

The Biosensors Using the Conducting Polymer Nanomaterials with Their Significance and Applications

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Abstract:

Polymers have long been used as insulating materials. For example, metal cables are coated in plastic to insulate them. However, there are at least four major classes of semiconducting polymers that have been developed so far. They include conjugated conducting polymers, charge transfer polymers, ionically conducting polymers and conductively filled polymers. The conductively filled conducting polymers were first made in 1930 for the prevention of corona discharge. The potential uses for conductively filled polymers have since been multiplied due to their ease of processing, good environmental stability and wide range of electrical properties. Being a multi-phase system in nature, however, their lack of homogeneity and reproducibility has been an inherent weakness for conductively filled polymers. Therefore, controlling the quality of dispersion to obtain homogeneous conducting polymer composites is critically important. Conducting polymers represent an important class of functional organic materials for next-generation electronic and optical devices. Advances in nanotechnology allow for the fabrication of various conducting polymer nanomaterials through synthesis methods such as solid-phase template synthesis, molecular template synthesis, and template-free synthesis. Nanostructured conducting polymers featuring high surface area, small dimensions, and unique physical properties have been widely used to build various sensor devices. The enhanced sensitivity of conducting polymer nanomaterials toward various chemical/biological species and external stimuli has made them ideal candidates for incorporation into the design of sensors. However, the selectivity and stability still leave room for improvement.

Keywords: conducting polymers; nanomaterials; chemical sensors; biosensors; polypyrrole; polyaniline; poly(3,4-ethylenedioxythiophene)

Introduction:

Conducting polymers have found a wide range of applications in the various fields of electronics, optics, energy devices, medicine, actuators, and composites as a viable alternative to metallic or inorganic semiconductor counterparts. In particular, there has recently been huge demand for developing flexible or wearable electronics, displays, and other devices, in which conducting polymers can ultimately be used as true flexible organic conductors or semiconductors. The most notable property of conducting polymers is their inherent electrical conductivity, which is closely connected to the charge transfer rate and electrochemical redox efficiency. Most conducting polymers act as semiconductors in terms of conductivity. Consequently, conducting polymers have had serious limitations in specific applications such as transistors and memories. For example, the performance of field-effect transistors (FETs) based on conducting polymers cannot rival that of FETs based on single-crystalline inorganic semiconductors, such as Si





and Ge, which have charge carrier mobilities that are about three orders of magnitude higher. The mobilities of FETs based on solution-processed conducting polymers [e.g., poly(3-hexylthiophene)] are generally found to be in the range of 0.1 cm² V⁻¹ s⁻¹, similar to those of amorphous silicon FETs. A mobility of 10.5 cm² V⁻¹ s⁻¹ has been the best reported thus far. Conducting polymer FETs are not practically suitable for use in applications requiring high switching speeds.

Another example of conducting polymer applications involves electrochromic devices for smart windows and flexible displays. Conducting polymers are one of the most attractive electrochromic materials because of advantages such as high coloration efficiency, rapid switching ability, and diverse colors. The switching time is one of the important parameters in display technology. The electrochromismof conducting polymers is based on reversible redox reactions accompanying ion exchange, and is completely different from the operating mechanism of FETs. The switching time is mainly affected by redox reaction efficiency, which depends on the ion diffusion rate and conductivity. Notably, it was recently reported that poly(3,4-ethylenedioxythiophene) (PEDOT) nanotubes with wall thicknesses of 10–20 nm exhibited fast switching speeds of less than 10 ms, though the color contrast was low. However, electrochromic displays based on conducting polymers have not yet been considered for commercialization. Stability, rapid response times, and efficient color changes are still critical parameters that need improvement.

Conducting polymers have also been used for sensor applications as a signal transducer. There are several important parameters in sensor technology, such as sensitivity, selectivity, and response time. In most cases, a response time on the order of seconds is enough for human recognition. Thus, it is easier to meet the requirements for response time than for other parameters. The sensing mechanisms of conducting polymers can involve redox reactions, ion adsorption and desorption, volume and weight changes, chain conformational changes, or charge transfer and screening. These polymers also share the strengths of polymers over other materials, including low-temperature synthesis and processing, large-area manufacture, flexibility, and cost effectiveness. As a result, conducting polymers can be competitive in sensor applications and sensors are therefore considered to be one of the most practical applications of conducting polymers.

