



Review article

Advances in photoelectrochemical glucose sensors: A critical review

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ABSTRACT

Diabetes mellitus (DM) remains a major global health burden, necessitating the urgent demand for advanced diagnostic devices that enable early detection and continuous monitoring of blood glucose levels. Photoelectrochemical (PEC) sensing has emerged as a powerful alternative to conventional glucose detection strategies, offering advantages such as high sensitivity, low background signals, ease of miniaturization, and cost-effectiveness. Although interest in PEC glucose sensors (PGSs) has grown substantially in recent years, the field lacks a comprehensive and integrative review that consolidates recent advances. This article addresses this gap by presenting a critical overview of the fundamental principles of PEC sensing, commonly employed photoelectrode nanomaterials, performance enhancement strategies, and methods for fabricating photoelectrodes for glucose detection. Furthermore, it systematically examines three major categories of PGSs, comprising enzymatic, non-enzymatic, and self-powered systems, spotlighting high-ranking material designs based on critical analytical performance metrics. It also explores the growing role of density functional theory (DFT)-based computational simulations and machine learning (ML) models in predicting, optimizing, screening, and understanding sensor performance. Despite notable advancements, existing PGSs remain at the prototype stage due to persistent challenges, including suboptimal sensitivity, narrow linear detection ranges, high detection limits, low power output, poor selectivity, fabrication complexity, weak operational stability, and the lack of device-level integration. Addressing these limitations requires coordinated advances in material innovation, device architecture optimization, system integration, and scalable manufacturing to enable reliable, high-performance, and commercially viable glucose sensors.

1. Introduction

Blood glucose is an integral component and a primary energy source in the human body. It is a clinical biomarker of diabetes, a chronic metabolic condition characterized by hyperglycemia, leading to severe long-term complications, including cardiovascular diseases, renal failure, stroke, blindness, nerve degeneration, and, in extreme cases, mortality [1]. The International Diabetes Federation (IDF) estimated that 537 million individuals were living with diabetes in 2021, a number projected to rise to 783 million by 2045. In the same year, the disease was responsible for 6.7 million deaths and incurred at least \$966 billion (USD) in healthcare costs [2]. Consequently, considerable strides have been made in developing high-performance glucose sensors for early diagnosis and effective management of diabetes and its complications.

Since Clark and Lyon introduced the first enzyme-based glucose electrodes in 1962 and Yellow Springs Instrument Company rolled out the first commercial electrochemical (EC) glucose sensor (Model 23 A

YSI analyzer) in 1975 using Clark's technology, tremendous efforts have been channeled toward developing glucose sensors for diabetes control and management [3]. Despite their high sensitivity and selectivity, inherent drawbacks, such as limited operational and storage stability, enzyme denaturation, complex immobilization procedures, and the high cost of enzymes [4] hinder the rollout of affordable and reliable devices. In addressing these shortcomings, the synthesis of mimetic nanozymes for developing non-enzymatic EC glucose sensors has gained traction in recent years, as they offer simpler fabrication procedures while maintaining the merits of enzymatic systems and further improving the performance of the devices. However, after 50 years, why has the promise of viable and cost-effective non-enzymatic glucose sensors still not been fulfilled? Researchers have ascribed this to challenges, including high operating potentials, poor selectivity, unpredictable redox reactions, slow electrokinetics, intermediate poisoning, and sub-optimal sensing performance [5]. It is therefore crucial to explore innovative glucose sensing approaches superior to conventional EC

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strategies.

Recently, as summarized in Table 1, the photoelectrochemical (PEC) technique, which combines photo-irradiation with electrochemical processes, has garnered considerable research interest for glucose detection due to its distinct merits over conventional EC methods, including low background noise, high sensitivity, enhanced stability, and facile miniaturization. In PGSSs, the biometric interaction between photoelectrode materials and glucose generates a photocurrent proportional to the analyte’s concentration. These systems are generally classified by semiconductor type-photoanode-based or photocathode-based, each with inherent limitations, such as poor anti-interference capability or low photocurrent output, respectively. Moreover, conventional PGSSs typically require an external bias to operate, which can introduce background signals, increase false positives, and compromise device portability. To overcome these challenges, self-powered PEC strategies have emerged, eliminating the need for external power sources while enhancing energy efficiency, wearability, and long-term operational stability, making them highly attractive for point-of-care (POC) diagnostics. Two main configurations are being actively explored: (i) the dual-photoelectrode system, which leverages the intrinsic Fermi level difference between a photoanode and a photocathode to drive internal charge separation, with the photoanode amplifying the photocurrent and the photocathode enabling bio-recognition [6], and (ii) the single-photoelectrode system, which combines a photoanode or photocathode with a suitable redox couple to generate a built-in photovoltage for autonomous glucose sensing. Unlike conventional PGSSs with simpler architectures and external voltage requirements, self-powered PGSSs demand careful selection and integration of complementary photoelectrode materials with aligned band structures. This distinction influences both the fabrication process, often involving advanced heterojunction design, and the measurement mechanism, which relies entirely on internally generated photocurrent. Specifically, choosing appropriate photoelectrode materials with excellent photoelectric properties is the key to building high-performance PGSSs [7]. Various nanomaterials, including transition metal oxides (TMOs), multinary oxides, metal sulfides (chalcogenides), carbon nanomaterials, organic compounds, and metal-organic frameworks (MOFs), have been investigated for glucose detection.

