol or tetrahydrofuran, to produce swelling. More generally, gels swell in organic solvents and undergo spontaneous motion when are placed in water. The driving force of the gel motion originates from the spreading of the inner organic solvent out of the material when it is placed in water.

If a water-swollen cross-linked polyelectrolyte gel is inserted between a pair of planar electrodes and a voltage difference is applied, the material can undergo anisotropic contractions and concomitant fluid exudations. Electrically induced contractions of the gel are caused by transports of hydrated ions and water in the network (electrokinetic phenomena). In fact, when an outer electric field is applied across a gel, both macro- and micro-ions are subjected to electrical forces in opposite directions. However, macro-ions are typically in a stationary phase, being chemically fixed to the polymer network, while counter ions are mobile and are capable of migrating along the electric field, dragging water molecules with them.

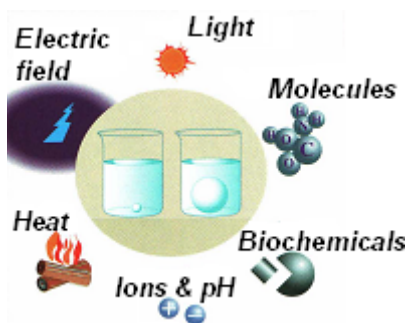


Figure 1: Stimuli enabling mechanical responses of polyelectrolyte gels.

Several active devices have been realized by using these phenomena with different actuating configurations, such as films, strips, membranes and fibres [1-5].

### 3 Ionic polymer metal composites

Most ionic polymeric membranes swell in solvents and are hydrophilic. This gives rise to the ability of the membrane of swelling in water, which can be controlled in an electric field, due to the ionic nature of the membrane. By placing two electrodes in close proximity of the membrane and applying a low voltage (below the threshold for electrolysis), the forced transport of ions within a solution through the membrane becomes possible at microscopic level. The occurring local swelling and de-swelling of the membrane can be controlled, depending on the polarity of the nearby electrode [1-5].

Such a basic principle is exploited in the so-called ion-polymer metal composites (IPMC) actuators. They are used to realize actuators showing large deformations in response to low applied voltages and offering low electrical and mechanical impedance. More in details, materials used for IPMC actuators (such as Nafion by Du Pont) have many ionizable groups in their molecular chain. These groups can be dissociated in various solvents, showing a resulting net charge, which is compensated by the presence of mobile counterions. The net charges of the network macromolecules are called polyions. Electrophoretic migrations (due to an imposed electric field) of the mobile ions within the macromolecular network can cause the network to be deformed accordingly. In fact, the shifting of ions of the same polarity within the network results in both electrostatic interactions with the fixed charges of opposite polarity (contained in the side groups of the polymer chains) and transport of solvent molecules. Both these factors concur to produce a stress gradient between the opposite sides of the membrane, where local collapse and expansion occur, causing a macroscopic bending of the structure. A schematic drawing of the resulting electro-chemo-mechanical activation is shown in fig. 2.

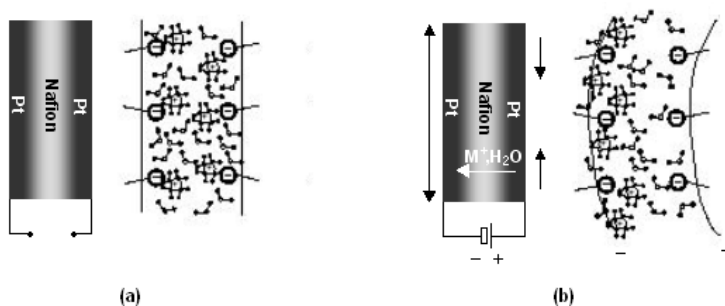


Figure 2: Working principle of IPMC actuators: (a) device at rest; (b) device under activation.

A typical material used to assemble IPMC actuators consists of film of Nafion-117 (Du Pont), an ion exchange membrane. Platinum electrodes are deposited on both sides of a film. The thickness of the actuator is typically of the order of 0.20 mm. To maintain the actuation capability, the film usually needs to be kept moist continuously. Structure and properties of Nafion membranes have been subjected to numerous investigations. An interesting property of this material is its ability of absorbing large amounts of polar solvents, i.e. water. Platinum ions, which are dispersed throughout the hydrophilic regions of the polymer, are subsequently reduced to the corresponding metal atoms.

