Recent Advances in Materials
for Enzyme-free Electrochemical Glucose Sensors

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Glucose is not only the main energy source of living cells but also an important metabolic intermediate product of organisms. Its detection and analysis are of great significance in the diagnosis and treatment of human diseases, especially diabetes. We discuss the progress in research on electrochemical enzyme-free glucose sensors in recent years and the electrochemical mechanism and glucose-sensing properties of various materials, including metals, metal oxides, and carbon nanomaterials and their nanocomposites. Finally, the prospects for the development of enzyme-free glucose sensors are outlined.

1. Introduction

Glucose plays a vital role in biology, not only as a major source of energy for living cells, but also as a metabolic intermediate in living organisms. The rapid and sensitive detection of glucose level has very important applications in clinical medical examinations, food safety analysis, biological fermentation control, and other fields, among which glucose detection in the blood is the most important. Abnormal levels of glucose in the blood can lead to many serious diseases, such as diabetes. According to the International Diabetes Federation, there were 415 million people with diabetes all over the world in 2015. It is predicted that by 2045, there will be 629 million people with diabetes worldwide. If not effectively controlled, diabetes can lead to stroke, coronary heart disease, blindness, and other complications. Therefore, as the only standard for the clinical diagnosis of diabetes, the detection of blood glucose is particularly important.

Over the past few decades, several glucose detection methods have been established, including acoustic, magnetic, optical, thermal, and electrochemical methods. In particular, electrochemical methods have been widely studied owing to their advantages of real-time detection, fast response, high reliability, low cost, easy operation, and miniaturization.[1] A

glucose sensor is mainly composed of a modified electrode and a transducer. The modified electrode selectively catalyzes the oxidation reaction of glucose on the electrode surface, and the transducer converts the chemical signal from the reaction into an electrical signal, which can be displayed by the instrument. Electrochemical glucose sensors can be roughly divided into two categories according to whether the modified electrode contains glucose oxidase (GOx), namely, enzymatic glucose sensors and enzyme-free glucose sensors. Sixty years after the first enzymatic glucose sensor reported by Clark and Lyons in 1962,[2] third-generation enzymatic glucose sensors are now being produced. The enzymatic glucose sensor is the most common glucose sensor at present, with good sensitivity, selectivity, and rapid response. This biosensor has been successfully commercialized since the 1980s and is now widely used in clinical diagnosis and personal health management. However, owing to the easy deactivation of enzymes and the effects of external conditions, including temperature, humidity, pH, and chemical environment, the stability of enzymatic glucose sensors is poor, as is their life, which is only about 30–40 days.[3,4] Moreover, the difficulty in fixing GOx and its high cost also limit its application.[5] Therefore, enzyme-free glucose sensors, which do not use any biological enzymes, have caught people's interests owing to their high stability, good reproducibility, ability to be manufactured in mass, and easy miniaturization.[6–10] Enzyme-free glucose sensors use sensitive nanomaterials directly loaded on the electrode surface to catalyze the reactions of glucose and can overcome the traditional dependence on enzyme activity. Owing to the rapid development of nanomaterials as well as other fields, enzyme-free glucose sensors have been developed fast.[11–15] We briefly summarize the development of these devices and elaborate on the opportunities and challenges presented by their practical application.

2. Latest Developments in Enzyme-Free Glucose Sensors

Many types of materials are involved in the design of enzyme-free glucose sensors, such as metals (Pt, Au, Ag, Ni, Cu), metal compounds (NiO, CuO, Co_3O_4), composites (Ni/Au, CuO/NiO), carbon materials, and others. These materials individually or in combination provide various advantages in the construction of enzyme-free glucose sensors, thereby improving their performance. The following sections introduce the progress in enzyme-free glucose sensors based on these materials.

2.1 Enzyme-free glucose sensors based on metals

Except for alkali metals, most metals are good conductors of heat and electricity. The properties of various metals differ greatly in terms of the metal–metal bond strength . At present, noble metals, such as Au and Pt, and transition metals, such as Co and Ni, are widely used in the design of enzyme-free glucose sensors.

2.1.1 Enzyme-free glucose sensors based on noble metals

Noble metals, the most commonly used sensitive materials by far, have stable physical and chemical properties and are good catalysts for glucose oxidation. The introduction of noble metals greatly improves the performance of glucose sensors. Therefore, noble metals such as Pt, Au, and Ag have been extensively studied.

2.1.1.1 Enzyme-free glucose sensors based on Pt

The noble metal Pt has excellent catalytic and electrochemical properties, resulting in its wide use in the catalytic, chemical, and electronics industries,[16] and it has excellent catalytic capacity for glucose in neutral and alkaline solutions. Malhotra *et al.* fabricated a simple non-enzymatic glucose sensor by immobilizing Pt particles on polyvinylferrocene-coated Pt electrodes (Pt/PVF/Pt). The Pt/PVF/Pt sensor showed a short response time of less than 3 s; its response was linear over a glucose concentration range of 0.1–11.0 mM. The sensitivity of the sensor was 327 μA mM^{-1} cm^{-2} with a low detection limit of 0.026 mM.[17] Although great progress has been made with enzyme-free glucose sensors based on Pt, they have several drawbacks that limit their applications. For example, in the presence of chloride ions in an electrolyte, the oxidation capacity of Pt for glucose decreases. Moreover, it is not as selective as GOx and is susceptible to interference by ascorbic acid (AA) and other agents. To overcome these shortcomings, different structures of noble metals, such as nanoparticles (NPs), nanowires, nanosheets, and nanoflowers, have been reported to increase the specific surface area and roughness. Multidimensional structures can not only provide effective mass transfer for the reactant molecules by increasing the active area but also improve the electron transfer rate. For instance, Chen *et al.* prepared Pt microsphere composites by electrodeposition, and the sensitivity of an enzyme-free glucose sensor based on these composites was 288.86 $\mu A mM^{-1} cm^{-2}$ in the linear range of 0–5 mM.[18] In addition, Yang *et al.* prepared multimodal nanoporous PtCu using a two-step method. An enzyme-free glucose sensor was prepared using a nanoporous PtCu-modified glassy carbon electrode (GCE), with a low detection limit of 0.1 μM and a linear range of 0.01–1.7 mM.[19] Wang *et al.* prepared Au@Pt core-shell nanoparticles integrated with carbon materials to construct a sensing interface for the enzyme-free electrochemical detection of glucose. This three-dimensional sensor interface was applied to detect glucose without enzymes at a markedly negative potential (at about −0.013 V vs Ag/AgCl), demonstrating a low detection limit of 42 nM and a wide linear range of 50 nm–2.5 mM.[20] Wang *et al.* developed an enzyme-free glucose sensor based on Ag&Pt hollow nanoparticles supported on TiO_2 nanotubes.[21] A wide range of Pt or Pt composite nanomaterials have been used in research on enzyme-free glucose sensors, which exhibit advantages such as excellent sensitivity to and good compatibility with biomolecules.

