

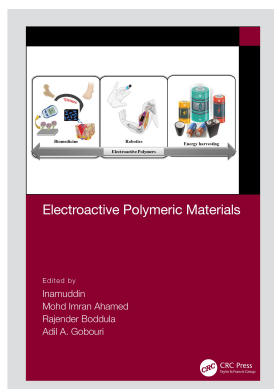
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Electroactive Polymeric Materials

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4 Intelligent Electroactive Polymers

Sapana Jadoun and Ufana Riaz

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4.1 INTRODUCTION

Before 1960, researchers prepared some conjugated polymers in the semiconductor field, which were used as insulators. Heeger *et al.* (1977) presented the first conducting polymer, iodine (I₂) doped poly(acetylene) (PAC). Its conductivity was 10³ S/cm. The synthesis of PAC and the enhancement in its conductivity after I₂ doping developed a new area of research (Inzelt, 2012). Hideki Shirakawa, Alan J. Heeger, and Alan MacDiarmid received the Nobel prize in Chemistry in 2000 for discovering the conductivity in PAC (Wallace *et al.*, 2008). This discovery opened a new area of research for the commercialization of these polymers as electrical conductors. These new conducting polymers have numerous applications in the field of sensors, actuators, molecular electronics, supercapacitors, electrochromic windows, corrosion protection, photovoltaics, and optoelectronics (Namsheer and Rout, 2021).

Electroactive polymers (EAPs) are materials that have conjugated π bonds in which electrons are delocalized along their polymeric backbone to conduct electricity (Guo and Facchetti, 2020). Intelligent EAPs are the class of polymers that can be excited by the electrical field that causes them to change their shape and size. These have received recent attention from industry and academia in polymer science and are designed based on a response or stimulus behavior. They can transfer electrons in the electric field, and therefore, have applications in various fields, such as biosensors, actuators, artificial muscles, soft robotic, and solar materials. They have several advantages including they are easy to process, have low density and mechanical flexibility, and can be mass produced. In addition, they can sense mechanical strain and produce electrical energy (Khan and Alamry, 2020).

A lot of biological systems have the ability of sensing, evaluating, and responding and are considered to be intelligent. A variety of intelligent behaviors can be seen in some types of materials, which are known as stimuli-responsive polymers. Some electroactive polymers possess conductivity and can monitor and respond to the surrounding chemical environment, which are known as intelligent EAPs. These polymers show a mechanical response to electrical stimulation and can respond to any change in the environment of living or non-living things, which is known as called actuation.

These polymers can be categorized as ionic or electronic. Numerous EAPs demonstrated a fast electromechanical response under electrical stimulation and mimicked natural muscles, and therefore, were called artificial muscles. They can change their shape and size in response to appropriate stimuli that can be electrical, thermal, chemical, or magnetic (X. Feng *et al.*, 2019; Perković *et al.*, 2020; S. Feng *et al.*, 2019; Wells *et al.*, 2019).

In this chapter, intelligent EAPs are summarized based on their classification and applications. In addition, conducting EAPs are discussed.

4.2 INTELLIGENT ELECTROACTIVE POLYMERS

Recently, polymers have become one of the most promising and attractive materials for biomedical applications due to their unique features and biocompatibility. The flexibility in their functionalization and properties makes them the best candidate for biodegradable thermoplastic polymers (Song *et al.*, 2018). In addition, various features allow numerous applications of these polymers. Of these, EAPs are considered to be a new generation of intelligent polymers that can respond to an electric field and change their properties for biomedical applications (Figure 4.1). As a polymeric material, these polymers include an anionic conductive hydrogel, percolated nanocomposites, and intrinsically conducting polymers (Kirillova and Ionov, 2019).