Although the PEC sensing field has been the subject of several review articles, such as those on enzymatic sensors [8], detection of ions in

water [9], analysis of mycotoxins in food [10], tumor marker detection [11], and pathogen detection [12,13]. Surprisingly, to the best of our knowledge, there remains a notable absence of a dedicated review focusing specifically on PGSSs. This gap is particularly striking considering the substantial volume of high-quality research published annually in this domain (Fig. 1). Therefore, to stay updated with these studies, we present a comprehensive review that introduces the fundamentals of PEC sensing, commonly employed photoelectrode materials, enhancement strategies, and photoelectrode fabrication methods for glucose detection. Furthermore, it systematically examines three major categories of PGSSs, comprising enzymatic, non-enzymatic, and self-powered systems, spotlighting high-ranking material designs, and the roles of computational simulation based on density functional theory (DFT) and machine learning (ML) models in predicting, optimizing, screening, and understanding sensor performance. Notably, current PGSSs remain at the prototype stage due to multiple performance and fabrication challenges, requiring coordinated advances in materials, device design, integration, and scalable manufacturing for commercial viability. This work serves as a valuable reference for materials and nanotechnology researchers, inspiring the development of cutting-edge smart glucose detection devices.

2. Fundamental principles of photoelectrochemical (PEC) sensing

Since Edmond Becquerel’s pioneering discovery of the photoelectric effect in 1839 [14], PEC sensing has emerged as a promising analytical technique that integrates photon absorption and electrochemical principles for detecting various target analytes. Its increasing appeal stems from distinct advantages, including a low background signal (i.e., high signal-to-noise ratio), high sensitivity attributed to the complete separation of the light excitation and signal detection platforms compared to conventional EC methods. As presented in Fig. 2A, a typical PEC sensing system contains three fundamental components: a light source, a detection system, and a signal acquisition unit. At the heart of the system lies the detection module, which comprises a standard three-electrode electrochemical setup. This includes the photoelectrode serving as the working electrode (WE), a platinum wire as the counter electrode (CE), and either an Ag/AgCl or a saturated calomel electrode (SCE) as the reference electrode (RE). The photoelectrode, often functionalized with photoactive materials, serves as the transducer, converting biochemical interactions into quantifiable photoelectric signals, and thereby plays a central role in the PEC detection process [15]. The PEC sensing mechanism is initiated upon photo-irradiation of the photoelectrode surface, wherein photons with energy greater than the semiconductor’s bandgap excite electrons from the valence band (VB) to the conduction band (CB), generating electron-hole pairs. These charge pairs are then rapidly

Table 1
Comparative Advantages of PEC Glucose Sensors (PGSSs) over Conventional EC Glucose Sensors.

Features	EC Glucose Sensors	PEC Glucose Sensors
Signal Quality & Sensitivity	Moderate sensitivity; affected by background noise and redox limitations	High sensitivity due to light-driven charge separation and reduced background interference
Power & Energy Requirements	Require an external power supply	Can operate at low bias or self-powered under light, reducing energy dependence
Selectivity & Detection Range	Limited selectivity; often requires modifications for specificity	Tunable selectivity via material and light modulation; broad and low detection range possible
Material & Enzyme Dependence	Often relies on noble metals or enzymes with stability issues	Employs diverse photoactive materials; both enzymatic and non-enzymatic formats are available
Stability & Practicality	Enzyme degradation reduces long-term use	Better stability in non-enzymatic formats; suitable for complex or wearable applications
Integration & Functionality	Well-established but limited in multifunctionality	Easily miniaturized; offers dual roles like energy harvesting and real-time glucose sensing simultaneously

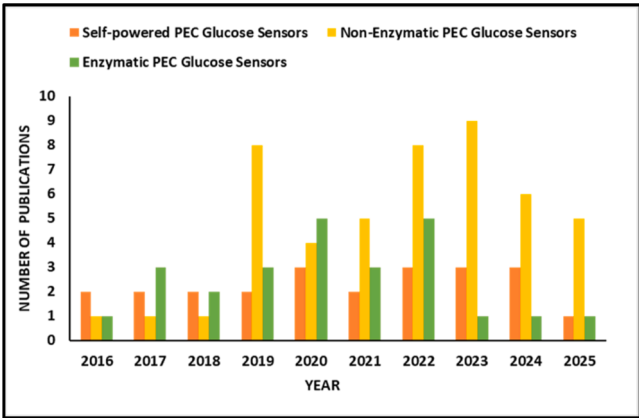


Fig. 1. Publication trends of PGSSs based on enzymatic, non-enzymatic, and self-powered systems (collated from Scopus’ database).