Theory: Conducting polymers have mostly been synthesized in the form of powder and film using chemical and electrochemical polymerization methods, respectively. It is important to precisely control the structure and morphology of conducting polymers during the synthesis process, since most of them are insoluble in common solvents and not thermoplastic. Particularly, a variety of metallic and inorganic semiconductor nanostructures have been fabricated, which have exhibited unique electrical, optical, and chemical properties. Nanostructured materials feature high surface-to-volume ratio and small dimensions, which are very beneficial for sensor applications. The high surface area facilitates enhanced interaction between the materials and analytes, which leads to high sensitivity, and the small dimensions enable fast adsorption/desorption kinetics for analytes in the material, which allows a rapid response time. Accordingly, recent years have





witnessed a shift in sensor technology towards more sensitive recognition elements, highly sophisticated architectures, and miniaturization due to the emergence of nanomaterials and nanotechnology. Conducting polymer nanomaterials also have strong potential for yielding enhanced sensor performance compared to their bulk counterparts. However, polymers are unstable at the nanometer scale due to the nature of covalent bonds, which makes their nanostructures unstable as well. There are several conducting polymers that have been extensively investigated for practical applications, which are summarized in Table (a). All of them possess high conductivity and good environmental stability, and their polymerization reactions are not only straightforward but also proceed with high yield. Polypyrrole (PPy) features low oxidation potential and good biocompatibility, and the advantages of polyaniline (PANI) include that the monomer is very inexpensive. Polythiophene (PTh) has many useful derivatives, one of which is PEDOT developed by Bayer AG (Leverkusen, Germany). PEDOT features optical transparency and can be soluble in water with polystyrene sulfonate (PSS). Owing to these desirable characteristics, PPy, PANI, PTh and their derivatives have become leading materials in various applications fields. This review addresses research efforts to fabricate and manipulate nanostructures mainly consisting of the representative conducting polymers for sensor applications, and highlights remarkable recent examples with a focus on materials functionalization, transduction mechanisms, and device characteristics in sensor applications.

Fabrication of Conducting Polymer Nanomaterials

Precise control over the size and morphology of conducting polymers at the nanoscale is essential to improving the performance of related sensors. The polymers are highly unstable at the nanometer scale, which is one of the greatest obstacles in building polymer nano architectures. Nevertheless, numerous efforts have been made to fabricate polymer nanomaterials with well-defined size and morphology, and various types of conducting polymer nanostructures have been fabricated in a controlled fashion. Conducting polymers have traditionally been synthesized via chemical or electrochemical oxidation polymerization. Chemical polymerization is advantageous for large-scale production at low cost, while electrochemical polymerization offers the possibility of in-situ formation, such as on an electrode for a sensor device. Conducting polymers can be obtained in the presence of various oxidizing agents. Oxidation polymerizations with acid or peroxide initiators result in insulating materials that require a post-doping process. Metal salts that can act as both oxidizing and doping agents are used to conduct the oxidation polymerization, which directly yields polymers in a conductive state. For example, ferric salts including $FeCl_3$ and $Fe(ClO_4)_3$ are widely employed. Owing to their electrical conductivity, conducting polymers can grow electrochemically without oxidizing agents on an electrode. It is possible to tailor the polymer thickness by controlling the applied potential, polymerization time, and electrolyte.

Conducting polymer nanostructures have been fabricated with the aid of templates during the polymerization process. The synthetic routes are traditionally classified into three classes depending on the kind of template: hard template synthesis, soft template synthesis, and template-free synthesis. It is sometimes





ambiguous to distinguish the hard and soft templates. For example, track-etched polymer membranes and polymer nanofibers are both soft, but they are categorized as a hard template and a soft template, respectively. Therefore, the templates are classified here as solid-phase and molecular templates as a new standard.

Solid-Phase Template Synthesis:

Organic/inorganic nanoparticles, anodic alumina membranes, track-etched polymer membranes, and mesoporous silica are examples of solid-phase templates. Anodic alumina and track-etched polymer membranes with parallel cylindrical nanopores have often been chosen for the production of nanotubes or nanorods [5]. The porous alumina membranes are formed by anodizing high-purity alumina disks in an acidic electrolyte, and the track-etched polymer membranes are produced by irradiating the membrane with high-energy heavy ions, followed by chemical etching. The use of such solid-phase templates is advantageous for simply tailoring the dimensions of nanomaterials, and it has thus been extensively studied for synthetic routes to obtain 1D nanostructures of organic and inorganic materials. Conventional electrode position and electrophoresis techniques can be simply applied to the templates to yield nanostructures. Because the geometry and morphology of the resulting nanomaterials are endowed by the template itself, precise control of the diameter and length is possible with the solid-phase templates. However, it is very hard to completely remove the template without degradation or irreversible aggregation of the resulting nanomaterials. Scale-up for commercial applications is also highly difficult due to the complicated synthetic process and high cost. Therefore, the solid-phase template synthesis might be suitable for fabricating high-value products in the form of composites without the removal of template.

