Recent Development of Morphology Controlled Conducting Polymer Nanomaterial-Based Biosensor

Sunghun Cho¹ and Jun Seop Lee ²,*

¹ School of Chemical Engineering, Yeungnam University, Gyeongsan 38541, Korea; shcho83@ynu.ac.kr
² Department of Materials Science and Engineering, Gachon University, 1342 Seongnam-Daero, Sujeong-Gu, Seongnam-Si, Gyeonggi-Do 13120, Korea
* Correspondence: junseop@gachon.ac.kr; Tel.: +82-31-750-5814

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Abstract: Biosensors are of particular importance for the detection of biological analytes at low concentrations. Conducting polymer nanomaterials, which often serve as sensing transducers, are renowned for their small dimensions, high surface-to-volume ratio, and amplified sensitivity. Despite these traits, the widespread implementation of conventional conducting polymer nanomaterials is hampered by their scarcity and lack of structural uniformity. Herein, a brief overview of the latest developments in the synthesis of morphologically tunable conducting polymer-based biosensors is discussed. Research related to the dimensional (0, 1, 2, and 3D) hetero-nanostructures of conducting polymers are highlighted in this paper, and how these structures affect traits such as the speed of charge transfer processes, low-working temperature, high sensitivity and cycle stability are discussed.

Keywords: conducting polymers; biosensors; polypyrrole; polyaniline; poly(3,4-ethylenedioxythiophene); aptamers; morphology control

1. Introduction

Biosensors are crucial for numerous applications in healthcare, medical science, agriculture, environmental monitoring, food, and biosecurity because of their extremely precise diagnostic and monitoring capabilities [1–5]. Analytes are detected with the aid of biological-sensing elements such as antibodies, cells, receptors, and aptamers, which are bound to transducers made from conductive nanomaterials [6–8]. Quantitative analysis of the target analyte is achieved by selectively converting the molecular recognition processes between the analyte and the biological element bound to the transducers from a nonelectrical domain to an electrical signal [9–12]. The binding of the analyte induces changes in the electrical properties of the transducers. An effective biosensor exhibits several crucial characteristics, namely, high sensitivity, good selectivity, fast response/recovery time, cycle stability, and the capability to operate at low temperatures. In addition to satisfying all of these prerequisites, conductive nanomaterial-based electrodes are renowned for their small size, high surface-to-volume ratio, and unique optical/electrical properties, thereby making them excellent candidates for biosensor-assisted applications [13–17].

Conducting polymers are often employed as transducer materials for biosensor applications because of the unique traits inferred by their unusual π-conjugated systems [18–22]. Conducting polymers contain polyconjugated chains consisting of alternating single (σ) and double (π) bonds that influence the electrical and optical properties of the sensing transducer [23–25]. Several parameters govern the physical properties of conducting polymers, including the length of the conjugation, the existing intra-/interchain interactions, and the extent of the disorder in the system [26,27]. Furthermore, research has shown that the partial or total inclusion of conducting polymer nanostructures in sensor transducers leads to significantly improved sensing performance [28–32]. The ability to selectively tune
defects, electronic states, and the surface chemistry has been the driving force behind diverse methods for fabricating morphologically tunable conducting polymer nanomaterials [33–36]. Additionally, assembling conducting polymer nanostructures into an ordered array is often necessary to render them functional [37,38]. Novel nanostructure-based sensing devices exhibit enhanced abilities that are generated through the development of new strategies for combining and assembling nanoscale units [39–42].

Various synthetic methods have been reported, of which the template method is the most promising and powerful tool for fabricating conducting polymer nanomaterials [43–51]. The template method involves the inclusion of inorganic or organic guest molecules in the voids of the host material. These voids act as the “template”, thereby defining the shape, size, and orientation of the compound being produced. Generally, the template method is classified into three different types, namely, hard, soft, and template free methods. Briefly, the “hard” template process involves the fabrication of 1D nanostructures such as nanotubes and nanorods using anodic aluminum oxide membranes, track-etched polycarbonates, and zeolites [52–55]. The “soft” template method involves the fabrication of various morphologies of conducting polymer nanomaterials. Here, the templates used include surfactants, liquid–crystalline polymers, cyclodextrins, and functionalized polymers [56–59]. Surfactants that include cationic, anionic, and non-ionic amphiphiles are commonly used to form micelles as nanoreactors. Relative to the other two methods, which provide facile, practical routes for the production of pure, uniformly sized, high-quality nanofibers, the “template-free” method encompasses various techniques such as dispersion polymerization and electrochemical, chemical, aqueous/organic interfacial syntheses [60–66].

In this review, recent trends in the development of biosensor electrodes based on conducting polymer nanomaterials are presented. Additionally, the synthetic routes that facilitate the production of morphologically tunable nanostructures are closely examined.

2. Polypyrrole-Based Electrodes

Polypyrrole (PPy), a class of conducting polymers possessing five-membered heterocyclic rings, is renowned for their high electrical conductivity, reversible redox activity, and environmental stability. Ease of synthesis and tunable conductivity make PPy particularly useful for applications involving nanoscale architectures [67,68] as their nanostructures can be readily prepared via electrochemical and chemical routes in both aqueous and non-aqueous solutions [69,70]. Electrochemical polymerization is commonly used to fabricate deposited films on substrates, whereas chemical polymerization is used to produce powdered products. Structurally, the PPy chain is intrinsically planar and linear because of α–α coupling. However, conformational defects (e.g., α–α bonds with non-regular rotation) and structural defects (i.e., the presence of α–β bonds, hydroxyl moieties, and carbonyl groups) induce structural changes that facilitate their application for the production of biosensors [71,72].

