

**Increasing the performance of dielectric elastomer actuators:  
a review from the materials perspective**

L.J. Romasanta, M. A. Lopez-Manchado and R. Verdejo\*

Instituto de Ciencia y Tecnología de Polímeros, ICTP-CSIC, Juan de la Cierva, 3 28006 Madrid, Spain

**Abstract**

Electro-active polymers (EAP) are emerging as feasible materials to mimic muscle-like actuation. Among EAPs, dielectric elastomer (DE) devices are soft or flexible capacitors, composed of a thin elastomeric membrane sandwiched between two compliant electrodes, that are able to transduce electrical to mechanical energy, actuators, and vice versa, generators. Initial studies concentrated mainly on dielectric elastomer actuators (DEAs) and identified the electro-mechanical principles and material requirements for an optimal performance. Those requirements include the need for polymers with high dielectric permittivity and stretchability and low dielectric loss and viscoelastic damping. Hence, attaining elastomeric materials with those features is the focus of current research developments. This review provides a systematic overview of such research, highlighting the advances, challenges and future applications of DEAs.

**Keywords:** dielectric elastomers, actuators, artificial muscle, dielectric permittivity

\*Corresponding author:

E-mail: rverdejo@ictp.csic.es; Tel.: +34 91 258 7633; Fax: +34 91 564 4853

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## Nomenclature

A	Area	ETAS	Ethyltriacetoxysilane
C	Capacitance	EVA	Ethylene-vinyl acetate
e	Energy density	FGS	Functionalised graphene sheets
E	Electric field or nominal electric field	HNBR	Hydrogenated acrylonitrile butadiene rubber
$M_w$	Molecular weight	HPU	Hyperbranched polyurethane
p	Electrostatic pressure	IIR	Polyisobutylene-co-isoprene
$\tan(\delta)$	Dielectric loss factor or loss tangent	IPGs	Ionic polymer gels
$T_c$	Curie temperature	IPMCs	Ionic polymer–metal composites
U	Electrostatic energy	IPN	Interpenetrating polymer network
V	Voltage	LCEs	Liquid-crystal elastomers
Y	Young's modulus	LDPE	Low density polyethylene
z	Film thickness	MWCNTs	Multiwall carbon nanotubes
$\epsilon_0$	Vacuum permittivity	NBR	Acrylonitrile butadiene rubber
$\epsilon$	Dielectric permittivity	NR	Natural rubber
$\epsilon'$	Real part of the complex dielectric permittivity, dielectric permittivity	PANI	Polyaniline
$\lambda$	Pre-stretch ratio	PDA	Polydopamine
BN	Boron nitride	PDMS	Poly(dimethylsiloxane)
BR	Butadiene rubber	PHT	Poly(3-hexylthiophene)
BT	Barium titanate $BaTiO_3$	PMMA	Poly(methylmethacrylate)
CB	Carbon black	PMN-PT	Lead magnesium niobate–lead titanate
CCTO	Calcium copper titanate $CaCu_3Ti_4O_{12}$	PSF	Polysulfone
CNTs	Carbon Nanotubes	PTFE	Poly(tetrafluoroethylene)
CPs	Conducting polymers	P(TrFE)	Poly(trifluoroethylene)
CR	Chloroprene rubber	PU	Polyurethane
CuPc	Copper-phthalocyanine	PVDF	Poly(vinylidene fluoride)
DBSA	Dodecyl benzene sulfonic acid	P(VDF-TrFE)	Poly(vinylidene fluoride-trifluoroethylene)
DE	Dielectric elastomer	PZT	Lead zirconate titanate
DEAs	Dielectric elastomer actuators	SBR	Styrene-butadiene rubber
DEGs	Dielectric elastomer generators	SEBS	Styrene-ethylene-butadiene-styrene rubber
DOP	Diethyl phthalate	SEBS-g-MA	Styrene-ethylene-butylene-styrene-grafted maleic anhydride
EAPs	Electro-active polymers	SWCNTs	Single wall carbon nanotubes
EGEs	Electrostrictive graft elastomers	TEOS	Tetraethoxysilane
EPR	Ethylene-propylene rubber		

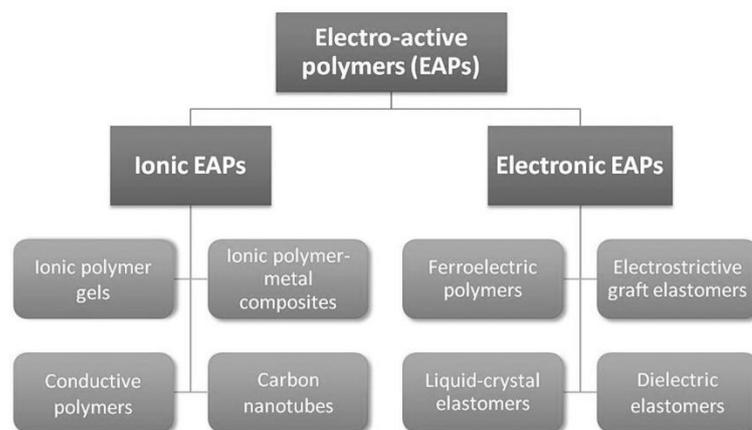
## 1. Introduction

Electro-active polymers (EAPs) that respond to an electrical field by changing their shape have the potential to mimic muscle-like behaviour enabling the development of lightweight, energy efficient and silent actuators, motors and force and strain sensors [1, 2]. Among the different EAPs, soft dielectric elastomers (DEs) have attracted much interest in recent years due to their outstanding deformation strains, in some cases greater than 100%. Such interest has not been restricted to the academic world and a number of enterprises [3-5] are already commercialising devices based on this technology.

The first investigations on the effect of electrical field on solids go back to the late 18<sup>th</sup> century when Italian scientist Alessandro Volta mentioned in a letter [6] that researcher Felice Fontana had observed volume changes in Leyden jars (the first electrical capacitors) [7, 8]. Subsequent work on a pre-stretched natural rubber band by Willem Röntgen [9], best known for the discovery of X-Rays that earned him the Nobel prize in 1901, showed length changes of several centimetres. Since then, further isolated work can be found on the strain response of dielectric materials to applied fields. However, it was not until 2000 when researchers from Stanford Research Institute reported for the first time large strains in dielectric elastomer in a paper published in Science [10]. Since that pivotal work, DEs are recognised to provide the best combination of electrical and mechanical properties for true muscle-like actuation with a work density of 10 to 150 kJ/m<sup>3</sup> for silicone and acrylic elastomers, respectively, compared to 40 kJ/m<sup>3</sup> in mammalian skeletal muscle [11]. The initial research in this field focused on identifying the appropriate DEs working conditions, designs and configurations. Over the last years, an increasing number of research groups have started to develop new formulations for DEs and a large body of research is currently available. This review provides a systematic overview of this recent research, highlighting the advances, challenges and future applications of DEs.

## 2. Electro-active polymers (EAPs)

EAPs encompass a large family of polymers that are broadly categorised into two major groups based on their method of actuation (Figure 1): materials which are driven by the mobility or the diffusion of ions- known as ionic EAPs-, and materials whose mechanical response is driven by electric fields or Coulomb forces- known as electronic EAPs [12]. This section provides a brief overview of the materials used as EAPs. A more detailed description can be found in the edited book by Yoseph Bar-Cohen [13].



**Figure 1.** Classification of electro-active polymers (EAPs).

### 2.1 Ionic EAPs

a) Ionic polymer gels (IPGs) consist of a cross-linked polymer network, typically a polyacrylic acid gel, with an electrolyte solution filling its interstitial spaces [14]. Generally, this type of materials can respond to different external stimuli; among them, the most commonly used is the pH. This change of pH is induced by a chemical process but can also be induced by an external electric field through an electrolytic reaction [15-17]. The main issues of these systems are their slow actuation rates and the need of an encapsulating material for the electrolyte.

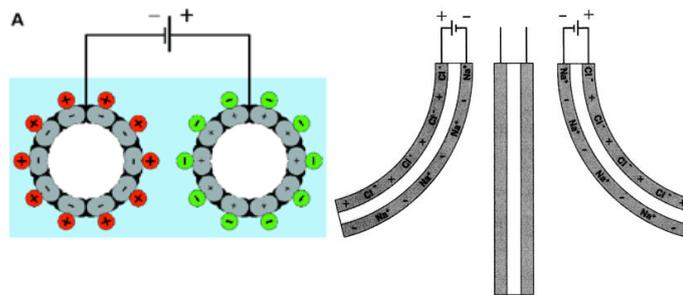
b) Ionic polymer-metal composites (IPMCs) consist of a solvent swollen ion-exchange polymer membrane (generally Nafion<sup>®</sup> or Flemion<sup>®</sup> from Dupont<sup>™</sup>) whose planar surfaces

are coated with a thin flexible layer of metallic particles (typically platinum or gold nanoparticles) [18]. The application of low driving voltages (1-5 volts) produces the migration of the ions within the film to the oppositely charged electrode. This causes, in turn, one side of the membrane to swell whereas the other side experiences contraction, thus resulting in a bending actuation. Although these systems have the advantage of requiring low voltages, their actuation strains are usually below 3% [19] that together with their hydration requirements represent their main drawbacks. Nevertheless, a promising development of a triple-layered Nafion composite membrane has recently shown that this configuration, as compared with traditional single-layered Nafion IPMCs, can remarkably enhance the bending rates, exhibiting 42% higher tip displacements at the maximum deflections [20].

c) Conducting polymers (CPs) are organic polymers, generally composed by C, H and simple heteroatoms, such as N and S, where the unique intrinsic conductive properties arise from the  $\pi$ -conjugation. To observe conductivity in this type of polymers, the polymeric backbone must be oxidised or reduced by a number of anionic or cationic species, called dopants, to introduce charge centres [21, 22]. Although CPs were discovered in 1977 [23], it was not until 1990 when the first actuators based on CPs were proposed by Baughman et al. [24]. Electro-mechanical actuation of CPs (generally, polypyrrole [25-27] and polyaniline [28, 29] is based on the dimensional changes that result from the uptake of counter-ions during electrochemical redox cycling, which implies that most CPs based actuators must be encapsulated due to the necessity of an electrolyte [30]. The actuation voltages for this type of actuators are typically low (on the order of volts) [30] while actuation strains can range from 1 to 40% [31, 32]. Future work on these materials is envisioned through the development of solid electrolytes, which could eliminate the need for their encapsulation.

d) Carbon nanotubes (CNTs) are allotropes of carbon with a cylindrical nanostructure. The application of a bias on single-walled carbon nanotubes suspended in an electrolyte

suspension, causes the electrolyte ions to migrate to the surface of the CNTs (Figure 2) [33]. This surface charge accumulation is compensated by a rearrangement in the electric charge within the tube. These effects together with the Coulombic forces produce the CNT actuation. CNTs present several positive characteristics for their use as electro-mechanical transducers, such as low actuation voltages (on the order of few volts), high operating stress (26 MPa) high effective power to mass ratios (270 W/kg), and fast response speed (on the order of milliseconds) [34, 35]. However, their applicability is limited based on their modest actuation strains (usually below 2%), cost and health and safety concerns over the use of CNTs.



**Figure 2.** Schematic illustrations of a) nanotube-based electromechanical actuator immersed in an electrolyte (background) and b) cantilever-based actuator operated in aqueous NaCl, which consists of two strips of SWCNTs (shaded) that are laminated together with an intermediate layer of double-sided Scotch tape (white) [33]. Reprinted with permission from American Association for the Advancement of Science, copyright 1999.