Molecular Template Synthesis

Molecular template synthesis has strengths compared to solid-phase template synthesis. The most widely used molecular templates include surfactants, liquid crystals, and polyelectrolytes. Because molecular template synthesis is comparatively straightforward and cost-effective, it is suitable for large-scale production. However, the molecular templates are not robust, static entities, leading to considerable difficulty in obtaining the desired nanostructures. Surfactant templating is a typical example of molecular template synthesis. Surfactants have the ability to self-assemble into ordered molecular structures called micelles. The micelles exist in various forms, and their dimensions generally range from a few to a few tens of nanometers, making them very suitable as templates. Micro emulsions consisting of thermodynamically stable micelles with size less than 50 nm have been used in polymerization techniques to prepare polymer nanoparticles. However, micelles are highly sensitive to the surrounding environment, which makes it very difficult to achieve stable micro emulsion systems for polymerization, preventing their widespread utilization for industrial products. One of the strategies for making stable micro emulsion systems is to use co-surfactants like long-chain alcohols.





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Template-Free Synthesis

Template-free synthesis, which is based on the self-assembly of molecular building blocks, is naturally very straightforward. However, extensive efforts are required to design and synthesize building blocks that are able to assemble into nanostructures under certain conditions. Fortunately, several species (precursors) can spontaneously form nanostructures without such artificial efforts. Typically, PANI has intrinsically 1D morphology at the nanometer scale when borne in aqueous solution, and thus, it has been intensively investigated to fabricate 1D PANI nanostructures in the absence of templates [5,6]. The formation of PANI nanofibrils has been studied extensively. The growth control of PANI nanofibrils in the presence of a steric stabilizer was reported, in addition to their morphologydependent electrochemical properties[9]. Wan *et al.* have actively reported the formation of PANI nanotubes under various synthesis conditions without templates [10,11,12]. It was found that subtle changes in polymerization parameters often result in drastic differences in the morphology of the resulting PANI product.

Biosensors

Biosensing is commonly conducted in a solution phase to maintain the biological activity of target species. Accordingly, it is critical to immobilize transducer materials on sensing platforms for continuously obtaining reliable signals. Compared to metals and ceramics, conducting polymers are more compatible with biological systems. Inorganic nanomaterials have been readily integrated into biosensor platforms using lithography and focused ion beam techniques. However, the integration of conducting polymer nanomaterials into biosensors has been limited due to their incompatibility with the traditional microfabrication processes. Chemical, thermal, and kinetic damage can possibly degrade conducting polymers during the microfabrication process. An alternative strategy was developed to circumvent this issue by Jang and Yoon [15]. Covalent linkages between PPy nanotubes and a microelectrode substrate were made to achieve reliable electrical contact in solution. PPy nanotubes with carboxyl groups were prepared and the carboxyl groups were chemically coupled with the surface amino group of the electrode substrate. A liquid, ion-gated field-effect transistor (FET) sensor could be successfully fabricated using this method. The structure of the FET sensor is similar to that of the normal metal-oxide-semiconductor FET, except for the gate, which incorporates the means of transduction from a chemical event to a voltage[16]. In the liquid ion-gated FET conFigure.uration, two metal electrodes, called the source and drain are deposited on a substrate. The gate electrode is electrically isolated from the two electrodes and immersed in an electrolyte. The gate potential affects the density of charge carriers in the semiconductor channel. The channel is normally modified with molecular or polymeric receptors for selectively recognizing the analyte of interest. The recognition of the analyte by the receptor can alter the gate potential being applied on the channel, which modulates the source-drain current. With this operating principle, many different types of FET sensors have been devised in order to detect glucose [17], odorants [18], tastants [19], hormones [20], and proteins [21]. Several critical parameters determining the FET sensor response have been also





investigated. For example, Mulchandani *et al.* fabricated single PPy nanowire-based FET sensors for real-time pH monitoring and examined how the diameter of the nanowire affects the sensor performance. PPy nanowires with three different diameters (*ca.* 60, 80, and 200 nm) were anchored on a pair of gold electrodes with different gap lengths (1 and 4 μ m). The FET sensors had higher sensitivity with lower diameter and higher length.