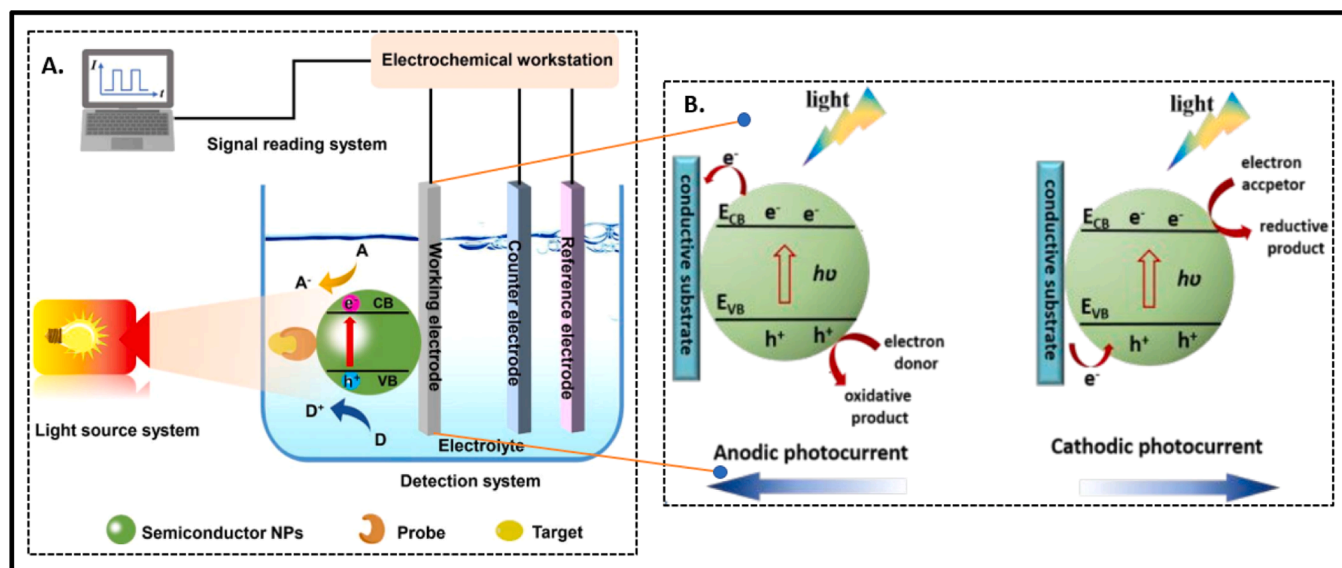


Fig. 2. The schematic illustrations of (A) a typical PEC detection system, (B) photocurrent generation mechanism (Adapted from [22,23]; with permission from the Royal Society of Chemistry).

separated by the internal electric field at the semiconductor–electrolyte interface [16]. Following charge separation, photoexcited electrons in the CB may migrate to the electrode or into the electrolyte, or they may recombine with holes in the VB. The fate of these photoelectrons determines the polarity of the resulting photocurrent [17]. As illustrated in Fig. 2B, in the presence of an electron donor (reductant), hole oxidation leads to electron transfer to the electrode, resulting in anodic

photocurrent generation. Conversely, in the presence of an electron acceptor (oxidant), the photogenerated electrons are captured, while electrons from the electrode recombine with holes, producing a cathodic photocurrent [13]. The widely accepted PEC mechanism comprises four sequential steps: (i) photon absorption, (ii) electron–hole pair generation and separation, (iii) migration and recombination of charge carriers, and (iv) participation of the separated charges in redox reactions.

Table 2

Classification of Photoelectrode Nanomaterials for PEC Glucose Sensing.

Material Class	Representative Material (Band Gap)	Merits & Features	Demerits	Reference
Transition Metal Oxides (TMOs)	TiO ₂ (~3.2 eV); ZnO (~3.3 eV)	Abundant, stable, non-toxic, strong oxidative power, high stability under illumination; High electron mobility, low cost, easy synthesis	Wide band gap limits visible light absorption; UV-only response, photocorrosion in aqueous media	[26,27]
	CuO (~1.2–1.7 eV), Cu ₂ O (~2.0 eV), WO ₃ (~2.6–2.8 eV), Co ₃ O ₄ (~1.5–2.0 eV)	Narrow band gap for visible light absorption, good catalytic activity, high absorption coefficient, good catalytic activity.	Poor stability, prone to photocorrosion, low carrier mobility	[28–31]
	NiO (~3.6 eV); α -Fe ₂ O ₃ (~2.1 eV)	p-type conductivity, chemical stability; Narrow band gap, earth-abundant, stable in aqueous media	Wide band gap, limited light absorption; Short hole diffusion length, poor conductivity	[32,33]
	Sn ₃ O ₄ (~2.5 eV est.); CeO ₂ (~3.0 eV)	Mixed valence states, broad spectrum absorption; Oxygen storage capacity, stability	Limited PEC reports, stability concerns; UV-active, poor visible response	[34,35]
	MnO ₂ (~1.3–2.0 eV); V ₂ O ₅ (~2.2 eV)	High catalytic activity, low cost; Good catalytic activity, visible light response; High conductivity, good catalytic activity	Low conductivity; Stability issues	[36,37]
Multinary / Ternary Oxides	BiVO ₄ (~2.4 eV), Bi ₂ WO ₆ (~2.7 eV); Fe ₂ V ₄ O ₁₃ , Fe ₂ TiO ₅ (~2.1 eV); MnV ₂ O ₆ ; BiPO ₄ (~4.0 eV); NiCo ₂ O ₄ (~2.1 eV); SrTiO ₃ (~3.2 eV)	Visible light absorption, high oxidative ability; narrow band gap, catalytic activity; high stability, oxidative ability; high conductivity, good catalytic activity; high stability, perovskite structure	Poor electron transport, recombination losses; limited PEC studies, low conductivity; limited reports, stability issues; UV-only	[4,38–44]
Metal Sulfides / Chalcogenides	CdS (~2.4 eV), ZnIn ₂ S ₄ (~2.4 eV), In ₂ S ₃ (~2.0 eV), CuInS ₂ (~1.5 eV), Bi ₂ S ₃ (~1.3 eV), CuS (~2.0 eV), PbS (~0.37 eV); MoS ₂ (~1.8 eV)	Strong visible light absorption, narrow band gap, good catalytic activity; Layered structure, catalytic edges	Toxicity, photocorrosion, stability issues; Low conductivity	[45–48,7, 49,50]
Bismuth Oxyhalides	BiOBr (~2.6 eV); BiOCl (~3.4 eV); BiOI (~1.8 eV)	Layered structure, visible light response; High stability, oxidative; Strong visible light absorption	Low conductivity; UV-active; Poor stability in aqueous media	[51,42,52]
Carbon-based Nanomaterials	g-C ₃ N ₄ (~2.7 eV); CNTs; CQDs, GQDs	Metal-free, stable, visible light active; High conductivity, surface area; Upconversion, electron transfer	Low surface area, recombination; not photoactive alone; Weak absorption alone, low intrinsic activity	[53,54]
Metal–Organic Frameworks (MOFs)	Polymer Phenylethynylcopper (PPhECu); Zeolitic Imidazolate Frameworks (ZIF–67)	Tunable band gap, high porosity; High surface area, catalytic	Low conductivity; Poor stability in water	[55,56]
Organic Compounds	Phthalocyanines, porphyrins; phenazines (poly-neural red)	Narrow band gap, strong light absorption; redox activity	Stability issues; prone to degradation	[45]