2.1. 0D Nanostructures

As stated earlier, the popularity of PPy nanoparticles as biosensor sensing materials is due to the ability to incorporate various diverse receptors for analyte detection thanks to the large surface to volume ratio of these polymers. In particular, spherical PPy, which are most commonly used for producing biosensors, can be prepared via numerous methods, namely, microemulsion polymerization, self-assembly, and dispersion polymerization. Of the three methods listed, microemulsion polymerization is the most extensively utilized means of fabricating diverse nanometer-sized conducting polymer particles at low temperatures [73,74]. In contrast, the self-assembly approach takes advantage of the evaporation processes occurring in the liquid reaction medium [75]. Dispersion polymerization features the synthesis of sterically stabilized PPy colloids using a tailor-made reactive polymeric stabilizer [76,77]. Hong et al. described the preparation of PPy nanoparticles with a diameter range of 20–100 nm using FeCl₃ in aqueous solutions containing PVA as a stabilizer [78]. Moreover, the generation of uniformly sized PPy nanoparticles
for biosensor transducers was best conducted when the carboxylated polypyrrole (CPPy) chain was synthesized via the co-polymerization of pyrrole and 3-carboxyl pyrrole monomers [79].

Oh et al. reported introducing uniformly sized nanoparticles to the electrode’s surface using the spin coating technique to solve the issues of limited surface area caused by nanoparticle aggregation. Additionally, amide coupling between the particle of interest and the electrode’s surface induces greater electrode stability (Figure 1a) [79]. In Oh’s study, the human parathyroid hormone receptor (PTH), which is vital for maintaining calcium homeostasis in mammals, was incorporated into the close-packed CPPy nanoparticle-based electrode. Irregularities associated with internal PTH levels result in potentially fatal diseases such as adenoma, hyperplasia, osteoporosis, and cancer. Here, the sensor electrode was extremely sensitive to human PTH and exhibited high stability because of amide formation reactions between the nanoparticle and the electrode. Lee et al. incorporated platinum particles onto the surface of CPPy to fabricate nonenzymatic biosensors capable of detecting the neurotransmitter, dopamine (Figure 1b) [80]. In conjunction with platinum catalysts, this biosensor consistently detected dopamine at concentrations as low as 100 fM without the involvement of other biomaterials. Oh et al. recently manufactured a biosensor electrode that could detect the odor molecule, cadaverine, in the gas phase using nickel-attached PPy nanoparticles (Figure 1c) [81]. Here, the strong binding interaction between the olfactory receptor embedded nanoparticles, namely, TAAR13c-embedded nanodisk, and nickel was vital for cadaverine detection. In this study, the TAAR13c-embedded nanodisk was introduced stably onto the electrode coated with the nanoparticles of interest. This artificial olfactory biosensor was capable of detecting low concentrations of cadaverine in both liquid (100 aM) and gaseous (10 ppb) phases.

Figure 1. (a) Schematic of CPPy nanoparticle-based biosensor electrodes for the detection of peptide hormones [79]. Copyright 2012, American Chemical Society. (b) Platinum-coated CPPy nanoparticle-based non-enzyme biosensors for dopamine detection [80]. Copyright 2015, Wiley. (c) A nanodisk-functionalized bioelectronic nose based on nickel-coated CPPy nanoparticles [81]. Copyright 2019, American Chemical Society.

2.2. 1D Nanostructures

One-dimensional nanomaterials are popular sensing elements in analytical systems as they enhance the efficiency and performance of the associated biosensors via the rapid transfer of electrical signals and their large surface-to-volume ratio. These nanostructures significantly decrease the detection
limit of biosensors by providing numerous immobilization sites, which, in turn, increases the number of biomolecular interactions [82]. Moreover, their high porosity facilitates fast penetration of the biomolecules, thereby reducing the detection time [83].

1D PPy-based nanomaterials are also synthesized using various manufacturing methods. Oh et al. synthesized human taste receptor-functionalized CPPy nanotubes to detect bitter compounds that produced phenylthiocarbamide and propylthiouracil [84]. CPPy nanotubes were fabricated as taste receptor templates using reverse microemulsion polymerization. Recently, Na et al. reported synthesizing CPPy nanotubes via a facile self-degradation method utilizing fibrillary templates composed of methyl orange and iron chloride (Figure 2a) [85]. Here, the carboxyl pyrrole monomer was incorporated into the fibrillar template via electrostatic interactions between the pyrrole monomer and the sulfite group of methyl orange. Simultaneously, there was a reaction between carboxyl pyrrole and Fe$^{3+}$ cations on the surface of the fibrillar template. The synthesized CPPy nanotubes were then applied as sensing transducers to detect 17$\beta$-estradiol, a renowned inhibitor that affects hormone immunity and other physiological processes.

Kim et al. fabricated platinum-coated CPPy nanofibers using an oxidative template and sonochemical methods (Figure 2b) [86]. Here, CPPy nanofibers were formed when the electrode underwent charge transfer processes via an oxidative template composed of cetrimonium bromide and ammonium persulfate. The oxidative templates exhibited lamellar micelle structures and were composed of folded, twin-tailed complexes of the cetylammonium cations and peroxydisulfate anions. The unique micelle structure generated by the cetylammonium cation–peroxydisulfate anion interactions resulted in the polymerized nanofibers’ fibrous shape. Next, composite nanomaterials were formed using a simple chemical reduction of the platinum precursors. The platinum-coated CPPy nanofibers were useful for nonenzymatic biosensors to detect oxalic acid, a notable indicator of renal tubular acidosis, hyperparathyroidism, medullary sponge kidney, hyperoxaluria, and Dent’s diseases.