Introduction to Biosensors

A biosensor is an analytical device, which converts a biological response into an electrical signal. It consists of two main components: a bioreceptor or biorecognition element, which recognizes the target analyte and a transducer, for converting the recognition event into a measurable electrical signal. A bioreceptor can be a tissue, microorganisms, organelles, cells, enzymes, antibodies, nucleic acids and biomimic and the transduction may be optical, electrochemical, thermometric, piezoelectric, magnetic and micromechanical or combinations of one or more of the above techniques.

The bioreceptor recognizes the target analyte and the corresponding biological responses are then converted into equivalent electrical signals by the transducer. The amplifier in the biosensor responds to the small input signal from the transducer and delivers a large output signal that contains the essential waveform features of an input signal. The amplified signal is then processed by the signal processor where it can later be stored, displayed and analyzed. Biosensors have been widely applied to a variety of analytical problems in medicine, the environment, food, process industries, security, and defense.

Generations of Biosensor

Depending on the level of integration, biosensors can be divided into three generations, i.e., the method of attachment of the biorecognition element or the bioreceptor molecule to the base of the transducer element. The three generations of a biosensor are depicted in *Figure. 2*.

In the first generation, the biorecognition element or the bioreceptor molecule is either bound to or entrapped in a membrane, which in turn is fixed on the surface of the transducer (based on Clark biosensors. The mediated or secondgeneration biosensors use specific mediators between the reaction and the transducer to improve sensitivity. It involves the adsorption or covalent fixation of the biologically active component to the transducer surface and permits the elimination of semi-permeable membrane. In the case of third-generation biosensors or direct biosensors, it is the direct binding of the bioreceptor molecule to the sensor element, and thus the bioreceptor molecule becomes an integral part of the biosensor. So no normal product or mediator diffusion is directly involved in this. Conducting polymer-based biosensors come under this category.

Classifications of Biosensor

Biosensors can be classified by their bioreceptor or their transducer type.





Bioreceptors:Classified into five different major categories. These categories include antibody/antigen, enzymes, nucleic acids/DNA/RNA, cellular structures/cells, and biomimetic. The enzymes, antibodies, and nucleic acids are the main classes of bioreceptors which are widely used in biosensor applications. Though the enzymes are one of the biorecognition elements, they are mostly used to function as labels than the actual bioreceptor.

Transducers:The transducer plays an important role in the detection process of a biosensor. In case of conducting polymerbased biosensor, the conductive polymer acts as a transducer that converts the biological signal to an electrical signal. Biosensors can also be classified based upon the transduction methods they employ. Although there are new types of transducers constantly being developed for use in biosensors, the transduction methods such as optical, electrochemical, and mass based are given importance here since these are the most popular and common methods. Each of these three main classes contains many different subclasses and they can be further divided into label and label-free (nonlabeled) methods, where, the labeled methods depend on the detection of a specific label (e.g., fluorescence) and the label-free detection is based on the direct measurement of a product developing during the biochemical reactions on a transducer surface.

Significance of Conducting Polymers to Biosensors

Conducting polymers have been used as a transducer in biological sensors due to its attractive properties and their use in biosensors has grown over the past decade. Some of the attractive features of conducting polymers for biosensor applications include:

- Availability of varied range of monomer types.
- Availability synthetic analogues of monomers.

• Composites can be prepared combing conducting polymers with nonconducting polymers or with nonpolymer materials such carbon, carbon nanotubes, metals, etc.

- It can be prepared both electrochemically and chemically.
- It can be prepared in a range of soluble and insoluble forms.
- It has unique electrical, electronic, magnetic and optical properties.
- Compliance with micro and nanoscale fabrication.

• Compatibility with diverse range of fabrication techniques such as electrochemical, optical, mass- based, etc.

• Biomaterials such as enzymes, antibodies, whole cells, and nucleic acids can be incorporated into the polymer matrix.

- Strong biomolecular interactions.
- Low detection limits.
- Enhanced sensitivity (when used as a composite material with nanoparticles).
- Reversible responses at ambient temperatures.





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• Cost effectiveness.

Sensor Applications

A variety of sensors have been formulated using conducting polymers in different transduction modes. The transduction modes can be divided into five main classes based upon the operating principle into conductometric, potentiometric, amperometric, colorimetric and gravimetric modes. The conductometric mode uses changes in electrical conductivity in response to an analyte interaction. The conductivity of a conducting polymer material bridging the gap between two adjacent electrodes is commonly measured as a function of analyte concentration, and it can be also monitored with a fixed potential in solution. Potentiometric sensing mode is based on analyte-induced changes in the chemical potential of a system when no current is flowing. The change in the opencircuit potential of the system is monitored, which is mostly proportional to the logarithm of the concentration of analyte. The chemical and diffusion processes have to be at equilibrium conditions in the potentiometric mode for a thermodynamically accurate signal to yield. Amperometric mode refers to either single-potential amperometry or variable-potential amperometry. The principle of amperometric sensing is to measure the current generated by the redox reaction of an analyte at a sensing (working) electrode, where the current is subject to Faraday's law and a dynamic reaction achieving steady-state conditions in the system.colorimetric and gravimetric modes.