This cascade of events not only produces measurable photoelectric signals at the electrode–electrolyte interface but also directly influences photoelectric conversion efficiency and signal output intensity [12]. Due to the potent redox capabilities of the generated electron–hole pairs, PEC systems exhibit reduced reliance on externally applied potentials, enhancing operational efficiency and system simplicity [18]. A key feature of PEC sensing is its ability to achieve selective and sensitive detection by incorporating biorecognition elements, such as enzymes, antibodies, or aptamers, on the electrode surface. These elements enable the modulation of photocurrent responses in the presence of specific target analytes [19,20]. In such systems, redox reactions between the immobilized biorecognition elements and target analytes induce changes in the photocurrent, which are directly correlated with analyte concentration. These interactions can affect charge transfer dynamics or alter the local electrode environment, thereby producing detectable shifts in the photoresponse [21].

2.1. Photoelectrode nanomaterials

Photoelectrode nanomaterials with outstanding photocatalytic properties are essential for developing high-performance, light-driven PGSs. The selection and engineering of appropriate nanomaterials has become a major research focus, requiring careful evaluation of key attributes such as strong visible light absorption, high photoelectric conversion efficiency, efficient charge separation and transfer, optical stability, and biomolecule immobilization ability [24,25]. As presented in Table 2 and Fig. 3, a wide range of nanomaterials with exceptional biocompatibility, high conductivity, and favorable optoelectronic properties, including transition metal oxides (TMOs), carbon-based nanomaterials, multinary/ternary oxides, metal sulfides (chalcogenides), bismuth oxyhalides, metal-organic frameworks (MOFs), and organic compounds such as phthalocyanines, porphyrins, and phenazines, have found application in the development of PGSs. Particularly, TMOs continue to attract significant attention due to their intrinsic advantages such as non-toxicity, natural abundance, low cost, excellent chemical stability, high specific surface area, strong photocatalytic activity, unique electronic band structures, and powerful oxidative capabilities. An ideal photoactive semiconductor TMO should possess the following characteristics: (i) an appropriate band gap to enable broad absorption of visible light; (ii) excellent PEC stability in electrolyte environments; and (iii) high charge carrier mobility, effective charge separation, and fast surface reaction kinetics. Thus, TMOs such as TiO_2 , ZnO , CuO , Cu_2O , NiO , $\alpha\text{-Fe}_2\text{O}_3$, Co_3O_4 , and MnO_2 , as well as multinary/ternary metal oxides like BiVO_4 , Bi_2WO_6 , NiCo_2O_4 , MnVO_2O_6 , Fe_2TiO_5 , and the perovskite-type oxide SrTiO_3 , have been investigated as photoelectrode materials, either as single-component or composites for glucose detection.

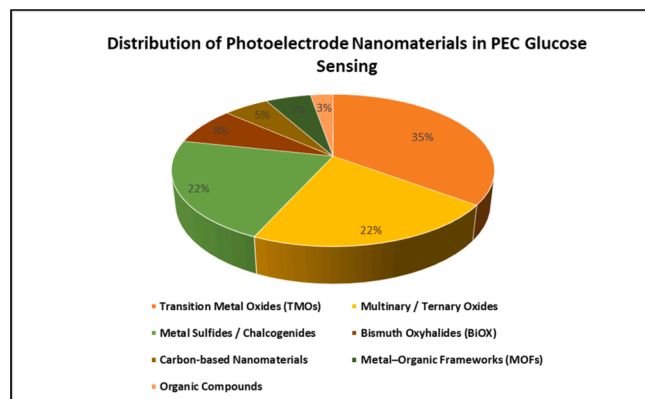


Fig. 3. Percentage distribution of photoelectrode nanomaterials in PEC glucose sensing.

