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Review Article

A review on nanosensors to detect diabetes

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ABSTRACT: Diabetes mellitus, a serious disease affecting millions of people worldwide, is a disease characterized by increased levels of glucose concentration in the blood. Monitoring blood glucose has been declared a crucial and important tool that makes diabetes management probable. A large number of suitable glucose biosensors have been developed so far. This research has particularly focused on covering achieving biocompatible and improved sensing platforms which are evolving with the contribution of novel materials. The motivation for writing this review is to discuss and review the recent advances in enzymatic and non-enzymatic glucose sensors evolved in the last few years.

KEYWORDS: diabetes detection; glucose sensors; nanomaterials; enzymatic glucose sensors; non-enzymatic glucose sensors

1. Introduction

Diabetes is one of the most common and challenging diseases in the 21st century globally. It is now considered a major death cause which seems to be an epidemic in many developing and newly industrialized nations. Complications from diabetes, such as coronary artery and peripheral vascular disease, stroke, diabetic neuropathy, amputations, renal failure, and blindness are resulting in increasing disability, reduced life expectancy, and enormous health costs for virtually every society[1]. Because of those, diabetes management which simply means maintaining a regular blood glucose level has developed rapidly. And monitoring the blood glucose level has turned into one of the most crucial tools in diabetes management. The normal range of blood glucose in a healthy body is found in the range of 4.9–6.9 mm. However, it can be increased by up to 40 mm in diabetic patients after glucose intake[2]. Although various types of glucose sensors have become commercially available, glucose biosensors have also made a huge improvement. Biosensors are devices that can analytically convert a biological response into an electrical signal.

Researchers are being inspired to fabricate affordable, accurate, and user-friendly glucose monitoring instruments by advancements in point-of-care (POC) sensor technologies, microfluidics, nanotechnology, and miniaturization. Accurate on-site and timely detection based on several kinds of glucose sensor platforms has been made possible by the coupling of many glucose sensing techniques with POC biosensors. For patients with abnormalities of glucose metabolism, the incorporation of smartphone-integrated electronic readers into such devices or their enhancement using 3D printing technology shows significant potential. POC biosensors enable people to monitor their blood glucose levels precisely and conveniently without the assistance of expert staff or hospital visit.

The industry has a persistent interest in cre-

ating novel glucose sensing devices since there is an enormous need for quick, affordable, and accurate ways to test blood glucose levels. Glucose sensors can be based on different types of transducers such as thermal, optical, electrochemical, acoustic and magnetic. Biosensors can be classified into different groups based on their transducer. Among all of them, the widely investigated one is electrochemical platforms which are divided into two main categories: enzymatic and non-enzymatic (as depicted in **Figure 1**). The categorizing factor is the presence of an enzyme as the sensing material in the biosensor.

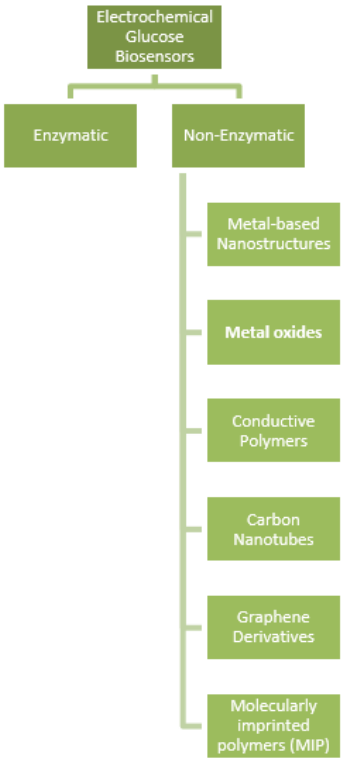

Figure 1. Different glucose biosensors based on their sensing material.

The enzymatic diabetes biosensors have an enzyme that plays a crucial role. A group of these biosensors is glucose oxidase (GOx) depended which uses GOx as the glucose-sensing enzyme. A thin layer of GOx is utilized over an oxygen anode in these electrochemical biosensors, where the oxygen is consumed by the enzyme-catalyzed response. The mechanism of GOx action leads to gluconic acid production (**Figure 2**). To execute this oxidation reaction by GOx, a redox co-factor is needed with the input of flavin adenine dinucleotide (FAD+) which is an electron acceptor. It can be reduced to $FADH_2$ by redox reactions. Subsequent reaction with oxygen that produces H_2O_2 regenerates the FAD+ at the anode, which can sense the number of transferring electrons that are correlated with the amount of H_2O_2 production and hence, the amount of glucose present[3].

Recently, nanomaterials are introduced into enzymatic glucose sensors to enhance electron transfer rates. A wide range of nanomaterials can be used for this purpose, including the nanoparticles of noble and transition metals, the nanostructured metal-oxides or metal sulfides, conductive polymers, carbon nanotubes, and graphene. **Table 1** summarizes the enzymatic biosensors[4].