Voltammetric mode is also a variant of amperometric mode. It monitors the change in current while varying the applied potential. Colorimetric sensors quantitate changes in optical absorption characteristics, which depend on the local electronic structure. The sensitivity of the bandgap of conducting polymers to analyte-induced changes provides a useful means to create this kind of sensor. Lastly, gravimetric mode takes advantage of a weight change in a conducting polymer as a result of analyte-polymer interaction. Minute weight changes in the polymer can be normally monitored using a quartz crystal microbalance. Numerous conducting polymer sensors based on these transduction mechanisms have been devised in order to detect various chemical and biological species.

Name	Structure
Polypyrrole (PPy)	
Polyaniline (PANI)	{<_>#
Polythiophene (PTh)	

Table. 1- Representative conducting polymers.





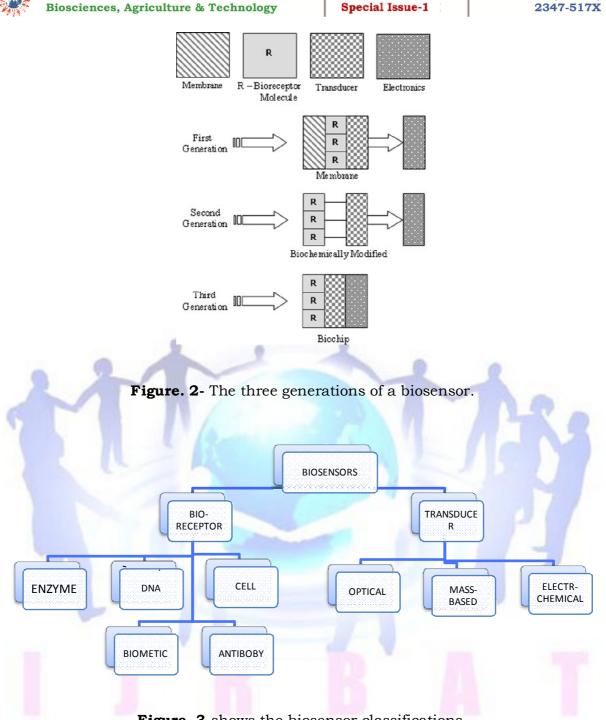
Name	Structure
Poly(3,4-ethylenedioxythiophene) (PEDOT)	

Table. 2-summarizes the templates frequently used for fabricating nanostructures and the possible resulting products.

Products	Ten	nplates	Templates	
	Soli	d -Phase	Molecular	
Nanospheres /Nanocapsule		ospheres	Spherical micelles	
,	00	1		
. 21	°	~		
Nanorods/fibe Nanotubes	ers/ Por	ous matrices	Rod like micelles	20
Nanofibers /N	anotubes Nar	orods	Liquid crystals	
	R	R		T
Tai	npurities get get lyte ioreceptor Transducer A		Record nd Display	

Figure. 1-shows schematic diagram of a biosensor.

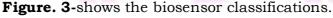




Feb. 2015

ISSN No. (Online)

International Journal of Researches In



Conclusion:

A number of studies have demonstrated that conducting polymer nanomaterials are promising candidates for building state-of-the-art sensors, due to their unique advantages over other materials. The most important parameters that determine the sensor performance include response/recovery time, sensitivity, selectivity, and stability. A great deal of effort has been directed toward enhancing these parameters over the past decades. The response and recovery times and the sensitivity have experienced impressive improvements with great advances in nanotechnology. However, selectivity is still a challenge. A conducting polymer by itself lacks the specificity or selectivity toward target species, and thus, it is crucial



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to judiciously functionalize the polymer with appropriate receptors. Another concern is that conducting polymers may degrade over time, even in dry, oxygenfree environments. New efforts should be aimed at improving the stability of the sensor response.Sensors play an ever-increasing role in environmental monitoring, medical diagnosis, industrial safety control, security, and so forth. Conducting polymer nanomaterials are believed to have much unexplored potential for sensor applications. Thus, future research into conducting-polymer nanomaterials-based sensors will offer great potential for the construction of next-generation sensor devices. In particular, it is anticipated that wearable or flexible high-performance sensors will be developed using conducting polymer nanomaterials in the near future.

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