In addition to this, carbon-based nanomaterials such as graphene, graphitic carbon nitride ($\text{g-C}_3\text{N}_4$), reduced graphene oxide (rGO), carbon nanotubes (CNTs), graphene quantum dots (GQDs) and carbon quantum dots (CQDs) have been utilized owing to their excellent electrical conductivity, large surface area, tunable band gaps, and superior light absorption properties. For instance, reduced graphene oxide (rGO) functioned as an electron transport medium in a $\text{TiO}_2/\text{rGO}/\text{PDA}$ -based PEC sensor, facilitating efficient charge transfer and enhancing the separation of photoinduced electron–hole pairs [57]. Metal sulfide (chalcogenide) nanomaterials, including CdS , ZnS , CuS , PbS , Bi_2S_3 , MoS_2 , In_2S_3 , CdSe , CuInS_2 , and ZnIn_2S_4 , have also demonstrated considerable potential as photoelectrode materials due to their low cost, high charge mobility, narrow particle size distribution, broad excitation spectra, excellent photostability, unique electronic structures, strong optical/electronic quantum effects, and tunable emission properties. A representative example is the integration of GP@CdS nanowires with CuO , which significantly enhances photoelectrocatalytic glucose oxidation in alkaline solution, showing a 2.45-fold increase in sensitivity under visible light compared to dark conditions [58]. Furthermore, bismuth oxyhalides (BiOX , where $\text{X} = \text{Cl}$, Br , or I) have also emerged as promising photocatalysts due to their appropriate band gaps and unique layered tetragonal crystal structures, which generate strong internal electric fields to facilitate effective separation of photogenerated electron–hole pairs [59]. These materials also exhibit low toxicity, high chemical stability, and low production costs [60]. For example, the construction of a BiOBr-TiO_2 p–n heterojunction not only broadens visible light absorption but also reduces electron–hole recombination, thus improving PEC performance [51].

Recently, metal–organic frameworks (MOFs), particularly zeolitic imidazolate frameworks (ZIFs), have emerged as promising photoelectrode candidates for PGS due to their highly ordered porous architectures, tunable chemical compositions, and abundant active sites, which enable strong light–matter interactions and selective glucose electrocatalysis [55]. Complementing these inorganic–organic hybrids, organic optoelectronic materials with extended π -conjugated systems, such as porphyrins, phthalocyanines, and phenazines, offer broad light absorption and efficient charge transfer. Among them, poly (neutral red) (PNR), a phenazine-derived polymer, exhibits notable photoelectric response and has recently been applied in PEC glucose sensing [45].

Despite the demonstrable promise of each class, single-component photoelectrodes rarely meet all performance requirements. Common shortcomings, including fast electron–hole recombination, limited visible-light absorption, low intrinsic conductivity, poor glucose oxidation kinetics, and photodegradation, motivate the widespread adoption of composite and interface-engineering strategies. In this context, heterojunction formation, morphological control, elemental and single-atom doping, and modification of electrocatalysts or co-catalysts are essential design levers.

2.2. PEC enhancement strategies for glucose sensing

2.2.1. Heterojunction formation strategy

The formation of heterojunctions by combining two semiconductors with unequal band gaps is a widely adopted strategy for enhancing the photocatalytic performance of PGSs. Typically, a narrow band gap semiconductor is coupled with a wide band gap counterpart to suppress the recombination of photogenerated electron–hole pairs and to promote visible light absorption, thereby improving the photocurrent response. Various types of heterojunctions have been developed based on charge separation and transfer mechanisms, including p–n heterojunctions, Z-scheme heterojunctions, Schottky junctions, and type-II heterojunctions. As described in Fig. 4A, the p–n heterojunction is particularly effective because when two dissimilar semiconductors come into contact, electrons diffuse from the n-type semiconductor, which has a higher Fermi level (E_F), to the p-type semiconductor with a lower E_F , until they attain equilibrium, generating a built-in electric field and