## 2.2 Electronic EAPs

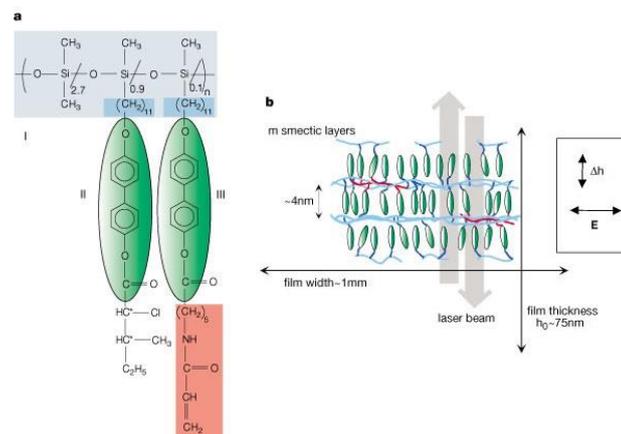
a) Ferroelectric polymers (FPs) are generally semicrystalline polymers with polar side groups. Thus, the application of an external electric field causes the polarisation of the dipoles and their alignment in the direction of the applied field. This field-induced polarisation is permanent and can only be removed by either applying a reverse electric field or by heating the polymer above the Curie Temperature ( $T_c$ ). Above the  $T_c$ , polymers exhibit paraelectric

behaviour and the dipoles do not align. This transition from ferroelectric to paraelectric is usually above room temperature and involves phase transformations that promote changes in the lattice constant accompanied by large strains [36, 37]. Poly(vinylidene fluoride) (PVDF) [38] is the classic example of this type of materials but other polymers, such as poly(trifluoroethylene) (P(TrFE)) [39] and certain odd-numbered nylons [40, 41], have also shown to possess ferroelectric properties. The main drawbacks of these materials are the high  $T_c$  and the characteristic hysteresis, present in any ferroelectric material (permanent polarization denoted in the plots of electric displacement  $D$  vs. electric field  $E$ ), which hinders a controlled response. A strategy to mitigate these two shortfalls have been the development of copolymers, such as P(VDF-TrFE). These copolymers reduce the size of the crystallites and, thus, lower the energy required for the transition between the paraelectric and ferroelectric state and their hysteresis [42].

b) Electrostrictive graft elastomers (EGEs) consist of a flexible non-crystallisable macromolecule chain with a grafted polar polymer that is able to crystallise. Thus, the application of an external electric field induces the reorientation of the polar crystallites, which results in bulk deformation of the materials. This field-induced orientation is maintained until the electric field is removed. This type of materials offer very low strain values (around 4%) but relatively high energy densities ( $\approx 247$  J/kg) [43, 44].

c) Liquid-crystal elastomers (LCEs) essentially consist of liquid crystal mesogenic groups attached to one another via a cross-linked elastic polymer. Thus, these materials combine the molecular mobility of liquid crystals with the elastic properties of elastomers. In absence of an external stimulus, the orientational range order of the mesogens forces the elastomeric backbone to be elongated along the direction of orientation of the mesogens (nematic phase) [45-47]. The application of a thermal [48, 49] or an electrical [50, 51] external stimulus produces the loss of the nematic order, allowing the elastomeric backbone to relax to its

coiled conformation (isotropic phase). Electrically activated LCEs (Figure 3) have shown faster response speed [51] than thermally activated while the required electric fields (1.5-25 V/ $\mu\text{m}$ ) are lower than for most of the EAPs. Their initial limitation is that they present very modest actuation strains, usually below 10%, which means that additional improvements in both strain and work density are needed for their application as actuators [37]. Future developments could come from targeting these materials for highly specialised movements where there is no need for large actuations and where other the properties of liquid crystals, like their known electro-optic or electro-chromic capabilities, are desired. A recent study has shown one such application by the development of an iris like LCE [52].



**Figure 3.** Schematic illustrations of an electrically activated LCE. a) Chemical structure of the sample, where each part has been colour coded, and b) measurement geometry using an interferometer to measure the electrically induced thickness modulation [51]. Reprinted by permission from Macmillan Publishers Ltd: [NATURE], copyright (2001).

d) Dielectric elastomers (DEs) are essentially compliant capacitors, where the dielectric medium is an elastomeric thin film with low elastic stiffness coated on both sides by compliant electrodes. Unlike their inorganic counterparts (e.g. shape-memory alloys [53], and piezoelectric ceramics [54]) DEs can induce several levels of strains (from 10 to 300%) but

they require large electric fields (usually around 100 V/ $\mu\text{m}$ ) [12]. In addition, DEs are relatively inexpensive, lightweight, mechanically robust, and scalable and can afford high energy densities ( $>8 \text{ MJ/m}^3$ ), high electro-mechanical coupling efficiencies ( $>90\%$ ), and low cycling hysteresis [36]. Their actuation mechanism is mainly related to the Maxwell stress effect, which is a consequence of a change in the electric field distribution inside the dielectric material. Nevertheless, some dielectric elastomers exhibit electrostrictive actuation together with Maxwell stress due to the presence of some crystallinity in their structure [55, 56]. This contribution can be easily checked by the increment in the dielectric permittivity of the elastomer when pre-strained [57, 58]. A detailed analysis of the current strategies to improve DEs performances is discussed below in section 3.

An alternative subdivision of electronic EAPs can be made based on their actuation mechanism: electrostriction and Maxwell stress effect. Therefore, the electro-mechanical response of a given polymer can be explained by either one of them or by a mixture of both. The typical materials of these actuation mechanisms are PVDF or liquid crystal elastomers for electrostriction and silicones for Maxwell stress. Meanwhile, polyurethanes and other thermoplastic elastomers like SEBS show both contributions.

### **3. Electro-mechanical transduction in dielectric elastomers (DEs)**

A feature of DEs is that they can not only convert electrical into mechanical energy, actuators (DEAs), but they can also transduce mechanical into electrical energy, generators (DEGs) [59]. The basic transduction principle of DEs is based on the electrical energy stored in capacitors,  $U$ , arising from the materials' capacitance,  $C$ , given by

$$U = \frac{1}{2} CV^2 \quad (1)$$

$$C = \varepsilon_0 \varepsilon_r \frac{A}{z} \quad (2)$$

where  $V$  is the voltage in the condenser,  $\epsilon_0$  is the permittivity of free space,  $\epsilon_r$  is the relative permittivity and  $A$  and  $z$  are the area and electrode distance, or thickness of the film membrane. Initial derivation of the fundamental equations was described in terms of the electrostatic Maxwell effect as an externally applied force to a linear elastic material [59, 60]. Further refinements of this initial model have later considered non-linear elastic materials [61, 62] or non-linear electro-mechanical field theory [63], among others. These refinements are mostly used in modelling oriented studies and the initial model in material oriented studies. Hence, we will provide a brief description of the main equations arising from this simplified model.

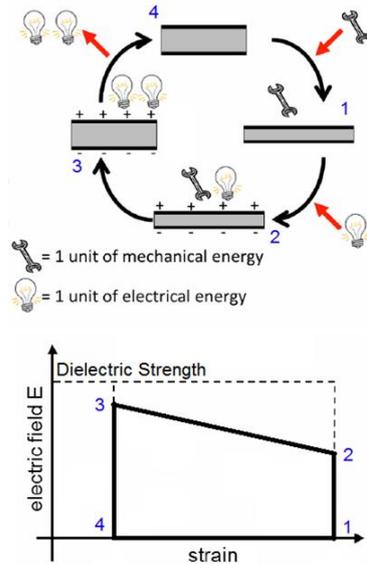
### 3.1 Working principles and equations of DE generators (DEGs)

In this mode, an input of mechanical energy (generally strain) plus a small electrical charge results in an overall energy gain as the DE relaxes. DEGs can be operated in three different energy harvesting cycles [64-66], constant charge, constant voltage, or constant field, being the first cycle the easiest to implement (Figure 4). This constant charge cycle presents four basic steps: first, an external source stretches the DE film (Figure 4.1), then, a voltage or electrical charge is applied to the surface of the DE film (Figure 4.2). At this point, the DE film is allowed to relax to a state of lower stretch (Figure 4.3), thus reducing its area while increasing its thickness. These geometric effects increase the electrical energy on the DE film, which is then removed from the DE film (Figure 4.4).

The basic equation of the generator mode is given by the energy harvested per cycle,  $e_h$ , and is calculated from the energy difference in the film between the stretched ( $s$ ) and relaxed ( $u$ ) states:

$$e_h = \frac{1}{2} C_u V_u^2 - \frac{1}{2} C_s V_s^2 = \frac{1}{2} C_s V_b^2 \left( \frac{C_s}{C_u} - 1 \right) \quad (3)$$

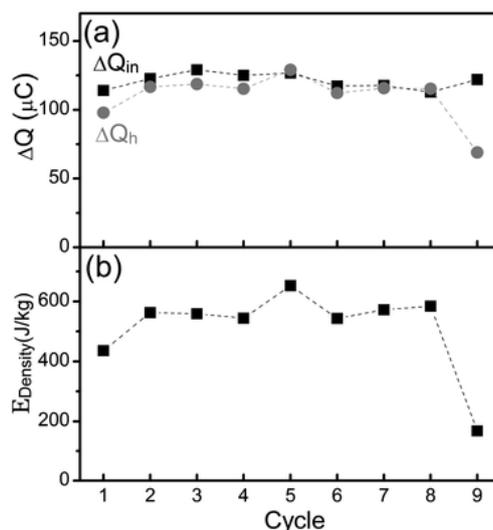
where  $V_b$  is the bias voltage.



**Figure 4.** Schema of the working principles of DEs in generator mode. Adapted and reprinted from [67] with permission from the authors.

The studies reported so far on DEGs from a materials perspective have mainly built a fundamental understanding on the experimental schemes to measure the energy harvesting while measuring commercially available polymer matrix, such as acrylic (3M VHB 4905 and 4910) [65, 66, 68-73], natural rubber (NR) [66], PU [74] and poly(dimethylsiloxane) (PDMS) [64] films. These studies have shown that the main parameters affecting the energy conversion are both experimental factors, such as the charging voltage, thickness of the film membrane, and the applied deformation, and material factors, such as the dielectric constant of the elastomer and its effective stiffness. The maximal specific electrical energies generated per cycle have theoretically been predicted to be 1300 to 1700 J/kg for NR and acrylic elastomers [66], respectively, that correspond to the maximum strain possible. However, this maximum strain is not normally applied to maximise the durability and reliability of the films [66, 68]. Hence, the reported energy densities are much lower than the theoretical predictions. So far, the reported values are normally around 100 J/kg to 300 J/kg for acrylic systems, and have recently reached 560 J/kg with a bi-axially stretched VHB 4905 film (Figure 5) [68].

However, this study also showed that the high viscous losses of acrylic films lowered the energy harvesting density, to less than 200 J/kg, and the conversion efficiency, to less than 10%, after the 8<sup>th</sup> cycle. Meanwhile, a comparative theoretical study between acrylic and NR films concluded that NR outperformed acrylics at low strains as DEG but it also required five times more stress to stretch [66].

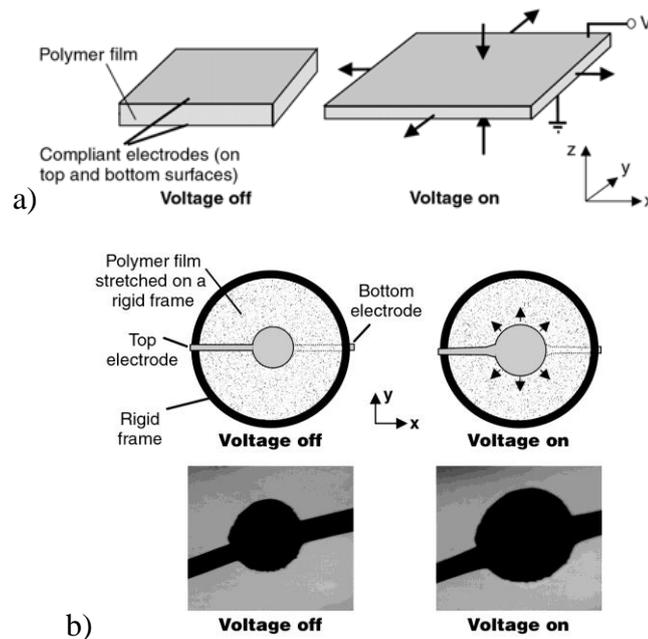


**Figure 5.** a) The input charge drawn from the power supply,  $\Delta Q_{in}$ , the harvested charge,  $\Delta Q_h$ , and b) the energy density,  $E_{Density}$ , in each cycle of a VHB acrylic film membrane [68]. Reprinted with permission from WILEY-VCH Verlag GmbH & Co, Copyright (2013).

