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Dielectric Elastomers

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Electroactive Polymers: Developments of and Perspectives for Dielectric Elastomers

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In memory of Werner Jenninger and Karsten Dierksen

compliant electrodes · dielectric elastomers · electroactive polymers · polyurethanes · silicones

We present the development and applications of dielectric elastomers. For the last 10 years the significance of this class of polymers has risen as more applications seem possible and first products have been commercialized.

1. Introduction

The emergence in the early 1990s of synthetic “electroactive polymers” (EAPs) that react to electrical stimulation with large changes in shape has inspired the creativity of many scientists and engineers and has widened the already broad spectrum of applications for polymer materials. Because the mode of action has some similarities with biological muscles, the term “artificial muscles” has entered the literature.[1–4] These soft, robust polymer materials typically provide larger strains than conventional piezoceramics, albeit at more modest stresses, making them better suited for many applications.[1]

The term EAP refers to a broad family of polymers. In general EAPs are classified as “ionic” or “electronic,” with different modes of action.[4–7] Ionic EAPs consist of materials such as polymer gels, conducting polymers, carbon nanotubes, and ionic polymer-metal composites, for which the mode of action involves electrically driven diffusion of ions within the bulk of the material. In contrast, electronic EAPs are bulk insulators that respond to surface charges carried by conductive electrodes patterned on them. The charges apply Coulomb forces to the materials that stress and strain them. Electronic EAPs are divided into electrostrictive elastomers, ferroelectric polymers, and dielectric elastomers (DE). All these EAP polymers offer advantages and disadvantages suiting them to different applications. Among them, dielectric elastomers offer good performance in an unusually broad range of applications covering actuators, generators, and sensors.[3] Accordingly this Minireview focuses on the dielectric elastomers.

2. Basic Principles of Dielectric Elastomers

The dielectric-elastomer actuation principle was first noted by Wilhelm Conrad Röntgen in 1880, when high voltage corona discharge produced airborne ions that settled on opposite sides of a rubber band and the band stretched in response.[8] As illustrated in Figure 1, a DE device comprises at least two compliant electrodes separated by a dielectric elastomer. On activation, an electric field is imposed between the electrodes which draws them together and causes the dielectric elastomer to expand in area. Dielectric elastomers are characterized by quick response, large actuation force, high mechanical energy density, and the ability to hold an induced displacement at constant voltage without consuming

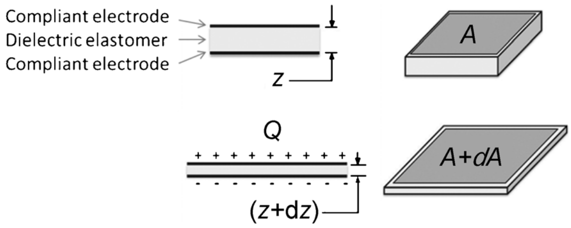

Figure 1. Dielectric elastomer actuation principle. Top: A DE transducer is a flexible capacitor with a pair of compliant electrodes sandwiching a dielectric elastomer film. Bottom: An electric field distributes charge Q and draws the electrodes together which causes the dielectric elastomer film to decrease in thickness z and increase in area A.

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electrical energy. Unfortunately, these materials work best at electric fields close to the breakdown voltage.[7]

A two-plate capacitor arrangement is used to make a dielectric elastomer (DE) convert between electrical and mechanical energy. The action is similar to that of a parallel-plate micro-electromechanical system (MEMS) electrostatic actuator but with two key differences. First, the solid DE typically withstands voltages that would break down an air gap, so higher forces can be developed between the capacitor plates. Second, the plates are elastic and typically patterned directly onto the DE material, which is essentially incompressible and changes shape much more readily than volume. This intimate arrangement of plates and an incompressible dielectric material imposes a geometric constraint on movement of the system. Bringing the plates together also increases their area, changing capacitance twice—once with the thickness and again with the area. Thus a DE transducer can convert energy more efficiently than an air-gap transducer. The polarizability of the dielectric, expressed as a dielectric constant ε_r, further enhances the electric energy storage. The elastic modulus Y of the dielectric governs the mechanical energy storage capacity.

The working principles of DE transducers can be derived in a number of ways.[9,10] One proposed by Pelrine et al.,[9] describes differential changes dU_e in electrical energy U_e in a plate capacitor of thickness z, and area A, that holds a charge Q across the capacitance C. Assuming constant volume ($A\,dz + z\,dA = 0$) leads to Equation (1).

$$dU_e = \left(\frac{Q}{C}\right)dQ - 2U_e\left(\frac{1}{A}\right)dA = \left(\frac{Q}{C}\right)dQ + 2U_e\left(\frac{1}{z}\right)dz \tag{1}$$

The first term on the right hand side is the change of electrical energy due to change of electrical charge in the capacitor (dQ). The second term represents mechanical work done on the electric field. For constant charge Q, any effect that increases film thickness (positive dz) converts mechanical work into electrical energy. Thus, the transducer works as a generator or sensor. Vice versa, any effect that thins the dielectric (negative dz) corresponds to conversion of electrical energy into mechanical, and the transducer works as an actuator. Equation (1) is the basis for the discussion of the fundamental operation modes at constant voltage, constant energy, or constant charge.

At constant charge, substituting the differential dz by infinitesimal mechanical work exerted on the elastomer by the conformational change dz, the above expression can be rewritten to obtain the physical dimensions of pressure, and further by introducing the energy content of the capacitor to obtain Equation (2)

$$p = \varepsilon_0 \varepsilon_r \left(\frac{V}{z}\right)^2 \tag{2}$$

In this equation, p is the Maxwell stress, ε_o and ε_r are the vacuum and the material permittivity (dielectric constant), V is the voltage applied across the thickness of the dielectric. If the DE response is linear, observing free boundary conditions and introducing Young's modulus Y, the expression for the free stroke S_z in the z direction of a single-layer DE actuator can be obtained as Equation (3).

$$S_z = \frac{1}{Y} \varepsilon_0 \varepsilon_r \left(\frac{V}{z}\right) \tag{3}$$

Using similar transformations and assumptions, governing equations can be derived for when a DE of a given geometry is used as a sensor or generator.

In the equations describing the performance of DE elements, the performance parameters are proportional to the dielectric constant and the square of the electric field in the capacitor. Increasing (V/z) and ε_r are therefore the most evident means to control the efficiency of a DE transducer device.

