Overview on lightweight multifunctional materials

Carlos M. Costa[1,2], Pedro Costa[1,3], Senentxu Lanceros-Mendez[4,5]

[1]Centro de Física, Universidade do Minho, 4710-057 Braga, Portugal

[2]Centro de Química, Universidade do Minho, 4710-057 Braga, Portugal

[3]IPC – Institute for Polymers and Composites, Universidade do Minho, Campus de Azurém, 4800-058 Guimarães, Portugal

[4]BCMaterials, Basque Center for Materials, Applications and Nanostructures, UPV/EHU Science Park, 48940 Leioa, Spain

[5]IKERBASQUE, Basque Foundation for Science, 48013 Bilbao, Spain

Abstract: Lightweight multifunctional materials represent an increasing field in materials science and engineering based on their technological applications in a wide variety of areas ranging from sensors and actuators, materials for structural and environmental applications, energy generation and storage, or biomedicine, among others. This chapter presents an overview on the main types, preparation techniques and applications of the most relevant lightweight multifunctional materials, as well as on relevant materials to be applied and/or implemented in lightweight structures.

Keywords: lightweight; smart materials; multifunctional materials; sensors; actuators; applications

Index

1. Introduction

Multifunctional materials as smart materials while, simultaneously, maintaining structural functions are increasing over the years in both academic and industrial areas [1, 2]. Novel developments in the overall properties of materials (physical and chemical modifications), processing methods and integration into applications allows to specifically design materials for wide variety of applications [2].

Considering the potential of these materials, the number of publications in this field has been strongly increasing in recent years, as shown in Figure 1.

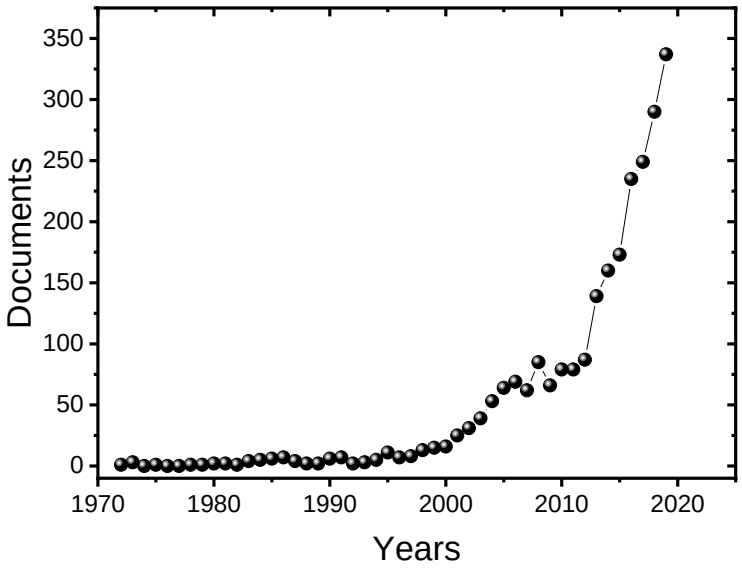

Figure 1 - Number of publications about lightweight multifunctional materials. *Source*: Scopus (keywords: Lightweight and polymer composites).

Additive manufacturing and related manufacturing technologies allow, nowadays, the development and implementation of novel materials for a variety of application, being one area of interest structural health monitoring (SHM), in which structural reinforcement in combined with non-structural functions [2]. Multifunctional structural materials are materials allowing both structural function and another (non-structural) function without the need for external devices. Structural function, including tailored strength, stiffness, fracture toughness and damping, can be combined with non-structural properties, such as the capability of providing noise and vibration control, electromagnetic interference (EMI) shielding, structural health monitoring, self-repair, thermo-chromism, self-cleaning, antibacterial, or energy harvesting/storage [2].

Multifunctional lightweight materials are a concept allowing significant improvement in the overall efficiency of the total space system, improving performance and environmental friendliness [3]. Lightweight materials are typically considered materials using polymers as host matrices or/and composites reinforced with fillers in order to address the specificity of each application or device. With polymer composites as material of choice, the intrinsic properties of the polymers (e.g., lightweight, flexibility and easy processing, etc.) can be combined with the unique properties of their reinforcements, such as electrical conductivity, high dielectric properties, or high mechanical stiffness and strength [4]. Polymer materials range from elastomers and thermoplastics to thermosets with a wide range of physico-chemical properties that can be tailor-made for applications. Further, in a broad sense, lightweight materials can also include inorganic high-density material that can be used in small amounts in lightweight structures to provide specific functions such as structural health monitoring or self-cleaning, without interfering in the lightweight characteristics of the overall structure.

This chapter will provide an overview on the multifunctionality of polymer composites reinforced with specific fillers (from micro- to nanofillers with specific morphologies) for energy, shielding, sensor or actuators materials. It does not pretend to be a comprehensive account of the field, which will be provided in the following chapters, but to highlight some of the main issues and concepts of this challenging and increasingly relevant field.

2. Multifunctional materials classification and types

Polymer-based material capable of providing structural and non-structural functions without the need of embedding or attaching external represent one of the main types of multifunctional structural material. This type of polymer-based materials can be classified by the following non-structural functions (Figure 2).

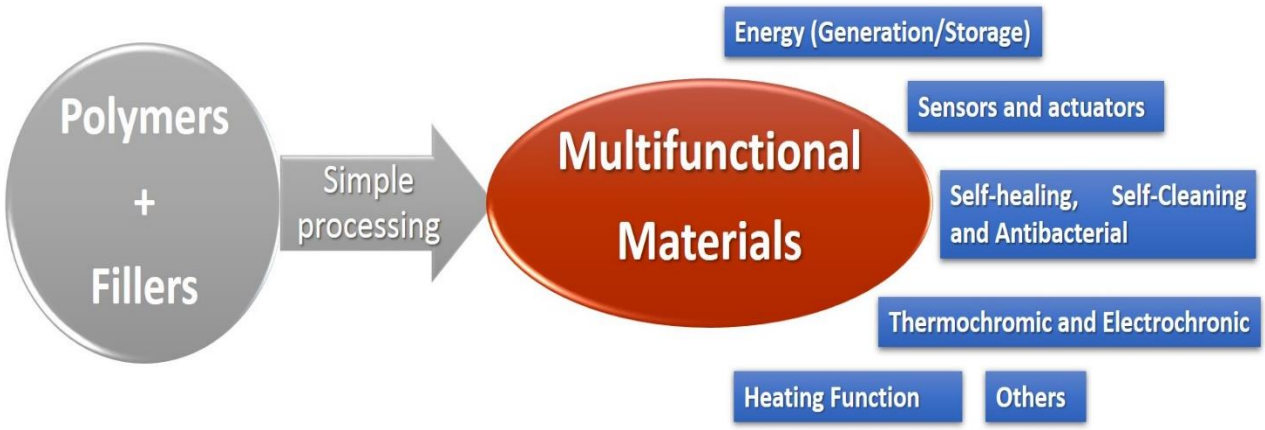

Figure 2 – Multifunctional lightweight materials representative application areas.

2.1. Energy generation and storage

Energy generation or harvesting refers to the conversion of mechanical, thermal or solar energy into electrical energy. Polymer-based materials for energy harvesting allow supplying energy to microdevices, replacing capacitors or batteries, reducing weight in the structures.