2.1.1.2 Enzyme-free glucose sensors based on Au

Owing to its outstanding electrochemical properties, excellent biocompatibility, and nontoxicity, and good stability, Au has been widely used in enzyme-free glucose sensors, resulting in an amplification of signals and an improvement in sensitivity.[15,22–27] However, the chemisorption ability of a Au electrode is lower than that of a Pt electrode. Under neutral and alkaline conditions, Au electrodes can be affected by chloride ions, and amino acids can bind to their surface, thus affecting the progress of the electrooxidation reaction. Owing to the poor chemical absorption of glucose on the surface of Au electrodes, the use of Au-based materials has been explored. The results show that the electrocatalysis of glucose oxidation is related to the specific surface area of Au electrodes. The larger the specific surface area, the greater the catalytic oxidation activity. Shen *et al.* prepared Au foam by electrodeposition with a dynamic gas template on a Au NP/Si substrate.[23] The enzyme-free glucose sensor prepared from the Au foam showed ultrasensitivity, high selectivity, and long-term stability in the quantitative detection of glucose and achieved a low detection limit of 0.14 μM over a wide linear range of 0.5 μM–12 mM. Zhong *et al.* developed a sensitive enzyme-free glucose sensing platform employing 3D hierarchical porous Au networks (HPANs) as electrocatalysts for glucose oxidization.[28] The HPAN-based electrochemical glucose sensor could be driven at a low potential (+0.1 V) and showed an outstanding performance for glucose determination within two linear ranges of 1–500 μM and 4.0–12 mM, a limit of detection of 0.2 μM, and a short response time of less than 2 s. In our previous work, we also demonstrated an ultrasensitive enzyme-free glucose sensor based on Ni/Au bilayer nanowire arrays, achieving an ultrahigh sensitivity of 5154.84 $\mu A\ mM^{-1}\ cm^{-2}$ in the range of 50 μM–10 mM and a sensitivity of 1750.16 $\mu A\ mM^{-1}$ cm^{-2} in the range of 10–65 mM.[25]

The preparation method for Au nanomaterials is simple, and their morphology and size are easily controlled. Owing to their excellent electrochemical properties, the combination of Au nanomaterials and other materials, such as metals, metal oxides, or metal hydroxides, their catalytic performance and stability can be greatly improved. Moreover, when Au nanomaterials combine with enzymes, they can not only improve the stability of enzymes but also further improve their own performance, resulting in improved biosensors.

2.1.1.3 Enzyme-free glucose sensors based on Ag

Ag nanomaterials have high extinction coefficient, electric field intensity, and physical adsorption capacity, show the highest electrical and thermal conductivities among all metals, and have relatively stable chemical properties. The technique of using Ag NPs in enzyme-free glucose sensors is being rapidly developed, owing to their great potential for sensing.[12,29–33] Arif *et al.* prepared an enzyme-free glucose sensor composed of a metal organic framework (MOF) composite decorated with Ag@TiO_2 nanoparticles.[12] The sensor based on Ag@TiO_2@ MOF showed a high sensitivity of 0.788 $\mu A\ \mu M^{-1}\ cm^{-2}$ with a linear concentration range of 48 μM–1 mM, a response time of 5 s, and an excellent detection limit of 0.99 μM. Gao *et al.* developed a highly sensitive enzyme-free glucose sensor via surface-enhanced Raman

spectroscopy (SERS) detection and electrochemical analysis based on Ag nanodendrites/Cu mesh that exhibited high SERS sensitivity and electrocatalytic activity.[31] Jadoon *et al.* investigated the sensing ability of a Ag-coronene complex for the enzyme-free electrochemical detection of glucose and H_2O_2.[33] Ag can be used not only in electrochemical enzyme-free glucose sensors but also in fluorescence glucose sensors. Han *et al.* developed a reliable and sensitive sensor for H_2O_2 and glucose detection based on the fluorescence enhancement effect when guanine-rich DNA sequences are in proximity to DNA-silver nanoclusters.[34] Their experimental results showed a linear relationship between the intensity and concentration of both H_2O_2 and glucose levels.

In summary, electrochemical biosensors prepared with noble metals show excellent comprehensive sensing performance with improved linear range, detection limit, selectivity, sensitivity, and stability. In addition to Pt, Au, and Ag, other noble metals such as Pd can be used as sensitive materials for enzyme-free glucose sensors.

2.1.2 Enzyme-free glucose sensors based on transition metals

Although noble metals can effectively accelerate electron transfer and improve the sensitivity of glucose sensors, they have a high cost, a narrow detection range, and poor glucose absorption which results in a low selectivity. In the detection process, the interface of noble metals is prone to poisoning, which adversely affects detection. Transition metals, by contrast, are cheap and can catalyze the oxidation of glucose directly, over a wide detection range. Therefore, transition metals are often used as highly efficient catalysts in the design of electrochemical biosensors. According to the literature, the transition metals most commonly used in enzyme-free glucose sensors are Ni, Co, and Cu.

2.1.2.1 Enzyme-free glucose sensors based on Ni

Nickel (Ni) is a common transition metal and has been widely used in glucose sensors,[10,13,35–38] because it has excellent electrochemical performance and high stability for glucose oxidation in an alkaline medium; in addition, it is not affected by the presence of chloride ions. Hu *et al.* designed a novel nanocomposite enzyme-free glucose sensor by fabricating hierarchically nanostructured metal nickel on titania nanowire arrays. Owing to the large surface area of the hierarchically nanostructured Ni and fast electron transfer of the TiO_2 nanowire array electrode, the electrodes exhibited a high sensitivity of 1472 µA mM^{-1} cm^{-2} with a linear range of 0.2–2 mM, a short response time within 5 s, and a low detection limit of 10 µM, in detecting glucose concentration.[36] However, Ni-modified electrodes also have many defects. For example, not only glucose but also other small molecules can be oxidized by Ni nanomaterials, resulting in poor selectivity for glucose. In addition, the narrow detection range of glucose and the low electron transfer efficiency limit the application of Ni. Glucose undergoes electrooxidation on the surface of Ni-based materials, with electron transfer between Ni^{3+}/Ni^{2+} redox pairs, independent of the original form of the Ni-based materials. Different from Au and Pt electrodes, the surface of Ni electrodes is immediately oxidized to a hydroxide at a

certain potential. Wang *et al.* used a chemical deposition technique to prepare a new 3D integrated electrode of amorphous Ni-B NPs grown on Ni foam (Ni-B/NF), thereby establishing an enzyme-free glucose sensor with excellent catalytic activity, a good sensitivity of 8.25 mAmM^{-1}cm^{-2}, a low detection limit of 5 μM, and a response time shorter than 10 s in the detection range of 4 μM–14.5 mM.[38]

