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Applications of Electroactive polymers: Review

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Electroactive polymers (EAPs), when subjected to electrical stimuli, undergo physical deformations. These polymers are broadly classified as electronic and ionic electroactive polymers, depending on the mode of transport of charges when the electric stimulus is applied. They are further classified into subcategories based on the way a particular material responds to electrical stimuli, and due to this diversification, EAPs find applications in many sectors. In this paper, the classification of Electroactive polymers is explained along with applications in a) Drug delivery due to biocompatibility, b) Actuators & sensors due to generation of high strain and adequate sensitivity to the electric field, c) Artificial muscles due to their low density and high strain capability, d) Coatings due to anticorrosive properties of conductive EAPs, e) Energy harvesting, are briefly discussed.

Keywords: electroactive polymers; drug delivery; artificial muscles; actuators; sensors.

1. Introduction

Polymers that change their size, shape upon an electrical stimulation are known as Electroactive Polymers. First studied by Rontgen in 1880, stretching of the rubber band is caused due to the repelling action of ions produced in the air as a result of high voltage application[1]. After that, for several years, the potential of these polymers was researched by researchers. A major development took place in 1969, Kawai reported the piezoelectric effect of polyvinylidene fluoride (PVDF)[2]. This provided an edge for researching more such materials which would show similar properties. Shirakawa et al discovered the first electrically conducting polymer in 1977[3]. They found that the conductivity of the polyacetylene polymer increases significantly when halogens are introduced – bromine, chlorine, iodine. In the case of Iodine, it increases up to seven folds. Later on, several such polymers were discovered. In the 90s-decade, the development of Ionic polymer-metal composites (IPMC) took place. IPMC showed deformations even at lower voltages – 1-2 volts. These materials have lower activation energy and they can be deformed more which was the advantage of these polymers over the previously developed EAPs. It was reported in one of the reports in the literature that IPMC exhibited 380% strain, much more than any other previously developed EAPs[4]. Later on, in 2005, during the conference of Electroactive Polymer Actuators and Devices, a challenge was held where an arm-wrestling match between a human and a robotic arm was held. Likewise, the development took place and the potential of EAPs is being explored to date. In the present study, some of the major applications of EAPs are explained. EAPs have applications in Pharmaceutics, Robotics, Sensing and Actuating devices, coatings, and a lot of other areas.

Acknowledging the versatility of EAPs, their applications in various sectors were looked at, a majority closely related to the medical field. From this preliminary survey, 5 areas of application – actuators, energy harvesters, drug delivery, artificial muscles, and coating applications were selected. The major findings from the past 2 decades have been briefly summarized under said categories of application.

Key characteristics examined while screening was 1) significant progress in terms of performance, safety, and reproducibility over past studies, 2) new features or properties relative to other studies chosen in the present paper, 3) potential for future relevance and development.

2. Classification of Electroactive polymers

EAPs can be broadly classified into two main subcategories[5,6] depending upon the mode of transport of charges – a) Electronic & b) Ionic. They can be further classified as shown in the hierarchy diagram Fig-1.