Piezoelectricity [5], thermoelectricity [6] and dielectric elastomer generators [7, 8] are some example of electrical harvesting mechanisms to generates energy. Triboelectric effect can be described as the electrification of two different objects or materials during friction, resulting in a remarkable flow of electrons from one material to the other, balancing the potential difference between both materials. The most extensively polymers used in piezoelectric energy harvesting applications are the ones form the poly(vinylidene fluoride) (PVDF) family (mainly PVDF-TrFE and PVDF-HFP), due to its higher output performance [9] and physico-chemical stability [9]. Conductive (carbonaceous or metallic) and ceramic nanofillers are also used as reinforcement materials, as well as blends with conductive polymers [9]. Carbon nanotubes and graphene as nanocarbonaceous, silver nanowires and zinc oxide particles as metallic

particles and barium titanate as ceramic are the most used materials for applications [9-11]. PVDF and copolymer or composites can generate some tens of μW output power for piezoelectric devices, 42 V output voltage for pyroelectric and tens of μW in magnetoelectric devices [9]. The peak output power density of some triboelectric devices has been reported to be as high as 500 W/m^2, through power conversion efficiencies up to 85% [12].

There are two main different ways to storage electrical energy: capacitors or batteries [1]. Typically, capacitors are more used in high power systems and batteries for lower power ones. Among capacitors, supercapacitors are electrochemical devices without the need of anode or cathode, as batteries. Although both of them need a separator to prevent the contact between the two electrodes and the electrolyte (ionic conductor).

Regardless of the energy storage system, lightweight materials are used as separator / electrolyte placed between the electrodes, the most used types being single polymers, polymer composites and polymer blends.

There are different polymer types used for this application and the most widely used , from synthetic to natural polymers, are poly(propylene), PP poly(acrylonitrile), PAN, poly(vinylidene fluoride), PVDF, polyimide (PI), poly (ether-ether-ketone) (PEEK), cellulose, chitin and poly(vinyl alcohol) (PVA) [9].

2.2. Sensing capabilities

The sensing function is essential in lightweight materials for structural health monitoring, wearables or materials for biomedicine [13]. As an example, in the area of structural health monitoring, strain/stress, bending or damage are monitored in the structure by multifunctional materials with resistive, capacitive, inductive and magnetic, acoustic and piezoelectric capabilities [4].

Resistive sensors are materials whose their electrical resistance is affected by a particular physical quantity [4]. The resistance can vary as a function of changes in material properties, geometry or a combination of both. Quantities typically measured using resistive effects include temperature, light, deformation and magnetic field strength, among others. Tailoring the materials properties, resistive sensors have been also used for measuring force, torque, pressure, distance, angle, velocity and acceleration [4].

In the following, potentiometric, strain gauges, piezoresistive and magnetoresistive will be briefly discussed, as well thermal and optical sensors based on resistive effects. Potentiometric, strain gages, piezoresistive, magnetoresistive, thermoresistive and light

dependent resistor (LDR) are typical sensor to measure the distinct stimulus applied to a material.

2.1.1. Potentiometric sensors

Potentiometric displacement sensors can be linear or angular, consisting in a main materials body which is either wire wound or covered with a conductive film.

In an ideal potentiometer, the voltage ratio (VR), defined as the ratio between the wiper voltage and the full voltage across the resistor is equal to x/*L*, where x is the distance between the initial and wiper positions and *L* is the electrical stroke. To measure the position of the wiper, the sensor is connected to a voltage source V_i with source resistance R_s; the output voltage on the wiper, V_o, is measured by an instrument with input resistance R_i. Ideally, the voltage transfer V_o/V_i equals the VR. Assuming that $R_s = 0$ and $R_i = \infty$ the output voltage of the sensor is, respectively, for linear (Eq. 1) and angular (Eq. 2) potentiometer given by:

$$V_0 = \frac{x}{L} V_i \tag{1}$$

$$V_0 = \frac{\alpha}{\alpha_{max}} V_i \tag{2}$$

where *L* is the total electrical length and α_{max} the maximum electrical angle.

Potentiometer applications include linear and angular displacements and is widely used in all kind of mechatronic systems to obtain information about angular positions of rotating parts. The potentiometers can also be used for the measurement of acceleration, force, pressure and level [4].

2.1.2. Piezoresistive sensors

Piezoresistive sensors are based on the electrical resistivity variation of one material when this is deformed (tension, compression or bending mode). This phenomenon was discovered in 1856 by William Thomson observing the variation in resistance with elongation in iron and copper [14]. Numerous materials show piezoresistive effect, but only those with a appropriate sensitivity are suitable to be applied in sensors [15]. Strain gages [15], polymer-based composites [16] and semiconductor [17] materials are the materials used for sensing using the piezoresistive effect. Semiconductors germanium and silicon were the first materials widely used as piezoresistive materials, reported the first

measurements of large piezoresistive coefficients in these semiconductor crystals in 1954 [14]. Semiconductor materials show low mechanical properties (fragile, small deformations) with larger piezoresistive sensibility [14, 18].

The piezoresistive sensibility of the material can be determined by the Gauge Factor (*GF*), which can be expressed in relative resistance change per unit of strain:

$$GF = \frac{dR/R}{dl/l} \tag{3}$$

where dR/R is the relative electrical resistance and dl/l is the relative deformation [16]. The GF ha two main contributions in materials under strain [15]:

$$GF = \frac{d\rho/\rho}{dl/l} + 1 + 2\nu \tag{4}$$

Where the Poisson coefficient, ν, of the material determines the geometric coefficient $(1 + 2\nu)$ and is always present in the sensors. The intrinsic resistance variation can be present in piezoresistive sensor or not, depending on the sample or type of measurement. An ideal rubber material shows a ν= 0.5, corresponding a *GF*= 2, as the highest geometric factor in the equation 4. Most metals or polymers show 0.2< ν <0.35 [15, 19]. Typically, the *GF* of strain gages (metallic filaments deposited over a plastic substrate) changes from 2 to 2.5 and is larger as 200 for semiconductor materials [4, 15, 19].

Piezoresistive sensors are suitable for the measurement of all kind of force-related quantities, for example normal and shear force, pressure, torsion, bending and stress. Polymer-based materials with large strain (elastomer-like matrices) are capable to measure from low to large strains, complementing the strain gages typical range of strains, with large piezoresistive sensibility [20-23]. Piezoresistive sensors with strain larger than 50% and piezoresistive sensibility similar to semiconductor crystal have been achieved [24].

2.1.3. Capacitive sensors

A parallel plane capacitor is defined as an insulator material between two conductive electrodes. The dielectric properties of the insulator and geometry of both electrodes determine the capacitance (C) of the sensor that for a parallel plate capacitor is given by:

$$C = \varepsilon_0 \varepsilon_r \frac{A}{d} \quad (5)$$

where ε_0 permittivity of vacuum, ε_r is the dielectric constant of the material, A is area of the conductive electrodes and d the thickness of the dielectric material.

Capacitive sensors for displacement and force measurements have several advantages, being very robust and stable and applicable at high temperatures and in harsh environments. Capacitive displacement sensors commonly form part of instruments measuring pressure, sound, or acceleration [25].

The main advantages of lightweight materials in capacitive sensors are their flexibility and stretchability, easy preparation and the possibility to be implemented in curves surfaces or unconventional geometries.

Highly sensitive capacitive pressure sensors have been developed based on highly porous boron nitrate (BN)/polydimethylsiloxane composite foams (BNF@PDMS), this lightweight composite showing excellent mechanical resilience, extremely high compressibility (up to 95% strain), good cyclic performance, and superelasticity [26].

Carbon black /polydimethylsiloxane composites were produced for pressure sensors showing a sensitivity exceeding 35 kPa^{-1} for pressures <0.2 kPa [27]. Capacitive sensors have been also developed based on polyvinylidene fluoride (PVDF) and polydimethylsiloxane (PDMS) polymers with different fillers from conductive (CNTs, graphene, carbon black) to ceramic fillers (zinc oxide (ZnO), barium titanate (BaTiO3) and lead zirconate-titanate (PZT)) [28].