EAPs have attracted attention because many electro-sensitive tissues are present in the human body, such as the skin, heart, bones, and blood vessels and they have been used in tissue engineering due to their advantageous properties. They can be prepared in numerous sizes and shapes with the desired morphology and have a wide range of chemical and physical properties. Many review articles have been published on conducting polymers (CPs) (Jadoun and Riaz, 2020a, b; Jangid, Jadoun, and Kaur, 2020; Jadoun *et al.*, 2017; Khokhar, Jadoun, Arif, and Jabin, 2020; Jangid *et al.*, 2020; Jadoun, Riaz, and Budhiraja, 2020; Bach-Toledo *et al.*, 2020; R Murad *et al.*, 2020; Dunlop

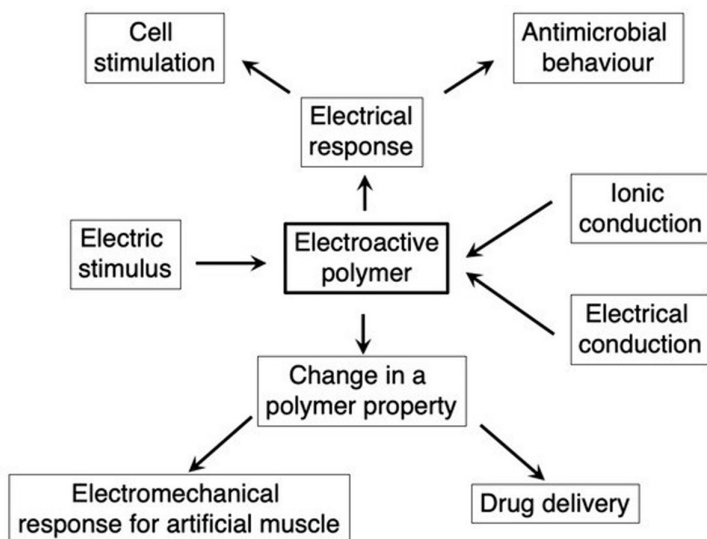


FIGURE 4.1 General representation of smart electroactive polymers: Its mechanism via ionic or electric conduction, mechanism proceed via electric current and producing cell stimulation or antimicrobial behavior or via the change in polymer property such as shape/size of polymer provided a response in electrochemical nature for artificial muscle and drug delivery.

(From Palza, Zapata, and Angulo-Pineda, 2019. With permission.)

and Bissessur, 2020) and piezoelectric polymers (Covaci and Gontean, 2020; Mishra *et al.*, 2019; Sappati and Bhadra, 2018). These intelligent materials can initiate suitable chemical responses when required and these chemical processes can be induced in CPs chemically or electrochemically.

To prepare these materials, the coating of non-conductive substance is carried out using a chemical polymerization method. During chemical polymerization, incorporation of counterions of oxidants occurs and there is the potential for counterion exchange; however, this type of synthesis restricted the chemical properties of the designed materials and electropolymerization offered a wide range of incorporations for counterion into the polymer. The advantage of electropolymerization is that it does not require an oxidant. Using anodic oxidation, a conducting film can be directly synthesized on the surface and these EAPs can be converted from an insulating to conducting state by various functionalization and doping techniques, which include charge injection at the polymeric interface, electrochemical doping, photodoping, chemical doping by charge transfer, and doping by acid–base chemistry (Pathiranage *et al.*, 2017; Guo and Ma, 2018; Zarren, Nisar, and Sher, 2019).

4.3 CLASSIFICATION OF ELECTROACTIVE POLYMERS

These polymers can be classified into two different groups:

1. Electronic electroactive polymers: these polymers are driven by Coulomb forces or electric and can be further classified into various types
 - a. Ferroelectric polymers
 - b. Dielectric electroactive polymers
 - c. Electrostrictive graft elastomers
 - d. Electrostrictive paper
 - e. Electro viscoelastic elastomers
 - f. Liquid crystal elastomer materials
2. Ionic electroactive polymers: these are driven by mobility or diffusion of ions and can be further classified into
 - a. Ionic polymer gels
 - b. Ionomeric polymer–metal composites
 - c. Conductive Polymers
 - d. Carbon Nanotubes
 - e. Electrorheological Fluids

These polymers experience dramatic changes in some of the properties when placed in an electric field. A general representation of the electrical and ionic conduction electroactive polymers are shown in Figure 4.1 (Palza, Zapata, and Angulo-Pineda, 2019). In this chapter, conductive EAPs that behave intelligently when subjected to an electric field are focused on.