2.2 Non-enzymatic nanobiosensors

Study on non-enzymatic biosensors as an alternative has started mainly due to the limited ranges of pH, temperature, and humidity conditions of the enzymatic glucose sensors. Metal-based glucose sensors are the sensors using

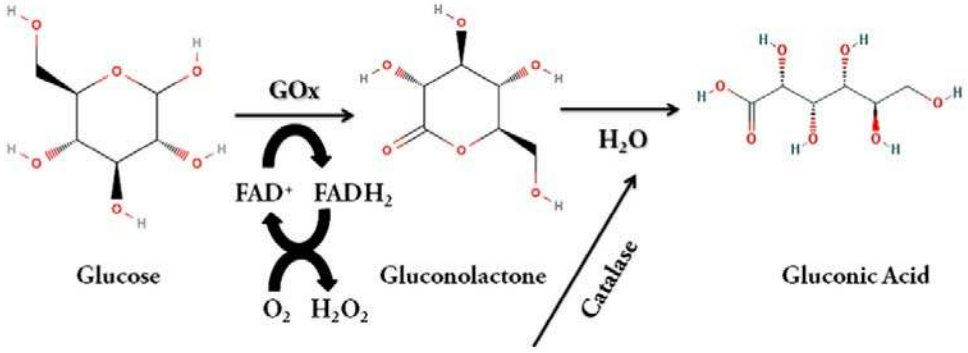

Figure 2. The transition of glucose to gluconic acid using glucose oxidase, reprinted with permission.

Table 1. A summary of the enzymatic biosensors for detecting diabetes

Sensing material	Linear range	Detection limit	References
Ternary-graphene-PANI-TiO_2	4–24 mm	–	[5]
3D-NiO-hollow sphere/RGO	0.009–1.129 mm	0.082 nm	[6]
Graphene/MnO_2	0.04–2 mm	10 μm	[7]
CHI-Pd@Pt core-shell nanocubes	1–6 mm	0.2 μm	[8]
RGO-Fe_3O_4	0.05–1 mm	0.1 μm	[9]
PANI-poly (ethylene oxide) nanocomposite	4–6 mm	820 μm	[10]
MnO_2/graphene nanoribbons	0.1–1.4 mm	50 μm	[11]
ZnO-nanorods/graphene	0.2–1.6 mm	–	[12]
GO_x/RGO	0.01–1 mm	5.8 μm	[13]
GO_x/3D graphene film	< 6 mm	200 μm	[14]
MoS_2/graphene aerogel	2–20 mm	290 μm	[15]
Pt@ CNOs	2–28 mm	–	[16]
MWCNTs/CSF	0–5 mm	210 μm	[17]
GO_x/Au-ZnO/GCE	1–20 mm	20 μm	[18]
GO_x/Cu-MOFs	9.1×10^{-3}–36 mm	2.73 μm	[19]
GOx/CHI/GS/PB	8.17×10^{-3}–1 mm	2.45 μm	[20]
Au/NS on carbon fiber	8.17–1 mm	2.45 μm	[21]
CHI-GOx/APTES	0.01–50 mm	10 μm	[22]

PANI: polyaniline, RGO: reduced graphene oxide, CHI: chitosan, GO_x: glucose oxidase, MOF: metal-organic frameworks, CNOs: carbon nano-onions, CNT: carbon nanotubes, GCE: glassy carbon electrode, PB: prussian blue, GS: graphene sponge, NS: nanostructures, APTES: (3-Aminopropyl) triethoxysilane, dPIn: dPIn doped-polyindole, CSF: carbonized silk fabric.

noble (e.g., Au, Pt) and transition (e.g., Ni, Cu) metals for glucose detection as they can facilitate electrocatalytic oxidation of glucose. The metals can be combined to form multi-metallic electrodes for better electrocatalytic performance because of their synergistic activity for glucose electro-oxidation. Nevertheless, the application of transition metal glucose sensors is not attractive enough due to the high cost of transition metals. These metals in oxide form can be a cost-effective alternative. To enhance their electronic conductivity, conductive support materials (e.g., Ni foam, Cu foam, 3D-KSCs) are combined with them. Another large group of non-enzymatic biosensors is a conductive polymer including glucose sensors. These polymers possess high electrical conductivity, as well as electron affinity and redox activity. Their electrocatalytic activity can be even improved by the introduction of metal or metal-oxide nanoparticles, CNTs, and graphene. Carbon nanotubes (CNTs) can be used for sensor applications. Their superior properties, for instance, high aspect ratio, large surface area, as well as remarkable optical and electronic properties, make them promising materials for glucose detection. Similarly, graphene has great properties to be used in electrochemical sensing applications. Besides, all the mentioned materials, molecularly imprinted polymers (MIP) are devised as artificial recognition elements that can recognize and bind target molecules specifically. Their functional monomer is polymerized within a template which is later removed. Glucose imprinted polymer can be utilized for sensor applications[23]. **Table 2** presents a comprehensive comparison between different materials used in electrochemical glucose sensors.