2.1.4. Piezoelectric sensors

The Greek word *piezein* meaning “to press” was adopted to describe the piezoelectric phenomenon. Piezoelectricity defines the ability of certain materials to develop an electrical potential variation under the application of a mechanical stimulus. The piezoelectric effect is divided into direct and the inverse and describes the capability of a material to transform mechanical into electrical energy and electrical into mechanical energy (Figure 3).

Materials with piezoelectric properties are crystals, ceramics and polymers. Crystalline dielectric materials composed by polar structures without a centre of symmetry will present piezoelectric behaviour. The direct and inverse piezoelectric effect are both characterized with electric charges movement in the material. direct effect is when a external stress is applied to a material, consequently the electrical dipoles in the crystal are oriented and the crystal develops positive and negative charges on opposite surfaces, resulting in an electric field within the material [10]. The inverse piezoelectric effect is the application of an electric field on a material, leading to a reorientation of the dipolar moments and, as a consequence, a deformation of the material, the strain being directly proportional to the electric field [10]. Depending on the polarity of the applied field, the strain of the piezoelectric material is extensive or compressive.

The direct piezoelectric effect was discovered by the Curie brothers in 1880 [10], while the inverse effect was deduced theoretically from the fundamental principles of thermodynamics in 1881 by Lippmann and confirmed in laboratory by the Curie brothers [10]. The piezoelectric effects, direct and inverse, are described respectively by the following constitutive equations [16]:

$$D_i = d_{ikl}T_{kl} + \varepsilon_{ik}^{T}E_k \tag{6}$$

$$S_{ij} = s_{ijkl}^{E}T_{kl} + d_{ikl}E_k \tag{7}$$

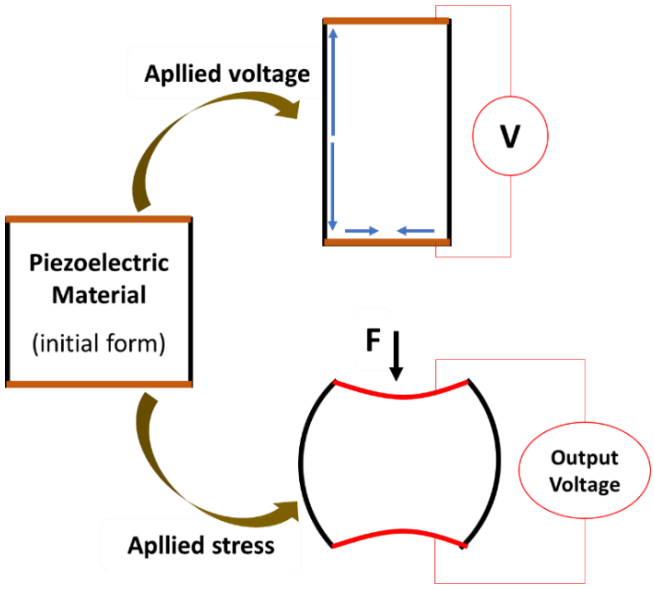

Figure 3 - Piezoelectric behaviour under force application.

Where the subscripts *i, j, k*, and *l* take values of 1, 2, and 3. The *S* and *T* are the strain and stress tensors, respectively, and *D* and *E* are the electric displacement and electric field vectors, respectively. Further, *s* is the elastic compliance matrix (at constant electric field), *d* the piezoelectric strain coefficient, and ε the permittivity values (at constant stress). In both piezoelectric effects, *d* represents the charge variation induced by a given force without electric field, or the geometric change caused by an applied voltage without an applied force [10].

The direct piezoelectric effect is used for sensor applications and the invers effect is typically used for actuators. The application areas are communications and control, industry, health and portable electronics [10]. Radio, television or radars, transducers, sensors or actuators, sonars, pumps, or motors are some examples of the interest in this type of materials [10]. Nowadays, these materials are largely implemented in microelectromechanical (MEMS) devices, smarts structures or biomimetic devices.

The most used piezoelectric coefficients for applications are the transverse and longitudinal coefficients, d_{31} and $d_{33,}$ respectively. The transverse coefficient, d_{31}, describes the electric polarization produced in a direction perpendicular to the applied stress in material. The longitudinal coefficient, d_{33}, defines the electric polarization produced in the same direction of the applied stress [29].

The conversion efficiency between electrical and mechanical energy (and reverse effect) are known as electromechanical coupling, *k*, being closely related to the bandwidth of resonant devices, and it is expressed as [29]

$$k_{ij} = \frac{d_{ij}}{\sqrt{\varepsilon_{ii}^{\sigma} S_{jj}^{E}}} \tag{8}$$

The coupling factor is related to vibration modes and can be measured experimentally from the resonance and antiresonance frequencies observed in the piezoelectric elements.

Materials with piezoelectric properties can be natural or synthetic. The most common natural piezoelectric material is quartz (SiO_2) [10]. Synthetic piezoelectric materials are more efficient than natural, and the most used ones are ceramic materials.

The piezoelectric polymer with the highest piezoelectric coefficient and dielectric constant is poly (vinylidene fluoride) and its copolymers (poly (vinylidene-co-

trifluoroethylene) fluoride (PVDF-TrFE) and poly (vinylidene-co-hexafluropropylene fluoride) (PVDF -HFP))) [11].

Regarding piezoelectric ceramic materials, the most used are Barium titanate ($BaTiO_3$) [30] and Lead zirconate titanate (PZT) [31].

Natural piezoelectric materials can be divided into two main groups: the mineral group, including quartz, Rochelle salt, topaz, and tourmaline, among others, and organics groups, represented by silk, wood, enamel, dentin, bone, hair, and rubber, among others [15].

Table 1 shows the density (ρ), dielectric constant ($\varepsilon/\varepsilon_0$) and the piezoelectric coefficient (d_{33}) of representative synthetic and natural piezoelectric materials.

Table 1 - Density, dielectric constant and piezoelectric coefficient for representative piezoelectric materials [32, 33].

Material	**ρ / g.cm^{-3}**	**$\varepsilon/\varepsilon_0$ at 1kHz**	**d_{33} / pC.N^{-1}**
Quartz	2.65	4.4	-2
Rochelle salt	1.77	350	275
PVDF	1.78	13	~ -20 - -30
PZT	7.5	1200	60-130
$BaTiO_3$	5.7	1700	149

Regarding bio-based organic piezoelectric materials, among the most prominent are Lysozyme, nucleobases, thymine, α-glycine, β-glycine and hydroxy-L-proline with piezoelectric coefficients up to 1 pC/N to 2 pC/N, in some cases [34].

2.1.5. Magnetoelectric sensors

Magnetoelectric (ME) materials respond with an electrical response to a variation of the magnetic field [35]. In last two decades, ME composite materials, combining ferroelectric and ferromagnetic components [36], have gathered increasing attention due their giant ME coupling at room temperature, allowing technological developments in the area of magnetic sensing [36]. Magnetic sensors have as main characteristic the wireless activation and response [35, 36], being used in electrical motors, generators, memories and biomedical devices, as well as in different energy fields [37, 38]. The work principle of ME composite materials is based on two distinct steps: when a magnetic field is applied

to a ME material, the ferromagnetic component suffers a distortion due to magnetostriction that is transferred to the ferroelectric component, inducing an electric charge through the piezoelectric effect.

The ME effect was predicted by Pierre Curie in 1894 and first observed in single crystals 1961 [36], and it evolved to particulate composites, to laminated composites, and finally to micro-/nano-thin films [36], as shown in Figure 4.