2.1.2.2 Enzyme-free glucose sensors based on Cu

Copper (Cu) is also a popular catalytic material for glucose detection because of its low cost, nontoxicity, and high electrochemical activity. Identical to Ni-based materials, Cu-based materials, such as Cu,[39–45] CuO,[46–50] Cu$_2$O,[51–54] Cu(OH)$_2$,[55] CuS,[56,57] and others, can be used for the electrocatalysis of glucose. Cu-based materials participate in the oxidation of glucose oxidation in various forms, such as nanocubes, nanorods, nanowires, nanosheets, and nanoflowers. Copper nanowires are usually used in electrochemical glucose sensors because of their high aspect ratio. Na *et al.* prepared an enzyme-free glucose sensor using Cu nanowires synthesized using a hydrothermal method. The device based on Cu nanowires achieved a low detection limit of 1 nM.[39] To prevent further oxidation of Cu and improve the performance of glucose sensors, Ye *et al.* proposed the use of a carbon shell structure wrapped around Cu nanocubes as a sensor.[40] The enzyme-free glucose sensor achieved an ultrahigh sensitivity of 2565 μAmM^{-1}cm^{-2} and a detection limit of 21.35 μM in the linear range of 0.04–40 mM.

2.1.2.3 Enzyme-free glucose sensors based on Co

Co-based materials are commonly used in the design of enzyme-free glucose sensors. Unlike Ni and Cu, Co has multiple valence states; therefore, the reaction mechanism is slightly more complicated. Various Co-based materials, including Co alloys,[35,58,59] Co$_3$O$_4$,[60–67] and Co(OH)$_2$,[68,69] have been used in nanostructured enzyme-free glucose sensors. Chu *et al.* fabricated a novel nonenzymatic glucose sensor based on a prism-like bimetallic alkaline carboxylate (CoNi-MIM). The electrochemical glucose oxidation by the sensor had a markedly high sensitivity of 5024.4 μA mM^{-1} cm^{-2}, a low detection limit of 56.1 nM, and a linear response of up to a concentration of 14.3 mM.[35] Wang *et al.* synthesized a glucose electrocatalyst by loading Co$_3$O$_4$ nanoclusters onto 3D kenaf stem-derived carbon and established an enzyme-free glucose sensor with a low detection limit of 26 μM and a linear range of 0.088–7 mM.[67] Wang *et al.* fabricated an enzyme-free glucose sensor based on Co(OH)$_F$ nanoflower/carbon cloth prepared using a microplasma-based synthesis method; the sensor had a high sensitivity of 1806 μAmM^{-1}cm^{-2} and a low detection limit of 0.75 μM.[69] Xie *et al.* reported the development of a cobalt nitride nanowire array on Ti mesh as an efficient catalyst electrode for glucose oxidation in alkaline solutions and H$_2$O$_2$ reduction in neutral solutions; they constructed enzyme-free glucose sensors with a high sensitivity of 3325.6 μAmM^{-1}cm^{-2} in the linear range of 0.1 μM–2.5 mM and a low detection limit of 50 nM.[70]

2.1.3 Enzyme-free glucose sensors based on other metals

Except for the common noble metals (Pt, Au, Ag) and transition metals (Cu, Ni, Co), most other metals have not been studied for use in enzyme-free glucose sensors because of their relatively poor electrocatalytic activity. However, several metals, such as Pd,[54,71–73] Fe,[74,75] and Mn, show a relatively high electrocatalytic activity and have also been studied. Wang *et al.* studied the sequential and transient electrocatalysis of D-glucose oxidation reactions using core-shell Pd@Pt particles.[73] Raza and Ahmad used Fe-doped ZnO to detect glucose on screen-printed electrodes, and the detection limit obtained was as low as 0.3 μM.[75] Gao *et al.* used Mn-doped NiO to construct an electrochemical sensor with a high sensitivity of 3212.52 $\mu AmM^{-1}cm^{-2}$ and a low detection limit of 0.8 mM.[76]

Moreover, how to overcome the defects and combine the advantages of noble metals and transition metals to prepare enzyme-free glucose sensors with high sensitivity, high selectivity, and low cost is an area of intense research interest. Enzyme-free glucose sensors based on alloy electrodes have not only integrated the advantages of each metal in the alloy but have also greatly improved the selectivity of glucose detection. Noh *et al.* prepared Au-M(M: Zn, Ni, and Co) alloy nanomaterials and demonstrated that the Au-Zn nanocomposite had the best catalytic performance for glucose oxidation and oxygen reduction.[77] Hsieh *et al.* fabricated an enzyme-free Ni-Au alloy nanowire electrochemical glucose sensor.[78] Li *et al.* chose to synthesize a Ni-Ag hybrid NP layer on SWCNTs as an electrocatalyst for glucose oxidation and then obtained a glucose sensor with high sensitivity and low detection rate.[37]

The working potential, sensitivity, selectivity, electrolyte solution, linear range, and detection limit of each metal-based enzyme-free glucose sensor are summarized in Table 1.

Table 1
Comparison of enzyme-free glucose sensors based on different metals.