When deriving the energy density inside the dielectric, it is of note that for small strains S_z, the energy density in the actuator—the quantity governing the efficiency of DE generators—scales as the square of S_z and thus with the 4th power of (V/z).[4,11] On the other hand, for large strains the elastic energy density scales approximately linearly with S_z,[4,11] which enables a great variety of different operation modes for actuation and generation. For actuators with high stroke, a small Young's modulus is crucial [see Eq. (3)]. Other optimization parameters for DE materials might be mechanical properties, creep, bulk resistivity, viscoelasticity[12] to name only a few, but each of these also imposes limits on the material properties. To increase the energy density, two principal methods are open: use of high electrical fields (V/z) or an increase in the dielectric constant ε_r. A limit to increasing (V/z) is however given by the material's dielectric breakdown strength (DBS). At electrical fields higher than the DBS, electric breakdown occurs, leading to failure of the

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DE transducer. The dielectric constant can be increased by introducing polar groups which have orientational degrees of freedom into the dielectric. To avoid dielectric breakdown, these dipoles need to have low translational mobility. These requirements can be met by use of an elastomer material with a polymer backbone in which polar moieties are incorporated.

An important fundamental limit to the electroactive effect is the "stability limit" $S_z = -1/3$, which appears for materials with pure linear elasticity, that is, $Y = \text{const}$. Beyond a thickness strain of 33 % induced by the Maxwell pressure, the film becomes unstable and collapses due to electromechanical instability, also called "pull-in failure".[13]

In many cases, linear models are not adequate to describe the mechanical as well as the electrical behavior of DEs.[14] In the literature, many non-linear models for the hyperelastic behavior of DE materials are used.[11,15–17] Unlike thermoplastic materials, elastomers show nonlinear behavior of Y at high strains which may avoid the above stability limit.

3. Chemistry and Materials Development of Dielectric Elastomers

According to Equation (3) storing a high amount of electrical energy in a given volume of material requires a material with both a high dielectric constant and high DBS. A high dielectric constant can be achieved by incorporating polar groups with a Debye moment greater than zero. Electron-withdrawing ligands, such as urethane groups, have a Debye moment of 2.4; SO_2, CO_2, chlorine, trifluoromethyl, acetamido groups have values of approximately 4 Debye; nitro and cyano groups have values of approximately 6 Debye. The combination of electron-withdrawing and electron-donating groups such as found in nitroanilines results in dipoles with values of approximately 10 Debye.[18–20]

To store electrical energy for a period of time, the polymer should have a very low specific conductivity. Materials with high leakage currents from ionic impurities and other electrical loss mechanisms are less efficient at energy conversion and may suffer from premature failures because of resistive heating. High electric fields in a polymer give rise to the accumulation of polymer specific amounts of space charge which may have an influence on the electrical aging behavior of the polymer.

Because the elastic capacitor should be easily deformed, the polymer should have a low glass-transition temperature. Reversible actuation requires a high mechanical elasticity, in particular a sufficiently large strain at break and a very low stress relaxation. For devices capable of large deformation under electric load, a very low modulus is essential, but the modulus should be high enough to prevent electro-mechanical instabilities. The polymer should also be tear resistant, chemically and thermally stable.

In practical material development, in many cases correlational behavior is observed, meaning that the optimization of one parameter often implies degradation of other properties. For example, generally an increase in dielectric constant correlates with a decrease in DBS owing to increased mobile charge density. This situation is observed in investigations of "electrical aging" of polymers.[21]

Many classes of materials have been investigated for DE applications including acrylates,[4,22] silicone,[6,11] urethanes,[6,11] rubbers,[23,24] latex rubber,[6,11] acrylonitrile butadiene rubber,[25] and olefinic,[6] fluorinated,[26] and styrenic copolymers.[27,28] Some of the most studied materials are described briefly below.

3.1. Acrylates

Acrylates were among the first materials investigated for EAP devices. Commercial adhesive films from 3M, the VHB family of acrylic transfer PSA tapes in particular, have been used extensively by the EAP community (for example see References [4] and [22]). They are readily available[29] with high film quality and low cost and exhibit excellent performance under certain conditions. High performance with deformations over 50% have been reported.[27] Figure 2 shows how deformation can be easily on the order of millimeters albeit with relatively high electric fields. These data are for a 50 mm diameter device with a 250 μm VHB film biaxially prestrained to 300% and biased out of plane.

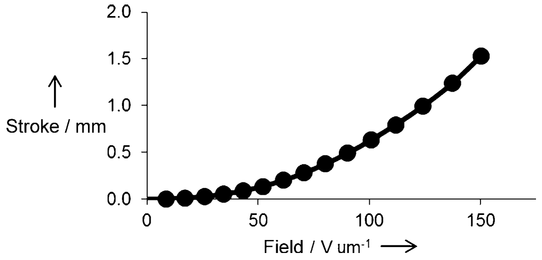

Figure 2. Actuator performance for an acrylate actuator as a function of operating field.[18]

Table 1 shows typical properties for the 3M acrylate film VHB 4910. VHB acrylates have a relatively high dielectric constant and dielectric breakdown strength (Table 1).

Table 1: Standard properties of 3M VHB 4910.[22,27,30]

Property	Value
Dielectric constant	4.7, 3.21@ 1 kHz
Dielectric breakdown strength	630 $\mathrm{V\,mm^{-1}}$ (25 $\mathrm{V\,\mu m^{-1}}$)
Volume resistivity	3×10^{15} ohm cm
Shear modulus	0.3–0.6 MPa
Young's modulus	1–2 MPa
Elongation	>600%
Long term use temperature	93 °C

As seen in Figure 3 the stress–strain curve for a 3M VHB acrylate film is very flat up to 300% strain. It shows extremely low moduli at high elongations. In contrast, self-reinforcing or strain-hardening materials, such as silicone, may show a pronounced inflection in the curves at lower strain, and the slope

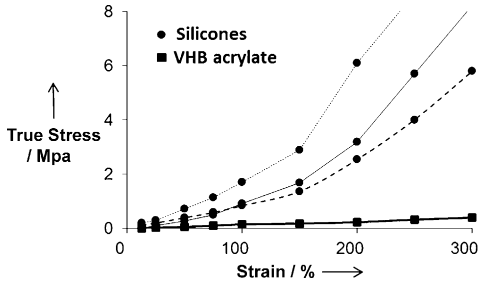

Figure 3. Tensile data of a 3M VHB acrylate adhesive film and experimental silicone films.[18]

of the silicone data after the inflection point is higher than that of the acrylate indicating a significantly higher modulus.