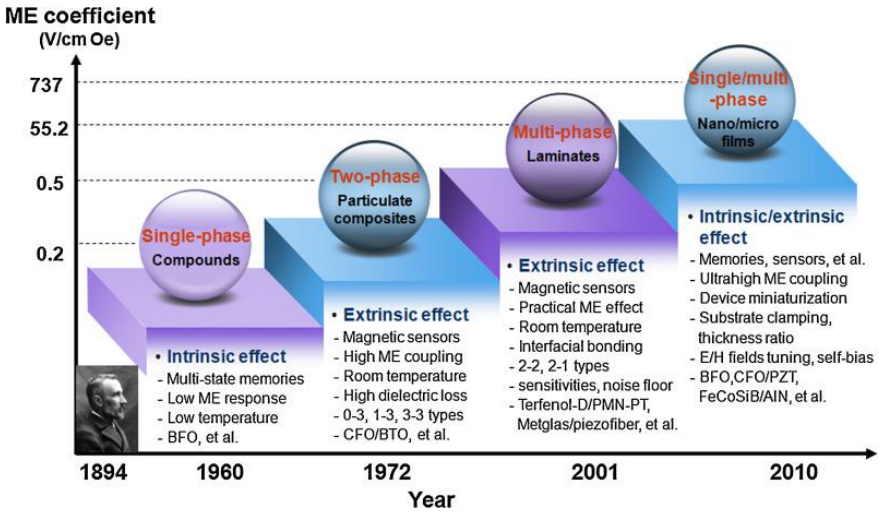

Figure 4 - Evolution on the development of ME materials: from single-phase compounds to multi-phase ferromagnetic/ferroelectric composites. Adapted from [36].

The overall properties of ME materials has been addressed in single-phase materials, rare-earths and nano/micro particles in composites, for laminate and particulate composites [36]. The high ME coupling has not yet been found in single-phase materials and most of them have low Curie temperatures [36]. Particulate composites show low ME coefficients and high dielectric losses, being difficult their use as sensing materials [36]. With laminate composites these limitations were overcome, with ME voltage coefficients up to 4.7 V/cm×Oe [36].

The most used materials in ME composites are magnetic ferrites, piezoelectric ceramics, magnetic alloys, piezoelectric materials (polymers or ceramics) and magnetic alloys [36].

2.2. Actuators

Actuators are materials that respond with a mechanical deformation when certain stimulus are applied: thermal, electrical or magnetic [1, 39, 40].

In the case of lightweight materials, the inverse piezoelectric effect is used to develop material actuators, using mostly PVDF as polymer or reinforced with high-dielectric materials [41].

Dielectric elastomer actuators (DEA) are a class of electroactive polymers that are capable of changing size and shape when an electric field is applied. This materials have found applications in artificial muscles, micro-robotics and micro air vehicles at industries [39].

Materials that exhibits a large reversible volume transition at a critical temperatures include also soft temperature-sensitive hydrogel [42]. Those materials have been applied by 3D printing of hydrogels that are both mechanically robust and thermally actuating [42]. Electroactive actuators based on poly (vinylidene fluoride) with IL have been intensively studied due to the low voltage response, easy processing and tailorable response by selection of polymeric matrix, anion and cation present in the IL [43]. It was observed that the cation alkyl chain size of the IL affects the electrical conductivity and consequently the actuator performance [43]. In addition, it was demonstrated for the IL 1-ethyl-3-methylimidazolium bis(trifluoromethanesulfonyl)imide, [Emim][TFSI], the effect of different polar fluorinated polymer matrix, the best polymer matrix with high actuator performance being poly(vinylidene fluoride-co-trifluoroethylene) (PVDF-TrFE) due to the low degree of crystallinity and high β-phase content [44]. These ILs based actuators can be suitable for a variety of applications, from electronic to biomedical applications.

2.3. Self-healing, Self-Cleaning and Antibacterial

Self-healing is the property of some materials have of recovering when damaged [45]. This property is very interesting because it extends the useful life and safety of the devices in which these materials are applied in a wide range of applications, from structural to biomedical applications. Self-healing materials are typically divided into polymeric and composite materials [46]. The most used polymers for this application are epoxy-based materials in which the resin and the hardener are mixed to cure the crack [47]. Poly(methyl methacrylate) (PMMA) and poly(methyl methacrylate-comethyl

ethylacrylate) (MMA-MEA copolymer) have been also used in this area [48]. Recently, a new regenerative polymer complex composed of poly(2-acrylamido-2-methyl-1-propanesulfonic acid), polyaniline, and phytic acid has been developed [49]. In addition, self-healing composites based on a polymer matrix with encapsulating healing materials, i.e., capsules containing healing agent where the capsules release the healing agent due to cracks formation [47]. Examples, of healing materials are tungsten chloride (WCl_6) [50], and dicyclopentadiene (DCPD) [51], among others.

The self-cleaning property is often inspired by biological materials such as lotus leaf, among others, an is related to the hydrophobic and hydrophilic behavior, finding applications in textiles, glass, and coatings [52].

Self-cleaning can be provided by surface morphology, as in the lotus leaf, or by chemical action, such as the one provided by self-cleaning composites based on TiO_2, ZnO and SiO_2 fillers dispersed on various polymeric matrices, such as poly (m-phenylene isophthalamide) (PMIA), poly (diallyldimethyl ammonium chloride) (PDDA), or chitosan, among others [53].

Antimicrobial property is the ability to inhibit or kill bacteria due to antimicrobial resistance [54, 55]. Polymers with this property are divided into passive and active polymers [56], and stand out poly (sulfobetaine methacrylate), poly (dimethyl acrylamide), polyphenols, acrylic based on poly (n, n-diethylene diamine-coyrosol), and polyurethane containing quaternary ammonium [57]. This property can be controlled through parameters such as molecular weight, chain length, functional groups, or hydrophobicity / hydrophilicity character [58].

2.4. Thermochromic and Electrochromic

Chromogenic are a family of materials changing their colour in response to some stimulus. In thermochromic and electrochromic materials the stimuli are temperature and electric field, respectively [59]. Thermochromic smart materials could reversibly vary colours and optical properties in response to a temperature change. These materials dynamically modulates the solar heat gain with environmental temperature, i.e., absorption differentially solar energy during the hot and cold seasons, respectively [60].

Thermochromic materials are widely used in asphalt and building constructions (windows and roofs) in order to maintaining controlled temperature in extreme conditions, saving

the equipment's or saving energy in climatizations [60-62]. The common used materials are titanium [60] and vanadium dioxide [59] coatings as coating or scattering materials.
Electrochromic materials are in an advanced states compared with thermochromic materials and are commercially available nowadays, used in innovative buildings [59]. The electrochromic materials include porous oxide films, typically tungsten and nickel oxides [59]. These materials are embedded between transparent and conductive materials, and, under some voltage can change their color.

2.5. Heating function

The Joule effect is explaining the conversion of electrical into thermal energy is commercially used in polymer-based composite materials for anti-icing and de-icing in aerospace, wind turbines and other structures [63].
Anti-icing prevents ice to accumulate on the object while de-icing removes the ice layer from the surfaces. The strategies can also be divided into active and passive. Passive methods use the physical properties of the surface to eliminate or prevent ice, whereas active methods use external systems [63, 64].
Carbon materials, such as, carbon black, carbon nanotubes, and graphene are more suitable as resistance heating element compared with metals, due to their electrical resistivity being higher than that of metals by some orders of magnitude [1]. Due the high electrical resistance of the carbon materials a heating element can be in a bulk form and not in a specific form as it is needed for metallic materials [1, 64].
These carbon materials have been used to prepare polymer composites with different polymer matrix such as, poly(amide12–b–tetramethyleneglycol) (PEBAX) [65], polyurethane (PU) [66], polycarbonate (PC) [67], Nylon 6 (PA6)/PVDF blends [68] in which the thermal conductivity depends on the type, content and dispersion of conductive nanoparticles in polymer matrix.