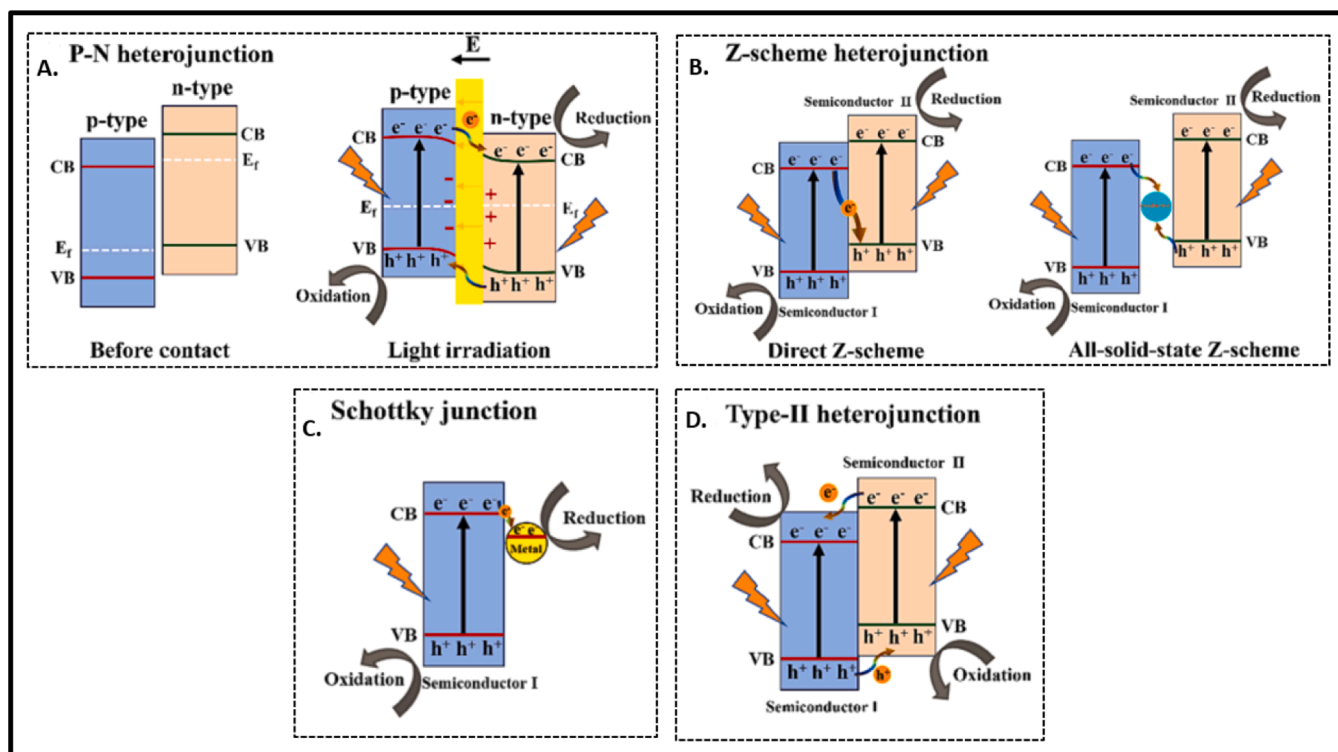


Fig. 4. Schematic illustrations of a (A) p-n heterojunction, (B) Z-scheme heterojunction, (C) Schottky junction, (D) type-II heterojunction (Adapted from [13]; with permission from Elsevier).

depletion region that promote charge separation in PEC processes. For example, constructing a CuO/TiO₂ p-n heterojunction, as shown in Fig. 5A, enables PEC detection under visible light and enhances photoelectric conversion efficiency, thereby improving glucose sensing performance [61]. Also, artificial Z-scheme heterojunctions mimic the natural electron transfer processes observed in photosynthesis. As illustrated in Fig. 4B, the Z-scheme system has two mainstream categories: direct and all-solid-state. These structures effectively couple two semiconductors via electron mediators to improve charge separation

and photocurrent conversion efficiency. A notable example is described in Fig. 5B, a Z-scheme TiO₂/Ti₃C₂T_x/Cu₂O heterostructure synthesized via a one-step hydrothermal process, which demonstrated excellent PEC activity for glucose detection [62]. Moreover, as depicted in Fig. 4C, coupling metal nanoparticles with semiconductor films to form Schottky junctions enhances light absorption via localized surface plasmon resonance (LSPR) and boosts photocurrent through catalytic effects. A case study provided in Fig. 5C is the enhanced performance of Au NPs/TiO₂ NTs nanostructures, where gold modification promotes TiO₂

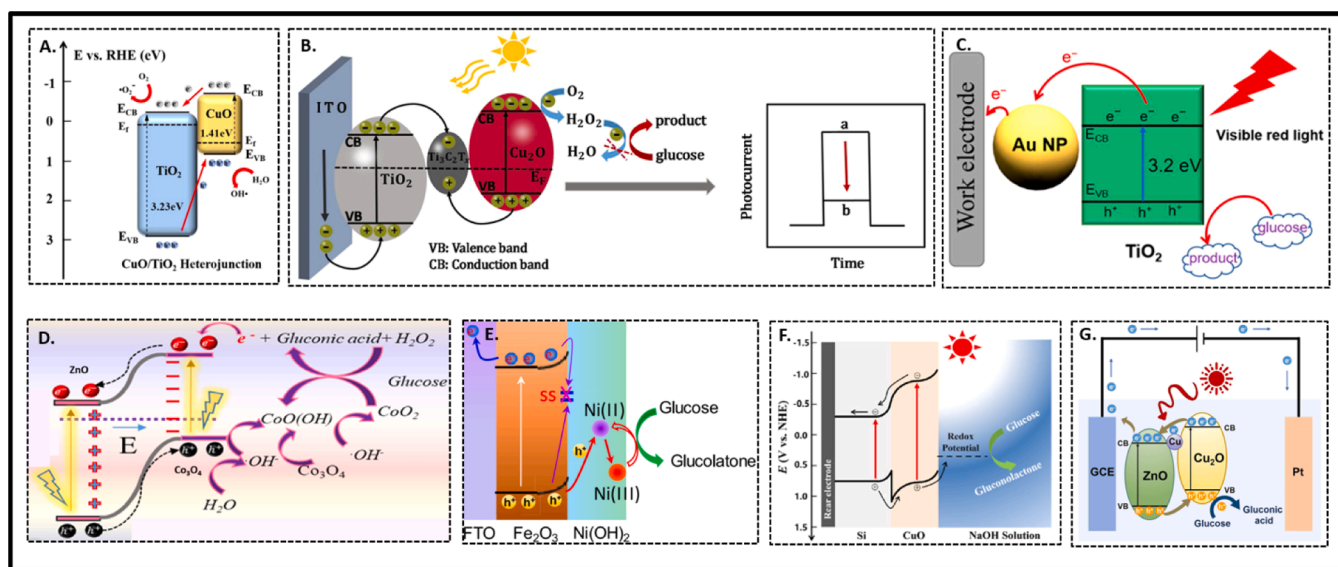


Fig. 5. Schematic illustrations of PEC sensing mechanism based on (A) CuO/TiO₂ heterojunction (Adapted from [61]; with permission from the American Chemical Society), (B) TiO₂/Ti₃C₂T_x/Cu₂O heterostructure (Adapted from [62]; with permission from Elsevier), (C) Au/TiO₂NTs composite (D) Z-scheme 3D Co₃O₄/ZnO composite (Adapted from [65]; with permission from Elsevier), (E) Ni(OH)₂/Fe₂O₃ composite (Adapted from [66]; with permission from Elsevier), (F) SiNWs-CuONSs heterojunction (Adapted from [67]; with permission from Elsevier), (G) Cu₂O-Cu-ZnO composite (Adapted from [68]; with permission from Elsevier).