2.3. 2D Nanostructures

Two-dimensional nanomaterials are widely used as sensor electrodes as they exhibit uniformed structures in a wide surface area, making them ideal for the production of thin films. Most PPy-based films are easily synthesized using electrochemical deposition techniques and can be used to generate biosensor electrodes via the introduction of biological receptors [87,88]. Electrochemical deposition is a simple method used to obtain conducting polymer thin films containing electrolytes incorporated as dopants. Another class of 2D nanostructure films, namely, composite-based conducting polymers, can be employed for various functions. Arora et al. synthesized double-stranded calf thymus DNA-functionalized PPy-polyvinyl sulphonate thin-films using electrochemical deposition for detecting 2-aminoanthracene [87]. Ozer et al. fabricated polymer composite films consisting of

![Figure 2. (a) Synthesis of methyl orange–iron chloride template CPPy nanotubes for 17$\beta$-estradiol-based biosensors [85]. Copyright 2016, Royal Society of Chemistry. (b) Platinum-coated CPPy nanofibers for the production of nonenzymatic biosensors capable of detecting oxalic acid [86]. Copyright 2018, Royal Society of Chemistry.](image-url)
PPy-dodecylbenzene sulphonate for the synthesis of cholesterol-sensing electrodes [88]. Weng et al. manufactured a composite ink made from PPy and polyvinyl alcohol mixtures that were then used to generate nanofilms for screen printing [89]. This conductive polymeric ink-based electrode was an effective biosensor for detecting glucose complexed glucose oxidase.

2.4. 3D Nanostructures

The detection capabilities of biosensor electrodes can be improved by maximizing the number of attached receptors per unit volume. This is achieved using 3D nanostructures possessing maximized surface areas as the biological receptor templates. Researchers have been actively involved in manufacturing 3D nanostructures using PPy for subsequent applications as biosensors.

As seen in one-dimensional structures, multidimensional PPy nanotubes with vertically coated bumps were manufactured using polyvinyl alcohol fibers as the mold via vapor deposition polymerization of the pyrrole monomer (Figure 3a) [68,90]. The 3D PPy nanotubes are ideal for monitoring cortisol hormone levels, thereby ensuring accurate regulation of blood pressure, glucose levels, and carbohydrate metabolism in response to environmental factors and the circadian rhythm. Relative to 2D nanofilms, 3D PPy films are manufactured via vapor deposition polymerization on the surface of the Co$_3$O$_4$ nanoplate attached perpendicular to the graphene electrode. The resulting thin film can be used to detect platelet-derived growth factor BB (PDGF BB), a bioindicator of blood cancer [91]. Cho et al. modified the electrochemical deposition technique to synthesize 3D PPy nanofilms with vertically coated nanorods on the electrode [92], thereby enabling the rapid transfer of electrical signals for sensing purposes. These nanofilms were ideal sensor electrodes for detecting HBsAg, a hepatitis B factor (Figure 3b). Relative to the 0D nanostructures discussed previously, urchin-like 3D PPy nanoparticles were fabricated using a combination of electrospray and vapor deposition polymerization of pyrrole (Figure 3c) [93]. Next, binding aptamers were introduced onto the particle’s surface to detect the environmental hormone, bisphenol A, as these aptamer-functionalized 3D nanoparticles were extremely sensitive environmental hormone sensors. The sensing performance of PPy-based biosensors are suggested in Table 1.

![Figure 3.](image-url)
Table 1. Comparative sensing performance and limit of detection of polypyrrole nanostructure-based biosensors.

| Nanostructure | Receptor | Analyte | Mechanism | LOD \(^1\) | Response Time | Working Temperature | Linearity | Cycle Stability | Reference |
|---------------|----------|---------|-----------|------------|---------------|---------------------|-----------|----------------|-----------|
| CPPyNP \(^2\) | hPTH 3   | hTPH 4  | FET 4     | 48 fM      | <10 s         | 25 °C               | 48 fM-480 pM | 2 weeks        | [79]      |
| Pt_CPPyNP     |          |         | Dopamine FET | 100 fM    | <10 s         | 25 °C               | 0.1 pM-1 nM | 4 weeks        | [80]      |
| Ni-CPPyNP 5   | TAAR13c 5-embedded nanodisk | Cadaverine | FET | 100 aM | <5 s | 25 °C | 0.1 fM-100 µM | 5 weeks | [81] |
| CPPyNT 17β-estradiol binding aptamer | hTAS2R38 6 | PTC 7, PROP 8 | FET | 1 fM | <5 s | 25 °C | 1 fM-1 µM | 1 week | [84] |
| Pt_CPPyNF     |          | Oxalic acid | FET | 100 fM | <10 s | 25 °C | 10 fM-100 pM | 8 weeks | [86] |
| PPy-PVS 9 film | CT-dsDNA 10 | 2-aminoanthracene o-chlorophenol | Amperometric | 0.01 ppm | 0.1 ppm | <30 s | 25 °C | 0.01-20 ppm 0.1-30 ppm | - | [87] |
| PPy-DBS 11 film | ChOs 12 | Cholesterol | Amperometric | 0.11 µM | - | 25 °C | 0.11 µM-1.9 mM | 30 days | [88] |
| PPy-PVA 13 GoD 14 | HRP | H2O2 Glucose | Amperometric | 10 µM | 1 mM | <10 s | 25 °C | 10 µM-10 mM 1-5 mM | - | [89] |
| 3D PPyNF 15 | F4P1A3 | Cortisol | FET | 100 aM | <5 s | 25 °C | 100 aM-10 nM | 30 days | [90] |
| 3D CPPy 16 plate-based film | PDGF-B binding aptamer | PDGF-BB 16 | FET | 1.78 fM | <10 s | 25 °C | 1.78 fM-17.8 pM | 4 weeks | [91] |
| 3D PPy 17 film | HBsAg-binding aptamer | HBsAg 17 | FET | 10 aM | <10 s | 25 °C | 10 aM-0.1 µM | 500 cycles | [92] |
| Urchin-like CPPyNP | BPA binding aptamer | BPA 18 | FET | 1 fM | <10 s | 25 °C | 1 fM-10 pM | 4 weeks | [93] |