3. Lightweight multifunctional materials

Lightweight multifunctional materials are increasingly being developed based on different single, blends and/or composite type polymers, from elastomers to conductive polymers, and applied in different areas including transportation and aircraft / aerospace, biomedical, energy storage and harvesting devices, among others [69]. Such polymer-based materials may have two or more functionalities that result from the combination of functions including actuation, sensing, shape morphing, optical modulation, self-heling, self-cleaning or stiffness variation. As an example, piezoelectric polymers with titanium dioxide (TiO_2) allow to develop piezoelectric sensors and actuators photocatalytic behaviour [70]. Typically, the impact of the polymer materials on multifunctionality can be divided in structural and functional properties, where the polymer material may have a structural function as a polymer matrix for composites or functional properties as in the case of conductive polymers through their electrical properties.

The stages of development of these materials are divided into: composition, processing, properties and performance [69-71]. Figure 5 shows the multifunctional polymer materials types and the function of each component.

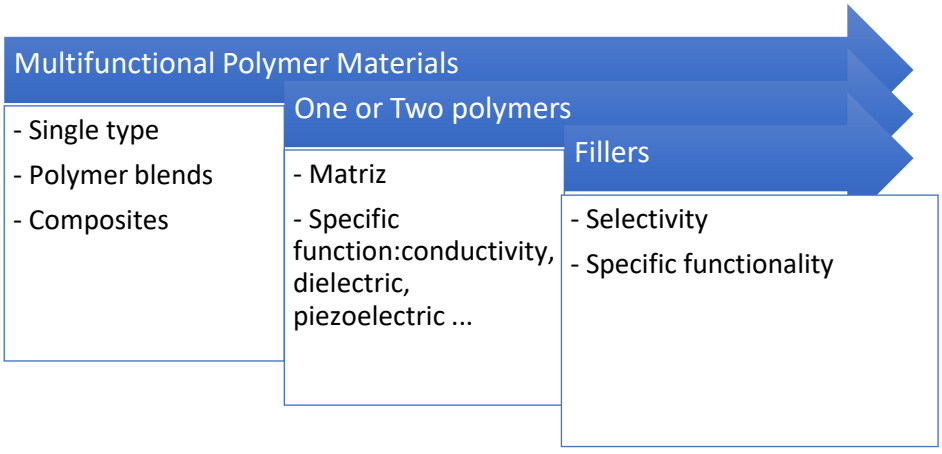

Figure 5 – Multifunctional polymer materials: components and each function.

These multifunctional materials based on natural or synthetic polymers can be custom designed, taking into account processing technology and applications in which their specific morphology and properties are controlled, with the following advantages:

- Solubility in many solvents;
- High thermal stability and long-term stability;
- Control of electrical properties;
- Excellent mechanical properties and their stability;

- Good adhesion on different surfaces;
- Low optical loss.

Given its advantages and applicability in the emerging area of the Internet of Things (IoT) concept [72], where easy-to-process, facile integration, and low-cost multifunctional materials are required, printing technologies of multifunctional materials [73, 74] are expected to play an increasing relevant role in this field.

4. Conventional preparation techniques

Multifunctional polymeric materials can be processed by different production methods, from coating techniques [75] to printing methods [76], as well as extrusion [77] or injection moulding techniques [77] for the processing of specific films, filaments or component. The main difference between coating and printing technologies is that printing techniques allow the production of more complex structures with less waste generation, where specific technique showing advantages and limitations, taking into account scalability and cost, output, resolution and size [78, 79]. Coating techniques are simple and inexpensive, with control over film thickness, whereas printing techniques are more complex and stand out for obtaining high resolution and patterned films. The most commonly used coating and printing techniques are doctor blade and spin-coating, and screen printing and inkjet technique, respectively [80].

4.1. Doctor blade and spin coating

The doctor blade technique is one of the most widely used for production of multifunctional polymer materials films, the film thickness being controlled by the gap size between the blade and the substrate. It can be applied on an industrial scale [81].
This procedure is shown schematically in Figure 6a. The solution is cast in front of the blade, which is dragged parallel to the substrate, leaving a film with a well-defined thickness. This technique allows high control over the film thickness (d), following the expression:

$$d = \frac{1}{2} w \frac{c}{\rho} \tag{9}$$

where w is the gap width between the blade and the substrate, c is the concentration of solids in the solution, and ρ is the film density. Typically, the final film thickness is almost half of the gap between the blade and the substrate due to solvent evaporation. The surface

tension of the solution is an important factor influencing the film processing. To ensure good film formation, the surface tension of the solution should be less than the surface energy of the substrate [82].

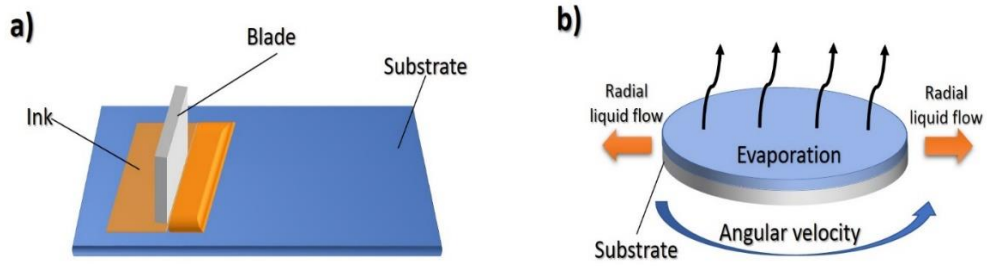

Figure 6 – Schematic representation of a) doctor blade and b) spin-coating techniques.

The main advantage of this technique is the reduced generation of polymer waste, below 5% [83].

The spin-coating technique is based on substrate rotation with solution deposition to create homogeneous films with well-defined thickness and high structural uniformity [84]. This procedure is depicted in Figure 6b.

This procedure is divided into two steps: first, the solution is casted onto a substrate at a low rotational speed to spread the solution on the substrate to prevent air bubbles. Then, after completing this step, the substrate is accelerated to a specific angular velocity, the acceleration leading to the ejection of most of the solution, leaving only a thin homogeneous layer on the substrate.

The thickness of the film (d) is controlled by the applied angular velocity according to the equation:

$$d = k\omega^{\alpha} \tag{10}$$

where ω is the angular velocity of the substrate, and k and α are empirical constants, related to the characteristics of the used materials [85].

The thickness of the film also depends on the solution viscosity, polymer concentration and solvent volatility.

4.2. Screen and inkjet printing

Screen printing is a very used printing technique in which inks are forced to pass through a screen over a wide range of ink viscosities [86]. The main procedure is shown in Figure 7a).

A mesh of woven material is glued to a frame and placed above the substrate. This screen is filled with an emulsion impermeable to the coating solution. Some areas of the screen are kept open without the emulsion to form a pattern of gaps that are permeable to the ink. The screen is then filled with the coating solution, and a squeegee is used to force the ink trough the gaps, leaving a printed pattern on the substrate. Thus, the squeegee characteristics, as material, shape, or angle of contact, have a significant influence in the printed film [87].