absorption under visible light via the SPR effect, while also facilitating charge-pair separation through the formation of a Schottky junction [63]. Lastly, as described in Fig. 4D, Type-II heterojunctions, formed by semiconductors with staggered band alignments, enable efficient charge separation as electrons move from semiconductor II to I and holes in the opposite direction under light irradiation. A representative example shown in Fig. 5D is a 3D $\text{Co}_3\text{O}_4/\text{ZnO}$ heterostructure, which exhibits broad-spectrum light absorption, efficient charge separation, and reduced carrier recombination, resulting in high-performance PEC glucose sensing [64]. Thus, the rational design and engineering of heterojunction architectures hold great promise for the development of highly sensitive and selective PGSSs with enhanced performance.

2.2.2. Electrocatalyst modification strategy

Electrocatalyst modification is a powerful strategy to enhance PEC performance by accelerating surface reaction kinetics, and facilitating efficient charge separation. Electrocatalysts not only introduce additional active sites for surface reactions but also passivate surface states, thereby enhancing interfacial charge transfer and suppressing carrier recombination. Transition metal oxides and hydroxides, such as Co_3O_4 , CoOOH , NiOOH , and $\text{Ni}(\text{OH})_2$, have been widely recognized as excellent electrocatalysts for glucose oxidation. For instance, NiOOH effectively improved the PEC and sensing performance of TiO_2 nanotube arrays (TNT) for glucose oxidation [64]. Elsewhere, as illustrated in Fig. 5E, the deposition of $\text{Ni}(\text{OH})_2$ on the surface of hematite (Fe_2O_3) significantly enhanced the photoelectrode's performance, with a photocurrent approximately five times higher than bare Fe_2O_3 . Detailed investigations attributed the improvement to the dual role of $\text{Ni}(\text{OH})_2$, which passivates surface states on Fe_2O_3 to suppress recombination and enhances band bending at the Fe_2O_3 /electrolyte interface for more efficient charge separation and transfer [66].

2.2.3. Morphology control strategy

Morphology control involves tailoring the structural characteristics of photoelectrode nanomaterials to optimize charge carrier transport and separation, as well as enhance the light-harvesting capabilities of PEC sensors. Based on their spatial dimensions, nanomaterials are classified into zero-dimensional (0D), one-dimensional (1D), two-dimensional (2D), and three-dimensional (3D) structures. The simplest forms, 0D nanomaterials, include quantum dots (QDs) and nanoclusters; 1D nanomaterials comprise nanotubes, nanorods, and nanowires; 2D nanomaterials include nanosheets, nanoplates, and nanodisks; while 3D nanomaterials encompass metal-organic frameworks (MOFs) and flower-like nanostructures. These nanomaterials exhibit unique physicochemical properties, such as high surface area, abundant active sites, excellent electrical conductivity, tunable bandgap, chemical and thermal stability, strong light absorption, efficient electron transfer, and superior biocompatibility. These attributes make them highly promising for the development of PGSSs [69]. Specifically, dense, burr-like CuO nanospikes (CuONSs) were grown on relatively sparse and ordered silicon nanowires (SiNWs). As demonstrated in Fig. 5F, the resulting SiNWs-CuONSs heterojunction exhibited excellent sensing performance, attributed to the large specific surface area of the SiNWs and the abundance of active sites provided by the CuONSs [67].

2.2.4. Doping strategy

Doping refers to the intentional introduction of impurities (dopants) into semiconductor materials to modify their electronic structure. Essentially, this section focuses on elemental doping and single-atom metal doping.

2.2.4.1. Elemental doping. Elemental doping of various metal and non-metal nanoparticles (NPs), such as Au, Al, Cu, Ag, Co, Bi, C, N, S, P, and B, onto photoelectrode supports represents an effective and promising strategy to enhance the performance of glucose sensors. This

approach leverages the exceptional properties of these nanoparticles, including high photocatalytic activity, excellent electrical conductivity, increased surface reactivity, biomolecule immobilization, and the SPR effect. Numerous studies have demonstrated that metal nanoparticle doping enhances the photoelectric conversion efficiency of photoactive materials by: (i) modulating band gaps to improve light absorption; (ii) altering morphology and crystalline structure; (iii) promoting the separation of photogenerated electron-hole pairs; and (iv) facilitating electron transfer while minimizing charge recombination, primarily through the LSPR effect [13]. For example, Au nanoparticles (AuNP) were deposited on ZnO nanorod arrays, exhibiting synergistic catalytic effects in glucose detection, with 1.29-fold improvement in sensitivity under light conditions [27]. Similarly, as illustrated in Fig. 5G, copper nanoparticles (Cu NPs) were incorporated into $\text{Cu}_2\text{O}/\text{ZnO}$ nanocomposites, possessing outstanding photocurrent response with excellent PEC performance under light illumination [68]. Likewise, by incorporating AuNPs into NiO_{1-x} , the unique structure and the generated plasmons on the Au surface improve the electron transfer in the nanowire arrays, providing excellent mass transport for glucose oxidation [70].