2.3 Noble and transition metals glucose nanobiosensors

Up to now, metal[13,30–32], metal-alloy[33–35], metal hydrate[36], metal sulfide[37,38], metal nitrides[39,40], and metal oxide[41–43] have been broadly applied as efficient nanostructures for their glucose oxidation ability in neutral and alkaline solutions. Amongst the various metals, Noble (e.g., Au, Pt, Pd) and transition metals (e.g., Ni, Cu, Co) are more commonly used for high-performance non-enzymatic glucose monitoring in the last decades. Nanostructured metallic materials with their exclusive physical, chemical, optical

Table 2. A comprehensive overview of different sensing materials used in electrochemical glucose sensors[24–29]

Sensing material	Pros	Cons
Enzyme	Good selectivity	Limited pH and temperature ranges
	Good sensitivity	Affected by humidity
		Deactivation by ionic detergents
Noble and transition metals	High stability	Poor sensitivity
	High electrocatalytic activity	Poor selectivity
		High cost
Metal oxides	High stability	Poor conductivity
	Low cost	Alkaline condition needed
Conductive polymers	Facile synthesis	Challenging confinement on the electrode
	Low cost	Low stability
	Adjustable properties	
Carbon nanotubes	High electrocatalytic activity	Challenging separation process
	High surface area	Degradation possibility of nanotubes
	High stability	
Graphene	Enhanced electrical conductivity	Heterogeneity of samples
	Biocompatibility	
MIPs	Low cost	Template removal stage
	Facile synthesis	
	Robustness	

and electrical properties such as high surface-to-volume proportion, huge specific surface area, high electrical conductivity, tunable optical property, and high electrocatalytic activity have been widely used in the fabrication of glucose biosensors[44–46].

These metals can be regarded as good electrocatalysts as a result of their capability to be in numerous oxidation states. By absorbing other compounds on their surfaces and acting as catalysts with a high number of surface atoms, intermediates are formed and the reaction process is facilitated by enhancing mass transport property[47]. Since the physicochemical properties of metal nanostructures such as size, shape, architecture, and composition can be varied and controlled, they can be regarded as appealing choices for electrocatalytic glucose sensing[48].To enhance the performance of detection, by investigating mass transport and electron transfer kinetics of metals, extensive attention has been paid to studying the electrocatalytic properties of metal nanostructures[49,50]. Based on the literature, most metallic nanomaterials used for non-enzymatic glucose sensing are noble metals (i.e., Au, Pd, and Pt) and their bimetallic nanostructures due to their high catalytic activity[51].

Despite the high sensitivity of noble metals towards glucose detection, surface fouling owing to the adsorption of intermediates has remained a challenging issue. Similarly, they do not have desired options for practical application due to their high costs because of inadequate supply. Considering cost-effectiveness, transition metals (such as Ni and Cu) and their bimetallic nanostructures (Cu/Ni) can be considered appropriate alternatives for non-enzymatic glucose sensors[40,52].

Moreover, the benefit of using transition metals instead of noble metals is the ability for oxidizing glucose at a constant potential and therefore simpler operation[53]. For the oxidation of glucose, Ni and Cu-based electrodes are commonly applied in the alkaline solution. Actually, by electron transferring between multivalent metal redox couples, the oxidation reaction of glucose is catalyzed by the transition metals. By immersing these metal electrodes (M) in an alkaline solution, $M(OH)_2$ compounds are formed at first, furthermore by the further oxidation reaction, MOOH is formed. Consequently, $M(OH)_2$/MOOH redox couple is the catalytic component in glucose detection[47]. Ni and Cu nanostructures have been used widely as glucose sensors in recent years.

As an alternative solution, to develop the

catalytic effect of metallic nanoparticles, bimetal alloy structures were utilized as electrode constituents. They are a novel class of nanomaterials that may have better technological performance compared to individual ones. They are synthesized between one element with filled d-orbitals and the other metal having empty d-orbitals. Their properties could differ along with the mixing pattern, in particular, their interfaces may play a pivotal role in controlling their behaviors[54].

Moreover, the applicability of these bimetallic nanostructures not only relies on their size and shape but also on the combination of the metals that participated (e.g., composition) and their fine structure. For instance, they can be formed by various hybrids of metals such as (noble/noble), (noble/transition), or both (transition/transition) metals[55,56].

Recently, considerable improvement has been reported on the fabrication of bimetallic nanostructures as electrochemical non-enzymatic glucose sensors owing to the synergistic effect of the two metals which can significantly enhance the electrocatalytic glucose oxidation reaction and minimize the interference and poisoning effect of the electrode[57]. **Table 3** summarized the non-enzymatic Noble and transition metals-based biosensors for detecting diabetes.

Table 3. A summary of the non-enzymatic Noble and transition metals-based biosensors for detecting diabetes

Sensing material	Linear range	Limit of detection	References
Ni/Cu nanocomposites	4×10^{-3}–5 mm	0.1 μm	[58]
Nafion/Cu (II) GCE	5×10^{-4}–2 mm	0.1 μm	[59]
Pd NPs porous GaN	1×10^{-3}–1 mm	–	[60]
La-Sr-Co-Ni-O nanofibers	0.1–1 mm	83 μm	[61]
bimetallic Co/Zn MOF	0.01–5 mm	6.5 μm	[62]
CuO/ZnO-DSDSHN	500–100 mm	3.575×10^{-1} μm	[63]
Ni-doped SnO_2	5×10^{-3}–0.825 mm	0.084 μm	[64]
Cu NPs HNT/PANI	0.01–5 mm	0.27 μm	[65]
$NiCo_2S_4$ nanoflakes	0.01–0.25 mm	0.01 μm	[66]
Cu/LAG	0.05–1.5 mm	0.01 μm	[67]
Ni-Cu ANPs/RGO	1×10^{-5}–3×10^{-2} μm	0.005 μm	[68]
Bimetallic Co-Zn-MOFs	0.001–0.255 mm, 0.255–2.53 m	4.7 μm	[69]
MoS_2-AuPt	0.005–3 mm	33 μm	[70]
Cu-Co-MOFs/Ni foam	–	23 μm	[70]
Cu-RGO	0.10–12.5 mm	65 μm	[71]
Ni-Co-S/CN/GCE	0.001–0.330 mm, 0.330–4.53 mm	467 μm	[72]