When equilibrated with aqueous solutions, the membranes are swollen and they contain a certain amount of water. Swelling equilibrium results from a balance between the elastic recovery force of the polymeric matrix and the water affinity to the fixed ion exchanging sites and the moving counter-ions. The water content depends not only on the hydrophilic properties of the ionic species inside the membranes, but also on the electrolyte concentration of the external solution. When an external voltage (usually of the order of 1 V) is applied to an IPMC composite film, it bends toward the anode. An increase of the voltage level causes a larger bending. When an alternate voltage is applied, the film undergoes movements like a swing. The displacement depends not only on the voltage magnitude, but also on the frequency (lower frequencies lead to higher displacements, according to the device bandwidth).

IPMC actuators usually operate best in a humid environment, even though they can be made as encapsulated devices to operate in dry conditions [1-5].

## 4 Conducting polymers

Conducting polymers (CP) are chemically characterized by the so-called conjugation, in which carbon double bonds alternate with carbon single bonds along a polymer backbone. Polypyrrole (PPy) and polyaniline (PANi) are highly renowned examples. Conducting polymers can be characterized by a high conductivity when doped with ions. Their conductivity can be reversibly changed by orders of magnitude, by changing the doping level. Unlike silicon, dopants can be easily inserted and removed from the spaces they occupy between the polymer chains. Moreover, in comparison with other semiconducting materials, the doping level can be very high: approximately one dopant counter-ion per three or four monomers.

To serve for actuation, conducting polymers have to be used as components of an electrochemical cell, whose basic structure includes two electrodes immersed in an electrolyte. The conducting polymer material constitutes one or both the electrodes of the cell. By applying a potential difference between them, red-ox reactions cause strongly anisotropic and reversible volume variations of the material (fig. 3) [1-5].

It has been found that the following three effects are responsible for dimensional and volume changes in conducting polymers: interactions between polymer chains, variation of the chain conformation and insertion of counterions.



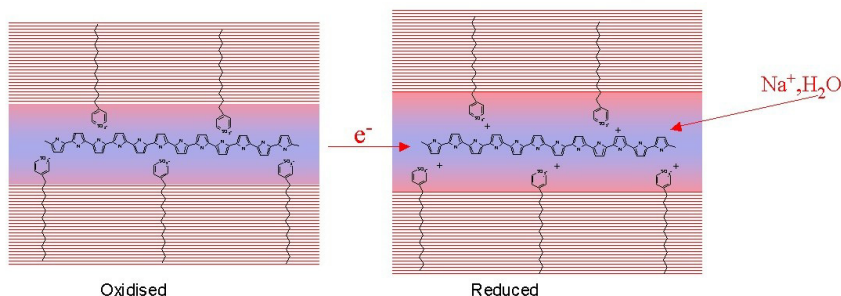


Figure 3: Working principle of conducting polymer actuators: (left) device at rest; (right) device under activation.

The third effect is generally considered to be the most dominant. In fact, the commonly accepted explanation of the observed deformations attributes the dimensional changes to the input/output of ions (exchanged with the surrounding media) into/from the polymer sample, driven by an applied voltage. In particular, the voltage produces a variation of the polymer oxidation state, causing the necessary modification of the number of ions associated to each chain, in order to maintain the global electro-neutrality.

The most diffused configuration for conducting polymer actuators is represented by the so-called unimorph bilayer bender. This kind of actuator consists of a film of active material coupled to a passive supporting layer. The bilayer structure is operated within an electrochemical cell, having a liquid electrolyte in which the device is immersed. The active polymeric layer of the actuator works as one electrode of the cell, while a counter electrode and a third reference electrode are separately immersed in the electrolyte. One end of the bilayer is constrained, while the other is free. The potential difference applied between the electrodes causes red-ox reactions of the conducting polymer. Since the CP and the passive layers are mechanically interlocked, when the polymer swells/shrinks the passive layer, which cannot modify its dimensions, transforms the CP linear displacement into a bending movement of the structure. Very similar is the bimorph structure. In this case the passive layer is substituted by a second CP film and they work in opposition of phase.

Fibre actuators made of conducting polymers have also been proposed, consisting of an extruded fibre, covered by a thin layer of solid polymer electrolyte and a counter-electrode of conducting polymer.

State-of-the-art CP devices need very low driving voltages (order of 1 V), producing strains of the order of 1-10% for linear actuators and rotations up to  $\pm 90^\circ$  for benders, with large active stresses (up to tens of MPa). Nevertheless, such interesting performances correspond to several drawbacks, such as high response times and short lifetimes, whose relevance has to be evaluated in relation to the specific application of interest [1-5].