The advantage of this flat behavior is that the acrylates can be used at much higher prestrains, on the order of 200–300%, as compared to the silicones and other elastomers for which prestrain may be limited to less than 50%. High prestrains may enable less-expensive manufacturing processes because films with higher initial thickness can be used. Prestrain can improve performance as seen in Figure 4 for DE transducers fabricated from dielectric films that have been biaxially stretched (prestrained). In addition, the dielectric breakdown strength of acrylates can increase dramatically with prestrain, which enables the use of higher electric fields which also leads to higher performance.

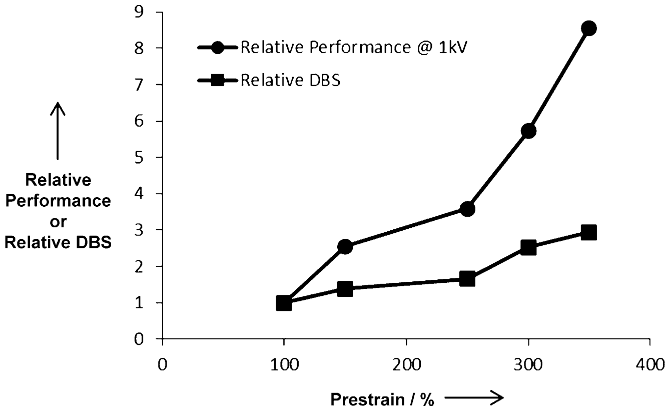

Figure 4. Effect of biaxial prestrain on the performance and dielectric breakdown strength of acrylate films; DBS = dielectric breakdown strength.[18]

For all their advantages which have contributed significantly to the development of EAP technology, acrylate films have not been successfully commercialized in EAP devices. A major reason for this is that acrylate devices may be very temperature sensitive which can limit the use temperature range significantly. As shown in Figure 5 for a 3M VHB adhesive (open circle data), performance at temperatures outside the range of 0–70 °C may be more than 25% lower than the performance at 20 °C. Motion drops to zero by −30 °C. This degradation in performance is generally reversible upon return to room temperature although there can be long-term material degradation at elevated temperatures.

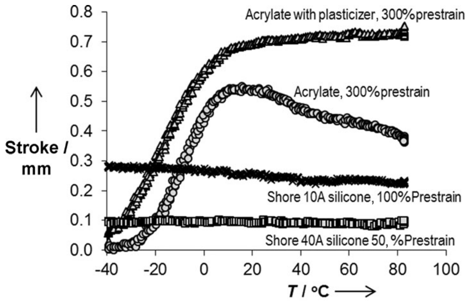

Figure 5. Performance of acrylate and silicone actuators as a function of temperature.[18]

Another issue with acrylate devices is they may show slow response times owing to the viscoelasticity of the polymer network.[31] As shown in Figure 6, an acrylate device may take a long time to reach a stable performance level; the performance of this device increases 25% over the course of a 1 s actuation.

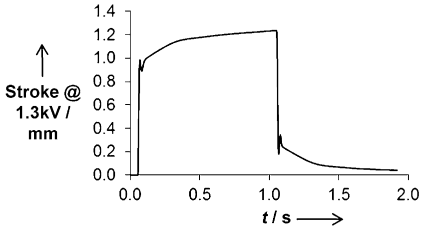

Figure 6. Acrylate actuator performance as a function of time. The actuating voltage is applied at 0.05 s and removed at 1.05 s.[18]

It takes a similar time to recover once the electric field is removed. This can severely restrict the use of these devices in applications, such as haptics or audio devices, which require high-frequency responses. Since some electrical energy is used to offset this viscous damping, the energy conversion of these materials can be low.

High viscoelasticity can also lead to a long-term relaxation of performance under constant field as the polymer chains relax over time. It has been reported that long term relaxation may also have a contribution from the high leakage current of these materials which increases power consumption and renders the material unable to hold strain with time.[32] Leakage current can increase with both temperature and humidity which can further exacerbate the environmental limitations of these devices.

Perhaps the most severe limitation of acrylate devices is their sensitivity to humidity. As stated above, the increase in leakage current which occurs with humidity decreases the charge-holding capability of the dielectric film and limits its long-term performance. However, a more critical failure mode appears at relatively modest humidity levels. Figure 7 shows the mean number of cycles to dielectric failure for acrylate devices tested at different relative humidities at 25 °C under the same actuation conditions. While no failures are observed after 25 000 cycles at 20% RH (relative humidity), mean lifetime drops rapidly above 50% RH. Thus for most

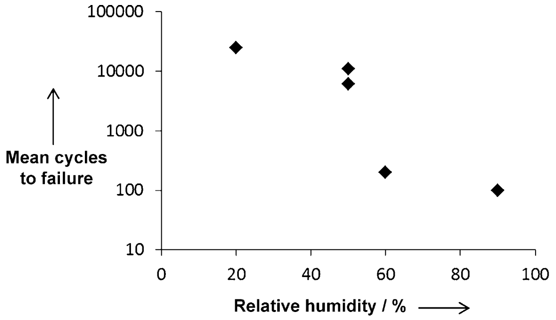

Figure 7. Effect of relative humidity on mean time to failure of acrylate actuators at room temperature.[18]

applications, acrylate devices must be operated in a humidity-controlled environment.

Much work has been done to improve the properties of acrylate EAP devices. Plasticizers can be incorporated or imbibed into acrylate films to widen the use temperature range of an acrylate EAP device as shown above in Figure 5.[27] This improvement is particularly important for applications where low-temperature operation is desired. The plasticizer can decrease the modulus and glass-transition temperature of the polymer network without sacrificing elasticity. However, plasticizers can also volatilize or migrate which can limit the lifetime or application of the device.

To prevent the limitations of plasticizer migration and to improve the mechanical stability of an acrylate film, reactive plasticizers and monomers can be introduced. These can either be grafted onto the existing acrylate network or allowed to react together in a separate network to form an interpenetrating network (IPN).[33,34] The use of IPNs can reduce the viscoelastic limitations of standard acrylate films such as creep and hysteresis. With the appropriate choice of additives, performance can be improved through higher dielectric constants and lower modulus.

Work has also been done to replace the proprietary acrylate used in 3M VHB films (e.g. References [33,35–37]). Acrylates are generally prepared by the radical polymerization of vinyl monomers as shown in Figure 8.

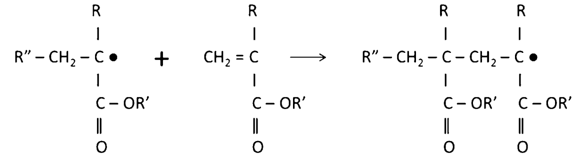

Figure 8. Radical polymerization of acrylate monomers.