Material	Working Potential (V)	Electrolyte solution	Linear range (mM)	LOD (μM)	Sensitivity ($\mu AmM^{-1}cm^{-2}$)	Selectivity	Ref.
Pt-MWCNTs/CSF	0.65	0.01 M PBS	0–5	0.05	288.86	AA, UA, AP	(18)
Nanoporous PtCu	0.4	PBS	0.01–2	0.1	3.16	AA, AA, DA	(19)
Au foam	0.1	0.3 M NaOH	5×10^{-4}–12	0.14	—	AA, UA, AP, DA	(23)
Au@MIPs	—	0.1 M PBS	10^{-7}–10^{-5}	3×10^{-6}	—	AA, UA, DA	(26)
3D HPANs	0.23	0.2 M NaOH	4–12	0.2	—	AA, UA, DA, AP	(28)
Ag@TiO$_2$@MOF	0.4	0.1 M NaOH	0.048–1	0.99	0.788	AA, UA, DA	(12)
Ni@ TiO$_2$ NWs	—	—	0.2–2	10	1472	—	(36)
3D Ni-B/NF	0.55	0.2 M NaOH	5×10^{-3}–2.66	5	8250	AA, UA, DA, Suc, Mal, Lac	(38)
Cu NWs	0.65	0.1 M NaOH	—	1×10^{-3}	—	NaCl, AA, UA, DA, Suc,	(39)
Cu@C nanocubes	0.6	0.4 M NaOH	0.04–40	21.35	2565	AA, UA, DA, NaCl, Gly, Try	(40)
CoNi-MIM	0.55	0.2 M NaOH	Up to 14.3	5.61×10^{-2}	5024.4	Urea, NaCl, KCl, AA, UA, Fru, Lcy, AP	(35)

NWs: nanowires, AA: ascorbic acid, UA: uric acid, DA: dopamine, AP: acetaminophen, Suc: sucrose, Mal: maltose, Lac: lactose, Fru: fructose, Gly: glycine, Try: tryptophan, and Lcy: L-cysteine.

2.2 Enzyme-free glucose sensors based on metal oxides

Noble metals and their alloys were first used as active materials for enzyme-free glucose sensor electrodes. The performance of glucose sensors based on noble metals as active materials is close to or even higher than that of third-generation enzyme glucose sensors in widespread use. However, there are several shortcomings that limit the application of noble metals in enzyme-free glucose sensors. First, the high price of noble metals increases the cost of glucose sensors. Second, when the electrocatalytic oxidation reaction between the electrode and glucose occurs, the intermediate products of the reaction accumulate in large quantities on the surface of the noble metal electrode, resulting in electrode poisoning, which impedes a continuous reaction and greatly reduces the linear range of the electrode.[79] Moreover, noble metals are also susceptible to the toxicity of Cl^-, which limits their stability.[21] In contrast, transition metal oxides, such as CuO, Co_3O_4, and NiO, have a significant cost advantage over noble metals due to their natural abundance. Transition metal oxides mainly catalyze glucose oxidation by valence transitions, which are usually reversible reactions. Therefore, the loss of active materials is almost negligible in the course of their use, avoiding electrode poisoning and greatly improving the stability of the electrode and the reproducibility of its response. In recent years, metal oxides have experienced renewed research interest because of their advantages, such as low cost, high stability, sensitivity, and fast response.

2.2.1 Enzyme-free glucose sensors based on NiO

Nickel oxide (NiO) has low toxicity, adequate natural abundance, low cost, excellent electrochemical activity, and stability; thus, it is expected to be used as a sensitive material for glucose sensors. Nickel oxide has also been extensively studied for its glucose oxidation properties in alkaline solutions. As early as the 1970s, carbohydrate oxidation on nickel oxide in an alkaline environment was studied by cyclic voltammetry.[80] Later, Shamsipur *et al.* reported an electrooxidation method used to deposit Ni oxide on MWCNTs and detect glucose in an alkaline environment.[81] Although relatively low performance was achieved (the detection limit was 0.16 mM and the linear range is 0.2 to 12 mM), this study was ground-breaking on the use of NiO to detect glucose. Subsequently, a series of NiO nanomaterials have been studied and prepared for use in enzyme-free glucose sensors. Ibupoto *et al.* prepared hollow cage NiO nanostructures (NiOHCs) using a hydrothermal method.[82] The sensitivity of the NiOHCs to glucose was 288.87 $\mu AmM^{-1}cm^{-2}$, their detection limit was 0.1 μM, and a wide linear range of 0.1–5.0 mM and excellent repeatability were observed. Zhou *et al.* synthesized NiO NPs on carbon cloth (NiO/CC) via a flame method and used it as an enzyme-free sensor for the detection of glucose.[83] This enzyme-free glucose sensor exhibited a response time as short as 3 s, a wide detection range of 5 μM–2 mM, a LOD of 7.45 nM, and a sensitive response of 4025 μA mM^{-1} cm^{-2}. Mugheri *et al.* presented a novel route towards the synthesis of NiO nanostructures via hydrothermal methodology assisted by a variety of amino acids.[84] The developed sensors based on NiO nanostructures demonstrated promising results in terms of sensitivity, selectivity, and stability, with a maximum sensitivity of 4930 μA mM^{-1} cm^{-2} and a

LOD of 0.01 μM. Yang *et al.* grew nanoflower-shaped Mn-doped NiO nanocomposites with high catalytic performance and excellent conductivity on 3D flexible carbon fiber cloth via hydrothermal and calcination methods to construct an efficient, flexible glucose-sensitive detection electrode, avoiding interference due to the oxidization of species present in real samples at higher redox potentials.[85] Other widely explored materials can also be compounded with NiO for enzyme-free glucose detection, for example, carbon materials. Subash *et al.* reported the ultrasonication–assisted synthesis of nickel oxide NPs anchored on graphene oxide nanosheets (NiO/GO).[86] The electrochemical results demonstrated that the NiO/GO-modified GCE had a higher electrocatalytic activity towards the oxidation of glucose than graphene oxide- and NiO-modified electrodes, achieving a lower detection limit of 0.17 μM in the range of 0.62 μM–2.4 mM.

Compared with Ni metal, NiO is low-cost and environmentally friendly, and displays fast redox kinetics, making it suitable for glucose detection. Since the mechanism of an enzyme-free glucose sensor depends on the redox pair of Ni(III)/Ni(II) on the electrode surface, the exposure of electrocatalytic active sites is highly beneficial in enhancing the sensing performance. However, NiO has poor conductivity, as do other metal oxides, and it is expected that Ni-based materials with high porosity or large specific surface area may be applied to enzyme-free glucose sensors to improve the charge transfer capability.