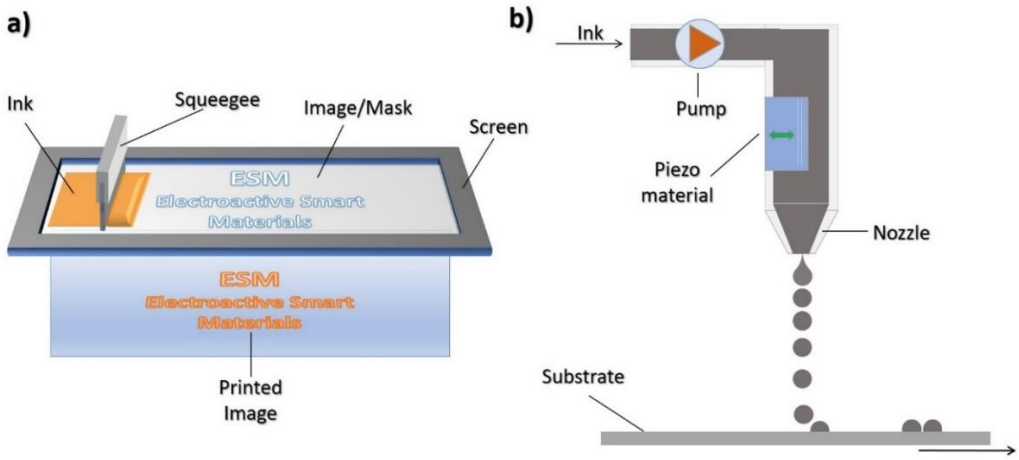

Figure 7 – Schematic representation of the a) screen-printing technique and b) inkjet-printing drop on demand by piezoelectric actuation.

This technique allows the production of thin films with two-dimensional printed patterns where the resolution of the printed films is limited to the mesh size and resolution and ink viscosity [88].

The inkjet printing technique is considered to be one of the most flexible techniques for obtaining high definition patterns. A cartridge is filled with the ink and placed in a printer which is set to print the desired pattern on the substrate. During printing, the ink flows into a chamber that is retracted, leading to the ejection of ink droplets through a nozzle into the substrate as shown in Figure 7b. The chamber retractions are controlled by pulses, allowing the design of the pattern and, after film printing, must be placed in an oven to evaporate the solvent [89].

5. Application areas

Lightweight multifunctional materials are applied on a wide range of technological applications including energy storage, production and conversion, water treatment, air purification, drug delivery systems, health monitoring, biomedical applications, structural health reinforcement and monitoring, among others, some representative ones being presented in Figure 8.

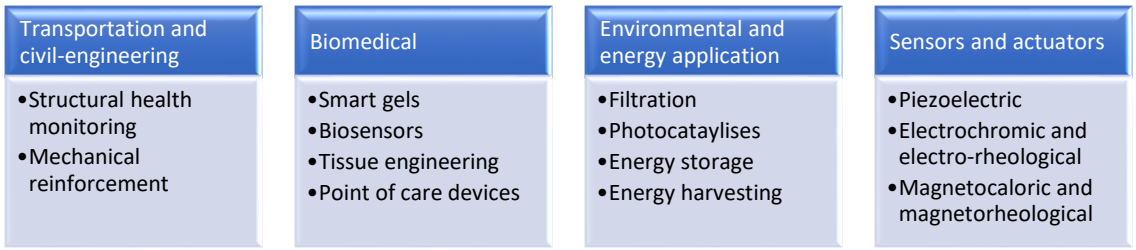

Figure 8 – Main representative applications of lightweight multifunctional polymer materials.

For these applications, these lightweight multifunctional mainly polymer-based materials are engineered to show different responses, depending on an external stimulus and/or physical/chemical process, readjusting their functionality, and performing sensing or diagnosis.

For civil and transportation applications, in addition to structural health monitoring and mechanical reinforcement, the additional properties that materials may have include cooling/heating regulation, UV and humidity resistance, air-cleaning, self-healing, antibacterial or self-cleaning [90]. For biomedical application, these materials in addition to being applied in various areas, they may include additional sensory functions such as hearing, somatosensitivity, thermoception, or nociception[91]. In relation to environmental and energy applications, green chemistry is essential to obtain more sustainable materials with the production more efficient energy generation and storage. On sensors and actuators, the materials are essential for applications including electronic skins or advanced wearable devices where advances in flexible, highly sensitivity, and transparent multifunctional materials are essential.

6. Conclusions

Lightweight multifunctional materials offer great opportunities for fundamental and engineering research and are increasingly being applied to a variety of. There exist many lightweight multifunctional materials types based on single polymer, blends with properties that can be specifically tailored for advanced applications.

These materials require an interdisciplinary approach based on chemistry, physics, biology, and materials science to optimize the path from optimization of the multifunctional response, to processing to integration into applications.

For new applications and technologies, new directions for research and development in this field are being developed through advanced fabrication procedures, multi-functionality integration, new architectures, and advanced composites, as it will be presented in the following chapters of this book.

Acknowledgments

The authors thank the FCT (Fundação para a Ciência e Tecnologia) for financial support under the framework of Strategic Funding grants UID/CTM/50025/2019, UID/FIS/04650/2019; and supported by FEDER funds through the COMPETE 2020 Programme under the project number PTDC/FIS-MAC/28157/2017 and POCI-01-0145-FEDER-007688. The authors also thank the FCT for financial support under grants SFRH/BPD/112547/2015 (C.M.C.) and SFRH/BPD/110914/2015 (P. C.). Financial support from the Basque Government under the ELKARTEK, HAZITEK and PIBA (PIBA-2018-06) programs is also acknowledged.

References

1. Chung, D.D.L., *A review of multifunctional polymer-matrix structural composites.* Composites Part B-Engineering, 2019. **160**: p. 644-660.
2. Gibson, R.F., *A review of recent research on mechanics of multifunctional composite materials and structures.* Composite Structures, 2010. **92**(12): p. 2793-2810.
3. De Sciarra, F.M. and P. Russo, *Experimental Characterization, Predictive Mechanical and Thermal Modeling of Nanostructures and Their Polymer Composites*. 2018: Elsevier Science.
4. Friedrich, K. and U. Breuer, *Multifunctionality of Polymer Composites: Challenges and New Solutions*. 2015: Elsevier Science.
5. Chen, J., et al., *Piezoelectric materials for sustainable building structures: Fundamentals and applications.* Renewable and Sustainable Energy Reviews, 2019. **101**: p. 14-25.