Acrylate chemistry is very rich with many monomers and multifunctional cross-linkers available. Since many of these are miscible, it is possible to tailor polymers and copolymers with a wide range of properties. There are also many manufacturing options since films can be prepared by solvent casting, thermal and/or photo-polymerization. Lombardi and Gasper provide an excellent introduction to acrylate polymers.[38]

Another approach to improving performance has been to use fillers, such as Janus particles,[39] alumina,[40] and titania,[41] which can increase the effective dielectric constant or improve creep resistance. However, in many cases, the modulus increases and the elongation and tear resistance decrease which may prevent the use of these composite materials in many applications.

3.2. Silicones

Silicones have also long been studied by EAP researchers.[6] They are the basis of the commercial products from Bayer MaterialScience[42] and the published devices under development by several other companies, such as Danfoss, CTS, and Optotune. Unlike acrylates where work has focused on 3M VHB adhesive films, researchers have cast their own films from many different commercial materials including: Nusil CF19-2186,[11] Dow Corning Sylgard186,[6,11] Dow Corning HS IV,[6] Dow Corning HS III,[6,11] and Dow Corning DC3481.[30,43] Dielectric constants are generally about 3; modulus ranges from 0.1 to 1 MPa. In general, dielectric breakdown strength rises with the modulus.[30]

In most cases, vinyl-addition cured silicones have been used to produce the films. In these materials, a silyl hydride is added across a vinyl group, generally in the presence of a platinum-based catalyst (Figure 9).

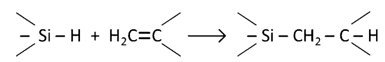

Figure 9. Silylhydration reaction.

The advantage of silylhydration systems is that they can be solventless and produce no byproducts upon reaction. Both solvents and byproducts can create defects as they are removed from the film during the coating process. These defects can lead to dielectric failures and greatly restrict the operating fields of the EAP device.

Material properties of silicones are more apt for DE transducer requirements. Electrical resistivity is on the order of 10^{13} ohm m and leakage currents can be an order of magnitude lower than those of acrylates. They can be highly elastic as seen in the narrower hysteresis loop for silicone in Figure 10. Elasticity is also evident in Figure 11 which shows

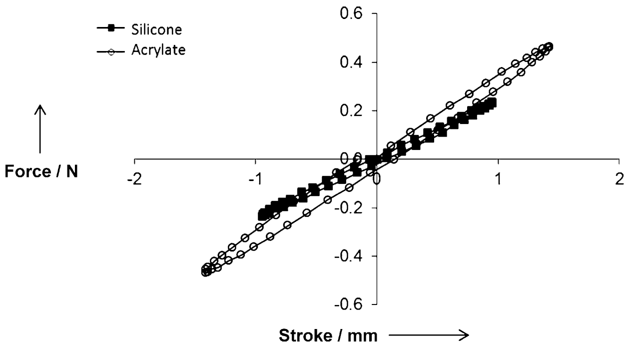

Figure 10. Operating cycle for EAP actuator where the operating voltage is applied and then released. The area within the loop is related to the hysteresis and indicative of the viscoelasticity of the material.[18]

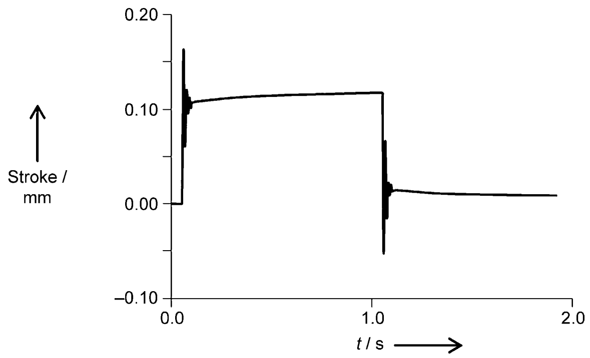

Figure 11. Performance of silicone actuator as a function of time. Operating voltage is applied at 0.05 s and removed at 1.05 s.[18]

very fast response times, and little change in the response with time.

Silicones have a broad temperature range with a response that can be constant from −40 °C to 80 °C although the response is in general lower than can be obtained with the acrylate films (Figure 5). Silicone-based EAP devices are less dependent on environmental conditions and show considerably better long-term reliability performance than acrylate devices. Devices have been tested to nearly 1 billion cycles under ambient conditions.[18] The mean time to failure can be in the 10 million to 100s of millions of cycles, even at high humidity. For example, the mean time to failure at 65 °C/85 % RH was over 20 million cycles for a set of silicone devices that had already experienced 7.6 million cycles at 25 °C/70 % RH.[18]

Despite their advantages, silicones have deficits that have impeded their application. These nonpolar polymers generally have lower dielectric constants than the acrylates and polyurethanes and thus lower actuator performance. Performance is also limited by constraints on the prestrain that can be used. As shown in Figure 3, many silicones show self-reinforcing behavior where the tensile stress and modulus (the slope of the stress–strain curve) rise quickly above 50–150 % strain. This behavior can obviate the advantages of increased prestrain as seen in Figure 12 where no performance gain is observed when the prestrain is increased from 50 % to 75 % for devices fabricated from the same base film and run at the same operating voltages. The low prestrain values in Figure 12 can also lead to manufacturing issues since thinner films must be fabricated and processed to reduce operating voltages.

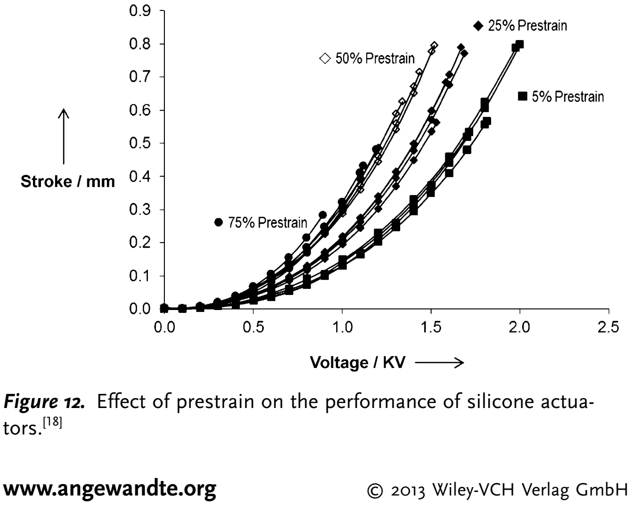

Figure 12. Effect of prestrain on the performance of silicone actuators.[18]

There are other manufacturing issues related to the use of silicone films. Their excellent chemical stability and low surface energy can make it difficult to coat them onto or get them to adhere to other materials that may be required in the final device. They are also significantly more expensive than most organic materials, such as acrylates, olefins, and polyurethanes.