2.2.2 Enzyme-free glucose sensors based on copper oxide (CuO) and cuprous oxide (Cu$_2$O)

Some pioneering studies have examined oxidation products on Cu electrodes covered with oxides in potassium hydroxide aqueous solutions by the dynamic scanning of potentials.[87] Cu(II) can be oxidized to Cu(III) in an alkaline environment and can be used to detect a variety of compounds, such as monosaccharides and disaccharides. Xie and Huber used the sensing properties of hydroxyl radicals to activate the active sites of metal oxides (CuO•OH) and studied the catalytic activity and response sensitivity of Cu$_2$O-modified GCEs in 0.1 M NaOH electrolyte.[88] They found that the actual sensing material of the electrochemical reaction was CuO produced by Cu$_2$O in an alkaline environment. In these early studies, the sensing mechanism of CuO or Cu$_2$O for monosaccharides and disaccharides was thoroughly explored and verified. The necessary steps to detect glucose involve the formation of hydroxyl and Cu$_x$O•OH intermediates, which act as catalysts for carbohydrate oxidation in an alkaline aqueous environment. Since then, many reports have appeared on enzyme-free glucose sensors based on CuO and Cu$_2$O, primarily focusing on improving the sensitivity, linear range, detection limit, and selectivity.[48–50,89–91] Identical to other metal oxides, CuO and Cu$_2$O have poor conductivity; therefore, carbon materials, such as carbon nanofibers, SWCNTs, MWCNTs, and graphene (GE), have been used to improve sensitivity by enhancing electron transfer. These studies further validated the sensing mechanism of hydroxide ions by activating CuO and then triggering glucose oxidation through intermediates. Owing to their flexible structure, large surface area, and large pore volume, MOFs have also been used recently in enzyme-free glucose sensors. An ultrasensitive enzyme-free glucose sensor was realized using an *in situ* synthesized

CuO NP-modified Ce MOF.[49] As a support material, MOFs have a large specific surface area, which can enhance the glucose oxidation reaction. The glucose sensor based on a CuO NP-modified Ce MOF had a sensitivity of 2058.5 $\mu AmM^{-1}cm^{-2}$ in a linear range up to 8.6 mM. The sensing mechanism of the glucose sensor based on CuO NPs/Ce-MOF/GCE was proposed to involve the oxidation of Cu(II) to Cu(III), after which Cu(III) was further used as the catalyst for glucose oxidation.

2.2.3 Enzyme-free glucose sensors based on Co_3O_4

In addition to the metal oxides described, Co oxides have also attracted considerable attention in the development of enzyme-free glucose sensors since the work of Ding *et al*.[66] Cobalt oxides, for example, cobalt tetroxide, have attracted much attention because of their excellent electrocatalytic activity. Cobalt oxides usually contain a certain redox couple and have excellent electrochemical performance. They have shown great potential in many applications, such as lithium-ion batteries, supercapacitors, and sensitive materials for sensors. Cobalt oxides with different shapes and 3D morphologies have been developed. Yang *et al*. demonstrated the preparation of hierarchical porous Co_3O_4@graphene microspheres by a one-step hydrothermal method to achieve a high electrocatalytic performance for enzyme-free biosensor applications.[60] Gao *et al*. used a hydrothermal method to grow needle-shaped Co_3O_4 nanotube arrays (ANTAs) and modified them with TiO_2 to prepare TiO_2/Co_3O_4 ANTA electrodes.[62] Compared with that of the Co_3O_4 ANTA electrode, the catalytic oxidation current of the TiO_2/Co_3O_4 ANTA electrode increased by 44% with a sensitivity of 2008.82 $\mu AmM^{-1}cm^{-2}$ and a linear range up to 3 mM. The electrode also had excellent selectivity, reproducibility, and stability. Han *et al*. prepared Co-Co_3O_4/carbon nanotube/carbon foam nanocomposites by soaking melamine foam and then used the nanocomposites to construct an enzyme-free sensor to detect glucose in alkaline solutions.[92] The sensor showed a detection range of 1.2 μM–2.29 mM with a detection limit of 0.4 μM and a high sensitivity of 637.5 $\mu A^{-1}\ cm^{-2}$. In our previous work, the synthesis and preparation methods for 3D layered Co_3O_4 nanobooks and cube-shaped Co_3O_4 NPs were studied.[65,93] The enzyme-free glucose sensors based on these materials showed high sensitivity and selectivity in the detection of glucose in human serum and saliva samples, thus confirming the potential for the application of Co_3O_4-based nanomaterials in the enzyme-free and noninvasive detection of glucose.

Similar to NiO and CuO, Co_3O_4 is also a poor conductor. The solutions to this problem include (1) the preparation of a special Co_3O_4 structure to increase the electrocatalytic activity potential and (2) the formation of a composite with metals, carbon materials, alloys, or other highly conductive materials to increase the conductivity, thus greatly improving the performance of glucose sensors.

2.2.4 Enzyme-free glucose sensors based on ZnO

ZnO is a functional semiconductor with high chemical stability and mechanical plasticity, which can be used as an efficient electron conduction catalyst and has been widely used in

enzymatic biosensors.[94] In recent years, ZnO enzyme-free glucose sensors have also attracted wide attention.[79,95–97] Hsu *et al.* synthesized ZnO nanowires on a glass substrate using a hydrothermal process, and then decorated the nanowires with Pt nanoparticles to fabricate a working electrode for an enzyme-free glucose biosensor.[79] Mahmoud *et al.* prepared copper-doped ZnO NPs by a sol–gel method.[95] The prepared Cu-ZnO sensor displayed high sensitivity and superior electrocatalytic activity in an ultralow linear detection range of 1 nM–10 μM with a detection limit of 1 nM. Although ZnO-based nanomaterials are promising electrocatalysts for glucose sensing, they still require relatively high working potentials, which may cause interference by, for example, AA and uric acid (UA) in real serum samples, leading to unexpected output signals. This behavior indicates that the selectivity of ZnO-based materials is a potential problem for high-performance glucose sensors.

2.2.5　Enzyme-free glucose sensor based on other metal oxides

In addition to the metal oxides already discussed, several metal oxides have been explored as electrocatalysts for enzyme-free glucose sensors, such as Fe_2O_3, Mn_3O_4, and MnO_2. Cao and Wang prepared Fe_2O_3 nanowire arrays and constructed an enzyme-free glucose sensor with a sensitivity of 726.9 $\mu AmM^{-1}cm^{-2}$ and a short response time of less than 6 s in the linear range of 0.015–8 mM.[74] Wang *et al.* prepared a 3D Ni/MnO_2 composite material using an electrochemical method.[98] The enzyme-free glucose sensor based on the 3D Ni/MnO_2 composites had a sensitivity of 1.04 $mAmM^{-1}cm^{-2}$ in the linear range of 0.25–3500 μM. Si *et al.* grew Mn_3O_4 on 3D graphene foam and constructed glucose and H_2O_2 sensors based on nanocomposites.[99] The synergistic effect produced by the electrochemical catalytic ability of Mn_3O_4 and the large specific surface area of 3D graphene foam resulted in the sensor having both a high sensitivity and a wide detection range. The sensitivities of the sensor to glucose and H_2O_2 were 360 $\mu AmM^{-1}cm^{-2}$ and 1.03 $mAmM^{-1}cm^{-2}$, respectively, and the linear detection ranges were 0.1–8 mM and 2 μM–6.5 mM, respectively.