Considering current energy harvesting solutions, DEs are able to convert 10 times more energy than electrostrictive polymers and 100 times more than piezoelectric composites [75]. The potential of DEs as energy harvesters is considerable and further research is needed on material development and device configuration to ensure high efficiency and reliability over a range of environmental conditions [65]. Most of the recent work on this area has so far focused on device configuration; among them, a new harvesting scheme has been proposed to optimise the energy density by controlling the rate of charge transport in an electromechanical cycle. Using such configuration, the authors reported an energy density of 780 J/kg and conversion efficiency of 30% with a commercial acrylic membrane 3M VHB 4905 [76].

### 3.2 Working principles and equations of DE actuators

The work principle in actuator mode is essentially the reverse of the generator mode. Here when an external electric field is applied across the electrodes, the electrostatic attraction between the opposite charges of both electrodes generate an electrostatic stress or pressure on the film, causing it to contract in thickness while expanding in area [10] (Figure 6).



**Figure 6.** Schema of a) the working principles of DEs in actuator mode and b) a circular-strain experimental set up where the photo shows 68% area expansion during actuation of a silicone film [10]. Reprinted with permission from American Association for the Advancement of Science, copyright 2000.

The fundamental equation that characterises DEAs is the electrostatic pressure, also known as Maxwell stress,  $p$ , generated upon application of a voltage between the electrodes, which is defined as the change in electrostatic energy,  $U$ , per unit area,  $A$ , per unit displacement of the film in the thickness direction,  $z$ , (the negative sign indicates that the generated stress is compressive):

$$p = \left(\frac{1}{A}\right) \left(-\frac{dU}{dz}\right) \quad (4)$$

Considering elastomers are essentially incompressible, so any decrease in thickness should result in a concomitant increase in the planar area, the electrostatic pressure exerted on a DE film subject to an electric field is expressed as follows:

$$p = \varepsilon_0 \varepsilon_r \left(\frac{V}{z}\right)^2 = \varepsilon_0 \varepsilon_r E^2 \quad (5)$$

Applying Hooke's law to an ideal elastomer characterised by an anisotropic Young's modulus  $Y$  independent of strain and a Poisson coefficient  $\nu=0.5$ , the compressive stress (or electrostatic pressure) can be related to the elastic compressive strain,  $s_z$ , as follows:

$$s_z = -\frac{p}{Y} = -\frac{\varepsilon_0 \varepsilon_r E^2}{Y} = -\frac{\varepsilon_0 \varepsilon_r}{Y} \left(\frac{V}{z}\right)^2 \quad (6)$$

Although this last equation is derived from a simplified case that assumes linear elasticity and small strains, it summarises the effect of the electrical and mechanical properties of the polymer membrane on the performance of DEA devices. An additional requirement of DEAs is that they should be able to withstand large voltages, in order for the actuation to be as large as possible. Hence, they should have high dielectric strength and low dielectric losses, to maximise the stored energy and efficiency, and avoid premature failures. The following sections describe, first, the commercial and unmodified materials tested as DEAs and their main properties (Table 1), and, second, the pursued strategies to enhance the electrical and mechanical properties to achieve an optimised material for DEAs.

#### 4. Elastomeric matrices for DEA devices

The two main elastomeric matrices initially identified to provide a balanced set of electro-mechanical properties were acrylic elastomers and silicones. Commercial acrylic films, in particular VHB series from 3M Corporation, are made of mixtures of aliphatic acrylate, which are based on the structure of a vinyl group and a carboxylic acid terminus. They possess

relatively high dielectric constant value ( $\epsilon' = 4.5 - 4.8$  at 1 kHz) and a theoretical energy density value of  $3.4 \text{ MJ/m}^3$ . They display an excellent electro-mechanical performance in terms of the actuation strain, in excess of 380% reported for highly pre-strained films. However, they also suffer from important viscoelastic effects, which result in long-term relaxations and slow response times, and are very sensitive to humidity and temperature, with an optimum operation temperature at around  $20^\circ\text{C}$  for un-modified acrylates [77]. Poly(dimethylsiloxane) (PDMS), commonly known as silicone, is a mixed inorganic–organic polymer based on silicon and oxygen backbone (Si–O), which provides low elasticity modulus and high mechanical dissipation factors. Silicones possess relatively low dielectric constant and energy density ( $\epsilon' = 2.5 - 3.0$  at 1 kHz, and  $e = 0.75 \text{ MJ/m}^3$ , respectively) and modest electro-mechanical actuation strain (about 120% of linear strain) when compared with acrylic elastomers. Nevertheless, they have the advantage of a lower viscoelasticity, resulting in a faster response time and lower losses. Moreover, they also exhibit the advantage of a wide temperature range (from  $-100$  to  $260^\circ\text{C}$ ) and have low rates of moisture absorption, which have resulted in their use in most commercial products.

In any case, the major advantage of DE technology is the wide range of materials that can be used since, in theory, any elastomer or soft thermoplastic may act as the dielectric layer. Indeed, a large number of elastomer materials have been tested for DE devices, including PU, isoprene or NR, fluoroelastomers and even thermoplastic copolymers [36, 37]. A comparison of the main properties of the DE materials was compiled by Brochu and Pei [37] in 2010. Here, such table has been updated and is summarised in Table 1.

**Table 1.** Updated comparison of the properties of the materials used for DEAs. Adapted from [37] with permission from WILEY-VCH Verlag GmbH & Co, Copyright (2010).

Polymer	Pre-strain (%)	Thickness Strain <sup>(a)</sup> (%)	Area Strain <sup>(a)</sup> (%)	Young's Modulus (MPa)	Electric Field <sup>(b)</sup> (V/ $\mu$ m)	$\epsilon'$ <sup>(c)</sup>	$\epsilon''$ <sup>(c)</sup>
Silicone (Nusil, CF19-2186) [72]	–	32	–	1	235	2.8	54
Silicone (Nusil, CF19-2186) [10]	(45,45)	39	64	1.0	350	2.8	6.3
Silicone (Nusil, CF19-2186) [10]	(15,15)	25	33	–	160	2.8	–
Silicone (Nusil, CF19-2186) [10]	(100,0)	39	63	–	181	2.8	–
Silicone (Dow Corning, HS3) [72]	–	41	–	0.135	72	2.8	65
Silicone (Dow Corning, HS3) [10]	(68,68)	48	93	0.1	110	2.8	79
Silicone (Dow Corning, HS3) [10]	(14,14)	41	69	–	72	2.8	–
Silicone (Dow Corning, HS3) [10]	(280,0)	54	117	–	128	2.8	–
Silicone (Dow Corning, Sylgard 186) [72]	–	32	–	0.7	144	2.8	54
Silicone (Burman, Cine-Skin ArBrC) [78]	(0,100)	11	–	0.04	15	4.8	0.2
Silicone (BJB, TC-5005) [79]	(0,100)	1.0	–	0.1	8.0	3.5	0.02
Silicone (Dow Corning, Sylgard 184) [80]	(0,10)	0.5	–	2.49	119	2.8	–
Silicone (Wacker Elastosil RT 625) [80]	(0,10)	12	–	0.303	75	3.2	–
Silicone (BlueStar, MF620U) [81]	(50,50)	–	3.5	0.29	56	3.1	0.01
Fluorosilicone (Dow Corning 730) [72]	–	28	–	0.5	80	6.9	48
Fluoro-elastomer (Lauren L143HC) [72]	–	8	–	2.5	32	12.7	15
PU (Dccrfield PT6100S) [72, 82]	–	11	–	17	160	7	21

PU (Polytek Poly74-20) [83]	(0,10)	1.2	–	0.292	–	7.6	17.6
NR latex [72]	–	11	–	0.85	67	2.7	21
Dr. School's gelactive tubing [84]	(140,0)	1.8	–	–	28	–	–
HNBR (Zetpol 3310, ACN content 25%) [85]	–	–	5.8	1.6	30	12.4	0.12
NBR [86]	–	–	2.2	1.3	36	9.71	–
Acrylic (3M VHB 4910) [87]	–	79	380	–	–	–	–
Acrylic (3M VHB 4910) [10]	(300,300)	61	158	3.0	412	4.8	90
Acrylic (3M VHB 4910) [10]	(15,15)	29	40	–	55	4.8	–
Acrylic (3M VHB 4910) [10]	(540,75)	68	215	–	239	4.8	–
SEBS 75 (GLS Corp) [88]	(300,300)	62-22	180-30	0.007-0.163	32-133	1.8-2.2	92-53
SEBS 161 (GLS Corp) [88]	Nominal	14	16.5	1.3	27	–	28
SEBS 217 (GLS Corp) [89]	(300,300)	71-31	245-47	0.002-0.133	22-98	1.8-2.2	88-40
SEBS 217 (GLS Corp) [88]	Nominal	16	18.8	1.1	29	–	28
SEBS (Elastoteknik AB Dryflex 500040) [90]	(0,100)	12	–	0.25	58	2.3	0.25
SEBS-g-MA(Kraton) [91]	–	0.4	–	2.144	136	2.0	–
IPN (VHB 4910-HDDA) [92, 93]	(0,0)	70	233	2.5	300	–	–
IPN (VHB 4905-TMPTMA) [94]	(0,0)	59.36	146	3.94	265.4	2.43	83.5
IPN (VHB 4910-TMPTMA) [94]	(0,0)	74.97	300	4.15	418	3.27	93.7
PTBA [95]	(50,50)	77	335	0.42	260	5.4	–

<sup>(a)</sup> Deformation at electric field breakdown or maximum deformation reported

<sup>(b)</sup> Electric field at breakdown or maximum electric field reported

<sup>(c)</sup> Measured at 1 kHz

## 5. Methods to enhance the actuation performance of DEA devices

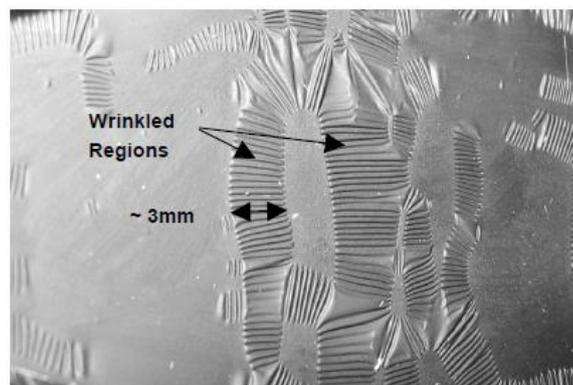
The strategies to enhance the actuation performance of DEs have to optimise the two elements that comprise the device: the electrodes and the elastomeric membrane. The main electrode requirements are that they should be highly compliant and conductive and should adhere well to the membrane. As for the membrane, its intrinsic factors are its dielectric permittivity,  $\epsilon_r$ , and elastic modulus,  $Y$ , as defined by Pelrine's model in equation 6. Hence,

the approaches to maximise the attained actuation strain are by: 1) reducing the DE film thickness 2) reducing the DE mechanical stiffness, and/or, 3) increasing the DE permittivity.