While there is not as much latitude in modifying the chemistry of silicones as there is with acrylates or polyurethanes, there have been a number of approaches to improve the performance of silicone EAP devices. In general, the silicon atoms are disubstituted with methyl groups. Poly(dimethylsiloxane) can be copolymerized or blended with poly(diphenylsiloxane) to increase the dielectric constant. These materials are not fully compatible at some molecular weights and phase separation may occur. Fluorosilicones have also been investigated which can increase the dielectric constant more than a factor of 2.[6,22] However, these compounds often exhibit high leakage current. They are also very expensive and restricted in their adhesive properties which render commercial adoption more difficult.

The dielectric constant of silicones can also be modified by grafting additives onto functional groups in the silicone network. For example, it has been found that the addition of fluorinated allyl compounds to residual Si−H groups in a silicone matrix could double the dielectric constant although at some cost to the DBS and mechanical strength.[18] Similar results have been reported by a number of groups such as Böse et al.[19] and Kussmaul[20] and co-workers who showed significant increases in dielectric constant and EAP performance by grafting allyl compounds with strong dipoles, such as aniline and cyano groups. In addition to their potentially adverse effect on the resistivity and DBS of dielectric materials, highly polar groups may also render them more susceptible to moisture uptake which can significantly reduce device lifetimes at higher relative humidities.[18]

Composites and blends have also been investigated by many groups. Some examples include high dielectric-constant additives, such as polyethylene glycol,[44] phthalocyanine oligomers,[43] metallic nanoparticles,[45] graphenes,[45] titania,[46] calcium copper titanate,[47] and barium titanate.[48] In all cases, the increased dielectric constant needs to be optimized against changes in the mechanical and electrical properties of the mixtures.

3.3. Polyurethanes

In great contrast to the strictly chemically cross-linked elastomers discussed in the preceding Sections, polyurethanes represent a unique class of elastomers, ranging from physically cross-linked thermoplastic polyurethanes (TPU) to physically and chemically cross-linked systems specifically developed for DE applications.

Polyurethanes (PUs) are generally polymerized by the reaction of isocyanates with alcohols as shown in Figure 13.

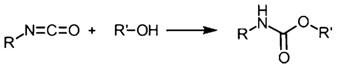

Figure 13. Condensation reaction to form a urethane linkage.

Figure 14. Left: Biuret group (based on HDI; HDI = 1,6-hexamethylene diisocyanate) $\mu = 3.3$. Right: Urethane (based on HDI) $\mu = 2.4$ Debye.[18]

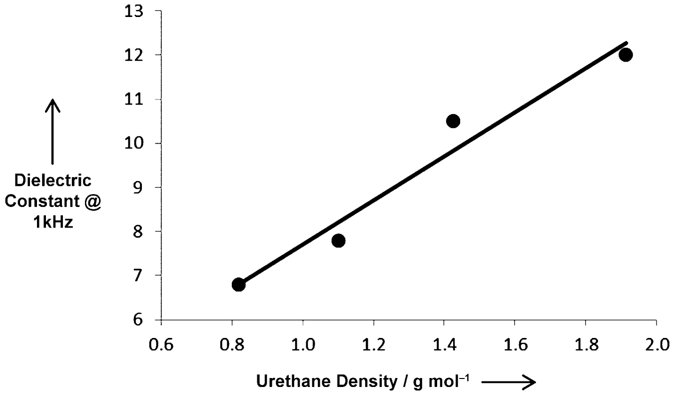

Figure 15. Influence of the microstructure of a homologous series of polyurethanes on the dielectric constant ε_r.[18]

There can be significant hydrogen-bonding and/or crystallization of the polyurethane groups which may segregate into regions of high modulus ("hard segments") to create a physically cross-linked network. This occurrence is the basis of thermoplastic polyurethanes (TPUs) which can be processed as polymer fluids at high temperatures but behave as elastomers upon cooling. In addition mixtures of multifunctional prepolymers can be used to tailor a chemically cross-linked network architecture.

The combination of chemical (covalent) and physical (van der Waals) cross-links in the network, with alternating structures of hard and soft segments, as well as the variety of suitable monomers ((poly)isocyanates, polyols, (poly)amines) enables almost unlimited possibilities to tailor the polymer backbone.[49] The controlled chemical structure of the network distinguishes polyurethanes from other elastomer materials giving them consistent mechanical properties over a broad temperature range.

Most elastomers have a low dielectric constant on the level of an insulator (2.5–4) and thus need additives to increase their dielectric constant. This results in a disorder of their polymer structure and in most cases a dramatic reduction of the DBS. Polyurethanes have an intrinsically high dielectric constant (above 7), which is based on the polar nature of the polyurethane fragments. These fragments may be separated into the hard segments, such as the urethane group or related modifications as biuret, trimer or allophanate, and soft segments mainly based on polyetherpolyol (C2–C4) or polyester (aliphatic or aromatic) building blocks.[50] The fragments can have significantly different dipole moments, as seen in Figure 14, which will impact the dielectric constant of the polyurethane material.

The influence of the PU microstructure on the dielectric properties is significant as seen in Figure 15 which shows a linear relationship between urethane density and dielectric constant.[18] Through careful choice of raw materials, a high dielectric constant can be achieved without influencing the elastomeric polymer properties.

Additional polarity may be incorporated into the soft segments, similar to the reported methods for silicone with nitro, nitrile, and fluorinated groups.[51,52] However, this often leads to inferior electrical and mechanical properties. It has been shown that such polar building blocks may result in significantly reduced electrical resistivity, up to two orders lower compared to silicones, or a decrease in DBS.[18]

Another key advantage of polyurethanes beside the high intrinsic dielectric constant is the easy processability of PU formulations which can be made into thin films by nearly all the methods known in the plastic industry.[49] Casting of 2 component systems in a roll-to-roll process allows the production of thin films (down to 5 μm) in high quality with high DBS.

In Table 2 the properties of a cast polyurethane Bayfol EA 102 which was especially adapted to the needs of DE application is compared to those of a standard extruded thermoplastic polyurethane (TPU), a typical silicone, an acrylate (3M VHB 4905), and a fluoropolymer (Solvay Solvene 250). It can be seen that the low concentration of chemical cross-links in the TPU and in the acrylate (evidenced by its extremely low modulus) result in a high level of creep or permanent deformation of these materials under actuation strain.