The working potential, sensitivity, selectivity, electrolyte solution, linear range, and detection limit of each metal oxide-based enzyme-free glucose sensor are summarized in Table 2.

2.3　Carbon-based enzyme-free glucose sensors

With the development of technology, various new materials have injected vitality into the study of enzyme-free glucose sensors. Carbon nanomaterials as the carrier are particularly outstanding, because they are electrochemically inert but have significant electron conductivity, which can effectively promote rapid electron transfer from the analyte to the electrode surface. They are relatively mature in terms of the development of biosensors.

Carbon nanomaterials usually refer to carbon nanotubes (CNTs), graphene, carbon nanofibers, graphene oxide (GO), and reduced graphene oxide (rGO), among which CNTs and graphene are most commonly used. Since CNTs were discovered by Iijima in 1991, they have received attention continuously because of their unique physical and chemical properties.[100] Ye *et al.* were pioneers in applying CNTs in enzyme-free glucose sensors and found that CNTs

Table 2
Comparison of enzyme-free glucose sensors based on different metal oxides.

Material	Working potential (V)	Electrolyte solution	Linear range (mM)	LOD (μM)	Sensitivity ($\mu AmM^{-1}cm^{-2}$)	Selectivity	Ref.
NiO/MWCNTs	0.6	0.1 M NaOH	0.2–12	160	—	—	(81)
NiOHCs	0.48	0.1 M NaOH	0.1–5	0.1	2476.4	AA, DA, UA	(82)
NiO/CC	0.57	0.1 M NaOH	0.005–2	7.45×10^{-3}	4025	AA, DA, UA, urea, Fru, NaCl	(83)
Mn-doped NiO nanoflowers	0.6	0.1 M NaOH	3–5166	0.28	—	AA, DA, ASA, Amp, NaCl, Gly	(85)
NiO/GO	0.5	0.1 M NaOH	6.2×10^{-4}–2.4	1.7×10^{-2}	0.33	AA, UA, DA, Suc, Fru, KNO3−	(86)
CuO/MWCNTs	0.4	0.1 M NaOH	up to 1.2	0.2	2596	UA, DA, AA, Suc, Fru, Lac	(47)
CuO nanowire arrays	0.58	0.1 M NaOH	0.1–6	1	1950	AA, UA, AP	(48)
CuO NPs/Ce-MOF	0.55	0.1M NaOH	5×10^{-6}–8.6	2×10^{-3}	2058.5	KCl, UA, DA, AA	(49)
Ag-doped CuO microflowers	0.45	0.1 M KOH	0.01–6	0.01	1527	UA, DA, AA, NaCl	(89)
Cu$_2$O/Cu/carbon cloth	0.6	0.1 M KOH	1×10^{-3}–1.555	6×10^{-2}	6952	UA, DA, AA, Fru	(90)
CuO nanorod/MOF	0.6	0.1 M NaOH	up to 1.25	1	1523.5	AA, DA, UA, NaCl, KCl	(91)
Co$_3$O$_4$@Graphene	0.55	0.1 M NaOH	0.02–8	3.8×10^{-2}	628	AA, DA, UA	(60)
TiO$_2$-modified Co$_3$O$_4$ ANTAs	0.5	1 M NaOH	up to 3	0.3396	2008.82	AA, UA	(62)
Co$_3$O$_4$ nanobook	0.554	0.1 M NaOH	up to 6	7.94	1068.85	DA, UA, AA, NaCl	(65)
Co$_3$O$_4$ NPs	0.535	0.1 M NaOH	up to 3	9.3×10^{-3}	2495.79	AA, UA, AA, NaCl	(93)
Cu-doped ZnO NPs	—	0.1 M NaOH	1×10^{-6}–0.01	1×10^{-3}	—	—	(95)
Fe203 nanowire array	0.5	0.1 M Phophate	0.015–8	6	726.9	AA, DA	(74)
3D Ni/MnO$_2$	0.45	0.02 M NaOH	2.5×10^{-4}–3.5	0.1	1040	AA, DA, UA	(98)
Mn$_3$O$_4$ /3DGF	0.4	0.1 M NaOH	0.1–8	10	360	AA, AP, UA	(99)

Ram: raminophenol, L-tyr: L-tyrosine, ASA: cetylsalicylic acid, Amp: acetaminophen, and Gly: glycine.

improved the sensitivity of the electrodes to glucose detection.[101] Baghayeri *et al.* prepared Ag NPs on metformin–functionalized MWCNTs using an electrochemical method. The enzyme-free glucose sensor based on the composites achieved a low detection level of 0.3 nM and a short response time of less than 4 s in the linear range of 1.0 nM–350 μM.[29] Wang *et al.* also prepared

$Cu/MnO_2/MWCNT$ nanocomposites using an electrochemical method. The resultant enzyme-free glucose sensor achieved a sensitivity of 494 $\mu AmM^{-1}cm^{-2}$, a low detection limit of 0.17 μM, and a short response time of 3 s in the linear range of 0.64 μM–2 mM.[44]

Graphene is a new two-dimensional carbon nanomaterial first obtained by mechanical stripping in 2004 and has been widely used in various fields since then. Graphene and its derivatives, including graphene, GO, and rGO, are promising electrode materials for enzyme-free glucose sensors because of their unusual electrical and thermal conductivities, large specific surface area, lightproof properties, and excellent mechanical strength. Tran *et al.* prepared layered graphene nanocomposites encapsulated with Au NPs by chemical vapor deposition and used them to construct an enzyme-free glucose sensor.[102] The sensor had excellent electrocatalytic activity, a low detection limit of 1 μM, and a sensitivity of 0.195 $\mu AmM^{-1}cm^{-2}$ in the linear range of 6 μM–28.5 mM. In addition, the device was not affected by AP, AA, and UA, and showed long-term stability. Xu *et al.* synthesized Ag-doped CuO microflowers on multilayer graphene (Ag-CuO@MLG) and studied their application to enzyme-free glucose detection.[89] The Ag-CuO@MLG electrode exhibited a sensitivity of 1527 $\mu AmM^{-1}cm^{-2}$ in a linear response range of 0.01–6.0 mM with excellent selectivity and long-term stability. Jiang *et al.* designed a novel non-enzymatic sensor based on a 3D nanostructured $Co(OH)_2$/rGO film to detect glucose at a low alkalinity.[68]

2.4   Enzyme-free glucose sensors based on nanocomposites

The materials used in recent years include not only simple metals or metal oxides but also composite materials, which integrate the beneficial properties of different materials. Glucose sensors based on nanocomposites have attracted wide attention owing to the synergistic effects of different materials, such as the high electrical conductivity of metals, the high selectivity of organic materials, and the high electrocatalytic activity of metal oxides.