\(^1\) Limit of detection, \(^2\) carboxylated polypyrrole nanoparticle, \(^3\) human parathyroid hormone, \(^4\) field effect transistor, \(^5\) olfactory receptor, \(^6\) bitter taste receptor, \(^7\) phenylthiocarbamide, \(^8\) propylthiouraail, \(^9\) polypyrrole-polyvinyl sulphonate, \(^10\) double stranded calf thymus DNA, \(^11\) polypyrrole-dodecyl benzene sulphonate, \(^12\) cholesterol oxidase, \(^13\) horseradish peroxidase, \(^14\) glucose oxidase, \(^15\) cortisol antibody, \(^16\) serum hepatis B surface antigen, \(^17\) platelet-derived growth factor, \(^18\) bisphenol A.
3. Polyaniine-Based Electrodes

The conductive states of polyaniine (PANI), which were first highlighted by Macdiarmid et al. are of research importance because of their redox behavior, simple doping/dedoping conditions, good environmental stability, facile synthesis, and lower synthetic costs compared to their counterparts [94,95]. PANI is particularly useful for analyte detection in a matrix of immobilized biomolecules, as it facilitates electron transfer processes in redox or enzymatic reactions. Therefore, PANI serves as a mediator for enzyme electrodes, as it undergoes redox cycling reactions and facilitates the direct coupling of enzyme active site electrons to the electrode’s surface. The size- and shape-dependent properties of PANI-based nanomaterials are crucial for fabricating high-performance sensors because of their unique charge-transport properties such as their doping and oxidation levels and the length of conjugation. These traits are strongly influenced by the shapes of the materials, whether they are nanofibers, nanowires, nanorods, nanotubes, or nanoparticles [96]. Extensive research is still needed to establish sustainable synthetic routes that enable researchers to adjust the morphology of the resulting PANI nanomaterials, thereby making them effective biosensor electrodes.

3.1. 0D Nanostructures

PANI-based nanoparticles, which are often available as pristine nanoparticles rather than functional coating materials, can be employed to improve binding attractions with the respective biomolecules. He et al. synthesized hollow carbon spheres coated with needle-like polyaniine to produce biosensor electrodes for the detection of malathion, an organophosphate insecticide (Figure 4a) [97]. Briefly, hollow carbon spheres were synthesized using the SiO$_2$ template method, after which PANI was coated on the surface via in situ oxidative polymerization of the aniline within the hollow carbon sphere. Acetylcholine esterase was then immobilized on the PANI’s surface by the amide group in the chain. As reported by Salimian et al. Au nanoparticles were attached to PANI’s surface to enhance binding attractions with the HBV binding aptamer biomolecules [98]. Conversely, Al-Sagur et al. fabricated lutetium phthalocyanine containing silica–PANI conducting nanobeads without the need to etch the core particle (Figure 4b) [99]. In this case, the SiO$_2$ core was vital for attaching glucose oxidase bioreceptors by introducing lutetium phthalocyanine. Thus, PANI served to interact with glucose oxidase and form thin films, thereby enabling glucose sensing.

**Figure 4.** (a) Hollow carbon spheres coated with needle-like PANI-based pesticide biosensors [97]. Copyright 2018, Elsevier. (b) Lutetium phthalocyanine incorporated silica–PANI nanobeads for use as glucose electrochemical sensors [99]. Copyright 2018, Elsevier.
3.2. 1D Nanostructures

One-dimensional PANI nanostructures are easily generated using various methods and exhibit excellent electrical signal transmission capacity. Lou et al. fabricated PANI nanowires using electrochemical polymerization, followed by the incorporation of Au nanoparticles onto the electrode’s surface (Figure 5a) [100]. This composite nanostructure was then bound to glucose oxidase for use as electrochemiluminescent glucose sensors. Additionally, Hui et al. coated polyethylene glycol (PEG) layers on the surface of PANI to enhance the antifouling properties of the nanofiber. The PEGlated PANI nanofibers were applied as breast cancer susceptibility gene aptamer sensors (Figure 3b) [101]. Additionally, Asmatuli et al. synthesized PANI-based nanofibers using a facile electrosprinning process to obtain uniformly sized polymer nanofiber webs (Figure 5c) [102]. The nanofibers were then applied to detect cyclooxygenase-2 related to tumor cell proliferation, differentiation, pain, and adhesion. By contrast, PANI nanotubes were generated through a different technique. Wang et al. synthesized PANI nanotube-based glucose enzyme sensors using self-assembly and molecular imprinting methods [103]. Conversely, Soni et al. fabricated PANI nanotubes using the MnO$_2$ nanofiber template to produce chronic myelogenous leukemia-sensing aptamer sensors [104].

3.3. 2D Nanostructures

Various methods were used to make PANI nanofilm-based biosensor electrodes with a large surface area. Oh et al. utilized the inkjet printing technique to create ammonium persulfate initiator solutions that were used to produce patterned PANI nanofilms via vapor deposition polymerization of aniline (Figure 6a) [105]. Inkjet printing is an efficient alternative to conventional photolithographic methods for the production of versatile CP-based micro- or nanoelectronics because of its cost-effectiveness, high-speed patterning capabilities, flexibility, morphological adjustability, and widespread applicability. The patterned PANI layer was applied for dopamine detection by monitoring the observed changes in the flowing current. Kaliasa et al. synthesized PANI nanosheet-based thin-films using the screen printing method to generate glucose electrochemical sensors [106]. Additionally, they synthesized
NiO-coated PANI nanosheets using in situ polymerization processes to facilitate glucose detection (Figure 6b) [107]. Conversely, Liu et al. attached 2D PANI layers directly onto silicon substrates using dilute oxidative polymerization reactions to fabricate B-type natriuretic peptide (BNP) biomarker FET sensors [108]. Note also that electrochemical deposition was effective for manufacturing polyaniline nanofilms. Yun et al. generated electrodeposited 2D PANI layers on a gold electrode pattern for use as FET BNP antibody FET sensors (Figure 6c) [109].