GCE: glassy carbon electrode, NPs: nanoparticles, MOF: metal-organic frameworks, DSDSHN: dumbbell-shaped double-shelled hollow nanoporous, HNT/PANI: halloysite nanotube/polyaniline, LAG: laser-ablated graphene, ANPs/RGO: bimetallic alloy nanoparticles reduced graphene oxide, MoS_2-AuPt: gold platinum bimetallic nanoparticles modified molybdenum disulfide nanosheet.

2.4 Metal oxides

Metal oxides are crystalline solids that contain a metal cation and an oxide anion. These materials show vast varieties of applications as gas sensors, catalysts, solar cells, optoelectronic devices, environmental protectors, and biosensors. Metal oxides have unique functional properties that essentially stem from their crystal structure, composition, indigenous defects, doping, etc. These properties provide them with chemical, optical, mechanical, and electrical characteristics[73]. In recent decades, different materials based on metal oxides such as metal-doped metal oxides, carbon nanotubes nanocomposites of metal oxides, and polymer composites of metal oxides have been explored broadly due to their cost-effectiveness and high sensitivity. These nanomaterials also show high selectivity when they are coupled with biorecognition molecules in analytical devices[74–77].

As mentioned, utilizing transition and noble metals, despite being desirable materials, has limitations for sensing glucose, because they are not

cost-effective. On the other hand, these materials in their oxide form have gained much attention and are considered a good alternative due to the ease of access and low cost. Especially, among these materials, oxides of metal elements of groups 7 to 12 in the Periodic Table have shown excellent performance in non-enzymatic glucose sensing[54]. To enhance the electronic conductivity and obtain higher surface area as well as more active sites, metal oxides are combined with other materials such as Ni foam, Cu foam, and three-dimensional Kenaf stem-derived carbon (3D-KSCs) that act as supports while possessing conductive properties[23].

Concerning the non-enzymatic glucose sensing, in the presence of hydroxide ions, different metal oxides can perform effectively at different pH ranges. The surface of metal oxides is activated by strong hydroxide ions and this process then results in the oxidation of glucose[78–80]. In addition, possessing a wide band gap makes metal oxides promising materials for detecting glucose[82]. Metal oxide-based non-enzymatic biosensors are listed in **Table 4**.

Table 4. A summary of the non-enzymatic metal oxides-based biosensors for detecting diabetes

Sensing material	Linear range	Limit of detection	References
Nanoporous CuO/ZnO	0.500–100 mm	358 nm	[63]
ZnO QDs on MWCNTs	10^{-5}–2.5×10^{-3} mm	208 nm	[82]
Laser-induced mesoporous NiO on Ni	0.005–1.1 mm	3.31 μm	[83]
NiO/Cu-TCPP	0.002–0.28 mm	0.95 μm	[84]
Co_3O_4 functionalized MoS_2-CNT	< 5.2 mm	0.08 μm	[85]
Ni-Co hydroxide nanosheets	0.002–0.8 mm	3.42 μm	[86]
PPy-CHI-Fe_2O_3	1–16 mm	234 μm	[87]
$NiMnO_3$	0.00005–1 mm	0.014 μm	[86]
CuO/NiO nanosheets	1.20–2.72 mm	0.0667 μm	[88]
MOF-CoO/CuO nanorod arrays	–	11.916 μa μm^{-1}	[89]
AuNPs-CuO NWs/Cu_2O	2.8×10^{-3}–2 mm	1.619 μm	[90]
TiO_2/Cu_2O/CuO CNFs	–	0.25 μm	[91]
CuO/NiO films	0.01–20 mm	1.86 μm	[92]

NF: nanofiber, QDs: quantum dots, MWCNTs: multi-walled carbon nanotube, TCPP: tetrakis (4 carboxyphenyl) porphrin, CNT: carbon nanotube, NWs: nanowires, CNFs: carbon nanofibers, CHI: chitosan, PPy: polypyrrole.