2.4.1   Enzyme-free glucose sensors based on metal/metal oxide nanocomposites

Many nanocomposites combine the high conductivity of metals with the high electrocatalytic activity of metal oxides, thereby enabling the preparation of enzyme-free glucose sensors based on metal/metal oxide nanocomposites. Fang *et al.* reported a sensitive enzyme-free glucose sensor based on Cu/Cu_2O composite nanoparticles decorated on a single carbon fiber by direct *in situ* electrochemical deposition.[51] The as-prepared electrode displayed superior catalytic performance and exhibited an ultrahigh sensitivity of 28071 $\mu AmM^{-1}cm^{-2}$ due to the combination of the advantages of Cu/Cu_2O nanoparticles and the excellent conductivity of carbon fiber. Hsu *et al.* synthesized Au/ZnO core-shell nanostructures decorated with Au nanoparticles to fabricate an optical enzyme-free glucose sensor.[103] Jiang *et al.* fabricated a flexible sensor based on Au nanoparticle-modified copper hydroxide nanograss arrays on flexible carbon fiber cloth (Au@$Cu(OH)_2$/CFC).[55] The excellent sensing properties were attributed to the collective effect of the superior electrochemical catalytic activity of $Cu(OH)_2$ nanograss arrays with a markedly increased electrochemically active surface area as well as the

mass transfer ability of Au. Yang *et al.* designed composite catalysts with Cu, Ni, and their oxides for the detection of glucose. The enhanced performance of a sensor based on the Cu-Ni/ CuO-NiO catalyst was considered to result from the synergistic effects between the two metals and their metal oxides.[104]

2.4.2 Enzyme-free glucose sensors based on metal/carbon nanocomposites

The combination of metals and carbon materials has become one of the trends in the design of electrochemical glucose sensors. Carbon cloth, CNTs, graphene, GO, and rGO have been widely used in enzyme-glucose sensors to improve their performance. Li *et al.* synthesized PdPt/rGO nanocomposites by an ionic liquid–assisted one-pot method to construct a new enzyme-free glucose sensor.[105] Zhao *et al.* loaded Pt-Ni NPs onto MWCNTs by electrodeposition.[106] Li *et al.* developed bimetallic MCo NP-doped carbon nanofibers by electrospinning for enzyme-free glucose detection.[59] Li *et al.* synthetized SWCNT-sandwiched Ni-Ag hybrid NP layers as an electrocatalyst for glucose oxidation.[37] Nguyen *et al.* developed an enzyme-free glucose sensor using the one-step electrodeposition of Au and Ru NPs on the surface of CNT-based Pt NPs nanocomposites.[3] All these studies demonstrated the potential of metal/carbon nanocomposites to improve the performance of enzyme-free glucose sensors. It should be mentioned that the size of carbon nanomaterials also affects the performance of the sensor, in addition to the type and amount of metal used. Ye *et al.* synthesized Pt NPs on an electrospun carbon fiber and investigated the effect of the diameter of the carbon fiber on the amount of Pt load, revealing that the larger the curvature and the smaller the diameter of the carbon fiber, the more Pt atoms were loaded on the surface. Thus, an enzyme-free glucose sensor was constructed with the optimal Pt NP load, with a linear range of 0.3–17 mM and a detection limit of 33 μM.[107]

2.4.3 Enzyme-free glucose sensors based on metal oxide/carbon nanocomposites

Transition metal oxides, such as NiO, CuO/Cu_2O, and cobalt oxide, have the advantages of high natural abundance, excellent electrocatalytic activity, and the existence of electrochemical redox pairs, and have attracted much attention. However, a single-component transition metal oxide has a basic problem of low conductivity, and it is difficult for one to meet the needs of enzyme-free glucose sensors. To solve this problem, composites of transition metal oxides and other materials have become a target of research interest. Carbon nanomaterials, such as CNTs, graphene, GO, rGO, and ordered mesoporous carbon, have good electrical conductivity, stable chemical properties, wide electrochemical windows, high specific surface areas, and good biocompatibility, making them ideal carriers for sensitive materials. On one hand, the addition of carbon nanomaterials can increase the dispersion of sensitive materials and the specific surface area of the materials, thereby improving their electrocatalytic efficiency. On the other hand, carbon nanomaterials can accelerate electrochemical reactions through a synergistic effect with sensitive materials. Lin *et al.* prepared Co_3O_4 and MWCNT composites by a single-step solvothermal method and constructed an enzyme-free glucose sensor based on them.[64] Zhang

et al. prepared a NiO-N-doped carbon/rGO nanocomposite using a hydrothermal method and constructed an enzyme-free glucose sensor that achieved an ultrahigh sensitivity of 4254 µAmM^{-1}cm^{-2} and an ultralow detection limit of 70.9 nM.[108] Pourbeyram *et al.* prepared a CuO/rGO nanocomposite and fabricated an enzyme-free glucose sensor, resulting in an ultrahigh sensitivity of 4760 µAmM^{-1}cm^{-2} and an ultralow detection limit of 91 nM.[109] These results confirm that glucose sensors based on metal oxide/carbon nanocomposites can achieve excellent sensitivity, outstanding selectivity, and long-term stability.

2.4.4 Enzyme-free glucose sensor based on other nanocomposites

In addition to the nanomaterials described, several other nanocomposites are being used to explore the construction of enzyme-free glucose sensors. Among them, MOFs have drawn much attention and have been widely used to construct glucose sensors in recent years, as the easily tunable metal ions and organic linkers enable them to produce multifunctional nanocomposites.[13] MOFs, such as Ni-MOF,[30,110,111] Cu-MOF,[41] Co-MOF,[112–115] and Ni-Co MOF,[58,116,117] and their derived composites, have all demonstrated outstanding performance for enzyme-free glucose detection.