3.4. 3D Nanostructures

Three-dimensional PANI nanostructures are most commonly obtained via the formation of hydrogels using crosslinking agents; this facilitates their application as diverse biosensor systems that are grounded in the working mechanisms governing the introduction of nanomaterials. Using amino trimethylene phosphonic acid as the crosslinking agent, Xu et al. added silver nanoparticles functionalized by a luminol derivative, N-(aminobutyl)-N-(ethylisoluminol) (ABEI), onto the surface of 3D PANI nanostructures to generate electrochemiluminescent sensors for xanthine detection (Figure 7a) [110]. Qing et al. employed phytic acid as the crosslinking agent to facilitate the introduction of photoactive materials such as poly[4,8-bis[5-(2-ethylhexyl)thiophen-2-yl]benzo[1-2-b:4,5-b’]dithiophene-2,6-diyl-alt-3-fluoro-2-[(2-ethylhexyl)carbonyl]thieno[3–4-b]thiophene-4,6-diyl] (PTB7–Th) to the electrode’s surface for subsequent use as photoelectrochemical guanine sensors (Figure 7b) [111]. Li et al. also used 3D PANI derived from phytic acid as uric acid electrochemical biosensors [112].
Additionally, research has been conducted on the production of unique 3D composite materials via a simultaneous synthetic route using PANI and other materials. Soni et al. fabricated PANI–MoS$_2$ nanoflower composites by taking advantage of the hydrothermal reaction between the PANI nanoneedle and the MoS$_2$ precursor [113]. Here, the resulting nanocomposites were functionalized for chronic myelogenous leukemia (CML)-binding aptamers as biosensors for impedance spectroscopy. Lakshmi et al. made PANI-graphene composite microflowers via the in situ polymerization of aniline and graphene layers for use as cholesterol detecting biosensors [114]. The sensing performance of PANI-based biosensors are suggested in Table 2.
Table 2. Comparative sensing performance and limit of detection of polyaniline nanostructure-based biosensors.

| Nanostructure                  | Receptor            | Analyte          | Mechanism       | LOD 1 | Response Time | Working Temperature | Linearity         | Cycle Stability   | Reference |
|--------------------------------|---------------------|------------------|-----------------|-------|---------------|---------------------|-------------------|-------------------|-----------|
| HCS@PANI 2                     | AChE 3              | Malation         | Electrochemical | 2.15  pM | -             | 25 °C               | 1.0 ng mL⁻¹–10 µg mL⁻¹ | -       | [97]      |
| HCS-PANI 1                     | Thiolated-probe DNA | Hepatitis B Virus| Electrochemical | 3.62  fM | -             | 25 °C               | 10 fM–1 nM        | -                 | [98]      |
| SiO₂ (LuPc₂) PANI (PVIA)-CNB 5 | Gox                 | Glucose          | Electrochemical | 0.1   mM | <2 s          | 25 °C               | 1–16 mM          | 45 days          | [99]      |
| Au/PANI NW                     | Gox                 | Glucose          | ECL 6           | 0.05  µM | -             | 25 °C               | 0.1–100 µM        | 40 cycles        | [100]     |
| PEClated PANI NF              | Aptamer             | BRCA1 7           | Electrochemical | 3.8   fM | -             | 25 °C               | 0.01 pM–1 nM      | -                 | [101]     |
| Electrosan PANI NFs           | COX-2 polyclonal antibody | COX-2 enzyme 8 | Electrochemical | 0.01 pg mL⁻¹ | -             | 25 °C               | 0.01 pg mL⁻¹–1 µg mL⁻¹ | -       | [102]     |
| PANI NTs                       | HRP 9               | H₂O₂             | Electrochemical | 8.1   fM | <200 s        | 25 °C               | 0.01–90 µM        | -                 | [103]     |
| PANI NTs                       | Aptamer             | CML 10           | Electrochemical | 0.1   fM | -             | 25 °C               | 0.1 fM–1 µM       | 40 days          | [104]     |
| 2D PANI                        | RGD 11              | Dopamine         | Electrical      | 2     nM | -             | 25 °C               | 2 nM–1 µM         | 48 h              | [105]     |
| PANI nanosheet                 | -                   | Glucose          | Electrochemical | 0.043 µM | <1 s          | 25 °C               | 1 µM–1 mM         | -                 | [106]     |
| NiO/PANI nanosheets            | -                   | Glucose          | Electrochemical | 0.06  µM | <10 s         | 25 °C               | 1 µM–1 mM         | 60 days          | [107]     |
| 2D PANI layer                  | Antibody            | BNT 12           | FET 13          | 50    pg mL⁻¹ | -             | 25 °C               | 50–1000 pg mL⁻¹   | -                 | [108]     |
| 2D PANI                        | Antibody            | BNT              | FET             | 100   pg mL⁻¹ | -             | 25 °C               | 100–1000 pg mL⁻¹  | -                 | [109]     |
| 3D PANI hydrogel/Ag/ABEI       | Xanthine oxidase    | Xanthine         | ECL             | 9.6   nM | -             | 25 °C               | 0.01–200 µM       | -                 | [110]     |
| 3D PANI hydrogel/PtB7-Th       | Xanthine oxidase    | Guanine          | PEC 14          | 0.02  µM | <2 s          | 25 °C               | 0.1–80 µM         | 2 days            | [111]     |
| 3D PANI hydrogel/Pt            | Uricase             | Uric acid        | Electrochemical | 1     µM | <3 s          | 25 °C               | 0.07–1 mM         | -                 | [112]     |
| 3D PANI-MoS₂ nanoflower        | Aptamer             | CML              | Electrochemical | 3     aM | -             | 25 °C               | 0.01 fM–1 µM      | -                 | [113]     |
| PANI/graphene microflower      | -                   | Cholesterol      | Electrochemical | 1.93  mg dL⁻¹ | -             | 25 °C               | 1.93–464.04 mg mL⁻¹ | -       | [114]     |