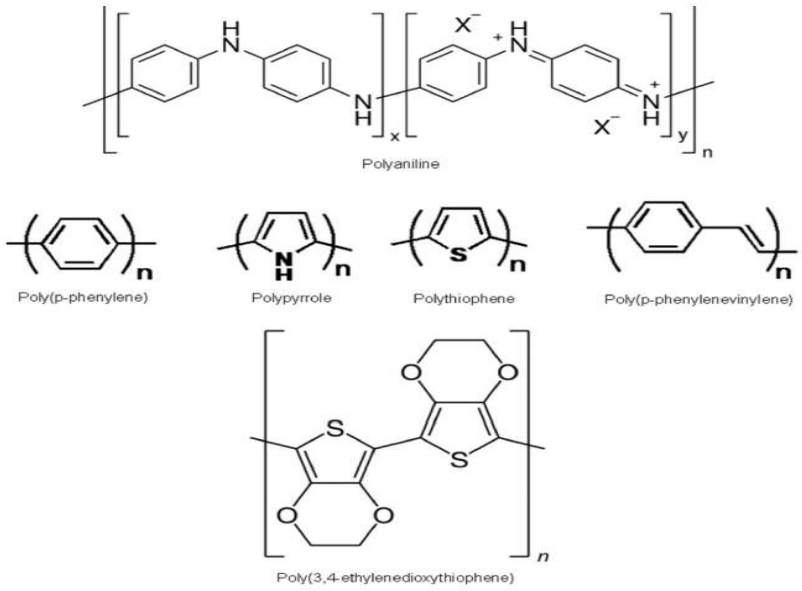

Figure 3. The chemical structures of typical conducting polymers.

2.5 Conductive polymers

By coupling the electronic properties of semiconductors with the properties of organic polymers, conducting polymers (CPs) emerged as

novel compounds attracting great importance nowadays. Since the discovery of CPs in 1977 by Shirakawa *et al*.[93], they have been utilized in rechargeable batteries[94], membrane separation[95], electrocatalysis[96], solar cells[97], supper capacitors[98], optoelectronic[55] extraction[99] and electrochemical biosensors. In **Figure 3**, some of the most commonly used CPs such as polyaniline (PANI), polypyrrole, and polythiophene have been represented[47].

These are used extensively due to their characteristics such as being cheap, ease of processing, and also facilitating the immobilization procedure[100]. In this type of polymer, alternative single and double bonds exist in the polymer chain, forming p-electron backbones which cause the uncommon properties of CPs. High electrical conductivity, mechanical flexibility, surface areas, electron affinity, electrocatalytic properties, and chemical stability in aqueous solutions, as well as low energy transitions are among the unique properties of these structures[101]. On the other hand, the conductivity of the polymer is affected by the length of conjugation, total chain length, and charge transfer to adjacent molecules[102].

Due to the porous structure of CPs, they are capable of acting as beneficial substrates for the immobilization of nanoparticles. By incorporating nanoparticles into CPs, the conducting polymer-based nanocomposite materials are formed. Additionally, due to the charge transfer which is facilitated between the dispersed nanoparticles, considerable enhancement in the conductivity of the hybrid systems is obtained. The incorporation of nanoparticles into the CPs causes higher performance for both CP and NPs, leading to different physical properties of the hybrid compound compared with the polymer and nanoparticle constituents, separately. **Table 5** summarized recent reports based on the application of conducting polymer-based nanocomposites in non-enzymatic biosensors.

Table 5. A summary of the non-enzymatic conductive polymer-based biosensors for detecting diabetes

Sensing material	Linear range	Limit of detection	References
Cu NPs/Graphene/PANI	0.5–15 mm	0.16 μm	[103]
Au NPs-PANI nanoarrays	1.026×10^{-3}–10 mm	3.08 μm	[104]
PPy/Co-decorated/2DMoS$_2$-AuNPs/GCE	10^{-7}–8×10^{-6} mm	8×10^{-5} μm	[105]
PPy/GQDs@PB	2×10^{-4}–5×10^{-2} mm	0.1 μm	[106]
NiO/CuO/PANI	2×10^{-4}–2.5×10^{-1} mm	2 μm	[107]
PANI/AuNPs	3×10^{-3}–2×10^{-1} mm	0.5 μm	[108]
PEDOT/IL	2×10^{-4}–3×10^{-2} mm	0.05 μm	[109]
NiNPs/PEDOT/RGO	10^{-3}–5.1 mm	0.8 μm	[110]
Ag-NPs-decorated PmAPNFs	0.1–8 mm	0.062 μm	[111]
CuO/PEDOT-MoS$_2$	3×10^{-2}–1.06 mm	0.046 μm	[112]
PPy@Cu(OH)$_2$NTs	1.78–6.53 mm	0.35 μm	[113]
Ni$_3$S$_2$ NW PEDOT-RGO HFs	1.5×10^{-2}–9.105 mm	0.48 μm	[114]
-NC-(CuS/NSC)	160–11.76 mm	2.72 μm	[115]
PAN/PANI/CuO	0.1–8 mm	0.062 μm	[111]

*PANI: polyaniline, PPy: polypyrrole, GQDs: graphene quantum dots, PB: prussian blue, PEDOT: 3,4-ethylenedioxythiophene, PmAPNFs: poly (m-aminophenol) nanofibers, NW: nano worm, RGOHFs: reduced graphene oxide hybrid films, NC: nanocomposite, ITO: indium-tin oxide, PAN: polyacrylonitrile, CHI: chitosan.

2.6 Carbon nanotubes

A carbon nanotube is an allotrope of carbon that resembles a tube of carbon atoms. Carbon nanotubes are extremely robust. Although it is hard to break them, they are still light. Large surface area, high aspect ratio, excellent thermal and chemical stability, and significant optical and electronic features, caused carbon nanotubes to be one of the most investigated materials[116].

Graphene is known as a single layer consisting of carbon atoms, forming a closely packed hexagonal lattice. Multiwalled carbon nanotubes (MWCNTs) consist of several layers of graphene

which are concentrated around the smallest nanotube, while for the synthesis of single walled carbon nanotubes (SWCNTs) only one layer of graphene is needed, which then results in a cylindrical shape with a nanometer-sized diameter[117].