The working potential, sensitivity, selectivity, electrolyte solution, linear range, and detection limit of nanocomposite-based enzyme-free glucose sensors are summarized in Table 3.

Table 3
Comparison of enzyme-free glucose sensors based on different nanocomposites.

Material	Working potential (V)	Electrolyte solution	Linear range (mM)	LOD (µM)	Sensitivity (µAmM^{-1}cm^{-2})	Selectivity	Ref.
Cu/Cu$_2$O NPs	0.6	0.1 M NaOH	1×10^{-3}–7.8	7.8	28071	DA, AA, UA, D-Fru, Gal, LA	(51)
Au@Cu(OH)$_2$/CFC	0.6	0.1 M KOH	0.1–3.3	2.7×10^{-2}	7350	Mal, Fru, UA, AA, DA, Cys, 4-AP, KCl and Na$_2$SO$_4$	(55)
Cu-Ni/CuO-NiO	0.7	0.1 M NaOH	0–0.35 0.35–9.85	5.4×10^{-2}	2637.282 923.084	KCl, AA, DA, D-Fru, LA, D-Mal, UA	(104)
Bimetallic MCo NP-doped carbon nanofibers	0.6	0.1 M NaOH	0.02–11	1	507	DA, AA, UA, AP, GA, LA	(59)
Ni-Ag/SWCNTs	—	0.1 M NaOH	0–2.5	0.084	2946	DA, AA, UA, AP	(37)
PtPd NPs/rGO	0	0.1 M PBS	0.1–22	2	1.47	DA, AA, UA	(105)
PtxNi1-x/MWCNTs	−0.3	0.1 M PBS	Up to 15	0.3	940	DA, AA, UA, AP, urea, Gal, Lac, Fru	(106)

Table 3
(Continued) Comparison of enzyme-free glucose sensors based on different nanocomposites.

Material	Working potential (V)	Electrolyte solution	Linear range (mM)	LOD (μM)	Sensitivity (μAmM^{-1}cm^{-2})	Selectivity	Ref.
Pt NPs/carbon fiber	−0.05	0.1 M NaOH	0.3–17	33	2.03	AA, UA	(107)
Au-Ru NPs/Pt NPs-CNT	−0.1	0.01 M PBS	1–10	68	23.47	AA, UA, AP, Fru, Suc, Lac	(3)
Co3O4-MWCNT	0.5	0.1 M NaOH	1×10^{-3}–0.122	0.28	2550	DA, AA, K+, Na+, Mg^{2+}, Ca^{2+}	(64)
NiO-N-doped carbon@rGO	0.5	0.1 M NaOH	5×10^{-4}–0.02	7.09×10^{-2}	4254	DA, AA, UA, KCl	(108)
CuO NPs/rGO	0.45	0.1 M NaOH	1×10^{-4}–0.15	0.091	4760	DA, AA, UA	(109)
Ni-MOF	0.728	0.1 M NaOH	1×10^{-3}–1.6	0.76	2859.95	—	(111)
Au@Ni-BTC MOF	0.55	0.1 M NaOH	5×10^{-3}–7.4	1.5	1447.1	urea, AA, Glu, proline, L-val, lysine, L-leu, NaCl	(13)
Cu-MOF/Cu foam	0.65	0.1 M NaOH	0.001–0.95	0.076	3.003×1010	AA, UA, DA, Fru, Lac, urea, NaCl	(41)
Cu@Co–MOF	0.6	0.01 M NaOH	0.005–0.4 0.4–1.8	1.6	282.89 113.15	D-Man, D-Fru, AA, DA, urea, UA	(114)
N-Co-MOF@PDA-Ag	0.55	0.1 M NaOH	1×10^{-3}–2	0.5	183	AA, UA, Fru	(115)
Ni-Co MOF	0.55	0.1 M NaOH	0.02–0.79	4.25	205.1	Lac, urea, DA, UA, AA, NaCl	(116)
Ni–Co MOF/Ag/rGO/PU	0.5	0.1 M NaOH	0.01–0.66	3.28	425.9	DA, LA, NaCl, UA, Cys.	(117)

LA: lactic acid, GA: glutaric acid, Gal: galactose, Cys: cysteine, 4-AP: 4-acetamidophenol, Glu: glusate, L:val: L-valine, L-leu: L-leu, D-Man: D-Mannitol.

3.   Conclusions

We reviewed the progress in research on enzyme-free glucose sensors over recent years and discussed their mechanisms of action. The performance of most enzyme-free glucose sensors is primarily related to the sensing materials themselves. Current research focuses on developing and improving the preparation of the sensing materials. In early research, metals (such as Au, Ni, Pt, Ni, Co, and Cu) and their metal oxides have been used to fabricate enzyme-free glucose sensors. Later, composites, such as metal/metal oxide composites and carbon nanocomposites, have emerged as electrode materials with excellent electrocatalytic ability. At the same time,

research on nanomaterials has also promoted the rapid development of electrochemical enzyme-free glucose sensors. Many new nanomaterials with specialized structures and excellent performance have been developed, providing broader concepts for the preparation of novel enzyme-free glucose sensors.

While encouraging progress has been made in recent years, there are many deficiencies still to be addressed. First, enzyme-free glucose sensors are still in the research stage, and the mechanism of glucose oxidation on various electrodes is still controversial, knowing that the mechanism is crucial to improving the performance of the sensor. Second, unlike the case with enzymatic glucose sensors, almost all enzyme-free sensitive materials cannot recognize glucose molecules as specifically as GOx, leading to poor specificity. Moreover, the contents of real-world samples such as blood and sweat are very complex and many substances interfere with glucose detection. Other organics, proteins, ions from inorganic salt and other substances may strongly adsorb to the electrode surface and impact the sensitivity, selectivity, and stability of the glucose sensors. Third, the test conditions for enzyme-free glucose sensors are mainly phosphate buffer or alkaline solutions, which are considerably different from the actual conditions where the sensors are used.

In summary, enzyme-free glucose sensors still face huge challenges. The future development direction of these sensors may be carried out in these areas: (1) studies of specific recognition mechanisms of glucose and how to improve the specificity and selectivity of enzyme-free glucose sensors; (2) work on noninvasive and low-concentration glucose detection in biological fluids such as sweat,[116,117] saliva, and tears; and (3) the integration of efforts in the technology and miniaturization of sensors with other devices or systems. All these issues support the continuous effort to improve enzyme-free glucose sensors and to revolutionize the glucose biosensor industry.

Acknowledgments

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