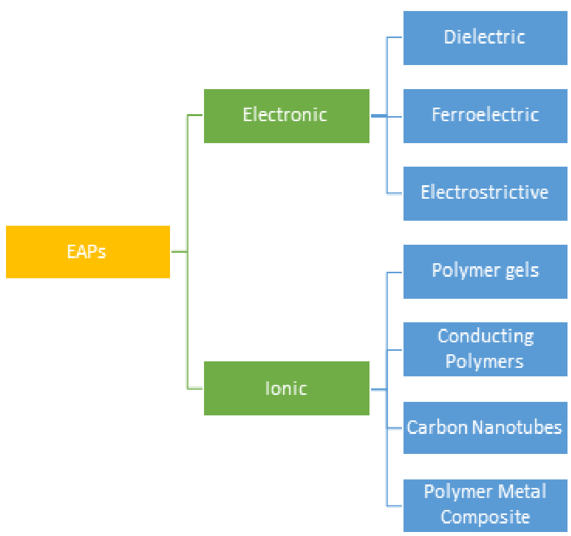

Fig-1: Classification of the Electroactive Polymers

In electronic polymers, the actual transfer of ions doesn't take place. The deformation takes place due to Coulomb forces, re-orientation of polarization, etc. Dielectric polymers are the polymers in which change in the dimensions takes place due to electrostatic action between two electrodes or charged regions. These polymers can possess high strains. When they are used as capacitors, their capacitance can change upon the application of voltage. High actuation voltage needs to be provided to these types of EAPs. Ferroelectric polymers are crystalline polymers. They are polar and retain the polarization i.e. permanent polarization[7,8]. Because of their pyroelectric characteristic, they are used in heat sensors, and because of their piezoelectric characteristics, they are used in actuators and transducers[9]. Electrostrictive EAPs consist of partial charges which rotate upon the application of the electric field. This causes an electrostriction effect[10]. In Ionic polymers, the actual flow of ions takes place. Conducting polymers are organic and conduct electricity and provide high conductivity[11,12]. The desired electrical properties of conducting EAPs can be obtained by appropriate synthesis and dispersion techniques[13,14]. Ionic polymer-metal composites are synthetic polymers consisting of anionic surface coated with conducting metals. Flemion and Neflon are some of the IPMCs which are readily used[15]. They possess high strains even at low voltage applications. They have a high potential for the application as actuators, sensors, energy harvesters & bio-mimetic purposes[16]. Polymer gels and carbon nanotubes are also classified under ionic EAPs. Gels are known for providing higher volumetric strains in solids[17]. They also can be used for actuators.

EAPs have proven to be having potential for a plethora of applications in the various domains. They have very high potential for the applications in futuristic technologies, some of them are listed in the Table.1 –

Table 1. Few futuristic, latest applications of EAPs

Application	Short notes	Reference
Application of EAPs in control system dynamics	Simulation study of use of DE (Dielectric) EAPs for control loops	18
In Microfluidic Pumps	Use of electrostrictive EAP – PVDF-TrFE as actuator material	19
In Braille & graphic display	Use of PVDF-TrFE-CFE for actuation	20
For a tactile display		21
Tunable lenses	Use of commercially available DE (Dielectric) type of EAP was done for actuation	22

For a refreshable tactile display	Use of bistable EAPs (BSEPs) who can have strain up to 335%	23
For Catheter	Use of conducting EAP as an actuator to drive the catheter	24
Artificial Flagellum of robot bacteria	Use of EAP actuators which can be operated in both wet and dry conditions, miniaturisation	25
For adaptive optic components	Tri-layer of EAP is used for actuation	26
For power generation from waves		27
For power generation from waves		28
For Organ on Chip device		29
MEESo robots	For the navigation of the natural lumens of the body of humans, IPMCs are used	30

3. Electroactive polymers as Actuators

In the electronic EAPs, the charges are stable and strain gets produced due to the coulomb's force whereas, in ionic EAPs, the ions are mobile and cause distortion. Dielectric Elastomers fall under the electronic EAPs category. In general, in these kinds of materials, change in the dimensions takes place as there is a development of charges on the surface of these due to the large voltage application[5]. There are various examples of such materials applied for actuation in the literature & they have great potential for actuation purposes as they could produce high strains, they are cost-effective, quick response, and highly efficient[31]. It was reported that silicone and acrylic actuators are capable of producing a linear strain of 117% & 215% respectively[32]. More studies report that polyurethanes[33,34], fluoropolymers[35,36] as DE. There are various designs of the DE actuators - Diaphragm actuators, Unimorph & Biomorph actuators, rolled actuators, Extender, Framed, Tube actuators, Bowtie, Spider[31]. Not just the DE EPAs but even Ion polymer-metal composites can be used for the actuation[37,38]. In this kind, a field is applied on one side of the polymeric membrane. This causes the ions to flow and accumulate at one location. Due to this, the membrane bulges on one side. One of the latest applications of this kind was reported in the literature to actuate the movement of aileron of an unmanned aircraft38. There is another report in the literature which has biomedical applications such as drug injection micro pumps in which they fabricate polypyrrole actuators using silicon bulk micromachining process with solid polymer electrolyte 102. Another report found in the literature states about the use of Ionomeric Polymers (IPMC) for transducing applications[39]. One of the plus points of this type of transducer was that it can be used at low voltages, typically below 5 Volts and the strain produced would be controllable. Even the sensitivity of these transducers was high. Albeit, it had issues of the breakdown of the solvents and stability issues in the air which was countered in the study.