4.4 CONDUCTIVE ELECTROACTIVE POLYMERS

Conductive EAPs work on the principle of counter ion exchange during the redox process. The exchange of ions with an electrolyte induces a volume change when reduction and oxidation occur at the electrodes. CPs have the desirable electric and optical properties of semiconductors to metals and are easy to synthesize (Naarmann, 2004). The initial conducting polymer was discovered as polyacetylene but now polyaniline (PANI) (Stejskal and Gilbert 2002; Boeva and Sergeyev, 2014) and their derivatives such as poly(o-phenylenediamine) (POPD) (Khokhar *et al.*, 2020; Khokhar *et al.*, 2021), polypyrrole (PPY) (Jain, Jadon, and Pawaiya, 2017), polythiophene (PTH) (Kaloni *et al.*, 2017), and poly(ethylenedioxy thiophene) (PEDOT) (Gueye *et al.*, 2020) have been

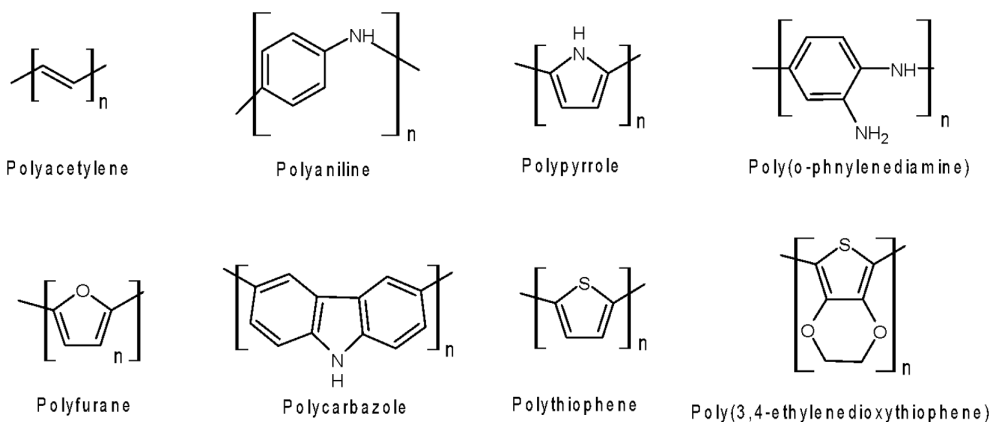


FIGURE 4.2 Structures of some CPs and their derivatives.

(From Khokhar, Jadoun, Arif, and Jabin, 2020. With permission.)

extensively studied for their electrical conducting. The structures of some CPs and their derivatives are shown in Figure 4.2.

4.4.1 POLYANILINE

Polyaniline (PANI) is one of the extensively studied polymers that exists in three forms: (1) fully oxidized (pernigraniline base); (2) half-oxidized (emeraldine base); and (3) completely reduced (leucoemeraldine base). The half-oxidized form of PANI emeraldine base exhibits the highest level of stability and conductivity and is inexpensive and has high processibility (Jangid, Jadoun, and Kaur, 2020). In addition, PANI exhibits the charge transport phenomena via doping and dedoping and is biocompatible for in vivo and in vitro applications. This polymer could be used in commercial applications, because it has good processibility, is stable and is inexpensive. However, chemical and electrochemical synthesis limit its solubility due to some structural defects that limit its practical applications (Ameen, Shaheer Akhtar, and Husain, 2010). Some researchers developed different approaches to overcome this problem and incorporated the conjugated and well-defined oligoaniline in the backbone of copolymers that resulted in the combined properties of an oligomer and polymer, such as film-forming ability and mechanical strength (Zhou *et al.*, 2006; Martin and Diederich, 1999). Huang *et al.* (2009) prepared electroactive polymers of polyimide via inserting amine-capped aniline trimers in between the backbone (Figure 4.3), which had higher electroactivity and could potentially be applied in anticorrosive coatings by forming a passive layer of metal oxide.

4.4.2 POLYPYRROLE

Polypyrrole (PPY) is the most explored CPs due to its easy synthesis (Jadoun, Biswal, and Riaz 2018), high electrical conductivity, and surface modification properties and it has shown high stability and stimuli-responsive properties. Due to its chemical stability, biocompatibility, and conductivity under physiological conditions, and high response in the electrical field, it is referred to as an intelligent electroactive material. Various studies showed its biocompatibility with osteoblasts and mesenchymal stem cells, nerve cells, myocardial cells, and endothelial cells. When repairing and regenerating damaged tissues, PPY was a promising material (Forciniti *et al.*, 2014). In the fabrication of a bending electroactive actuator, a sandwich of PPY and other conducting polymer electrodes with an electrolyte was used (Nezakati *et al.*, 2018; Pei and Inganäs, 1992).