Carbon nanotubes are found in diverse applications as cost-effective materials. They are utilized in energy conversion and storage[118], water filters[119], thin-film electronics[120], and coatings[121]. Due to possessing a large surface area, these materials have shown great performance in pharmacy and medicine to adsorb or conjugate a wide range of medicinal and diagnostic substances[122].

Since CNTs have unique optical, electrical, and structural properties, they are appealing materials for applications such as drug delivery and biosensing[123]. They have efficient capabilities to be used in treating varieties of diseases. These materials also exhibit effective performance when they are utilized for monitoring blood levels as well as other chemical properties of the human body[124,125].

Electronic conductivity and high surface area make CNTs good candidates for detecting glucose electrochemically. It is reported that MWCNTs can act as efficient support in the process of glucose detection[126]. Also, nanowires can be imported into these materials for amperometric glucose detection[127]. **Table 6** shows a summary of CNTs applications for non-enzymatic glucose sensing.

Table 6. summarized the non-enzymatic CNT-based biosensors for detecting diabetes

Sensing material	Linear range	Limit of detection	References
FTO-CNTs	0.07–0.7 mm	7×10^4 μm	[128]
PdSWCNTs-GCE	0.5–17 mm	200 ± 50 μm	[129]
GCE/CNT/MoS_2/Ni-NPs	0.05–0.65 mm	200 μm	[130]
CNT	10^{-3}–50×10^{-3} mm	1.708×10^{-6} μm	[131]
β-CD/SPCE/CNTs	10^{-6}–3×10^{-3} mm	5×10^{-4} μm	[132]
Pd-NPs-GNPs/MWCNTs/GCE	2.5×10^{-2}–10 mm, 10–100 mm	83.0 μm	[133]
Ag-NPs-MWCNT	10^{-6}–3.5×10^{-1} mm	3×10^{-4} μm	[126]
Ni(TPA)-MOF	2×10^{-2}–4.4 mm	4.6 μm	[134]
Cu-MWCNTs	< 7.5 mm	1.0 μm	[135]
CHI-(CuS/NSC)	1.6×10^{-1}–11.76 mm	2.72 μm	[115]
Au@Pt-NPs-MWCNTs	5×10^{-5}–0.1 mm	4.2×10^{-2} μm	[136]
Ni-Co-NPs-MWCNT	–	2.6×10^{-1} μm	[137]
QCM-CNT	0.5–120 mm	150 μm	[138]

CNT: carbon nanotubes, FTO: fluorine-doped tin oxide, SWCNTs: single-walled carbon nanotube, GCE: glassy carbon electrode, β-CD: β-cyclodextrin, SPCE: screen printed electrodes, MWCNTs: multi-walled carbon nanotube, (Ni (TPA)) nickel(II)-terephthalic acid MOF: metal-organic framework, QCM: quartz crystal microbalance, CHI: chitosan.

2.7 Graphene

Various nanoscale compounds such as metal nanowires, nanoparticles, carbon nanotubes, and graphene have been remarkably used as sensing materials in biosensors. Graphene particularly has attracted extensive attention after obtaining its single layer by mechanical exfoliation in 2004[139]. Furthermore, extra information about the unexpected properties of graphene was discovered as the Nobel prize in 2010[140]. Graphene is a single atomic layer of carbon atoms with sp^2 hybridization which is organized into a closely packed hexagonal lattice. The exclusive properties of graphene correspond to the pi orbitals of carbon atoms forming π bonds[141].

Graphene demonstrates special mechanical, electrical, thermal, and optical properties. Biocompatibility, high values of thermal conductivity, electron mobility, optical transmittance, and mechanical flexibility along with large specific surface area enable its application in electrochemical sensors[142]. Owing to the atomic thickness of graphene layers, carbon atoms presented in its structure could entirely interact with analytes. Graphene can be regarded as an ideal compound

in electrode fabrication because of its high surface area, wide potential window, great flexibility in addition to its robustness, and lower resistance against charge transfer compared to carbon nanotube structures[143].

Apart from carbon nanotubes, graphene also shows high purity as a result of the absence of transition metals like Fe, Ni, etc.[144]. Using graphene in its fabricated or functionalized form (graphene incorporated with metal or metal oxide nanoparticles or conducting polymer such as chitosan) in the modification of the electrode improves the electrochemical performance and detection time of the electrode. The first graphene-based glucose biosensor was constructed using graphene coupled with ionic liquid functionalized polyethyleneimine-modified electrodes. This biosensor displayed a wide linear range, high stability, and good reproducibility[145].

Additionally, as a result of the reduction of graphene oxide, reduced graphene oxide (RGO) is formed. Before the reduction step, the surface of graphene oxide is fabricated by oxygenated functional groups for instance carboxyl and epoxy groups. With the aid of Van der Waals interactions, these functional groups can affect the solubility of graphene oxide in solvents. The reaction rates are determined by the accessibility of unsaturated bonds presented on the surface of the graphene structure. Owing to the elimination of oxygenated functional groups, the surface of RGO have no free oxygen molecules, facilitating the participation of the surface in any surface activity. RGO can be considered a beneficial choice in electronic components, especially catalytic sensors. This is a result of the presence of oxygen-based functional groups and structural defects such as carbon vacancies.