4. Electroactive Polymers as energy harvesters

Another potential application of EAPs is for harvesting energy[40–44]. It's an opposite process of that of actuation that is by use of electroactive polymer transducers mechanical energy gets converted to electrical. Various studies in the literature have reported this aspect. In a study, Acrylic DE was tested and 0.4 J/g of output was recorded[40]. Silicone-based DE EAP was investigated in another study for energy harvesting[41]. In a report in the literature, a comparison of 6 types of polymers which are electroactive, was done for their use for energy harvesting and scavenging purpose[45]. It was found that dielectric polymers have high energy density and better performance for passive materials. Their ability to scavenge energy is 10 fold more than electrostrictive type and 100 fold more than piezo-electric type. Applications of such DE generators are reported in the literature to convert energy from walking[46], wind[47], waves[48–52] and harvest that energy. Another application of EAP is in devices used in order to achieve electrically stimulated generations of colors from electroactive polymers[53].

5. Electroactive polymers in Pharmaceutics:

Ever since electroactive polymers were proved to be compatible with a plethora of vital biological components, cell lines of the human body, namely bone, endothelial, glial, neural and fibroblast among others, the potential of these electrically simulated materials in the field of drug delivery has increased exponentially. Consequently, a large variety of pharmacologically active compounds have and are being tested to study the drug release profiles against a number of polymers, which are either ionic in nature or have the property of intrinsic conductivity[54,55]. According to Tandon et al, drug release and loading from these polymers follows three different mechanisms[56]. While the optimum method of drug loading depends on the properties of the drug itself, the process of drug release depends mainly on the ionic character (cationic/anionic/neutral) of the molecule[57–60].

In this part of the review, we look at a variety of drug delivery systems (DDS) that have been crafted by use of various electroactive polymers, and the most common has been observed to be transdermal drug delivery (TDD) patches. In a recent paper, natural rubber (NR) was used to prepare the matrix, which was loaded/doped with Ibuprofen. NR was chosen for its elastic, flexible nature coupled with strength required to maintain integrity of the patch even when applied on high friction/high motion areas of the body. Studies showed that the storage modulus values were high enough to serve this purpose. By changing the amount of trimethylolpropane tris-(3-mercaptopropionate) (TMP TMP), it was found that permeation-release of Ibuprofen decreased with increasing crosslinking ratio [61]. Aloin was loaded into PPV (poly (p-phenylene vinylene))/polyacrylamide-hydrogel where PPV was used as a conductive polymer, to form a TDD patch which was tested for drug release using pig skin and Franz Diffusion Cell apparatus. Here too, the crosslinking ratio was varied, and it was found that mesh size decreased with increasing ratio[62]. A similar hydrogel, poly(acrylic acid)(PAA) was used in combination with poly-pyrrole as a conductive polymer, and advantage of such a combination over neat hydrogel drug delivery systems was observed in the form of higher diffusion coefficient (observed between 5.06×10^{-9} and 4.92×10^{-8} cm^2/s in pristine PAA hydrogel, and increased to fall within 1.97×10^{-8} and 7.30×10^{-7} cm^2/s in system incorporating electroactive polymer calculated using the Higuchi equation) of the combination, where 5-sulfosalicylic acid was used as dopant[63]. In all these studies, the common observation was that the drug release/ diffusion coefficient increased as the applied electric field potential was increased[61–63]. In a patent, a slight variation of patches i.e., films were prepared on glass substrates, where dexamethasone phosphate (DMP) release was observed and among the preferred embodiments were poly(caprolactone) diols with average molecular weight of 500-2000 kDa and alcohol-terminated poly(ethylene glycol)s[64]. Recently, researchers have started replacing synthetic polymers with natural polymers such as dextran, hyaluronic acid among others, for better biocompatibility and non-toxicity. Jin et al.[65] combined dextran with hexamethylene diisocyanate (HDI) and aniline tetramer (AT) to form a biocompatible hydrogel while model drugs used were dexamethasone and indomethacin. Desired biocompatibility was confirmed by carrying out Toluidine blue, and Hematoxylin and eosin staining via insertion of prepared hydrogel in rats, subcutaneously. Chances of delamination of the polymer is one of the major disadvantages[66,67]. There is a good possibility of natural polymers overcoming this because of the non-toxicity offered by them.