6. Guo, L. and Q. Lu, *Potentials of piezoelectric and thermoelectric technologies for harvesting energy from pavements.* Renewable and Sustainable Energy Reviews, 2017. **72**: p. 761-773.
7. Yang, Y., et al., *Enhanced energy conversion efficiency in the surface modified BaTiO3 nanoparticles/polyurethane nanocomposites for potential dielectric elastomer generators.* Nano Energy, 2019. **59**: p. 363-371.
8. Chiba, S., et al., *Consistent ocean wave energy harvesting using electroactive polymer (dielectric elastomer) artificial muscle generators.* Applied Energy, 2013. **104**: p. 497-502.
9. Costa, P., et al., *Recent Progress on Piezoelectric, Pyroelectric, and Magnetoelectric Polymer-Based Energy-Harvesting Devices.* Energy Technology, 2019. **7**(7): p. 1800852.
10. Nunes-Pereira, J., P. Costa, and S. Lanceros-Mendez, *3.9 Piezoelectric Energy Production*, in *Comprehensive Energy Systems*, I. Dincer, Editor. 2018, Elsevier: Oxford. p. 380-415.
11. Martins, P., A.C. Lopes, and S. Lanceros-Mendez, *Electroactive phases of poly(vinylidene fluoride): Determination, processing and applications.* Progress in Polymer Science, 2014. **39**(4): p. 683-706.
12. Dharmasena, R.D.I.G. and S.R.P. Silva, *Towards optimized triboelectric nanogenerators.* Nano Energy, 2019. **62**: p. 530-549.
13. Narayana, K.J. and R. Gupta Burela, *A review of recent research on multifunctional composite materials and structures with their applications.* Materials Today: Proceedings, 2018. **5**(2, Part 1): p. 5580-5590.
14. Barlian, A.A., et al., *Review: Semiconductor Piezoresistance for Microsystems.* Proc IEEE Inst Electr Electron Eng, 2009. **97**(3): p. 513-552.
15. Regtien, P.P.L., *4 - Resistive Sensors*, in *Sensors for Mechatronics*, P.P.L. Regtien, Editor. 2012, Elsevier: Oxford. p. 57-100.
16. Dios, J.R., et al., *Carbonaceous Filler Type and Content Dependence of the Physical-Chemical and Electromechanical Properties of Thermoplastic Elastomer Polymer Composites.* Materials (Basel), 2019. **12**(9).
17. Morris, A.S. and R. Langari, *Chapter 13 - Sensor Technologies*, in *Measurement and Instrumentation (Second Edition)*, A.S. Morris and R. Langari, Editors. 2016, Academic Press: Boston. p. 375-405.
18. Gonçalves, B.F., et al., *Green solvent approach for printable large deformation thermoplastic elastomer based piezoresistive sensors and their suitability for biomedical applications.* Journal of Polymer Science Part B: Polymer Physics, 2016. **54**(20): p. 2092-2103.
19. Falletta, E., et al., *Development of high sensitive polyaniline based piezoresistive films by conventional and green chemistry approaches.* Sensors and Actuators A: Physical, 2014. **220**: p. 13-21.
20. Dong, W.K., et al., *Piezoresistive properties of cement-based sensors: Review and perspective.* Construction and Building Materials, 2019. **203**: p. 146-163.
21. Niu, D., et al., *Graphene-elastomer nanocomposites based flexible piezoresistive sensors for strain and pressure detection.* Materials Research Bulletin, 2018. **102**: p. 92-99.
22. Costa, P., et al., *Mechanical, electrical and electro-mechanical properties of thermoplastic elastomer styrene–butadiene–styrene/multiwall carbon nanotubes composites.* Journal of Materials Science, 2012. **48**(3): p. 1172-1179.
23. Teixeira, J., et al., *Piezoresistive response of extruded polyaniline/(styrene-butadiene-styrene) polymer blends for force and deformation sensors.* Materials & Design, 2018. **141**: p. 1-8.

24. Amjadi, M., et al., *Stretchable, Skin-Mountable, and Wearable Strain Sensors and Their Potential Applications: A Review.* Advanced Functional Materials, 2016. **26**(11): p. 1678-1698.
25. Yao, S. and Y. Zhu, *Wearable multifunctional sensors using printed stretchable conductors made of silver nanowires.* Nanoscale, 2014. **6**(4): p. 2345-2352.
26. Tay, R.Y., et al., *Lightweight, Superelastic Boron Nitride/Polydimethylsiloxane Foam as Air Dielectric Substitute for Multifunctional Capacitive Sensor Applications.* Advanced Functional Materials, 2020. **30**(10): p. 1909604.
27. Pruvost, M., et al., *Polymeric foams for flexible and highly sensitive low-pressure capacitive sensors.* npj Flexible Electronics, 2019. **3**(1): p. 7.
28. Xu, F., et al., *Recent Developments for Flexible Pressure Sensors: A Review.* Micromachines, 2018. **9**(11): p. 580.
29. Ramadan, K.S., D. Sameoto, and S. Evoy, *A review of piezoelectric polymers as functional materials for electromechanical transducers.* Smart Materials and Structures, 2014. **23**(3): p. 033001.
30. Acosta, M., et al., *BaTiO3-based piezoelectrics: Fundamentals, current status, and perspectives.* Applied Physics Reviews, 2017. **4**(4): p. 041305.
31. Adair, J.H. and E. Suvaci, *Submicron Electroceramic Powders by Hydrothermal Synthesis*, in *Encyclopedia of Materials: Science and Technology*, K.H.J. Buschow, et al., Editors. 2001, Elsevier: Oxford. p. 8933-8937.
32. Fraga, M.A., et al., *Wide bandgap semiconductor thin films for piezoelectric and piezoresistive MEMS sensors applied at high temperatures: an overview.* Microsystem Technologies, 2014. **20**(1): p. 9-21.
33. Schwartz, M., *Smart Materials*. 2008: CRC Press.
34. Guerin, S., S.A.M. Tofail, and D. Thompson, *Organic piezoelectric materials: milestones and potential.* NPG Asia Materials, 2019. **11**(1): p. 10.
35. Correia, D.M., et al., *Low-field giant magneto-ionic response in polymer-based nanocomposites.* Nanoscale, 2018. **10**(33): p. 15747-15754.
36. Wang, Y., J. Li, and D. Viehland, *Magnetoelectrics for magnetic sensor applications: status, challenges and perspectives.* Materials Today, 2014. **17**(6): p. 269-275.
37. Cui, J., et al., *Current progress and future challenges in rare-earth-free permanent magnets.* Acta Materialia, 2018. **158**: p. 118-137.
38. Chernenko, V.A., et al., *Chapter 1 - Fundamentals of magnetocaloric effect in magnetic shape memory alloys*, in *Handbook of Magnetic Materials*, E. Brück, Editor. 2019, Elsevier. p. 1-45.
39. Panahi-Sarmad, M., B. Zahiri, and M. Noroozi, *Graphene-based composite for dielectric elastomer actuator: A comprehensive review.* Sensors and Actuators A: Physical, 2019. **293**: p. 222-241.
40. Ceyssens, F., et al., *Actuators: Accomplishments, opportunities and challenges.* Sensors and Actuators A: Physical, 2019. **295**: p. 604-611.
41. Huang, X., et al., *High-k polymer nanocomposites with 1D filler for dielectric and energy storage applications.* Progress in Materials Science, 2019. **100**: p. 187-225.
42. Bakarich, S.E., et al., *4D Printing with Mechanically Robust, Thermally Actuating Hydrogels.* Macromolecular Rapid Communications, 2015. **36**(12): p. 1211-1217.
43. Correia, D.M., et al., *Ionic Liquid Cation Size-Dependent Electromechanical Response of Ionic Liquid/Poly(vinylidene fluoride)-Based Soft Actuators.* The Journal of Physical Chemistry C, 2019. **123**(20): p. 12744-12752.
44. Dias, J.C., et al., *Improved response of ionic liquid-based bending actuators by tailored interaction with the polar fluorinated polymer matrix.* Electrochimica Acta, 2019. **296**: p. 598-607.
45. Wu, D.Y., S. Meure, and D. Solomon, *Self-healing polymeric materials: A review of recent developments.* Progress in Polymer Science, 2008. **33**(5): p. 479-522.