### *5.1 Reducing DE film thickness*

This reduction of the thickness has been pursued through a pre-stretching of the membrane, which was a key aspect of the improvement observed in the Science paper [10] compared to previous works [60]. Pre-stretching has shown to effectively enhance the breakdown field in certain commercial acrylic elastomers [57, 96, 97], being in some cases by more than an order of magnitude. Thus, for instance, in the case of the acrylic elastomer film VHB 4910, it has been reported that the breakdown electric field increases from 18 to 218 MV/m when equi-biaxially stretched up to 6 times [57]. However, the mechanism underlying this improvement is not yet entirely understood. The initial studies to clarify this issue ascribed it to changes in the dielectric permittivity with pre-stretch ratio. In this sense, Kofod *et al.* [57] reported that the dielectric permittivity of acrylic VHB 4910 film (measured with a dielectric analyser) was only slightly modified (from  $\epsilon' = 4.7$  to  $\epsilon' = 4.5$ ) with pre-stretch ratios higher than  $\lambda = 6$ . Nevertheless, a different study showed that the VHB dielectric permittivity (measured with an LCR meter) can decrease from its nominal value of  $\epsilon' = 4.7$  to  $\epsilon' = 2.6$  at a pre-stretch of ratio of  $\lambda = 5$  [98]. Qiang *et al.* [99] have recently corroborated this result. The authors have shown that the dielectric constant of VHB 9410 drops linearly from a value of  $\epsilon' = 4.4$  to  $\epsilon' = 2.4$  with increasing equal biaxial pre-stretching. This reduction in the dielectric constant was ascribed to a reduction of the dipoles freedom to align themselves, thus decreasing their ability to gather polarization charge. Nevertheless, the authors didn't address the influence of this observation on the electro-mechanical performance of the acrylic membrane, which still leaves the issue unresolved.

This conflicting information as to whether the electro-mechanical performance of VHB materials depends or not on the pre-stretching degree has led to the search for alternative explanations. Several authors [96, 100-102] have demonstrated that the improvement on the actuation performance with pre-stretch ratio is related to the suppression of a failure mechanism known as pull-in instability (Figure 7). Further studies have also evidenced that the pre-stretching increases the dielectric strength or breakdown field [57, 96, 97]. Therefore, nowadays, it is accepted that, in the case of acrylic membranes, any improvements in the actuation performance as a function of the pre-stretch ratio are attributed to both the suppression of the pull-in instability and the increase in the elastomer dielectric breakdown strength. All these theoretical and experimental studies have mainly focused on acrylic membranes and cannot be directly extrapolated to other DE, as recently shown by Akbari *et al.* [103]. These authors have shown that, in the case of PDMS, it is more effective to develop a thinner membrane rather than a highly pre-stretched one. Moreover, they have also shown the existence of an optimum pre-stretch ratio that suppresses the pull-in instability. Further stretching above this optimum value, only stiffens the elastomer increasing the actuation voltage necessary to achieve certain actuation strain, thus decreasing the electro-mechanical performance.



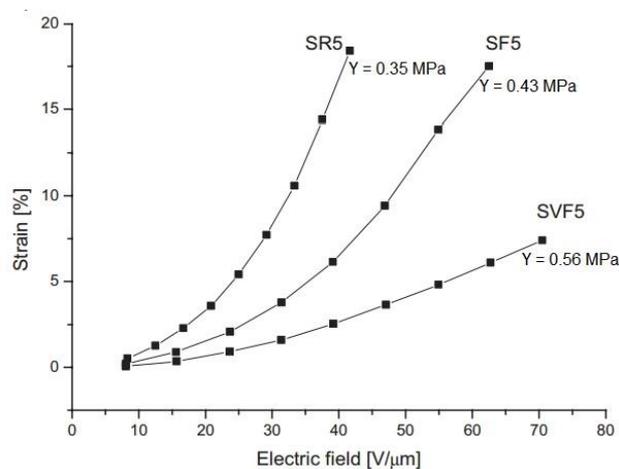
**Figure 7.** Pull-in wrinkles observed in an acrylic film [104]. Reprinted with permission from the authors.

In spite of the benefits detailed above, the main drawback of the pre-straining technique is that a rigid frame must be used to maintain the tension in the DE film, which increases the total weight and space of the DEA, thus eventually reducing the effective work density, and increasing the fatigue of the resulting actuators. Moreover, some issues regarding long-time stress relaxation of the DE devices may also promote their premature failure. To bypass this rigid frame, Ha *et al.* [92] developed an interpenetrating polymer network (IPN) to produce a similar actuation performance as highly pre-stretched DEAs. They sprayed a monomer together with an initiator onto a highly pre-strained VHB acrylic film. The monomer was then polymerised, thus forming a second elastomer network within the VHB elastomer host that preserved most of the film pre-strain after removal of the frame. These IPNs have shown strains up to 300% in area expansion at an electrical field of 420 V/ $\mu\text{m}$ , thus enabling the design of high performance actuators without the constraint of the frame.

### 5.2. Reducing DE mechanical stiffness

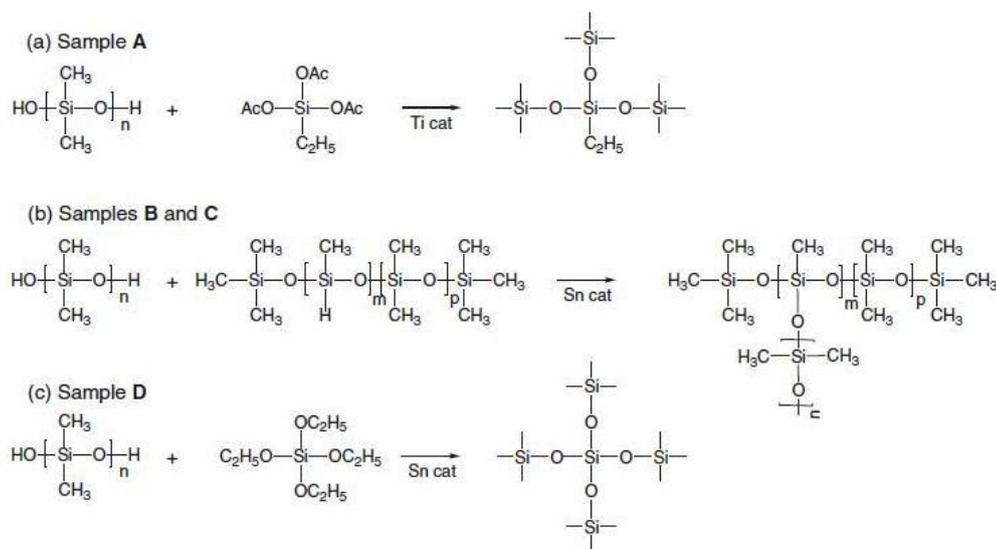
Generally, the mechanical properties of a DE can easily be modified through the use of additives, namely plasticisers, which lower the Young's modulus of the material. In this sense, Löwe *et al.* were the first authors to investigate the effect of both the concentration and type of different hardeners (81-R, 81-F and 81-VF from Suter-Kunststoffe) on the dielectrical, mechanical and electro-mechanical properties of a commercial silicone elastomer (DC3481 from Dow Corning) (Figure 8) [105]. They improved the actuator performance by tuning the Young's modulus with the type and concentration of hardener while keeping the dielectric permittivity value constant. This strategy has also been pursued on acrylonitrile-butadiene rubber (NBR) membranes through the addition of dioctyl phthalate (DOP) plasticiser [86]. The study showed an actuated radial strain of 1.6% at 20 V/ $\mu\text{m}$  for NBR loaded with 100 phr (parts per hundred of rubber) of DOP, compared to 0.13 at 20 V/ $\mu\text{m}$  without the plasticiser.

Nevertheless, this type of plasticiser is toxic to humans and the European Farmacopea has restricted its use in medical applications. The same authors have later studied the effect of non-toxic epoxydised soybean oil on both dielectric and mechanical properties as well as in the electro-mechanical performance of hydrogenated acrylonitrile-butadiene rubber (HNBR) [85]. The inclusion of 30 wt.% of oil slightly decreased the dielectric permittivity of HNBR matrix (from  $\epsilon' = 12.5$  to  $\epsilon' = 11.5$ ), but it also decreased the HNBR Young's modulus from  $Y = 1.6$  MPa to  $Y = 0.75$  MPa. These two facts led to an increase of about 100% in the actuated strain (at 30 V/ $\mu\text{m}$ ) over that of pure HNBR. Furthermore, the addition of 30 wt.% of oil in a 40 wt.% TiO<sub>2</sub>/HNBR composite was also able to weaken the molecular interaction between HNBR molecules and to break up the TiO<sub>2</sub> filler network, which increased the composite actuated strain in almost 170% at 30 V/ $\mu\text{m}$ . The main drawback for the use of plasticisers is their volatile or migrating nature that would affect the durability of the actuators or the application of the device.



**Figure 8.** Effect of three different hardeners, 81-R (SR5), 81-F (SF5) and 81-VF (SVF5), in the electro-mechanical properties of a silicone film. The figure includes the reported Young's modulus of the films. Reproduced from [105] with permission from WILEY-VCH Verlag GmbH & Co, copyright (2005).

A different strategy to modify the mechanical properties of elastomers is through the chemical modification of their molecular weight, stoichiometry and cross-linking degree, among others. In this sense, new DE films have recently been developed via UV curing of precursors comprising a mixture of acrylate co-monomers [106]. Varying the amount of cross-linker, the authors were able to control the stress-strain relationship and achieve high strain performance while suppressing the electro-mechanical instability. Again, the introduction of plasticizers resulted on elastomers with lower viscoelasticity and hence, improved strain sensitivity. Opris *et al.* [107] reported the formation of silicone networks with adjustable mechanical properties through reacting a hydroxyl end-functionalised PDMS ( $M_w = 139.000$  g/mol) with three different cross-linkers (Figure 9): (ethyltriacetoxysilane (ETAS) (sample A), (25-35%) methylhydrosiloxane-dimethylsiloxane copolymer (sample B and C) and a tetraethoxysilane (TEOS) (sample D)) and cured via standard procedures. The results showed that silicone networks with the copolymer as a cross-linker displayed the best electro-mechanical response, with a lateral actuation strain of 10.8% at 30 V/ $\mu\text{m}$ . Silicone elastomers prepared with ETAS and TEOS as cross-linkers showed less than 2% actuated strain at the same voltage. A recent study has described a two-step controlled addition reaction with the aim of synthesising silicone elastomers with low Young's modulus [108]. The authors reported Young modulus values in the order of 20-30 kPa without adversely affecting the viscous dissipation through the development of a bimodal network. All these works have shown that the stiffness of elastomers can effectively be decreased through the use of different approaches; nevertheless, it is worth to mention that very soft elastomers tend to exhibit strong viscoelastic effects, which may cause hysteresis and relaxation, thus decreasing the reliability of the DEAs, with low output force and breakdown strength values [85, 86].



**Figure 9.** Cross-linking reactions used for the formation of room temperature vulcanised silicone rubber. Reproduced from [107] with permission from WILEY-VCH Verlag GmbH & Co, Copyright (2011).