Researchers have also delved deeper to fabricate DDS other than films/patches, one of the most promising have been electroactive polymer nanotubes, which increase effective surface area and decrease impedance of electrodes[68]. Mohammad et al.[68] incorporated dexamethasone in nanofibers of poly(lactide-co-glycolide) (PLGA) which were produced by electrospinning. These were then coated with poly(3,4-ethylenedioxythiophene) (PEDOT) to form nanotubes, which were tested for their ability to release the drug in a controlled manner, due to electric simulation as well as the slow biodegradation of

PLGA. In another paper, TiO_2 nanotubes were modified with polydopamine, and incorporated with ibuprofen. Polydopamine has multiple amino and phenolic groups which were expected to help in increasing drug-polymer interactions and hence assist in sustained delivery of the drug. Release kinetics showed a burst of drug release initially (first order kinetics) followed by sustained release (which fit the Higuchi equation)[69]. Helin et al.[70] prepared freeze-dried microgels by inverse mini-emulsion polymerization method with colloidal stability in aqueous medium, by combining hydroquinone with chitosan, using doxorubicin as the model drug. The electrochemical behavior as well as pH dependence was observed, whereas degradation studies showed that different degradation pathways were followed when urea and an enzyme - lipozyme were used. Microspheres using electroactive polymer Poly (vinylidene fluoride-co-trifluoroethylene) were prepared, which showed crystalline characters required for biomedical applications. This was carried out by displacement of non-solvent by polymer solution, followed by deposition of electroactive polymer to form aforementioned microspheres[71]. Such a technique of microsphere/microcapsule formation has been carried out many times with the incorporation of a drug[72]. Hence, fabrication of such DDS among others, for sustained delivery via electroactive polymers, is the next foreseeable research in this field.

6. Application of EAPs for artificial muscles

Human muscles cannot be termed as one particular kind of actuator; they show spring-like behavior with elasticity that can be tuned as per requirement[73]. Along with that from the mechanical point of view, human muscles display the following properties[74] -

(i) Peak stress level 150-300 KPa developed at 25% strain

(ii) Their maximum power output is- 150-225 W/Kg

(iii) Average power- 50 W/Kg

(iv) Energy density- 20-70 J/Kg

Hence while making an actuator, the following properties would serve as a baseline[74].

The requirements of an actuator are[75] –

(i) Robustness
(ii) Compactness
(iii) High Force Density
(iv) Less response time
(v) High power/weight ratio
(vi) High efficiency while the conversion of energy takes places

Electro Active polymers are known for their low density and high strain capability, along with these it shows the required amount of toughness, large actuation strain and they have the characteristic property of controlling the vibrations produced by inherently damping them. Due to these properties, they are referred to as artificial muscles, and hence they could be used to replace damaged human muscles which in turn could be considered as the first step towards “Bionic Human”[73]. Various types of EAPs could be considered as an option for actuators. They have the capability to respond to change in potenz of hydrogen[76,77], heat[78,79], light[80,81] and electric field[82] and hence produce muscle like contraction and relaxation.

Polyelectrolyte gels are observed to show large alteration in volume. The example of some of these gels are poly (vinyl alcohol)-poly (acrylic acid) (PVA-PAA), polyacrylamide (PAM), and poly(2-acrylamido-2-methylpropane) sulfonic acid (PAMPS). When these gels are subjected to an electric field, they contract and swell, hence these can be converted into mechanical work[83]. The major drawbacks of using these gels is[83] -

(i) Mechanical incompetence
(ii) Inability to hold large amount of weight

Polyacrylonitrile is found to show a more prominent change in length and they are mechanically stronger[83]. Hence this ionic polymer gel based EAP could be used and it is activated by the change in potenz of hydrogen[83,84]. Changing the potenz of hydrogen from acidic to alkaline, it becomes dense or swollen respectively. When they become activated, these gels bend as the cathode side becomes more alkaline and the anode side is more acidic[84,85].

The preparation of an actuator starts from the inelastic polymer, polyacrylonitrile which is annealed at temperatures between its transition temperature (approximately 110 Celsius) and the thermal degradation temperature (approximately 250 Celsius). Due to this annealing, crosslinking takes place in the strands of this polymer. Further, the remaining nitrile groups are made to go through saponification with a strong and carboxyl group replacing them[83]. The annealing process determines[86,87] -

(i) Degree of cross linking

(ii) Modulus of elasticity

(iii) Mechanical Strength

The PAN fibers are found to contract over half of its initial length, and hence are considered to be superior than all the other options available[83].

The chemical activation using acid and bases is a drawback because of lack of feasibility. Hence attempts are being made in order to make artificial muscles that can be controlled electrically and are fast acting as well[88,89].