Recently, extensive studies have been conducted to use nanocomposites in the fabrication of enzymatic or nonenzymatic electrodes for monitoring glucose. In this regard, an effective mixture of graphene with metal oxide nanopowder has been used as the substrate for glucose sensors. This is because of the exclusive properties of carbon atoms, such as conductivity toward heat, great surface area, biocompatibility, high possibility of functionalization, and inertness. Besides, graphene shows high mechanical properties in which sheets are considerably stronger than steel with a thickness of a million times smaller than human hair. In addition, another key property of graphene is its higher electrical conductivity causing a high rate of electron transfer and low resistance against charge transfer. Last but not least, herein, the incompatibility between graphene and metal oxide is not an issue[142].

In the case of metal oxide, excellent selectivity, large surface-to-volume ratio, and outstanding catalytic activities are the key advantages for applying them in non-enzymatic sensors. In this regard, the noble and transition metals and also their alloys are used as nanomaterials in combination with graphene. Nevertheless, the key issue with applying Nobel gases like platinum, gold, or palladium in sensor fabrication is their high prices, so they can be mixed with graphene to improve the efficiency of the sensor and decrease the operating costs. Furthermore, surface poisoning and surface fouling against chloride ions are two probable problems with noble metals, owing to the intermediates absorbed onto the electrode. As a consequence of the negative charge, hydrophilicity, and smooth surface of graphene, this downside of noble metals can be successfully compensated. Accordingly, hydrophilic and negatively charged interference are prevented from the surface of this fabricated sensor.

In contrast, transition metals are cost-effective and present in different oxidation states, the latter means that with the aid of their unpaired d-electrons, they can form more than one ion (for example Fe^{2+} and Fe^{3+}). As a result, improvement of the rate of adsorption and desorption of the analyte on the electrode surface, much more current responses, and also a smaller amount of interferences (experienced by noble metals) are amongst the most remarkable advantages of

transition metals[60]. For that reason, transition metals can be considered ideal preferences for sensor fabrication. With the aid of the high electrical conductivity of graphene in combination with Nano-metal oxides, a synergistic effect has been observed, and excellent results for the oxidation of glucose acquired, compared to both of them individually[146,147]. On the other hand, it should be emphasized that the values of metal oxide used in the electrode fabrication, should be optimized precisely since these metal oxides can cause high resistance and inadequate surface-active sites for glucose oxidation. **Table 7** summarized recent reports on the application of graphene and also its various modification in non-enzymatic biosensors.

Table 7. A summary of the non-enzymatic graphene-based biosensors for detecting diabetes

Sensing material	Linear range	Limit of detection	References
RGO-Au-CuO-NPs	10^{-3}–12 mm	0.01 μm	[148]
Nafion/Cu-NWs-MOFs-GO	2×10^{-2}–26.6 mm	7 μm	[149]
N-doped graphene	1.3×10^{-5}–14 mm	14.52 μm	[150]
Pt-NPs-MnO_2-RGO	2×10^{-3}–133 mm	1 μm	[151]
N-RGO-Mn_3O_4-NPs	10^{-3}–3.295×10^{-1} mm	0.5 μm	[152]
RGO-Pt-NiO	0.008–14.5 mm	2.67 μm	[153]
Ag-Pt-RGO	0.003–7.72 mm	1.8 μm	[154]
GO-MIP	10^{-7}–10^{-6} mm	10^{-3} μm	[155]
CuO nanoflakes/RGO	10^{-3}–2 mm	0.19 μm	[156]
Co/graphene/IL/SPCE	0.01–13 mm	0.67 μm	[157]
Co/Fe/N-doped graphene	0–32.5 mm	37.7 μm	[158]
PAN-RGO	7.5×10^{-1}–12 mm	600 μm	[159]
NiPcNRs-N-doped RGO	–	1.34 μm	[160]

RGO: reduced graphene oxide, NPs: nanoparticles, NWs: nanowires, MOF: metal-organic framework, GO: graphene oxide, MIP: molecularly imprinted polymers, IL: ionic liquid, SPCE: screen printed electrodes, PAN: polyacrylonitrile, NiPcNRs: nickel phthalocyanine nanorods.

2.8 MIPs

Molecularly imprinted polymers (MIPs) are a class of polymer materials that are produced by the polymerization of functional monomers and cross-linker molecules. Functional monomers are molecules possessing reactive groups in their structure. They are utilized for synthesizing macromonomers or improving the functionality and performance of polymer chains. These molecules are the major participants in the process of preparing MIPs, due to forming complexes with templates which results in the production of recognition sites in polymers. Cross-linkers are molecules having two or more reactive sites. These reactive sites can be functional groups like primary amines, sulfhydryls, etc. Cross-linkers are involved in a chemical reaction to link the polymer chains.