### 5.3 Increasing DE permittivity

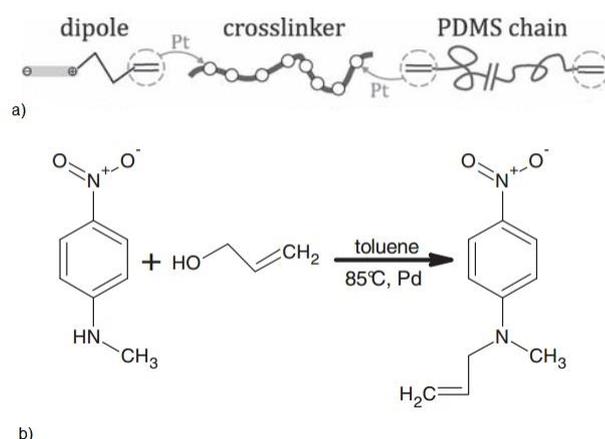
The need for high permittivity materials is shared by different applications, from condensers for insulators for mid and high power electrical systems to materials for batteries, and, hence, has heavily been pursued in the literature. In DEs, the increase of the film permittivity produces a direct increase in its capacitance, thus reducing the required electric field intensity. This strategy has been investigated through three different approaches, chemical modification of the elastomer backbone, elastomer composites and blends.

#### 5.3.1 Chemical modifications of the elastomer backbone

A suitable strategy to obtain elastomers with specific dielectric properties is by means of either the synthesis of new molecular structures or the chemical modification of existing elastomers. This last approach has been pursued through the chemical grafting of a  $\pi$ -conjugated conducting macromolecule, namely polyaniline (PANI) doped with dodecyl

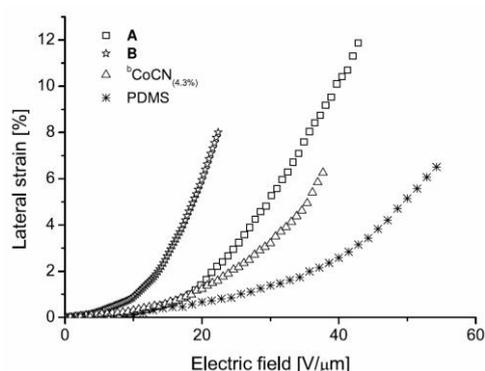
benzene sulfonic acid (DBSA) to an elastomer backbone [91]. The idea behind this approach was to encapsulate the PANI chains through chemical bonding, such that the conductivity in the composites remained low. The authors have shown that PANI grafting up to 2.0 vol.% to a thermoplastic copolymer (polystyrene-co-ethylene-co-butylene-co-styrene-grafted maleic anhydride (SEBS-g-MA)) did lead to a large increase on the dielectric permittivity of about 470% over the raw matrix. The chemical modification hardly reduced the breakdown strength value (from 140 to 120 V/ $\mu\text{m}$ ) with the subsequent increase in the electrostatic energy value. These two effects, combined with a slight reduction in the mechanical stiffness, resulted in a significant improvement in the electro-mechanical response as well as in the maximum actuation strain achieved. Nevertheless, a transition from non-conductive to conductive behaviour was observed for concentrations above 2.0 vol.% of grafted PANI. Composites above this percolation threshold showed an increase in the dielectric permittivity accompanied by a drastic increase in the conductivity and a decrease in the Young's modulus due to the DBSA dopant acting as a plasticiser. In spite of the conductive character of these composites, they showed enhanced electro-mechanical performance but with a drastic reduction in the breakdown strength value, thus limiting their application. The same concept was further studied by Kussmaul *et al.* [109], using in this case a push-pull dipole (N-allyl-N-methyl-p-nitroaniline) as high dielectric grafted molecule in a PDMS matrix. The novelty of this work was that the selected dipole possessed a vinyl functionalisation, which enabled the grafting of the dipoles and the formation of the silicone network in one step (Figure 10). The grafting of the push-pull dipole, in concentrations below 13.4 wt.% to a silicone cross-linker, caused the relative permittivity at 1 kHz to increase from 3.0 to 5.9 while decreasing the elastic modulus from 1900 to 550 kPa. Again, this combined effect was found to result in an improvement of the electro-mechanical response of more than six times that of the raw matrix. These authors exploited this strategy in a later work changing the silicone matrix employed [80]. The

grafting of N-allyl-N-methyl-p-nitroaniline dipoles to two commercial silicone networks (Sylgard 184 from Dow Corning and Elastosil RT 625 from Wacker) in concentrations ranging from 0 to 10.7 wt.% increased the dielectric permittivity (from  $\epsilon' = 2.84$  to  $\epsilon' = 6.15$  for Sylgard 184, and from  $\epsilon' = 3.17$  to  $\epsilon' = 5.57$  for Elastosil RT 625) and decreased the Young's modulus (from  $Y = 2490$  to  $850$  kPa for Sylgard 184 and from  $Y = 303$  to  $Y = 142$  kPa for Elastosil RT 625) [80]. Samples with the highest dipole content in both silicones displayed enhanced electro-mechanical performance as compared with neat silicone, although the breakdown value decreased in both cases. These authors have also modified a typical silicone cross-linking molecule with a highly polarisable dipole (allyl cyanide) and a compatibilising agent (allyltrimethylsilane) to reduce the possible precipitation of the allyl cyanide group during the synthesis [110]. The incorporation of up to 19.4 wt.% of the dipole led to an enhancement in the dielectric permittivity while decreasing the mechanical stiffness. Nevertheless, this strategy often results in an adverse decrease of the breakdown strength which might limit the application of the materials. This issue worsens with larger amounts of dipole content and can render the materials prone to moisture uptake [77].



**Figure 10.** a) Schematic representation of the direct grafting of a dipole to the cross-linker of a hydrosilylation reaction of a vinyl-terminated PDMS. b) Push-pull dipole (N-allyl-N-methyl-p-nitroaniline) with vinyl functionalization [109]. Reproduced with permission from WILEY-VCH Verlag GmbH & Co, copyright (2011).

The idea of chemical modification of a given silicone network with highly polarisable groups has recently been further studied by Racles *et al.* [111]. In this case, the inclusion of cyanopropyl groups in the silicone network was pursued through three different synthesis paths: hydrosilylation of allyl cyanide on a methylhydrosiloxane-dimethylsiloxane copolymer, redistribution reaction of a PDMS with 1,3,5,7-tetra(3-cyanopropyl)-1,3,5,7-tetramethylcyclotetrasiloxane ( $D_4^{CN}$ ) and finally, cationic ring opening copolymerisation of  $D_4^{CN}$  and octamethylcyclotetrasiloxane ( $D_4$ ). The prepared polymers were further functionalised with hydroxyl end groups to enable their later cross-link. Results showed that the last synthesis route was the most effective method to increase the silicone dielectric permittivity from  $\epsilon' = 2.4$  to  $\epsilon' = 6.5$ , with 23% of cyanopropyl groups. Unfortunately, most synthesised films had low molecular weights and were too brittle to use as DEA membranes. The only sample with the appropriate properties was achieved via the redistribution reaction with 4.3% of the polar groups in the elastomer backbone. This sample presented a slightly higher dielectric permittivity (from  $\epsilon' = 2.4$  to  $\epsilon' = 3.0$ ) and lower Young's modulus as compared with the neat silicone, leading to a 6.2% maximum actuation strain (which was three times over the neat silicone) at  $37.5 \text{ V}/\mu\text{m}$  (Figure 11).



**Figure 11.** Electro-mechanical properties of films of PDMS with 4.3% cyanopropyl groups synthesised through a redistribution reaction and different amounts of cross-linker (films **A** and **B**) [111]. Reproduced with permission from IOP, copyright (2013).

Recent studies building on this strategy have also evidenced its feasibility to obtain new and improved elastomeric systems. For example, the functionalization of a siloxane copolymer by attaching a pendant polar group, 1-ethynyl-4-nitrobenzene via copper-catalysed azide-alkyne 1,3-dipolar cycloaddition reactions, produced a 70% increment of  $\epsilon'$  without compromising either the electrical breakdown strength or the dielectric losses [112]. Likewise, Dünki *et al.* used a high molecular weight polymethylvinylsiloxane as a functionalizable elastomeric chain where the vinyl groups were the starting point for the introduction of highly polarisable moieties, nitrile groups from 3-mercaptopropionitrile. The authors used these moieties not only to increase the permittivity, but also as crosslink points to tune the final mechanical properties of the matrix with different catalyst [113]. In this case, the best material showed  $\epsilon' = 10.1$  at  $10^4$  Hz with an elastic modulus of 154 kPa at low strains, thus enabling an electro-mechanical performance of 20.5 % at low electric fields (10.8 V/ $\mu\text{m}$ ). This material also showed a very attractive self-healing ability after a breakdown at low actuation voltages, which might enhance both lifetime and reliability.

In summary, all these results have shown that upon chemical modification it is possible to obtain improved elastomeric systems with reasonable high permittivity values while maintaining other properties, such as dielectric losses and electric breakdown strengths. Both chemical synthesis and modifications protocols enable the fine-tuning and control of elastomeric network structures. These strategies will open the pathway for the mass-production of the next-generation of elastomers ready to be used in real applications.

### 5.3.2 Elastomer composites

The dielectric permittivity enhancement for a given elastomer can also be accomplished using the more conventional strategy of elastomer composites. The idea of this approach is to combine the high dielectric permittivity values of certain fillers with the high breakdown

strength of polymers and presents the advantage of being easily scalable. Nevertheless, it also has to overcome additional limitations, since the addition of fillers is prone to increase both the dielectric loss and the tensile stress values, which can decrease the electro-mechanical performance for DEA applications. Therefore, composites satisfying these three characteristics (high dielectric permittivity, low dielectric losses and low Young's modulus) are difficult to attain and the best solution will be a compromise among them.

#### *5.3.2.1 Conductive filler/elastomer composites. Sub-percolative approach*

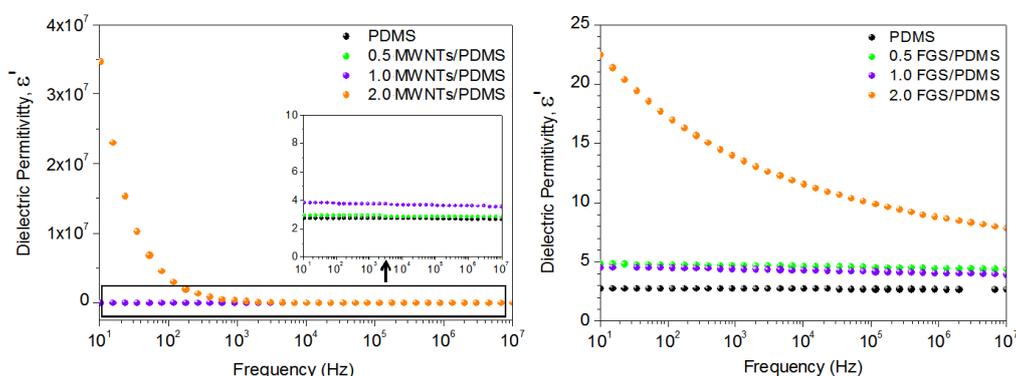
In general, the system composed of conductive filler/polymer exhibits a non-linear increase of the permittivity and electrical conductivity as the concentration goes above the percolation threshold ( $p_c$ ). According to the percolation theory [114], at low filler concentration the conductive particles are separated from each other and the electrical properties of the composites are still dominated by the matrix. As the concentration increases, the fillers start to form a three dimensional network through the matrix, giving rise to a huge increase in the electrical conductivity. The idea of the sub-percolative strategy is to maintain the dielectric character of the matrix, by working at concentrations below the percolation threshold, while taking advantage of the increase in the dielectric permittivity.

The search for high permittivity elastomer composites has been pursued with different conductive fillers, from metal [115-117] to carbon-based micro particles [118-120]. Thus, for instance, in 2009, Stoyanov *et al.* reported the dielectric properties of a poly-styrene(co-ethylene-co-butylene-co-styrene) (SEBS) matrix loaded with carbon black (CB) as conducting filler [118]. Experiments revealed a significant increase in the dielectric permittivity value in the entire frequency range near the percolation threshold while maintained low dielectric losses. Above the percolation threshold ( $p_c = 4.62$  vol.% of CB), composite samples showed a

drastic reduction in the breakdown strength values due to the increase in the conductivity and the appearance of CB agglomerates on the micrometre range.