1 Limit of detection, 2 hollow carbon spheres coated with needle-like polyaniline, 3 acetylcholine esterase, 4 hollow carbon spheres decorated with polyaniline, 5 lutetium phthalocyanine incorporated silica-polyaniline conducting nanobeads, 6 electrochemical luminescence, 7 breast cancer susceptibility gene, 8 cyclooxygenase-2, 9 horseradish peroxidase, 10 chronic myelogenous leukemia, 11 arginine-glycine-aspartate peptide, 12 B-type natriuretic peptide, 13 field effect transistor, 14 photoelectrochemical.
4. Poly(3,4-ethylenedioxythiophene)-Based Electrodes

Poly(3,4-ethylenedioxythiophene) (PEDOT), which is a polythiophene (PT) derivative, is an outstanding conducting polymer for practical applications because of its small bandgap, good optical properties, and high electrical conductivity compared to other conducting polymers [115]. PEDOT exhibits excellent electroactivity in phosphate buffers, making it ideal for biosensor applications, particularly in enzyme matrices for immobilization processes. Most importantly, its mode of action features the direct transfer of electrons during detection [116]. Given these findings, numerous studies have attempted to synthesize PEDOT nanostructures using different electrochemical and chemical oxidative polymerization methods.

4.1. 0D Nanostructures

The functional groups of PEDOT-based 0D materials facilitate the formation of composites containing various components and moieties in the polymer chain’s structure. Jiang et al. synthesized Pt-coated PEDOT nanospheres using a Pt precursor (H$_2$PtCl$_4$) as an oxidant during polymerization [117] for subsequent application as H$_2$O$_2$ non-enzyme sensors. Liu et al. fabricated Pt-coated PEDOT nanospheres using CaCO$_3$ sphere template-assisted chemical polymerization to generate electrochemical glucose sensors (Figure 8a) [118]. Kim et al. synthesized MnO$_2$-coated PEDOT nanoellipsoids by conducting reverse microemulsion reactions following reduction processes. The composite nanoellipsoids were then used to detect catechol amine in PC-12 cells (Figure 8b) [119]. Additionally, commercial PEDOT:PSS has been used to generate the associated composite nanoparticle biosensors. Wang et al. synthesized nanobeads composed of polyethyleneimine binding with ferrocene (BPEI–FC) and PEDOT:PSS via chemical coupling reactions to create glucose biosensors (Figure 8c) [120]. Zhang et al. produced PEDOT:PSS-based micelle structures via electrostatic interactions with chitosan for subsequent application as H$_2$O$_2$ biosensors (Figure 8d) [121].

4.2. 1D Nanostructures

Park et al. synthesized carboxylated PEDOT nanotubes using microemulsion polymerization as templates for immobilizing human dopamine receptor nanovesicles [122] that were used to coat extremely sensitive PEDOT-based dopamine sensors. These sensors had a low limit of detection of 10 fM. Commercial PEDOT:PSS offers the advantage of easy processing, and is integral to the formation of 1D structures via numerous manufacturing methods. Jiang et al. generated PEDOT:PSS/silk fibroin core/shell wires for detecting ascorbic acid using wet-spinning and photo-crosslinking methods (Figure 9a) [123]. Wang et al. fabricated PEDOT:PSS nanowires via soft-lithography before introducing biotin-derivatized poly(L-lysine)-grafted oligo-ethylene glycol (PLL-g-OEG-Biotin) into the doped polymer chain [124]. The PEDOT:PSS-based nanowires were ideal electrochemical biosensors for detecting streptavidin. Moreover, PEDOT offers the advantage of expanded functionality since the surface of the 1D materials can be covered using PEDOT-coated PAN nanofibers via the VDP of EDOT. Next, the PAN/PEDOT core/shell nanofibers were employed as electrodes for electrochemical glucose sensors [125]. 1D PEDOT nanostructures were also fabricated using a simple electrochemical polymerization method; in this case, Chen et al. synthesized Au–hemoglobin–PEDOT nanowhisker composites for H$_2$O$_2$ detection (Figure 9b) [126].
4.3. 2D Nanostructures