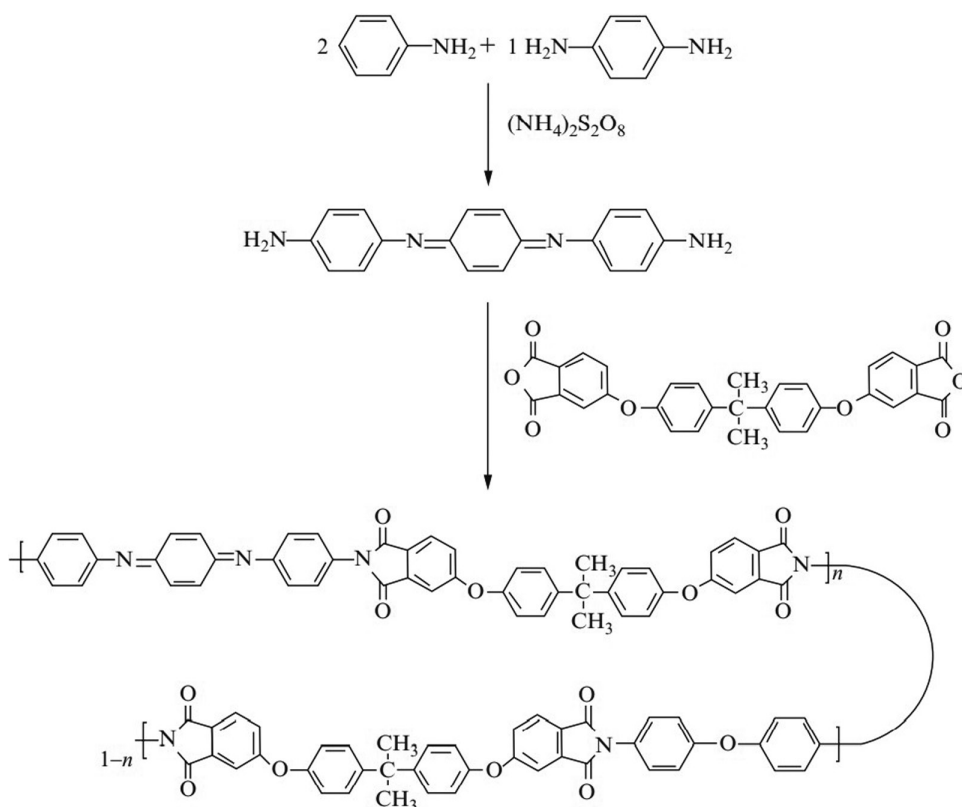


FIGURE 4.3 Representation for the synthesis of aniline trimers and polyimides.

(From Huang *et al.*, 2009. With permission.)

4.4.3 POLY(3,4-ETHYLENEDIOXYTHIOPHENE)

Poly(3,4-ethylenedioxythiophene) (PEDOT) is a very interesting CP that can be synthesized by chemical or electrochemical polymerization that uses an EDOT monomer. It is a derivative of polythiophene (PTH) that has superior properties to PTH, such as its redox potential, lower bandgap and it is more conductive and thermally established, which provides it with environmental, chemical, and electrical stability (Higgins *et al.*, 2012; Nambiar and Yeow, 2011). PEDOT has been used for preparing electrochemical transistors for biosensing (Mabeck and Malliaras, 2006), and it is a useful regenerative material due to the development of a neurotransmitter delivery system (Simon *et al.*, 2009). PEDOT has been employed to prepare biomaterials that have electrical conductivity and can make an electrical interface with tissues that are electrically responsive, such as the nervous system, heart, and skeletal muscles. In addition, PEDOT was used to coat neural cells and resulted in the fabrication of PEDOT-live cell electrodes and microelectrodes (Ghasemi-Mobarakeh *et al.*, 2011).