According to the theory, the dielectric behaviour of nanofillers would differ from the micrometre fillers and would not only depend on the nature and quantity of the used nanoparticle but also on the nanofiller-polymer interfaces [121]. Hence, there is an increasing body of research on the use of nanofillers to modify the dielectric behaviour of polymer [122]. Among them, most studies reported the use of graphitic nano-materials, in particular multiwall carbon nanotubes (MWCNTs) [123-127] but some initial work has also been done with graphene layers [128, 129]. Park *et al.* [123] were among the first to show that the addition of 0.5 wt.% of commercial MWCNTs increased the dielectric permittivity of a PDMS matrix from  $\epsilon' = 3$  to  $\epsilon' = 5$ , in both low (50 Hz) and high (100 kHz) frequencies. Meanwhile, the alignment of MWCNTs in polysulfone (PSF) nanofibres by means of electrospinning produced a dielectric permittivity of up to  $\epsilon' = 58$  for a sample with 25 vol.% of MWCNTs [126]. The authors also reported that the filled samples had a similar loss tangent value ( $\tan(\delta)$ ) with high breakdown fields, thus indicating the suitability of these composite sheets as energy density capacitors and flexible high dielectric components. A subsequent study aimed at improving the dispersion and modifying the interface region used an anionic surfactant, in particular DBSA, on the separation and distribution of MWCNTs in a PDMS matrix [127]. The dielectric permittivity of both neat MWCNT/PDMS and MWCNT-DBSA/PDMS composites increased as the filler concentration increased. Indeed, both composites displayed the same dielectric permittivity value at low volume fractions (i.e. 1.5 vol.%). However, at 4 vol.% of filler, the dielectric permittivity of neat MWCNT/PDMS was  $\epsilon' = 2500$ , indicating a percolated behaviour, whereas for MWCNT-DBSA/PDMS the dielectric permittivity was  $\epsilon' = 20$ . Unfortunately, no information was provided on the dielectric breakdown of the materials.

The use of graphene layers in elastomeric matrices with the aim of improving the dielectric permittivity was first reported by Romasanta *et al.* [128]. The authors compared the dielectric permittivity of functionalised graphene sheets (FGS)/PDMS and MWCNT/PDMS composites at room temperature. A similar filler concentration of 2.0 wt.% led to different behaviour, while FGS maintained the dielectric nature of PDMS increasing ten times the permittivity value (to  $\epsilon' = 23$ ) at low frequencies, MWCNTs showed a percolated behaviour with an increase of six orders of magnitude (Figure 12). The reason for this different trend was ascribed to the presence of functional groups on the graphene surface that improved the filler/matrix compatibility and, thus, reduced the Maxwell-Wagner-Sillars polarisation at low frequencies. A similar behaviour has subsequently been observed in reduced graphene oxide (RGO) and expanded graphite (EG)/polyisobutylene-co-isoprene (IIR) composites [129].



**Figure 12.** Dielectric permittivity of (left) MWCNTs/PDMS composites and (right) FGS/PDMS composites at various filler concentrations (wt.%) [128].

A recent study has addressed possible restacking of the graphene layers during sample preparation by self-assembling graphene oxide (GO) on carbon nanospheres (CNS) [130]. These hybrids nanoparticles, where GO forms the shell and the CNS the core, enabled the formation of a dielectric network within a XNBR matrix, thus increasing the dielectric permittivity from  $\epsilon' = 25$  (raw XNBR matrix) up to  $\epsilon' = 400$  (0.75 vol% of nanohybrids) at

$10^3$  Hz. The presence of the CNS also facilitated the formation of a segregated filler network, thus avoiding any sharp increases in the dielectric losses ( $\epsilon'' < 0.65$  at  $10^3$  Hz). Although the actuation strain increased from 2.69% (pure XNBR) to 5.68% (composite with 0.5 vol% of nanohybrids), the breakdown strength still showed a strong decrease from 14.3 to 7.0 V/ $\mu\text{m}$ , which could limit the applicability of these materials only where low electric fields are required

Besides the reduction on the breakdown strength, generally, the mechanical properties of this type of composites are improved with the nanoparticle loading due to their reinforcing character. In most of the cases, the tensile stress at low strains shows a sharp increase, which is not desirable for DEAs applications. Hence, several studies for DEA devices proposed a strategy to minimise those two effects: the development of a passivation layer around the conductive particles. The first study by Molberg *et al.* [131] showed that the encapsulation of PANI particles with an insulating polymer shell prior to their dispersion in a PDMS oligomer increased the breakdown strength of the system while enhancing the PANI/PDMS compatibility; thus, overcoming the filler agglomeration and the dielectric losses increase. This strategy led to composites with enhanced dielectric properties as compared with the neat PDMS while maintaining its dielectric nature. Nevertheless, as a result of the improved PANI/PDMS compatibility the Young's modulus showed a twentyfold increase for the composite loaded with 31.7 vol.% of PANI particles. As a consequence, all composites displayed lower actuation strains as compared with PDMS, except for a composite with 13.6 vol.% of filler, which showed identical electro-mechanical behaviour in all the electric field range. In any case, all samples showed electric field strengths of more than 57 V/ $\mu\text{m}$ , which indicated that the encapsulation of conductive particles is a viable mean to obtain high dielectric strength composites.

This encapsulation strategy was also employed recently with MWCNT to reduce their tendency to aggregate and, thus, minimise the dielectric losses while achieving again a better filler/elastomer compatibility [83]. In this study, the MWCNT surface was chemically modified by grafting either a polyacrylonitrile or a diurethane polyacrylate prior to their incorporation into a PU matrix. This non-conductive shell maintained the dielectric loss of the functionalized MWNTs/PU composites at the same levels of those of the raw matrix, thus indicating that no percolation phenomena occurred. Moreover, these samples showed a lower elastic modulus than PU matrix, due to a competing activity of the filler surface with the chemical mechanism in the curing process. All developed composites were found to actuate due to their combination of good dielectric and mechanical properties. In fact, a composite containing 0.5 wt.% of MWCNTs grafted with diurethane polyacrylate showed an electro-mechanical response two times higher than neat PU matrix.

A second approach to create a passive layer has been the development of a secondary network that would disrupt the conductive network. Zhao *et al.* [132] have shown that the incorporation of barium titanate, BaTiO<sub>3</sub> (BT), to an acetylene-CB/PDMS composite successfully interfere the electrical conductivity and decrease the dielectric loss. The presence of CB, up to an optimal mass fraction of  $m_{CB}=0.04$ , decreased the elastic modulus of the three component nanocomposites, since it was also able to destroy the network formed by the BT filler and enlarge its packing volume fraction due to the differences in size. These factors, together with the observed increase in the permittivity value synergistically contributed to an increase of the electro-mechanical performance of the composites, obtaining the maximum area strain of around 8% with 0.03 wt.% CB and 0.60 wt.% BT.

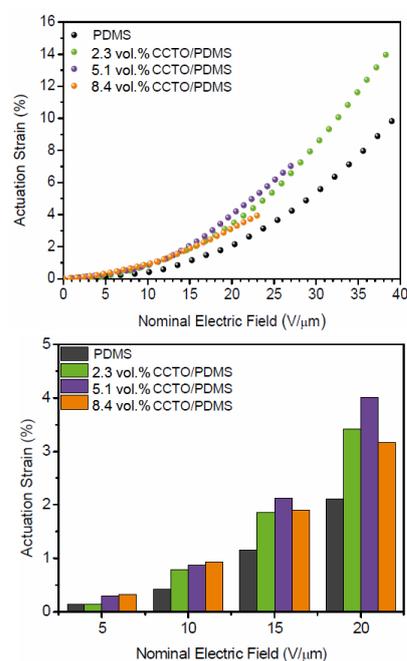
The above-mentioned studies mostly focused on the increase of the dielectric permittivity via a composite approach of conductive nanofillers below the percolation threshold. While they showed such an increase, they also reported decreases in the dielectric strength and

increases in the dielectric losses. However, several authors have also observed increases of the dielectric strength and loss reductions in nanocomposites (see the reviews [133, 134]) ascribed to dispersion or agglomeration issues during composite preparation. Further research is needed to fully understand the dielectric behaviour of nanocomposites and the role of the filler-polymer interface.

#### 5.3.2.2 Ceramic filler/elastomer composite

The second strategy has been the use of inorganic fillers of high permittivity values since, in principle, they should increase the dielectric permittivity of a given elastomer while maintaining its dielectric nature. The addition of common inorganic fillers, such as organically modified montmorillonite [135], titanium dioxide  $\text{TiO}_2$  [136-138],  $\text{BaTiO}_3$  (BT) [139-142], boron nitride (BN) [143] and lead magnesium niobate–lead titanate  $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{--PbTiO}_3$  (PMN-PT) [144], have already shown to be a suitable strategy to improve the dielectric permittivity of a wide range of elastomers (PDMS, PU, SEBS, EVA, among others). One of the first works on this topic showed that the addition of 7.3 vol.% of rutile-type  $\text{TiO}_2$  to a silicone rubber improved its dielectric properties while reducing its Young's modulus [78]. This composite showed a transverse actuation strain of 11%, more than eight times higher than that observed for pure silicone. Nevertheless, although the observed dielectric loss was preserved in the composite, the breakdown strength was reduced as compared with the neat silicone. A later work has shown that the surface modification of  $\text{TiO}_2$  with a PDMS oil coating in a SEBS film membrane raised the breakdown strength from 44  $\text{V}/\mu\text{m}$  to 54.8  $\text{V}/\mu\text{m}$  [145]. This study showed that the addition of 15 vol.% of these oil-coated  $\text{TiO}_2$  particles caused a 27% reduction of the driving electric field and a maximum actuation strain of about 12%. Recently, Romasanta *et al.* [81] proposed the use of calcium copper titanate,  $\text{CaCu}_3\text{Ti}_4\text{O}_{12}$  (CCTO), as an appropriate filler for DEA devices, due to its

frequency and temperature independent giant dielectric constant of  $10^4$  (DC to  $10^6$  Hz). Loading fractions of CCTO in PDMS, up to 5.1 vol.%, exhibited a remarkable combination of dielectric and mechanical properties, increasing their dielectric permittivity while maintaining both the dielectric loss value and tensile stress values at different strains. The electro-mechanical behaviour of these composites showed improvements in the actuation strain of about 100% and a reduction of 25% in the electric field to reach the same strain compared to the raw PDMS matrix (Figure 13).



**Figure 13.** Experimental electro-mechanical performance of PDMS and CCTO/PDMS actuators showing: a) actuation strain (%) as a function of the nominal electric field (each curve is the average of at least 5 independent actuator devices), and b) actuation strains (%) at nominal electric fields of 5, 10, 15 and 20 V/μm. Reproduce from [81].

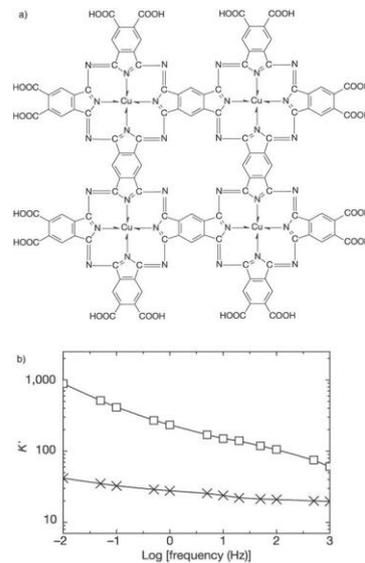
Nevertheless, other works with BT [146], PMN-PT [144] and lead zirconate titanate (PZT) [147] have also reported that the use of these ferroelectric ceramics is not always an appropriate strategy for DEA devices. These studies showed no real improvements in the theoretical or experimental electro-mechanical performance of the elastomer even though all

reported an increase in the permittivity values. The authors attributed these results to several factors: i) the increase in the dielectric permittivity was counterbalanced by an increase in the elastic modulus, ii) the electrical breakdown strength was considerably reduced, which limited the maximum performance and, finally, iii) the dielectric loss dramatically increased with the filler content cancelling the electro-mechanical performance of the actuator. A further issue, which was not considered by the authors, is the ferroelectric and piezoelectric nature of these fillers that may introduce an additional electro-mechanical coupling and could be affecting the composite response. Also, these fillers present a dependency of their dielectric permittivity with temperature that would limit the DEAs application range. The encapsulation of BT particles with a polydopamine (PDA) thin layer [148] has also shown to partially solve these issue by improving the dielectric permittivity and decreasing the dielectric loss. Nevertheless, and although the developed composites displayed identical or slightly higher actuation strain than pure HNBR, they were not able to withstand high electric fields.