Even though electrochemical polymerization is most commonly employed to manufacture conducting polymer films, the use of PEDOT tends to be more widespread since it results in uniformity in the large surface area and high stability. David et al. fabricated PEDOT:PSS-based “layer-by-layer” films consisting of PEDOT:PSS/chitosan/glucose oxidase + N-graphene molecules for glucose detection [127]. Briefly, PEDOT:PSS was obtained via electrodeposition method before the positively charged chitosan layer was adsorbed onto the negative PSS chain. A third layer consisting of glucose oxidase + N-graphene was introduced via physical adsorption onto the positively charged chitosan layer. Kim et al. generated Au nanoparticles coated with PEDOT films using a one-pot modified electrodeposition method [128]. Next, anti-vascular endothelial growth factor (VEGF) antibodies were added for detecting VEGF, which is associated with severe vision loss due to exudative age-related macular degeneration, advanced diabetic retinopathy, and retinal vascular occlusive disease. Moreover, Scotto et al. fabricated layer-by-layer PEDOT/graphite nanosheet-based film using simple spin-coating method [129]. In detail, EDOT and graphite composite solution was deposited by spin-coating process and then exposed oxidant (Fe (III) tosylate) with high temperature. Then, glucose oxidase (GOx) and osmium-modified polyallylamine (OsPA) were coated layer-by-layer on the films surface to detect glucose. In the sensing performance, incorporated graphite nanosheet in the PEDOT structure not only improves the voltammetric response of the resulting all-polymer electrodes but also produces a better integration of the electrochemically active supramolecular assembly allowing the effective glucose sensing. In addition, Wang et al. suggested low fouling electrochemical biosensor
using zwitterionic polypeptide doted PEDOT film by simple electrodeposition process to detect breast cancer biomarker (BRCA1) detection [130]. Especially, the polypeptide in the film structure acted as a bifunctional material that not only inhibits the nonspecific adsorption of proteins but also allows for the usage of its carboxylic to fix DNA probes (terminated with amino group).

Figure 9. (a) PANI:PSS/silk fibroin core/shell wires derived via wet spinning and photo crosslinking reactions for use as ascorbic acid biosensors [123]. Copyright 2019, MDPI. (b) PEDOT nanowhiskers–hemoglobin composite electrodes for \( \text{H}_2\text{O}_2 \) detection via electrodeposition [126]. Copyright 2013, Royal Society of Chemistry.

4.4. 3D Nanostructures

PEDOT-based 3D nanostructures can be manufactured by modifying conventional synthetic routes. Here, Jia et al. fabricated 3D Au–PEDOT–graphene aerogels using the in situ polymerization of EDOT on 3D graphene aerogels and the electrodeposition of Au on the surface [131]. This composite nanostructure is a highly sought after material with a 3D PEDOT-based surface via PEDOT coating of the 3D graphene skeleton. Antibodies for detecting prostate cancer factors were introduced on top of this material to generate highly sensitive biosensor electrodes. Meng et al. synthesized PEDOT nanofiber-based 3D networks using tetrabutylammonium perchlorate (TBAP) soft templates (Figure 10a) [132]. The nanofibers’ network structure possessed a more active surface area to facilitate the binding of lactate dehydrogenase. The group noted improved sensitivity of the electrode to the target analyte (i.e., lactate). Park et al. modified VDP of 3,4-ethylenedioxythiophene (EDOT) on the
PMMA surface (Figure 10b) by controlling the temperature and pressure conditions of the reaction, thereby generating nanorods on the fibers’ surface [133]. The resulting 3D PEDOT nanofiber-based membranes were applied for dopamine detection through the immobilization of the human dopamine receptor (hDRD1). The sensing performance of PEDOT-based biosensors are suggested in Table 3.

Figure 10. (a) The 3D PEDOT nanofiber network from the tetrabutylammonium perchlorate (TBAP) soft template for lactate detection [132]. Copyright 2020, Elsevier. (b) Multidimensional PEDOT nanofiber membranes via a modified vapor deposition polymerization method for dopamine detection [133]. Copyright 2016, American Chemical Society.
Table 3. Comparative sensing performance and limit of detection of PEDOT nanostructure-based biosensors.

| Nanostructure                        | Receptor       | Analyte     | Mechanism    | LOD 1 | Response Time | Working Temperature | Linearity        | Cycle Stability | Reference  |
|--------------------------------------|----------------|-------------|--------------|-------|---------------|---------------------|------------------|----------------|-----------|
| Pt/PEDOT nanosphere                  |                | H$_2$O$_2$  | Electrochemical | 2.84 µM | <10 s         | 25 °C               | 2.5 µM–mM       | 3 weeks         | [117]     |
| Pt/PEDOT microsphere                 | GOx            | Glucose     | Electrochemical | 1.15 µM | <10 s         | 25 °C               | 0.1–10 mM       | 12 days         | [118]     |
| PEDOT/MnO$_2$ nanoellipsoid          |                | Catechol amine | Electrical  | 0.25 mM | <20 s         | 25 °C               | 0.25–25 mM      | -              | [119]     |
| BPEI-FC 7/PEDOT/PSS nanobead        | GOx            | Glucose     | Electrochemical | 2.4 mM  | <20 s         | 25 °C               | 0.5–5 mM        | -              | [120]     |
| PEDOT:PSS/CS micelle                 | HRP            | H$_2$O$_2$  | Electrochemical | 30 pM    | -             | 25 °C               | 0.1 nM–10 µM    | 35 days         | [121]     |
| PEDOT NT                             | hDRD1 3        | Dopamine    | FET 4        | 10 fM   | <1 s          | 25 °C               | 10 pM–10 nM     | -              | [122]     |
| PEDOT:PSS-silk fibroin core-sheath wires |                | Ascorbic acid | Electrical  | 1.14 µM | <20 s         | 25 °C               | 1.14–800 µM     | 4 weeks         | [123]     |
| PEDOT/PSS                            | PLL 5-g-OEG 6-Biotin | Streptavidin    | Electrochemical | 1 fg mL$^{-1}$ | -           | 25°C               | 1–1000 fg mL$^{-1}$ | -               | [124]     |
| PEDOT NF                             | GOx            | Glucose     | Electrochemical | 2.9 µM  | <3 s          | 25 °C               | 2.9 µM–25 µM    | 25 cycles       | [125]     |
| PEDOT nanowisker                     | Au-Hb 7        | H$_2$O$_2$  | Electrochemical | 0.6 µM  | <20 s         | 25 °C               | 1 µM–1.1 mM     | 60 days         | [126]     |
| PEDOT/PSS-based LBL film             | GOx            | Glucose     | Electrochemical | 41 µM   | <20 s         | 25 °C               | 0.1–1.4 mM      | -              | [127]     |
| PEDOT/Au                             | Antibody      | VEGF 8      | Electrochemical | 0.5 pg mL$^{-1}$ | -           | 25°C               | 1–20 pg mL$^{-1}$ | -               | [128]     |
| Au-PEDOT-Graphene Aerogel            | Antibody      | PSA 9       | Electrochemical | 0.03 pg mL$^{-1}$ | -           | 25°C               | 0.1 pg mL$^{-1}$–50 ng mL$^{-1}$ | -               | [131]     |
| 3D PEDOT NF network                  | LDH 10        | Lactate     | Electrochemical | 0.05 mM | <10 s         | 25 °C               | 0.05–1.8 mM     | -              | [132]     |
| 3D PEDOT NF membrane                 | hDRD1         | Dopamine    | FET 4        | 100 fM  | <2 s          | 25 °C               | 0.1–100 pM      | -              | [133]     |