Table 2: Properties of several DE films.[18,53,54]

Elastomer material	Film thickness [μ]	Strain at break [%]	Young's modulus at 50% [MPa]	Creep [%]	ε_r at 1/8 Hz	DBS [V μm^{-1}]
Polyurethane TPU LPT 4210 UT50	50	421	3.36	24	6.0	218
Polyurethane Bayfol EA102	50	300	1.44	2.9	7.1	130
Silicone proprietary formulation	45	422	0.25	16	2.4	80
Acrylate 3M VHB 4905	498	879	0.04	70	4.5	31
Fluoropolymer Solvene 250	300	525	800		14	250

There are still a few drawbacks impeding the use of polyurethanes in DE applications. The high polarity of polyurethanes can lead to a slightly increased sensitivity towards humidity compared to silicones. Also, most commercially available materials exhibit higher Young modulus. Some systems based on polyether polyols suffer from low electrical resistivity while others (mainly polyester based) have too high glass-transition temperatures. However, recent results indicate that polyurethane formulations suitable for DE applications are achievable.[18]

The combination of intrinsic positive properties and the huge versatility of this chemistry make the polyurethanes a very suitable candidate for DE development. The high number of combinations to create a perfect polymer architecture gives the materials scientist almost unlimited options to modify the polyurethane chemistry although there is still a huge challenge to find the right electromechanical properties.

3.4. Fluoropolymers

Fluoroelastomers, in particular copolymers of poly(vinylidene fluoride) with trifluoroethylene, tetrafluoroethylene, and/or esafluoropropylene, have been investigated for use in EAP devices.[26] Solvay Solexis has commercialized these systems under the tradename Solvene[53,54] and Strategic Polymer Systems is developing devices using these materials.

Like acrylates, fluropolymers are generally synthesized by radical polymerization. However, whereas acrylates are often processed from neat monomer systems, fluoropolymers are usually prepared from emulsion or suspension polymerization. Copolymers have been developed to reduce crystallinity and improve the elastomeric properties. They also enable lower temperature melt-processing or solution processing in many cases. Ameduri provides a good Review on fluoropolymer chemistry in Reference [55].

Fluoroelastomers are characterized by very high dielectric constants (ca. 15), dielectric breakdown strengths greater than 200 Vμm^{-1}, excellent temperature stability, electrical resistivity, chemical stability, and toughness.[53] Very thin films can be formed by solvent casting to enable the use of lower operating voltages. However, the modulus of these materials is very high, on the order of 0.5–10 GPa, and performance may be low unless the applied electrical fields are extremely high, for example, 1% strain at 100 Vμm^{-1}. Also, these materials are generally semicrystalline so devices may suffer from creep and relaxation issues. Finally, as with most fluorinated polymers, the cost of these compounds is high and special corrosion-resistant equipment may be necessary to process them.

Work to mitigate the deficits of these materials has included optimization of the copolymer formulations and incorporation of high dielectric constant compounds, such as conductive polymers (e.g. polyaniline (PANI))[56] and oligomers of copper phthalocyanine.[57] The copper work reports dielectric constants above 200 and a strain of 2% at 13 Vμm^{-1}. Analysis indicates that the results may not be due solely to Maxwellian forces responsible for the properties of the dielectric elastomers such as the acrylates and silicones discussed above. Instead, there may be contributions arising from a non-uniform electric-field distribution caused by the heterogeneous composite structure of these blends. The semicrystalline matrix material itself has a heterogeneous morphology and in addition is expected to have an electrostrictive response as a result of the polarization of the polymer chains.

4. Compliant Electrodes

Together with the dielectric membrane, electrodes play a key role in enabling the widespread use of DEs for actuators, sensors, and generators. The electrodes must adhere to the elastomer membrane and “follow” its deformation without constraining it either mechanically or electrically, that is, they should be able to sustain large deformations without losing conductivity. For this reason these electrodes are often defined as compliant. Ideally, compliant electrodes should fulfill the following requirements:

1) be conductive (sheet resistance under 1 kOhm sq^{-1}, depending on application)
2) sustain large deformations while remaining conductive (10–100% depending on application)
3) adhere well to the dielectric membrane
4) be softer than the elastomer (Young Modulus under 1 MPa)
5) be easily patterned
6) not alter the membrane’s properties
7) have low mechanical and electrical losses
8) be self-clearing
9) sustain millions of cycles without loss of performance.

Herein we will highlight only the most significant approaches for compliant electrodes reported to date. For more exhaustive information the reader should refer to the recent Review by Rosset and Shea and references cited therein.[58]

Carbon-based electrodes (usually based on carbon black and graphite) are the most widely used, mainly because of their low cost, low impact on stiffness and ease of fabrication. They are generally used as loose particles (carbon powder), dispersed into a viscous media (carbon grease), or dispersed into an elastomer (conductive rubber). Among these, the conductive rubber has some interesting advantages over the other two approaches, the most valuable being its robustness, low mechanical loss, and good adhesion to the dielectric membrane. High-aspect-ratio carbon and other conductive particles are particularly effective for electrode formulations.[59] The concentration of fibers and flakes needed to achieve a given degree of conductivity is much lower than that of low-aspect-ratio particles. The mechanical properties of the electrode composite can thus better reflect the properties of the matrix material. A detailed comparative study of several carbon-based electrodes was performed by Carpi et al. in 2003.[60]

A common alternative to carbon-based electrodes are metallic thin-films, deposited by standard processes, such as electron-beam evaporation and sputtering. These electrodes have the advantage of being thin, patternable, and highly conductive. The low thickness may give them the advantage of self-clearing or fault-tolerance.[61] During self-clearing, the electrode is burnt off locally where an electrical short thorough the dielectric film takes place, effectively insulating the short-circuited zone. Clearly, self-clearing electrodes can significantly improve the lifetime of EAP actuators and are therefore very attractive.

The use of thin-film metal as compliant electrodes is not straightforward however, since they are barely stretchable (2–3%) and are generally very stiff (Young's Modulus in the range of GPa). To overcome these limitations, the most successful solutions rely on corrugated or patterned electrodes.[59] Corrugated electrodes can be fabricated by deposition of a thin metal layer on a pre-corrugated membrane or by out-of-plane buckling of the electrode as a consequence of the relaxation of a pre-strained or pre-heated membrane. A successful approach developed by Benslimane et al. is based on fabricating a corrugated membrane by applying a silicone elastomer onto a mold with a sinusoidal or periodic profile.[62] Subsequently, a thin film of silver is deposited on top of the corrugated surface of the elastomer membrane. For the final capacitor configuration, two of these films are then laminated together with their flat sides in contact. In this way, elongations up to 80% can be obtained without cracking of the electrode. The main constraint of this approach is the unidirectionality of the actuation direction which may limit adoption in some applications.