### 5.3.3 Elastomer blends

The idea is similar to the composite approach but using two polymers, miscible or immiscible, to obtain novel materials with attractive properties. Indeed, Zhang *et al.* [149] explored for the first time this concept by developing composites fabricated using an organic oligomer (copper-phthalocyanine (CuPc)) (Figure 14) dispersed in an electrostrictive polymer matrix (poly(vinylidene fluoride-trifluoroethylene), P(VDF-TrFE)). CuPc oligomers have shown dielectric permittivity values around  $10^5$  due to the conjugated  $\pi$ -bonds within the molecule. However, this electron delocalisation also produces high dielectric losses. In this work, the authors demonstrated that the long-range intermolecular hopping of electrons can be reduced with the polymer matrix forming insulating layers around the CuPc, thus blocking the space charge conduction and leading to composites with both enhanced dielectric and electro-

mechanical properties. Subsequent research with this same oligomer analysed the effect of adding a conductive polymer, in particular polyaniline (PANI), to CuPC/polyurethane (PU) blends [150]. This three-component composite showed an enhanced dielectric permittivity value with an electro-mechanical response of 9.3% under an electric field of 20 V/ $\mu\text{m}$ .



**Figure 14.** a) Schematic of the copper-phthalocyanine (CuPc) oligomer and b) real part of the dielectric constant ( $K'$ ) as a function of frequency of the P(VDF-TrFE) (crosses) and its composite containing 40 wt.% CuPc (squares) [149]. Reprinted by permission from Macmillan Publishers Ltd: [NATURE], copyright (2002).

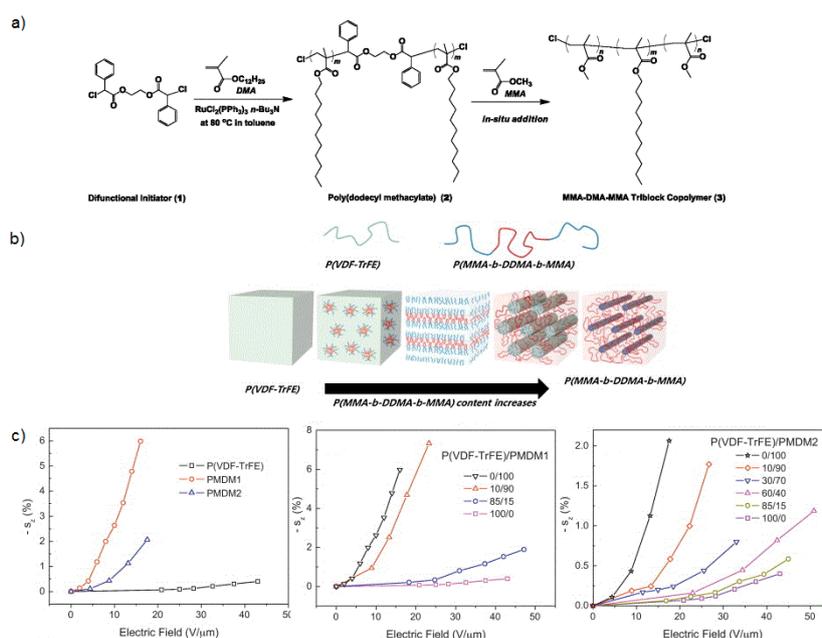
This concept has been further studied by blending a commercial PDMS (TC-5005 A/B-C, BJB Enterprises Inc., USA.) with very low percentages (1-6 wt.%) of poly(3-hexylthiophene) (PHT), a highly polarizable conjugated polymer [79]. The developed blends showed a relatively large increase in dielectric permittivity with a small increase in the dielectric loss and an unexpected reduction of the tensile elastic modulus. Such unexpected reduction of the Young's modulus, from 100 kPa for PDMS to 20 kPa with 1 wt% PHT in PDMS, was ascribed to a reduced cross-linking degree of PDMS matrix due to the high morphological integration between matrix and PHT, resulting in a partial hindrance of the cross-linking

behavior of the curing agent. The combination of high dielectric permittivity and low elastic modulus increased the electro-mechanical strain response of all the blends, which was found to be the largest for a 1 wt.% content of PHT. The increase in the PHT concentration led to a progressive electro-mechanical performance decrease, due to an observed increase in the dielectric loss. These authors extended this approach to blends of PDMS/PU for DEAs applications [151]. The simple mixing of these two elastomers in 60/40 (v/v) ratio gave rise to a strong interfacial polarisation (Maxwell-Wagner-Sillars relaxation) and, thus, producing a marked increase in the dielectric constant at low frequencies, which was unexpectedly higher than the dielectric permittivity of PU and PDMS. The blend showed higher actuation strains for any given electric field but it also had lower dielectric strength, which was ascribed to the observed interfacial polarisation.

More recently, the approach of enhancing the interfacial polarization in elastomeric blends has further been explored by incorporating a low molecular weight polyethylene glycol (PEG) into a thermoplastic polyurethane (TPU) matrix [152]. The addition of this oligomer had a synergic effect on both dielectric and mechanical properties. Thus, the observed increment in  $\epsilon'$  was ascribed not only to the PEG ionic conductivity but also to its ability to disrupt the hydrogen bonds on TPU, which, in turn, enhanced TPU chain polarization. Besides, Young's modulus was largely decreased due to PEG plasticizing effect in the TPU soft segments. Although the breakdown strength decreased with PEG content (from 25 V/ $\mu\text{m}$  for pure TPU to 3 V/ $\mu\text{m}$  for TPU with 80 phr of PEG), the developed blends showed remarkable actuation strains enhancements (7500% increase at 3 V/ $\mu\text{m}$ ) at low electric fields, thus making this material suitable for applications where low electric fields are desirable.

The development of a blend of an electrostrictive polymer, in particular poly(vinylidene fluoride-co-trifluoroethylene) P(VDF-TrFE), with a methacrylate-based block copolymer, i.e. poly(methylmethacrylate-*b*-dodecylmethacrylate-*b*-methylmethacrylate), has

shown to be a versatile approach to fabricate dielectric elastomer actuators with adjustable properties [153]. The idea behind this study was to use selectively the miscibility of P(VDF-TrFE) with only one of the methacrylate blocks. The authors showed that they can then tune the dielectric, mechanical and electro-mechanical properties by varying the molecular weight of the methacrylate block copolymer and the concentration of the (P(VDF-TrFE)/P(MMA-b-DDMA-b-MMA)) blends (Figure 15).



**Figure 15.** Schematic of the a) synthesis procedure of the P(MMA-b-DDMA-b-MMA) triblock copolymer, and b) microstructure evolution of the P(VDF-TrFE)/P(MMA-b-DDMA-b-MMA) blends as a function of P(MMA-b-DDMA-b-MMA) concentration. c) Tuned electro-mechanical strains of the (left) three pure polymers: PVDF, PMDM1 ( $M_w = 144,000$  g/mol) and PMDM2 ( $M_w = 126,000$  g/mol), (middle) P(VDF-TrFE)/PMDM1 blends and (right) P(VDF-TrFE)/PMDM2 blends, as a function of the applied electric field [153]. Reprinted by permission from Elsevier, copyright (2012).

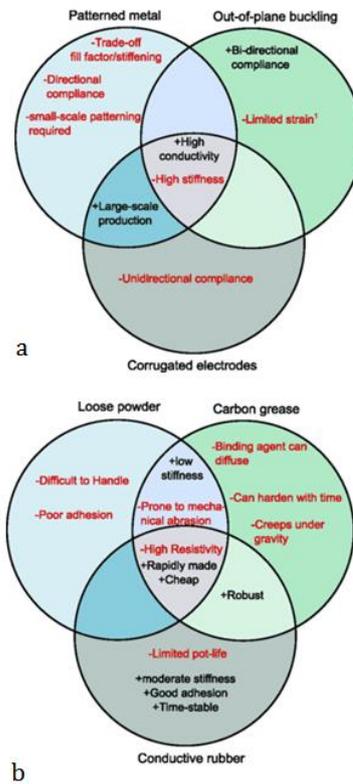
Finally, one recent study taking advantage of the polymer blend strategy has looked at the possibility of controlling the directional actuation of DEAs. The authors incorporated aligned

and unidirectional high dielectric PU fibres within VHB-4905 membranes [154]. The developed blends showed improved electro-mechanical performance due to the coupling effect between the mechanical anisotropy, by the unidirectional position of the PU fibres, and the increment in the dielectric permittivity value upon the incorporation of the PU fibres.

In summary, the increase of the dielectric permittivity has been achieved through the proper manipulation of the elastomer backbone and straight forward mixing procedures. The former is an elegant solution enabling the development of tailor-made DE film membranes but involves time-consuming and rather expensive reactions, which could hinder a timely scale-up of the developed materials. Meanwhile, the composite and blend approaches require a careful selection of the filler or polymer to minimise mechanical reinforcing, dielectric losses and, also, the reduction of the breakdown strength. The results suggest that a suitable method to reduce the accumulation of charges at the particle/polymer interface and minimise leakage currents is the addition of a passivation layer via the encapsulation of the inclusions.

#### *5.4 Developing highly compliant electrodes*

Identifying or developing a compliant electrode is also a key factor in DEA devices and a number of electrode materials have been explored to achieve good electro-mechanical performance. Original tests were performed using thin metal films, which provided good electrical conductivity but also limited strains to approximately 1%. Therefore, good electrode materials for DEA devices should not impede its mechanical motion, resulting in the term “compliant electrodes”. Moreover, they should maintain high conductivity at large strains and good stability, have negligible stiffness, and be fault tolerant at the same time [155]. We present below a brief overview of the two main electrode types: metal thin film and carbon-based electrodes (Figure 16), the reader can find a comprehensive and recent review on their characteristics and approaches in reference [155].



**Figure 16.** Main characteristics of the two principal electrode types. a) Metal thin film and b) carbon-based electrodes [155]. Reprinted with kind permission from Springer Science+Business Media.