## 5 Carbon nanotubes

Carbon nanotubes (CNT) can be described as a graphite monoatomic sheet rolled to form a tube. Carbon nanotubes have lengths about 1000 times higher than their width (typical diameters are of the order of 1 nm, while typical lengths about 1 micrometer). Moreover, they are typically combined in bundles with diameters of 10 nm. Carbon nanotubes can be divided in two classes: single-walled and multi-walled. A single-walled CNT consists of a single film rolled to make a tube, while a multi-walled CNT is made of several films rolled together. Mechanical performances of multi-walled tubes are predicted to be lower, with respect to those predicted for the single-walled ones, according to the lower forces between the layers.

CNT actuators can be realized by using sheets of single-walled nanotubes [1-4]. Their actuation properties have been demonstrated by employing electrochemical cells with at least one CNT electrode (fig. 4), which is characterised by a very high surface area. A change of the applied cell voltage results in a double-layer charge injection for this electrode, with a related deformation. The actuating principle is represented by this charge-injection, which is able to produce dimensional changes in the CNT structure.

Early investigations on CNT bending actuators showed active strains of the order of 0.2%, depending on the experimental conditions, when an applied voltage was limited to the electrochemical stability of the electrolytes ( $-1$  V to  $+1$  V, versus Saturated Calomel Electrode, for aqueous electrolytes). CNT have been used to demonstrate bending film actuators. Recently, high-quality CNT thin fibres and yarns have been demonstrated, opening new opportunities for fibre actuators [1-3].

## 6 Piezoelectric and electrostrictive polymers

Piezoelectricity is a well known effect, largely used nowadays in commercial devices based on piezoceramics. Polymer representatives of piezoelectric materials, such as PVDF, are also well studied. Unlike piezoceramics, wherein the crystal structure of the material creates the piezoelectric effect, in piezoelectric polymers the intertwined long-chain molecules attract and repel each other when an electric field is applied [1-5].

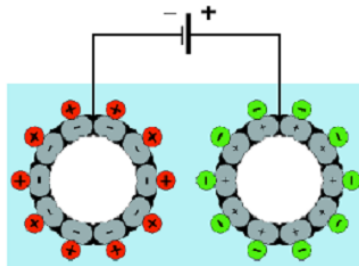


Figure 4: Use of carbon nanotubes for actuation.

Ferroelectrics are an important group of piezoelectric polymers because they exhibit a much higher piezoelectric response than the non-ferroelectric materials. Ferroelectricity, which is defined by the appearance of a switchable spontaneous polarization, has been observed in many polymers, such as PVDF, copolymers of VDF with trifluoroethylene (TrFE) or tetrafluoroethylene (TFE) and odd-numbered polyamides (nylons). Due to the appearance of ferroelectric domains, ferroelectric polymers have to be poled to obtain piezoelectricity. The poling condition, including poling electric field and temperature, is critical to the piezoelectricity of the polymer. Among all ferroelectric polymers, PVDF and P(VDF-TrFE) are the best known and exhibit the highest piezoelectric response ( $d_{33} \sim 35$  pC/N and  $d_{31} \sim 28$  pC/N,  $k_{33} \sim 0.15$ ,  $k_{31} \sim 0.15$ ,  $\epsilon_{33}/\epsilon \cong 13$ ,  $Y = 2.5$  GPa,  $\nu = 0.39$  and  $\rho = 1.78\text{--}1.80$  g/cm<sup>3</sup>) at room temperature. Although the physical mechanism is different, so-called electrets exhibit an apparent piezoelectric response. The piezoelectric constant of some electrets can be comparable to ceramics. In fact, a very high piezoelectric response ( $d_{33}$  up to 1000 pC/N,  $Y = 1\text{--}10$  MPa) has been reported in these porous polymers, in which each air bubble can be charged by discharging air, which makes an air bubble a dipole with a very large dipole moment. More interestingly, these porous electrets can exhibit an apparent ferroelectricity due to the charging and discharging in the air bubbles in the porous polymer; they are, therefore, named ferroelectrets.