46. Bekas, D.G., et al., *Self-healing materials: A review of advances in materials, evaluation, characterization and monitoring techniques.* Composites Part B: Engineering, 2016. **87**: p. 92-119.
47. Wang, Y., D.T. Pham, and C. Ji, *Self-healing composites: A review.* Cogent Engineering, 2015. **2**(1).
48. Yuan, Y., *Self healing in polymers and polymer composites. Concepts, realization and outlook: A review.* Express Polymer Letters - EXPRESS POLYM LETT, 2008. **2**: p. 238-250.
49. Lu, Y., et al., *Ultrastretchable Conductive Polymer Complex as a Strain Sensor with a Repeatable Autonomous Self-Healing Ability.* ACS Applied Materials & Interfaces, 2019. **11**(22): p. 20453-20464.
50. Kamphaus, J.M., et al., *A new self-healing epoxy with tungsten (VI) chloride catalyst.* Journal of The Royal Society Interface, 2008. **5**(18): p. 95-103.
51. Sharma, A., et al., *Effect of Self-Healing Dicyclopentadiene Microcapsules on Fracture Toughness of Epoxy.* Materials Today: Proceedings, 2018. **5**(10, Part 1): p. 21256-21262.
52. Liu, K. and L. Jiang, *Bio-Inspired Self-Cleaning Surfaces.* Annual Review of Materials Research, 2012. **42**(1): p. 231-263.
53. Ragesh, P., et al., *A review on 'self-cleaning and multifunctional materials'.* Journal of Materials Chemistry A, 2014. **2**(36): p. 14773-14797.
54. Kamaruzzaman, N.F., et al., *Antimicrobial Polymers: The Potential Replacement of Existing Antibiotics?* International Journal of Molecular Sciences, 2019. **20**(11): p. 2747.
55. Alfredo, N.V. and J. Rodríguez-Hernández, *Chapter 4 - Antimicrobial Polymeric Nanostructures*, in *Nanostructures for Antimicrobial Therapy*, A. Ficai and A.M. Grumezescu, Editors. 2017, Elsevier. p. 85-115.
56. Huang, K.-S., et al., *Recent Advances in Antimicrobial Polymers: A Mini-Review.* International Journal of Molecular Sciences, 2016. **17**(9): p. 1578.
57. Arora, A. and A. Mishra, *Antibacterial Polymers – A Mini Review.* Materials Today: Proceedings, 2018. **5**(9, Part 1): p. 17156-17161.
58. Ergene, C., K. Yasuhara, and E.F. Palermo, *Biomimetic antimicrobial polymers: recent advances in molecular design.* Polymer Chemistry, 2018. **9**(18): p. 2407-2427.
59. Granqvist, C.G., *Electrochromics and Thermochromics: Towards a New Paradigm for Energy Efficient Buildings.* Materials Today: Proceedings, 2016. **3**: p. S2-S11.
60. Hu, J. and X. Yu, *Performance evaluation of solar-responsive asphalt mixture with thermochromic materials and nano-TiO2 scatterers.* Construction and Building Materials, 2020. **247**: p. 118605.
61. Fabiani, C., V.L. Castaldo, and A.L. Pisello, *Thermochromic materials for indoor thermal comfort improvement: Finite difference modeling and validation in a real case-study building.* Applied Energy, 2020. **262**: p. 114147.
62. Aburas, M., et al., *Thermochromic smart window technologies for building application: A review.* Applied Energy, 2019. **255**: p. 113522.
63. Parent, O. and A. Ilinca, *Anti-icing and de-icing techniques for wind turbines: Critical review.* Cold Regions Science and Technology, 2011. **65**(1): p. 88-96.
64. Dalili, N., A. Edrisy, and R. Carriveau, *A review of surface engineering issues critical to wind turbine performance.* Renewable and Sustainable Energy Reviews, 2009. **13**(2): p. 428-438.
65. Droval, G., et al., *Simulation of Electrical and Thermal Behavior of Conductive Polymer Composites Heating Elements.* Journal of Thermophysics and Heat Transfer, 2005. **19**(3): p. 375-381.
66. Cheng, C., K.-C. Ke, and S.-Y. Yang, *Application of graphene–polymer composite heaters in gas-assisted micro hot embossing.* RSC Advances, 2017. **7**(11): p. 6336-6344.
67. Kim, H.S., et al., *Thermal conductivity of polymer composites with the geometrical characteristics of graphene nanoplatelets.* Scientific Reports, 2016. **6**(1): p. 26825.

68. Zhang, Z., et al., *Improvement of the thermal/electrical conductivity of PA6/PVDF blends via selective MWCNTs-NH2 distribution at the interface.* Materials & Design, 2019. **177**: p. 107835.
69. Riggs, J.P., H.A. Goldberg, and J.B. Stamatoff, *Multifunctional Polymers — An Industrial Perspective*, in *Frontiers of Polymer Research*, P.N. Prasad and J.K. Nigam, Editors. 1991, Springer US: Boston, MA. p. 27-44.
70. Gan, Y.X., et al., *Piezoelectric Behavior of Self-Organized Titanium Dioxide Nanotubes.* Nanoscience and Nanotechnology Letters, 2012. **4**(8): p. 801-807.
71. Curtis, P.T., *Multifunctional polymer composites.* Advanced Performance Materials, 1996. **3**(3): p. 279-293.
72. Lu, T. and W. Neng. *Future internet: The Internet of Things*. in *2010 3rd International Conference on Advanced Computer Theory and Engineering(ICACTE)*. 2010.
73. Oliveira, J., et al., *Polymer-based smart materials by printing technologies: Improving application and integration.* Additive Manufacturing, 2018. **21**: p. 269-283.
74. Kuang, X., et al., *Advances in 4D Printing: Materials and Applications.* Advanced Functional Materials, 2019. **29**(2): p. 1805290.
75. Roth, B., R.R. Søndergaard, and F.C. Krebs, *7 - Roll-to-roll printing and coating techniques for manufacturing large-area flexible organic electronics*, in *Handbook of Flexible Organic Electronics*, S. Logothetidis, Editor. 2015, Woodhead Publishing: Oxford. p. 171-197.
76. Ngo, T.D., et al., *Additive manufacturing (3D printing): A review of materials, methods, applications and challenges.* Composites Part B: Engineering, 2018. **143**: p. 172-196.
77. Isayev, A.I. and M. Modic, *Self-Reinforced melt processible polymer composites: Extrusion, compression, and injection molding.* Polymer Composites, 1987. **8**(3): p. 158-175.
78. Eckertová, L., *Methods of Preparation of Thin Films*, in *Physics of Thin Films*. 1977, Springer US: Boston, MA. p. 14-51.
79. George, J., *Preparation of Thin Films*. 1992: Taylor & Francis.
80. Prudenziati, M. and J. Hormadaly, *Printed Films: Materials Science and Applications in Sensors, Electronics and Photonics*. 2012: Elsevier Science.
81. Ribeiro, C., et al., *Electroactive poly(vinylidene fluoride)-based structures for advanced applications.* Nature Protocols, 2018. **13**(4): p. 681-704.
82. Teichler, A., J. Perelaer, and U.S. Schubert, *Inkjet printing of organic electronics – comparison of deposition techniques and state-of-the-art developments.* Journal of Materials Chemistry C, 2013. **1**(10): p. 1910-1925.
83. Bagen, S., et al. *Next generation coating technologies for low-cost electronics manufacturing*. in *Nineteenth IEEE/CPMT International Electronics Manufacturing Technology Symposium*. 1996.
84. Sahu, N., B. Parija, and S. Panigrahi, *Fundamental understanding and modeling of spin coating process: A review.* Indian Journal of Physics, 2009. **83**(4): p. 493-502.
85. Nguyen, N.-T., *Chapter 4 - Fabrication technologies*, in *Micromixers (Second Edition)*, N.-T. Nguyen, Editor. 2012, William Andrew Publishing: Oxford. p. 113-161.
86. Lin, H.-W., et al., *The rheological behaviors of screen-printing pastes.* Journal of Materials Processing Technology, 2008. **197**(1): p. 284-291.
87. Novaković, D., et al., *15 - Screen Printing*, in *Printing on Polymers*, J. Izdebska and S. Thomas, Editors. 2016, William Andrew Publishing. p. 247-261.
88. Suikkola, J., et al., *Screen-Printing Fabrication and Characterization of Stretchable Electronics.* Scientific Reports, 2016. **6**(1): p. 25784.
89. Nayak, L., et al., *A review on inkjet printing of nanoparticle inks for flexible electronics.* Journal of Materials Chemistry C, 2019. **7**(29): p. 8771-8795.
90. Ferreira, A.D.B.L., P.R.O. Nóvoa, and A.T. Marques, *Multifunctional Material Systems: A state-of-the-art review.* Composite Structures, 2016. **151**: p. 3-35.

91. Lendlein, A. and R.S. Trask, *Multifunctional materials: concepts, function-structure relationships, knowledge-based design, translational materials research.* Multifunctional Materials, 2018. **1**(1): p. 010201.