7. Application in anticorrosive and other coating applications

Conductive polymers are one of the main classes of the electroactive polymers. The goal of this section is to review the various applications of electroactive conducting polymer and its application in anticorrosive and other coating applications. Electrochemical impedance is one of the important techniques which is used to study the electron conducting polymer film coated electrodes. There is one report in the literature which describes the problem of the transfer of diffusion motion in a homogeneous electroactive polymer film between a metal electrode and an electrolyte solution when vibration of an electrode with a low polarity is required [90]. The polymer film is stable enough to be able to study the electrochemical behavior of the film and dissolved particles in a wide potential range and over a long period of time. Platinum electrode modified with a stable surface made by applying an electroactive polymer, a platinum surface coated with adsorption or precipitation from a polyvinyl ferrocene platinum electrode PVF polymer behaves like a chemically modified electrode [91].

The various electroactive conductive polymers are used in secondary batteries application as electrode coatings. Various oxidizable conductive polymers with high operating voltage and capacity is proof of high gravimetric-energy-density materials. On the other side, reduced hydrocarbon polymers provide thermal stability and development of high life cycles in electrochemical cells [92]. There is one report about the synthesis of a 32 m thick PVDF film using centrifugation technology and electrochemically applied a polypyrrole (PPy) layer on both sides of this thin film to create an ultrathin polymer drive. In this study, a bending micro drive smaller than 1 mm in size, consisting of two layers of polypyrrole (PPy), separated by a polyvinyl fluoride (PVDF) membrane, was prefabricated and operated in the air and Water environment. Similarly, since the membrane is very porous and about 3 times thicker than the PVDF film, these factors affect ionic conductivity, so charge transfer in thicker drives is faster than in thin drives [93].

Ionic electroactive polymer actuators (IEAPAs) fabricated with this easy-to-scale-up method of spray-coating in combination with layer-by-layer self-assembly technique. There is observation of generating higher strain at a faster rate, improved response time and strain with low Young's modulus [94]. Ionic polymer metal Composites (IPMC) is one of the metal coated ion exchange membranes. But there is one problem associated with it: difficulty in bending control of IPMC. By using dehydration treatment and silver coated Nafion type IPMC gives better electrical bending controllability [95]. Recent studies show that poly (n-vinyl carbazole) (PVK) thin film contain specific properties like electronic character it has many applications in microelectronics.in this article study the relationship between the solvent characteristics and film properties, including surface roughness and structure, film thickness, and density. The spin-coated polymer thin films properties are affected by the dynamics of the spin-coating process [96].

Polyaniline (PANI) is one important type of electroactive Conductive polymer which has commercial applications like to produce batteries, sensors, electronic devices, anticorrosive Coatings products but there are drawbacks that are limited to solubility in many solvents. This is overcome by Nanocasting Technique to Prepare Lotus-leaf-like super hydrophobic electroactive polyimide (SEPI) as Advanced Anticorrosive Coatings from a natural Xanthosoma sagittifolium leaf [97]. Superhydrophobic electroactive epoxy (SSE) coating is used for corrosion protection by coating on the cold-rolled steel surfaces using nanocasting technique from the Xanthosoma sagittifolium leaf [98].

This review by Spinks et al[99] explained the effect of electroactive conducting polymer coatings on the corrosion rate and mechanism of corrosion protection of iron, steel and stainless steel. This study specifically concentrated on the polypyrrole and polyaniline coating on iron and steel and its coating type, preparation and test methods. This work illustrated the corrosion-testing environments electrically active polymers provide corrosion protection to steel substrates. Due to the polymer reduction, ion diffusion through the polymer is difficult so corrosion rate also decreases[99,100]. Inhibition or barrier coatings slow down the corrosion rate. PANi or PPy coatings are used for the corrosion protection of steel. PPy-oxalate coated film of steel reduces corrosion rate by factor of 20 as compared to bare steel. For the same electrolyte the corrosion potential of PANi coated metal is greater and the corrosion current is lower than bare metal[98–100]. The several research groups developed new techniques for fabricating electroactive polymers by using Photolithographic methods. A report demonstrated Some methods for patterning electroactive material for fabricating various devices which have rare electronic, chemical and optical properties[101].. Nanoparticle containing electroactive polyamide (EPA) coatings material used for the production of advanced anticorrosive coatings. The EPA coating was used to enhance corrosion protection effects on cold-rolled steel (CRS) electrodes as compared to non-electroactive polyamide (NEPA) coating by using electrolyte solution[103].

8. Conclusion

The present study summarizes the applications of Electroactive polymers in

- drug delivery systems,
- artificial muscles,
- sensing and actuating devices,
- energy harvesting,
- coatings and some other areas.

It can be realized that EAPs possess great potential for applications in different fields. Exploration of EAPs and their properties, applications is still in progress and there are a lot of opportunities and scope of research about EAPs.

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