Different from the piezoelectric effect, the electrostrictive effect occurs in all substances, whether crystalline or amorphous, solid or liquid. The electrostrictive strain response can be written as:

$$x_{ij} = Q_{ijkl} P_k P_l = M_{ijkl} E_k E_l \quad (1)$$

where  $P$  is the electric polarization,  $Q_{ijkl}$  is the charge-related electrostrictive coefficient,  $E$  is the electric field and  $M_{ijkl}$  is the field-related electrostrictive coefficient. Both  $Q_{ijkl}$  and  $M_{ijkl}$  are fourth rank tensors.

For an isotropic polymer under an electric field along the direction 3 (i.e.  $E = E_3$ ) with a polarization response  $P$  along the direction 3 (i.e.  $P = P_3$ ) we have:

$$x_3 = Q_{33} P^2 = M_{33} E^2 \quad \text{and} \quad x_1 = Q_{13} P^2 = M_{13} E^2 \quad (2)$$

where  $x_3$  is the strain along the  $P$  direction, known as the longitudinal strain, and  $x_1$  is the strain perpendicular to the  $P$  direction, known as the transverse strain. That is, different from the linear relationship between the field-induced strain and the electric field observed in piezoelectric polymers, the relationship in the electrostrictive material is quadratic, as shown in fig. 5.

Newly developed electrostrictive polymers exhibit an electrostrictive strain of more than 5% [1-4].

It is experimentally and theoretically proven that  $Q_{33} (M_{33}) < 0$  and  $Q_{13} (M_{13}) > 0$  for an isotropic polymer and that  $Q_{33} (M_{33}) > 0$  and  $Q_{13} (M_{13}) < 0$  for an isotropic crystal or ceramic. Hence, an increase in  $E$  or  $P$  will result in a contraction of polymers, but an extension of crystals/ceramics, along the polarization direction.



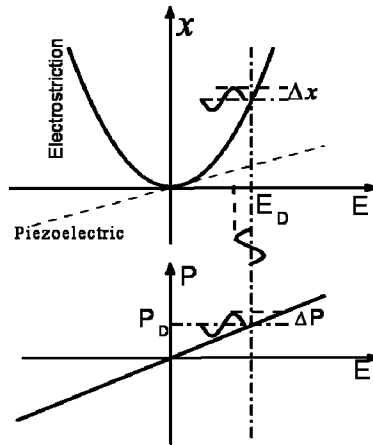


Figure 5: Schematic view of strain response in piezoelectric and electrostrictive polymers.

Different electrostrictive polymers have been developed in the last decades. One of the most studied materials with a high strain response is the high-energy-electron irradiated P(VDF-TrFE) copolymer. It is found that the irradiation converts the polymer from a normal piezoelectric to an electrostrictive state. The  $Q_{33}$  of the irradiated P(VDF-TrFE) ranges from  $-4$  to  $-15 \text{ m}^2/\text{C}$  (the  $M$  is greater than  $2 \times 10^{-18} \text{ m}^2/\text{V}^2$ ), depending on the processing conditions. Additionally, irradiated copolymers exhibit a high dielectric permittivity ( $\epsilon/\epsilon_0 \sim 60$ ) at room temperature. It is also found that the irradiated copolymers are actually relaxor ferroelectrics. Similar results were also observed in the high-energy-proton irradiated P(VDF-TrFE) ( $M_{33} = -1.83 \times 10^{-18} \text{ m}^2/\text{V}^2$ ). By varying the film processing conditions, transverse strain in the irradiated P(VDF-TrFE) copolymer can be tuned over a big range. For instance, the transverse strain response in unstretched films is relatively small ( $\sim +1\%$  at  $\sim 100 \text{ MV/m}$ ) with an amplitude ratio of the transverse to longitudinal strain of less than 0.33, while the transverse strain in stretched films reaches  $+4.5\%$  along the stretching direction [1-5].

## 7 Dielectric elastomers

Macromolecular actuators made of dielectric elastomers (DE) are stimulating a growing interest in the EAP field, due to their excellent electromechanical properties [1-6]. These materials consist of dielectric polymers with a low elastic modulus, which can present significant electrically-induced strains. In particular, a dielectric elastomer actuator consists of a thin layer of an insulating rubber-like material sandwiched between two compliant electrodes (e.g. made of carbon conductive grease), which are electrically charged by a high voltage difference. Following the electrical activation, the material undergoes an electric field-sustained deformation at constant volume, consisting of a thickness squeezing and a related surface expansion (fig. 6).