4.4.4 FUNCTIONALIZED CONDUCTING POLYMERS

The advantages of CPs can be enhanced by modifications or functionalization of them by composite formation, copolymerization, or by doping to make them more biocompatible for biomedical applications (Jadoun, Ashraf, and Riaz, 2018; Jadoun, Verma, and Riaz, 2018; Jadoun, Sharma, *et al.*, 2017; Riaz, Ashraf, *et al.*, 2017). Functionalization has enhanced the solubility, stability, electrical conductivity, mechanical stability, and the power to dispense biological moiety (Jadoun,

Ashraf, and Riaz, 2017; Riaz, Jadoun, *et al.*, 2017; Riaz *et al.*, 2016). For example, Riaz *et al.* (2019) modified POPD by doping it with dyes to make the materials water-soluble and they could be applied for the detection of BSA and bioimaging. Some researchers used PVDF as fillers with PANI and PPY enhancement in processability (Merlini *et al.*, 2014; Saïdi *et al.*, 2013). A lot of in vitro studies have been performed on these functionalized conducting materials, which showed responses after interacting with cells and tissues, which made them suitable for applications in the biomedical field (Riaz *et al.* 2018). Where electrical conductivity is needed for cell growth, these are promising for use as medical implant materials, such as in sensing, neural stimulation, and regenerating tissues.

4.5 APPLICATIONS

Intelligent EAPs are widely applied in the biomedical fields and other fields, such as corrosion protection, and aerospace applications. The applications of intelligent EAPs in numerous biomedical fields, such as sensors, drug release, robotics, artificial muscles, tissue engineering, and actuators, are shown in Figure 4.4.

The intelligent EAPs provide various types of electrical stimulation to cells, and therefore, are potential candidates for tissue engineering and the healing of bones and nerve regeneration. These polymers recently attracted a lot of attention due to their use in many sensitive tissues in the human body, such as nerves, skin, vessels, heart, and bones (Guo and Ma, 2018). PPY has shown stimuli-responsive properties and high stability under environmental conditions, which proved that the polymer was a biocompatible material that could be used in tissue engineering applications (Ravichandran *et al.*, 2012). In neural and cardiac tissue engineering, PPY was suitable due to its electrical conductivity. In addition, some two-dimensional substrates, and fibrous or porous scaffolds were promoted for cardiac differentiation using PANI or PPY (Kai *et al.* 2011; Qazi *et al.* 2014). PANI is suitable for in vivo applications (Mattioli-Belmonte *et al.*, 2003). Nanofibers of PANI and gelatin were synthesized by blending them to favor H9c2 cell adhesion and growth (Guo *et al.*, 2012), which demonstrated the use of PANI for nerve and cardiac tissue regeneration. Therefore, using CPs or their composites are good alternatives for materials in tissue engineering applications. Some potential applications of intelligent EAPs in tissue regeneration are shown in Figure 4.5.

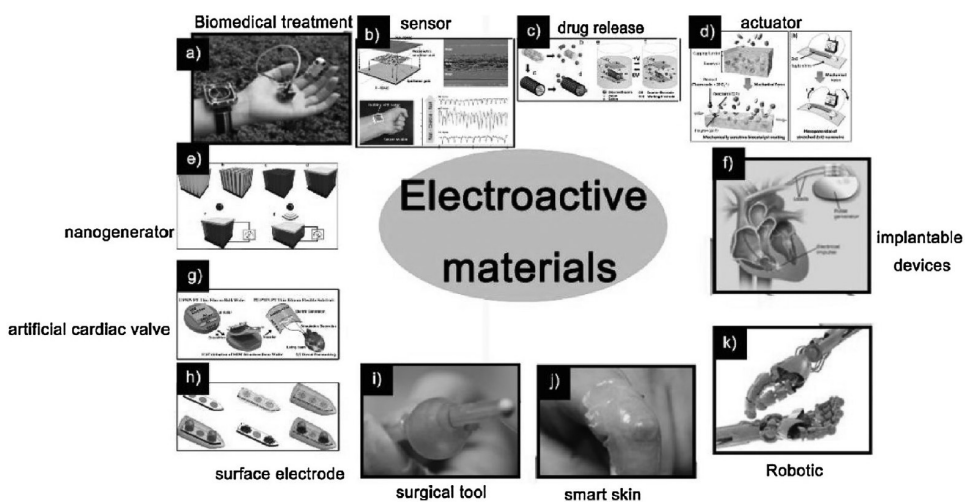


FIGURE 4.4 Applications of intelligent electroactive polymers in various biomedical fields.