- *Metal thin-film electrodes.* The development of microelectronics and MEMS industry has put on the market a broad range of microfabrication technologies (i.e. electron beam evaporation, cathodic sputtering, electroplating, photolithographic processes...) that deposit thin metal layers even in the nanometre scale [155]. Nevertheless, there are three major drawbacks that still hinder their widespread use: i) the high Young's modulus of metals (several orders of magnitude higher than that of membrane) limits the actuation strain of the DE, ii) their low elasticity (typically around 2-3%) causes cracks above this limit resulting in conducting areas separated by non-conductive elastomer, and iii) metals have a tendency to form an insulating oxide layer at the surface. Current research efforts have developed new

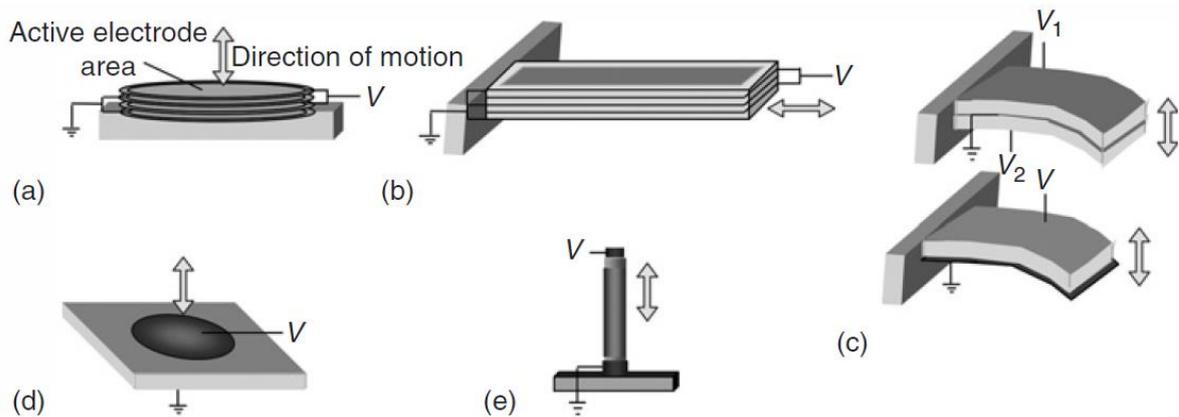
solutions to use metallic thin films for DEAs applications such as patterned electrodes [72, 156, 157], out-of-plane buckled electrodes [158, 159], and corrugated membranes [160, 161].

- *Carbon-based electrodes*. These are the most commonly used electrodes as they provide good conductivity even at very high strains. They can be divided in three main categories. 1) Carbon powders, such as carbon black and graphite, are directly deposited on the elastomeric membrane. The main advantage of this type of electrodes is that they do not contribute to the stiffness of the membrane on which they are applied, mainly due to the absence of strong binding forces between the particles. However, they could get detached from the DE surface reducing the electrode's durability. For this reason, these powders are generally found in combination with acrylic elastomers and silicones since they are intrinsically sticky and the particles become bound to their surface. 2) Carbon greases consist of carbon particles dispersed into a viscous media, such as oil. DEA devices developed using this electrode sustained large strains while remaining conductive, but they also showed long-term instability issues due to drying or diffusion into the DE. 3) Carbon compounds are formed by the dispersion of carbonaceous particles, generally carbon black, in an elastomer and cross-linked after being applied on the membrane. Since the filler concentration must be sufficiently high to percolate, the contribution of these electrodes to the DE stiffness cannot be neglected (as compared with the other two). Additionally and although the electrode bounds to the DE membrane during the cross-linking process, some delamination issues have been reported to reduce the electrodes' life expectancy [162]. O' Brien *et al.* [163] characterised the three main types of carbon-based electrodes for a resistive self-sensing device. In this work, the three electrodes were deposited up to measuring a surface resistance of about 9 k $\Omega$ /square. The electrodes were then subjected to uni-axial strain while monitoring the change in resistance. Carbon powder electrodes produced the best electrical and mechanical characteristics out of the several possibilities tried.

Recent studies of carbon compounds have used carbon nanotubes and graphene sheets, resulting in transparent compliant electrodes that presented self-healing characteristics, thus increasing both lifetime and reliability. This self-healing behaviour was first reported by Yuan *et al.* [164] using sprayed SWCNTs on an acrylic film membrane. They mechanically damaged the membrane with a wood pin and still observed the capacitive properties of the device with only a minimal reduction in the actuation strain. These authors proposed that, in the vicinity of the area where the dielectric breakdown occurs, the electrode becomes non-conductive due to the degradation of SWCNTs. However, they suggested that the SWCNTs have the ability to self-clear and, hence, isolate the fault from the rest of the active area, which can actuate normally. This self-healing behaviour has also been observed in a conductive and stretchable PDMS composite based on 20 wt.% of reduced graphite nanoplatelets [162]. Other transparent electrodes have been reported using Ag nanowires [165] and, more recently, by replacing the electrodes with soft, ionic hydrogels [166].

## **6. Applications of DEAs**

The versatility of conformable elastomers allows shaping them into a wide range of dimensions, thus, enabling the actuator configuration to be tailored with the desired/needed characteristics. Although most actuator designs use the framed/in-plane actuator configuration, nowadays a vast variety of actuators' configuration can be found in the literature (Figure 17) [11, 77, 167-169].



**Figure 17.** Basic dielectric elastomer configurations: (a) stack, (b) extender, (c) bimorph and unimorph bending beam actuators, (d) diaphragm and (e) rolled actuators [11]. Reprinted with permission from Elsevier, copyright (2008).

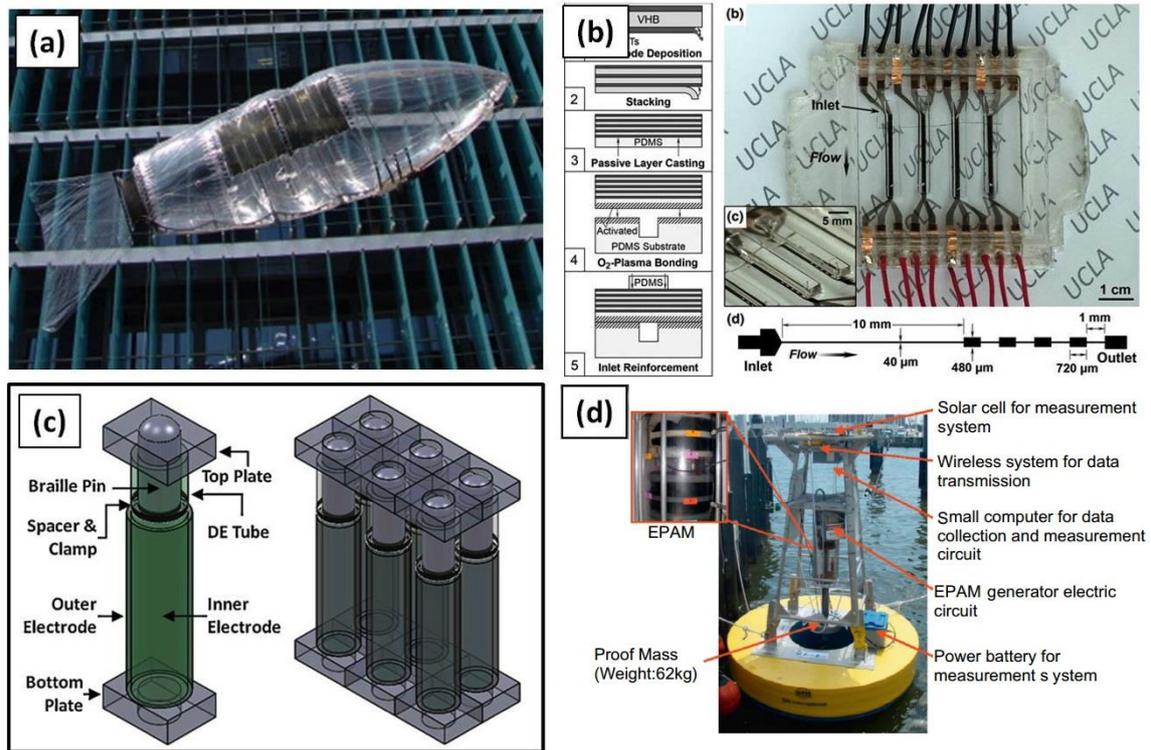
- *Multilayer/stacked actuators.* The elastomer films are stacked forming a series of capacitors electrically connected in parallel with alternating polarities on each layer. This configuration produces devices with large force and deformation characteristics [170, 171].
- *Unimorph [172, 173] and bimorph [174] bending beam actuators.* Unimorph actuators are made of a DE bonded to a flexible substrate; whereas in bimorph actuators the flexible substrate is sandwiched between two DE films. These configurations provide large out of plane displacements.
- *Diaphragm actuators.* These actuators are made with a planar construction to produce out of plane motion [175-177]. This type of configuration is employed in pumps with direct energetic coupling to an external load [167].
- *Cylindrical/rolled actuators.* The coated DE film is rolled around an axis with a core, such as a spring (“spring roll”), or without a core [178-181]. This configuration is employed in muscle-like devices due to its relatively large output force.

The latest configuration development has analysed an electrode-free configuration by reproducing Röntgen 1880’s study. The authors charged the surface of an acrylic membrane

using a corona discharge and reported that the material endured much higher voltages and, thus, presented higher deformations than the currently used system. They suggest that this could lead to new actuator designs and complex 3D geometries [7].

As a result of the relatively soft polymer composition and large strain response shown by DEAs, especially those with rolled structures, this type of electronic EAPs are also known as “artificial muscles”. Thus they could find applications in biologically inspired robots, as well as human prosthetic or orthotic devices. Examples of DEA bio-mimetic robots include a fish-like propulsion airship [182] and inchworm robots [183], complex walking robots [184], transparent “active skins” [185] and motors for the eyeballs of an android face [186]. In fact, in 1999, Yoseph Bar-Cohen (Jet Propulsion Lab, NASA) created the “Arm-wrestling match of an EAP robotic arm against a human” challenge to demonstrate the potential of the EAP actuator technology in the field of robotics. An arm wrestling robot based on rolled DEAs presented by the Swiss Federal Laboratories for Materials Testing and Research (EMPA) in 2007 demonstrated very promising performances, however it still lost the contest against a human [178]. Although the promise of muscle-like actuation technology has not yet been completely fulfilled, the upsurge interest in the development of robotic devices spurred by DARPA Robotics Challenge will bring further developments in DEAs devices.

Finally, DEAs devices have already been commercialised by a subsidiary of Bayer MaterialsScience [3] on a haptic feedback for computer gaming (<http://www.vivitouch.com/>) and other applications include (Figure 18), refreshable Braille and tactile displays [187-189], motion capture suits [190], micro-pumps [191-194], loudspeakers [195], variable focus lenses [196-199], active isolation devices for cancelation of vibrations [200, 201] and even shape controllers of lightweight mirrors [202]. It is envisioned that, as DE materials and design improve, this technology will find further applications in the medical and industrial sector.



**Figure 18.** Potential applications of DEAs. a) Fish-like propulsion airship [203]. Reprinted with permission from Elsevier, copyright (2010). b) Overview of an electro-adaptive microfluidic device showing the fabrication steps involved [193]. Reprinted with permission from Springer Science+Business Media, copyright (2013). c) Schematic diagram of the proposed DE based Braille cell showing the single actuator and its components and their assembly in a Braille cell [187]. Reprinted with permission from Elsevier, copyright (2012). d) DE generator prototype designed to provide on-board power to a navigation buoy [204]. Reprinted with permission from Elsevier, copyright (2013).

## 7. Conclusions and outlook

The intrinsic properties of elastomers provide a good starting point for the development of electro-active polymers as transduction devices. They offer large actuation strains with high energy densities while providing lightness, noiseless, good damage-tolerance and low cost. This field attracted the attention of researchers back in the year 2000 and has since shown its potential in a wide range of applications, having already been commercialised in 2011.

However, a number of issues still need to be addressed; being their major drawback the high driving voltages required to obtain a large response. This review has comprehensively presented the progress on the different pursued approaches to improve the DE properties and solve this main issue.

Further developments on both the devices materials and configurations will definitely result in widespread implementation of DEAs in everyday applications. Additionally, the capacitive nature of DE devices not only enables mimicking muscle-like actuation but also transduce mechanical into electrical energy. Additional potential applications will arise from the current exploration of these systems as clean energy harvesters from renewable and environmental resources.

### **Acknowledgement**

The authors gratefully acknowledge the financial support of the Spanish Ministry of Science and Innovation (MICINN) through the project MAT 2010-18749.

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