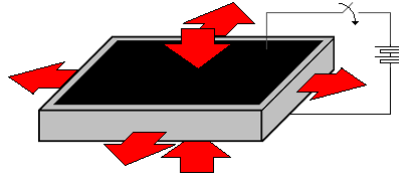


Figure 6: Working principle of a dielectric elastomer planar actuator.

This deformation is mainly due to a Coulombian effect, arising from the electrostatic interactions among the electrode free charges. The stress of the Coulomb force acting between the electrode free charges is responsible for the so-called Maxwell stress for this electromechanical phenomenon. This kind of stress acts in any kind of dielectric material subjected to an applied electric field. However, the corresponding deformations are emphasized by the eventual compliance of the electrodes, as well as by the polymer softness. These key-features basically distinguish actuating devices made of dielectric elastomers from those based on different electric-field-driven dielectrics, such as piezoelectric or electrostrictive materials.

Thickness strains  $S$  can be analytically described, by assuming that the dielectric elastomer is a linearly elastic body, with a Young's modulus  $Y$  and a relative dielectric constant  $\varepsilon_r$ , as follows

$$S = -\frac{1}{Y} \varepsilon_0 \varepsilon_r E^2 \quad (3)$$

where  $\varepsilon_0 = 8.85 \times 10^{-12}$  F/m is the dielectric permittivity of vacuum.

This equation shows that such materials exhibit a quadratic dependence of the strain on the applied field, as it happens for electrostrictive polymers. However, in comparison with these polymers, dielectric elastomers are capable of significantly larger deformations, even though at reduced forces, according to the lower elastic modulus.

Acrylic and silicone rubbers are the most significant types of the dielectric elastomers used for actuation. Such kinds of polymers comprehend representative materials which can be very compliant, being able of showing the highest actuating deformations among all EAP. High-level actuation capabilities have been reported for certain types of acrylic polymers (or acrylates): thickness strains up to 60-70% at 400 V/ $\mu$ m, area strains up to 200% at 200 V/ $\mu$ m and corresponding stresses of some MPa. Such performances are enabled by low elastic moduli and high dielectric strengths (dielectric breakdown can occur at electric fields up to about 500 V/ $\mu$ m). The highest active performances have been achieved by prestretching the material: this operation was demonstrated to increase the dielectric strength, allowing higher electric fields to be applied.

Beyond acrylates, silicones (mainly poly-dimethylsiloxanes) offer attracting characteristics: they are easily processable (by spin coating, casting, etc.) and permit the realization of rubber-like dielectrics with suitable elastic properties,

arising from the flexibility of the material molecular chains. Certain silicone elastomers have been actuated with electric fields up to 100 - 350 V/ $\mu\text{m}$ , enabling thickness strains up to 40-50% and area strains up to 100%, with related stresses of 0.3-0.4 MPa.

Owing to the excellent figures of merit shown by several dielectric elastomers (high actuation strains, considerable stresses, very fast response speeds, high efficiency, stability, reliability and durability), this class of EAP is considered today one of the most outstanding for polymer actuation. Nevertheless, some drawbacks still affect this performing technology. The most significant is certainly represented by the high driving electric fields needed (order of 10-100 V/ $\mu\text{m}$ ). For a definite polymer thickness, such field levels can be reached by applying high voltages, which may be disadvantageous in several applications. In order to reduce such driving fields, polymers with unusually high dielectric constants would be advantageous.

Several configurations for DE actuators have been proposed and demonstrated, including planar, tube, roll, extender, diaphragm, bimorph and unimorph bender. Linear (i.e. working along a line) actuators can be obtained in the forms of tubes and rolls (which elongate upon activation), as well as helical structures, multilayer stacks and folded devices (which contract upon activation). These and many other configurations are currently being used to develop a huge number of applications of this performing technology [1-6].

## 8 Final remarks and conclusions

This paper has briefly highlighted key issues related to the development of electroactive polymer actuators. According to their structure, polymerase gels and ionic polymer metal composites typically offer high strains but low stresses, while an opposite behaviour is shown by conducting polymers. These materials can advantageously be driven by low voltages, although they are limited by a low response speed, due to an ionic diffusion control, and poor efficiency and durability, due to underlying electrochemistry. Similarly, carbon nanotubes present low strains while interesting potential forces, even though their technological development is not mature. On the other hand, piezoelectric and electrostrictive polymers show small strains at high forces and large bandwidth, while dielectric elastomers offer a unique combination of attractive electromechanical properties, although at the cost of high driving fields. As such, each EAP category shows a different potential in specific areas of use.

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