(From Ning *et al.*, 2018. With permission.)

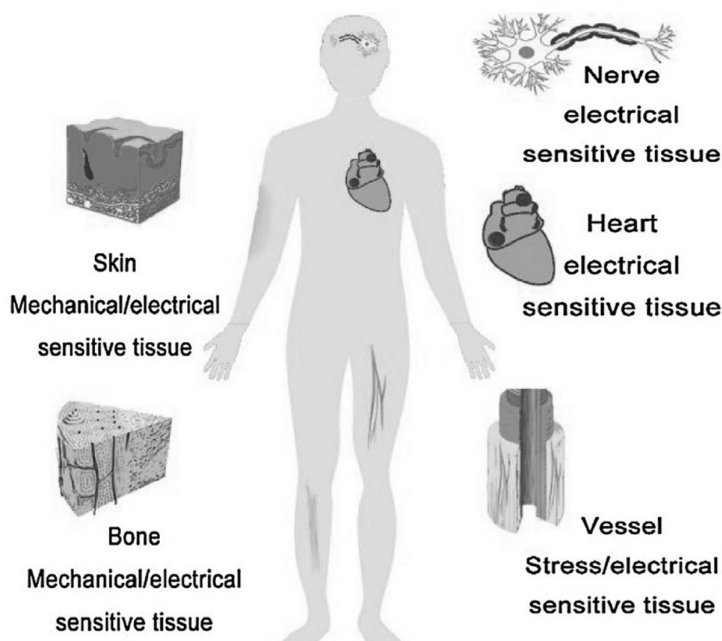


FIGURE 4.5 Potential applications of intelligent electroactive polymers in tissue engineering.

(From Ning *et al.*, 2018. With permission.)

When chemical interactions between materials occurs with the surrounding atmosphere, the deterioration of the materials takes place, which is known as corrosion. The protection of materials can be carried out by various coatings and intelligent EAPS are one of these best substitutes (Chang and Yeh, 2015). In these polymers, an amine-capped aniline trimer was first evaluated for corrosion protection. It was prepared by an oxidative coupling reaction using the one-step method with aniline and p-phenylenediamine in an acidic aqueous solution (Huang *et al.*, 2009; Peng *et al.*, 2011).

Intelligent EAPs are very convenient and practical for actuation using electricity, because they can easily emulate biological muscles. These polymers provide approximately 20 J/cm^3 of mechanical energy densities, which is quite high, and therefore, counted as effective actuation materials. Some researchers fabricated an effective actuator by sandwiching solid polymer electrolytes between two PEDOT electrodes. In addition, Ribeiro *et al.* (2018) studied solid-state CPs actuators. During the redox cycle, these actuators work via the insertion and expulsion of reversible counter ions (Otero *et al.*, 1993). Oxidation and reduction at the anode and cathode occurred because of the applied voltage between the electrodes, which resulted in the change in volume due to ion and electrolytes exchange (Madden, 2018) and by ions migration between the electrodes and electrolytes, and the electric charge was balanced. These ions swelled the polymer and after removing the ions, swelling reversed and resulted in the sandwich bending. Cui *et al.* (2020) reviewed the polymers-based bioinspired actuators, which included their design strategies, capabilities, and applications in robotic and biomedical areas.

4.6 CONCLUSION AND FUTURE PERSPECTIVES

Intelligent EAPs have shown significant potential for the development of biologically inspired unique devices. In addition, they have shown many potential advantages in tissue regeneration, actuating, and corrosion protection coatings. This chapter provides the reader with an understanding

of general intelligent EAPs and their classification. It focused on conducting EAPs and summarized their applications in various fields. In the future, research should focus on the long-term stability and performance of EAPs by designing a water-resistance surface, which will prevent water loss from the polymers and decrease the positive counter ion loss during the functioning of them in an aqueous environment.

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