A related approach consists in out-of-plane buckled electrodes which are usually formed by compressive stress of a pre-heated[63] (or pre-stretched)[64] dielectric membrane; once coated with a metallic thin film, the membrane is allowed to cool down (or relax), resulting in a contraction which compresses the electrode and forces it to buckle out-of-plane.[61] Electrodes fabricated with this method can undergo bi-axial strain; limited strains up to 28% have been reported.[64] Similar results can be obtained with patterned electrodes that are carefully designed to allow a certain degree of stretchability. Several designs (e.g. zigzags and horseshoes) have been investigated and strains up to 100% without loss in conductivity have been reported.[65] Most of these electrodes are also limited to unidirectional deformation.

In addition to the aforementioned methods for compliant electrodes, other more unusual techniques have been described, including carbon nanotubes (CNTs),[66–68] silver-based electrodes,[69,70] Pt salt reduction,[71] conductive PANI nanofibers,[72] and the implantation of metallic nanoclusters.[73,74]

Yuan et al. have shown that single-walled CNTs as well as conductive PANI nanofibers are capable of forming highly compliant electrodes with a fault-tolerant (or self-clearing) behavior.[68,75,76] PANI electrodes may, however, de-dope under high electric fields, which may limit their long-term lifetime and application.[18]

The approach to compliant electrodes followed by Rosset et al. is based on the implantation of metallic nanoclusters in the first tens of nanometers below the elastomer surface.[73,74] By this method, the electrode's compliance as well as its adhesion and robustness are highly improved compared to a plain thin film. In addition, metal-nanocluster electrodes have no restriction on the strain direction.

Delille et al. have developed an electrode based on a Pt salt dispersed into a photosensitive elastomer matrix.[71] This electrode is photopatterned with UV light and then immersed in a reducing agent which reduces the Pt salt on the surface to metallic Pt. They reported of strains up to 150% before losing conductivity, however, the resolution of this patterning method still needs to be improved.

The recent advances on stretchable electrodes have been a key enabler for improving the performances of EAP compliant electrodes and some products are already on the market. Nonetheless, particular efforts need to be focused on improving both the compliance and the conductivity of electrodes, not just through the electrode composition but also through optimization of the fabrication process. To illustrate the impact of electrode resistance on the time response of EAP actuators, the strokes of two actuators made with standard-resistance and high-resistance electrodes have been plotted in Figure 16.

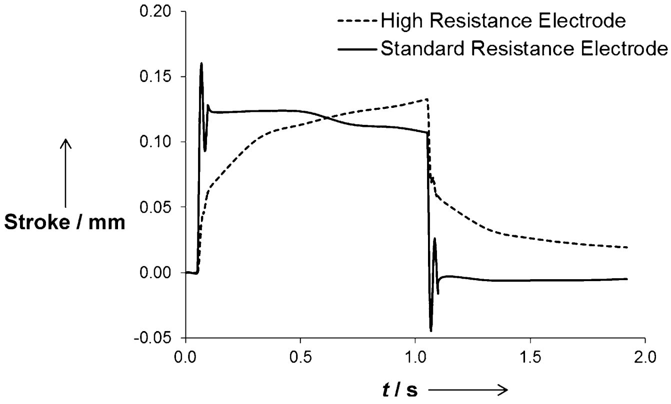

Figure 16. Effect of electrode resistance on the time response of EAP actuators.[18]

High-resistance electrodes can limit the kinetics of charging the electrodes when imposing the operating voltage and increase the time required to observe full motion and recovery of the actuator. Electrode resistance can be reduced by improved formulations and also often by increasing the thickness of the deposited electrode. However, thicker electrodes can significantly decrease device performance, as seen in Figure 17.

This loss of performance becomes more pronounced as the dielectric film thickness is decreased to enable the use of lower operating voltages. Improving the electrode's intrinsic properties and enabling the use of thinner electrodes (under 300 nm) will help reduce the impact on stiffness and allow the possibility of a beneficial self-clearing behavior.

Lastly, the main requirements for compliant electrodes may differ depending on the final application. In addition to actuators (discussed above), electroactive polymers can be used as sensors and as generators for energy-harvesting devices. For sensors, a good electrode would mostly need

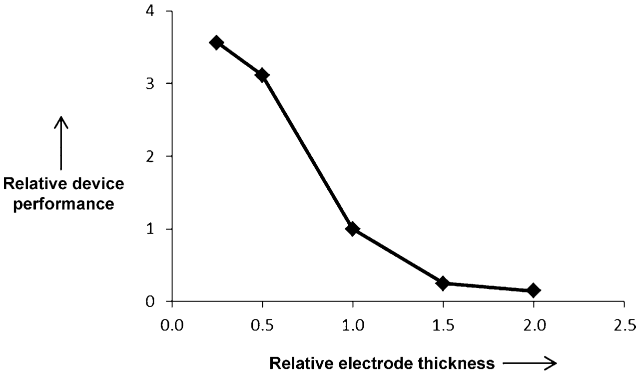

Figure 17. Effect of electrode thickness on performance.[18]

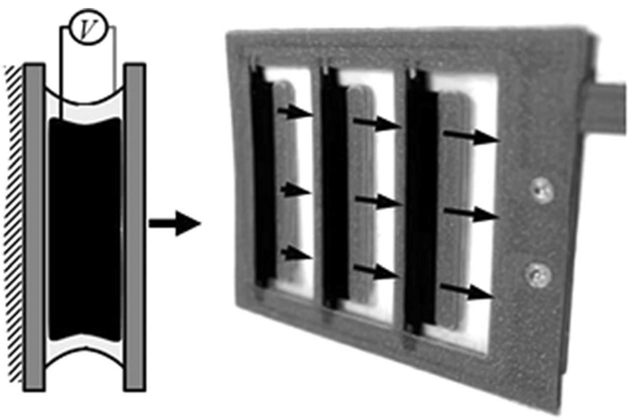

Figure 18. Two membrane actuators, the "trench" (left) and "bar array" (right).

a well-controlled surface resistance, limited degradation upon cycles, and low hysteresis of resistance versus strain. This implies that, for example, conductive rubber should be preferred over carbon powders, carbon grease, or metallic thin-film electrodes. On the other hand, for energy-harvesting purposes, compliant electrodes need to sustain very large elongations (over 50%) and millions of cycles while having high conductivity to minimize Joule losses. To date, the most successful approach for large-scale energy-harvesting devices has been the use of metallic thin films on corrugated membranes, as shown by the company SBM Offshore.[77]

5. Mechanical Design of Transducers

In this Minireview it is only possible to list some basic designs for DE transducers. Detailed designs and frameworks for analysis can be found elsewhere.[2,78,79]

5.1. Actuator Mode

DE actuators can be roughly classified as membranes, stacks, and rolls.