In the process of producing MIPs, a template which can be an atom or ion, a molecule, a complex, or a macromolecular assembly including micro-organisms, is present as a pure stereochemical compound. By removing the template, cavities are formed in the polymer matrix correlatively to the parent template molecules. MIPs are used as recognition elements to design sensor devices[161,162]. These low-cost materials possess unique properties. They have predictable structures and can specifically recognize the target. Plus, high physical stability, robustness, and ease of preparation are their significant features. Thereby, MIPs have been utilized in diverse applications such as chemical separation[163], selective extraction[164], molecular sensing[165], drug delivery[166], and catalysis[167]. Compared to other biological receptors, they are more stable at high thermal conditions, and also have other superior properties such as reusability and selectivity.

There are two main methods for the production of MIPs; covalent and non-covalent imprinting. In the covalent imprinting approach, more amounts of cross-linkers are needed to

produce an insoluble network with good rigidity. This approach reduces the probable interactions which are not desirable because it results in the production of polymers whose bonding groups are exactly located in the imprinted cavities. MIPs produced by this approach possess higher selectivity than the MIPs prepared by the non-covalent method. Also, it is reported that the distribution of their bonding sites is more homogeneous. Covalent imprinting suits the polymer for acting as a catalyst[168]. The disadvantages of covalent imprinting are the limitation in choosing a functional monomer and template, the need for synthesizing the template monomer before polymerization, and lower efficiency in recognizing target molecules[169].

In non-covalent imprinting, there is a non-covalent bond between the polymer network and the template, so the whole synthetic procedure is demonstrated to be easier. Simple extraction processes used in this approach to remove the template from the polymer network are another advantage of this method. The major disadvantage of non-covalent imprinting is the excess need for functional monomers which are necessary for binding with template molecules. As a result, lots of binding sites are formed while many of them are not needed[170]. MIPs can be utilized for sensor applications including non-enzymatic glucose sensing. In **Table 8**, some examples of MIP-based sensors for detecting glucose are listed.

Table 8. A summary of the non-enzymatic molecularly imprinted polymers-based biosensors for detecting diabetes

Sensing material	Linear range	Limit of detection	References
EMMIPs	10^{-8}–10^{-6} mm	3×10^{-6} μm	[171]
MI-QCM	1.38×10^{-9}–1.72×10^{-6} mm	2.7×10^{-7} μm	[85]
Nano-MIP/SPPE	5×10^{-8}–2×10^{-6} mm	8.1×10^{-8} μm	[172]
MIP-cryogel/MWCNTs/Au	5×10^{-11}–1.40×10^{-9} mm	3.3×10^{-8} μm	[173]
MIP/nCD	5×10^{-6}–4×10^{-5}, 5×10^{-5}–6×10^{-4} mm	0.09 μm	[174]
GO-MIP	10^{-5}–6×10^{-3} mm	0.02 μm	[175]
MIP-coated microwave	8.32×10^{-11} mm	2.7×10^{3}–2.22×10^{-4} μm	[176]
MIP-Au-SPE	3.32×10^{-6} mm	24.15–2415 μm	[177]
MIP	19.4×10^{-6} mm	19.4–330 μm	[178]
MIP-PANI	1.0048×10^{-3} mm	2.2×10^{3}–1.11×10^{4} μm	[179]
PPy-MIP	1.25×10^{-8} mm	10^{-5}–10^{3} μm	[180]
MIP-CS/Co_3O_4	4.01×10^{-6} mm	12.17–2.3×10^{3} μm	[181]

EMMIPs: electromagnetic molecularly imprinted polymers, QCM: quartz crystal microbalance, NanoMIP: molecularly imprinted polymer nanoparticles, SPPE: screen printed platinum electrode, nCD: nano-carbon-dots, GO-MIP: graphene oxide, olecular imprinted polymer, SPE: screen printed electrode, PANI: polyaniline, PPy: poly-pyrrole, CHI: chitosan.

3. Conclusions

In this paper, the recent trends in enzymatic and non-enzymatic glucose sensor applications have been thoroughly reviewed and discussed. Among the different types of glucose sensors, enzymatic sensors have pH, temperature, and humidity limitations, however, they possess high sensitivity and specificity. So, non-enzymatic glucose sensors have been developed to cover their weaknesses. They function as the electrode material on which the glucose is oxidized. Recent efforts have been focused on exploiting different materials and fabrication processes for these electrodes, such as metal-based nanostructures, metal oxides, conductive polymers, carbon nanotubes, graphene derivatives, and molecularly imprinted polymers.

Unique nanostructures and approaches have been created to modernize glucose sensors as a result of the recent rise in nanotechnology research. Metals or metal oxides with nanostructures provide a higher surface area for the oxidation of glucose. The utilization of carbon nanotubes and graphene has also attracted a lot of interest because the high conductivity of these materials boosts the electron transfer rate and hence enhances their sensitivity. The majority of nanotechnology research has been concentrated on the creation and integration of nanomaterials

to enhance the performance of electrochemical glucose sensors. Before these approaches may be commercialized, there are still several obstacles to overcome with regard to their application to the human body. Due to their poor biocompatibility, high cost, and labor-intensive preparation procedures, the majority of glucose sensors based on novel materials have been restricted. Thus, the challenge of developing materials with high selectivity, low detection limit, a wide detection range, and quick response time still exists when attempting to prepare these sensors for continuous glucose measurement. Future research is expected to continue toward detecting infinitesimally small concentrations of glucose in forming sensors that are capable of being incorporated into small portable devices.

Conflict of interest

The authors declare no conflict of interest.

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