1 Limit of detection, 2 polyethylenimine binding with ferrocene, 3 human dopamine receptor nanovesicle, 4 field effect transistor, 5 biotin-derivatized poly(L-lysine), 6 grafted oligoethylene glycol, 7 hemoglobin, 8 vascular endothelial growth factor, 9 prostate specific antigen, 10 lactate dehydrogenase.
5. Others

As noted throughout this review, researchers have taken advantage of the unique electrical and chemical properties offered by the alternative structures of conductive polymer chains to synthesize more effective biosensor electrodes. Soylemez et al. synthesized poly (2,5-di(furan-2-yl) thiazolo [5,4-d][thiazole]) (PTTzFr)-based films using electrodeposition before introducing glucose oxidase for electrochemical glucose detection (Figure 11a) [134]. The excellent electrochromic properties of this polymer chain facilitate easy detection, as noted by the obvious color changes of the film material. One clear example of this can be seen in the study conducted by Liu et al. in which colorimetric thrombin aptasensors were generated using poly [3-(3’-N,N,N-triethylamino-1’-propyloxy)-4-methyl-2,5-thiophene hydrochloride] (PMNT) films via intrinsic peroxidase-like activities that catalyzed the reaction of the peroxidase substrate 3,3’,5,5’-tetramethylbenzidine (TMB) in the presence of hydrogen peroxide ($\text{H}_2\text{O}_2$). As a result, a notable blue color was observed (Figure 11b) [135]. Voccia et al. synthesized “layer-by-layer” films consisting of polythiophene/streptavidin/biotinylated captured probes to detect small RNA (e.g., miR-221 microRNA) biomarkers of lung and breast cancer cell lines [136]. The sensing performance of other conducting polymer-based biosensors are suggested in Table 4.

![Figure 11. (a) The PTTzFr film-based electrochemical sensors for glucose detection [134]. Copyright 2019, Elsevier. (b) The poly [3-(3’-N,N,N-triethylamino-1’-propyloxy)-4-methyl-2,5-thiophene hydrochloride] PMNT-based colorimetric thrombin aptasensors [135]. Copyright 2017, Elsevier.](image-url)
Table 4. Comparative sensing performance and limit of detection of other conducting polymer nanostructure-based biosensors.

| Nanostructure     | Receptor | Analyte   | Mechanism     | LOD 1 | Response Time | Working Temperature | Linearity     | Cycle Stability | Reference |
|-------------------|----------|-----------|---------------|-------|---------------|---------------------|--------------|-----------------|-----------|
| PTTzFr 2 film     | GOx 3    | Glucose   | Electrochromic| 12.8 µM | <10 s         | 25 °C               | 12.8–500 µM  | -               | [134]     |
| PMNT 4 film       | Aptamer  | Thrombin  | Colorimetric  | 4 pM  | -             | 25 °C               | 0.01–0.1 nM | -               | [135]     |
| PT-based LBL 5 film| Biotin   | miR-221   | Electrochemical| 0.7 pM | -             | 25 °C               | 0.7–100 pM  | -               | [136]     |

1 Limit of detection, 2 poly(2,5-di(furan-2-yl) thiazolo[5,4-d]thiazole), 3 glucose oxidase, 4 poly[3-(3′-N,N,N-triethylamino-1′-propyloxy)-4-methyl-2,5-thiophene hydrochloride], 5 polythiophene-based layer-by-layer.
6. Conclusions and Outlook

Various conducting polymers are of interest for biosensor applications because of their excellent electrical, chemical, and physical properties. Researchers have utilized these properties to actively optimize the performance of these biosensors by controlling the structure of the conducting polymer in nanoscale such as 0D, 1D, 2D, and 3D. In the case of the 0D structure, most researches were conducted to generate uniformly sized conductive polymer nanoparticles using template-based methods (microemulsion and hard template) and then applied to the biosensor electrodes by attaching inorganic components or bio-receptors to the surface. In the case of 1D structure, the tube or fiber structure-based textiles were manufactured using methods such as electrospinning, electrodeposition, and hard template and applied them as a flexible biosensor electrode by attaching various receptors. Using simple electrodeposition, spin coating, and screen printing methods, the 2D structure manufactured a flexible, large-area film and used it as a conductive polymer film-based sensor electrode. Finally, in order to expand the active surface area of the nanomaterials to the analyte, various manufacturing methods were modified to generate the 3D-conducting polymer nanostructure, and applied as a highly reactive biosensor electrode material. Insights gained from this field will promote the development of flexible, wearable sensors that could have far-reaching consequences in the healthcare industry.

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