5.1.1. Membranes

The "trench" configuration (Figure 18, left) is the most basic membrane actuator. Charging an electrode-covered area (Figure 18, black region), stresses and thins the membrane, which increases the separation of the bars at the edges. It is convenient for laboratory testing, but the free elastomer edges are vulnerable to tearing. A commercially produced "bar array" (Figure 18, right) stabilizes the edges with a frame.[18] Each output bar adjacent to the active regions can be coupled to a suspended load such as an inertial mass which moves when the actuator is activated. Multiple bars are bundled together for additional force, which scales with the total length of the bar edges.[80] It is a thin design and can be readily manufactured by the methods of printed electronics.

From this starting point follow a range of alternatives. For example, when the suspended load is a central disk it can be ringed with electrode segments that provide bi-axial planar movement. Biasing the central disk out of plane produces the "Universal Muscle Actuator", useful for pumps and valves because the membrane centers the disk, reducing the need for additional mechanical guiding components.[81]

Other membrane actuators are produced by lamination to an inextensible substrate. The unimorph and bimorph geometries with a dielectric layer laminated to either one or both sides of the substrate, respectively, provide large out-of-plane bending movements with low force. High fields on a laminated membrane can produce "crease-to-crater" actuation that spontaneously pits the membrane surface to dislodge biofouling.[82] Alternately, when the substrate is provided with fluid-filled pockets, then "zipper mode" actuation can bow the membrane into the pockets, pumping fluid at modest pressure.[83] When the substrate has round apertures and the membrane is biased with pressure, it creates the "buckled diaphragm" configuration, useful for tactile displays,[84] braille,[85] and liquid lenses.[86]

5.1.2. Stacks

One method to efficiently build a higher force device in a small volume is to stack many layers. In these designs the load is effectively coupled to the face rather than the edge of the electroded membranes, so force scales with the stack area. Several methods of alternating layers of electrode and dielectric have been used to make stacked actuators including folding,[87] layering elastomer films between perforated metal electrodes,[88] cutting and stacking electroded films,[89] and alternately coating dielectric and patterning electrodes.[90] Arrays of actuators can be created to make tactile displays and peristaltic pumps. Higher-displacement, lower-force output is provided by the addition of soft passive slabs, which are also thinned upon activation and act as a solid-state transmission.[91]

5.1.3. Rolls

Rolls are another method for packing many dielectric and electrode layers into a small volume.[92] In some designs the films are rolled to form a hollow core;[93] in others they are

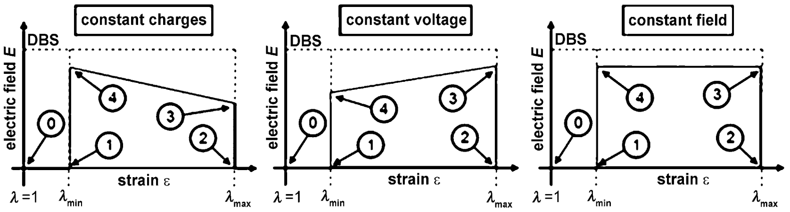

Figure 19. Three generator work cycles.[96]

wrapped around a bias spring.[94] Rolls are typically used in tension or compression along the core axis.

5.2. Generator Mode

Dielectric elastomer devices act as generators as well as actuators. In these devices, mechanical energy stored by stretching or deforming an elastomeric film is converted into electrical power through increased charge density on the surfaces of the film when the film returns to its undeformed state. Generators have been built at a variety of length scales ranging from wearable energy scavengers[95] to large tubular wave-energy converters.[77] Many different generator work cycles[96] are possible within the energetic limits of the material,[97] however, the cost and complexity of high-voltage electronics typically favors the "constant charge" cycle (Figure 19). In autonomous applications, self-priming electronics can work charge up to high voltage to provide the "seed" that begins the work cycle.[98]

5.3. Sensor Mode

As sensors, dielectric elastomer transducers allow for high strains and flexible, non-planar configurations.[99] They are more sensitive and nonlinear than air-gap capacitive sensors. Since displacements that thin the dielectric by a factor $\lambda = z/z_o$ simultaneously enlarge the area of the capacitor plates, and since both effects increase capacitance, the capacitance scales with an inverse square law, as $C = C_0(1/\lambda^2)$. When it is important to keep the number of leads to a minimum, the capacitance of a dielectric elastomer transducer can be sensed through the same leads that actuate it,[100] facilitating closed-loop control.

6. Application Areas

DEs can be used for a large variety of applications that use small electrical motors, actuators, force and strain sensors, and electrical-energy generators or even a combination of these. In the 1990s, EAP research activities were mainly funded by military institutions. Applications such as small robots for intelligence operations or small portable energy harvesters (e.g. heel strike generators[4]) were the focus.

EAP-based small electrical motors allow lighter, quieter, more energy efficient, fast responding, and better controllable systems than the traditional electromagnetic motors. The soft, flexible character of an EAP film allows completely new form factors and configurations of motors and actuators.

EAP actuators were first commercialized in 2011 and provided high-definition haptic feedback in a computer gaming application.[42] The application exploited the small form factor, energy efficiency, large frequency range and, most importantly, fast response time of EAP actuators. Other applications which are close to commercialization include optical positioning systems, pumps, valves, and disposable sensors. Solenoids in positioners and valves powered by DE actuators allow a reduction in energy consumption by up to 80% and fewer moving parts to reduce operating and maintenance costs. Optical applications, such as aperture and lens positioning actuators, especially for small mobile devices, allow the combination of small form factor, precise displacement, and energy efficiency.

Bayer MaterialScience has developed EAP actuators that enhance the sound dimensions of headphones for a more realistic audio experience. This same application may evolve towards a small form factor hearing aid. In the future, more powerful versions of these actuators might drive prosthetic limbs.[101] The industry is just beginning to leverage the potential of new, disruptive actuator designs.

In the next 3–5 years the advantages of EAP actuators are to be used in low displacement, low force, lightweight, energy conscious, and noiseless applications. As the EAP material and actuator design evolve, the technology will penetrate more demanding applications and thereby increase its share in the growing actuator market.

EAP sensors allow large-scale and high-precision strain and force sensors at a fraction of the cost of today's products. They are to be introduced into the market for medical and industrial applications where precise sensing and disposable sensors are often an unmet need for applications.

EAP based generators are being explored as a promising option to generate energy from ocean waves. Early results indicate the promise of this concept.[77]

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