2.2.4.2. Single-atom metal doping. The deposition of single-atom metals (e.g., Ni, Pt, Co, Fe, Ir) on photoelectrode supports is a promising strategy for enhancing the performance of PGSSs. Since the seminal work by Zhang and co-workers in 2011 [71], single-atom catalysts (SACs), characterized by isolated, well-defined active sites uniformly anchored on support materials, have attracted significant attention in glucose sensing due to their exceptional catalytic activity, high atom-utilization efficiency, large specific surface area, selectivity, stability, tunable coordination environment, strong metal-support interactions, and quantum size effects. Various nanomaterials have been employed as supports for SACs, including carbon-based materials (carbon black, carbon nanotubes, carbon fibers, graphene), metal oxides, metal-organic frameworks (MOFs), and covalent organic frameworks (COFs) [72]. Among these, metal oxides are notable for their tunable surface pH and redox properties [73], while carbon-based supports are favored for their excellent conductivity and doping flexibility. More recently, SACs have been explored as enzyme mimics, offering strong potential in sensor development [74]. Although their application in PEC systems is still emerging, their proven performance in electrochemical glucose sensing and photocatalysis suggests they hold great promise in this domain [75]. Incorporating SACs into photoelectrodes can modulate the band and electronic structures of photoactive materials, enhancing light absorption, promoting photogenerated charge separation and transport, and accelerating surface catalytic reactions [76]. A critical advantage of SACs lies in their suitability for theoretical investigations to pinpoint active sites, providing insight into atomic-level catalytic mechanisms and structure-performance relationships [77]. However, due to their high surface energy, single atoms are prone to migration or agglomeration, making SACs synthesis challenging. To address this, several methods have been developed, including chemical vapor deposition, atomic layer deposition, pyrolysis, wet chemistry, photochemical methods, and atom trapping. Among these, electrochemical deposition is particularly advantageous, as it overcomes the limitations of requiring specific support materials or metals. In this method, metal ions are deposited under an applied electric field, with optimal atomic loading kept below 4.7 wt% to prevent aggregation into nanoparticles or multilayer structures. A case study is the cathodic and anodic electrochemical deposition of Ir species depicted in Fig. 6(A & B) [78].

2.3. Photoelectrode fabrication methods for PEC glucose sensing

The fabrication of highly efficient photoelectrodes is fundamental to advancing the frontiers of PGSSs, as their structural and photoelectric properties directly influence sensor performance. The choice of

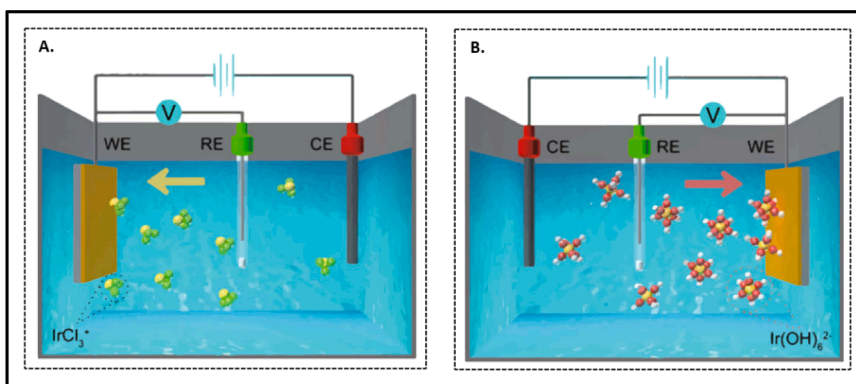


Fig. 6. Schematic illustrations of the mechanisms of the electrochemical deposition of Ir species (A) cathodic, (B) anodic (Adapted from [78]).

photoelectrode substrate, such as fluorine-doped tin oxide (FTO) glass, indium-doped tin oxide (ITO) glass, glassy carbon electrode (GCE), or metal foils (Ti), is crucial because it affects the conductivity, stability, and adhesion of the photoelectrode. To meet clinical requirements for glucose sensing, various fabrication strategies have been utilized. This section discusses the common fabrication methods, including direct methods- hydrothermal, solvothermal, successive ionic layer adsorption and reaction (SILAR), electrochemical anodization, electrodeposition techniques, and the indirect method- sol-gel method.

2.3.1. Direct methods

2.3.1.1. Hydrothermal and solvothermal methods. The hydrothermal method is a simple, cost-effective approach for synthesizing photoelectrodes. It uses an aqueous solvent in a sealed vessel (typically a Teflon-lined autoclave), where precursors react under high temperature and pressure in a muffle furnace to form crystalline materials. This method allows control over morphological, optical, and structural

properties and enables the formation of complex nanostructures like nanorods, nanoflowers, and hierarchical architectures, with benefits such as low processing temperatures and high yield [79]. Typically, the synthesis lasts for about 6–48 h, during which dissolution, nucleation, and crystallization of otherwise poorly soluble or insoluble materials occur [80]. Based on the feeding method, hydrothermal synthesis can be classified into continuous and one-pot reactions. In continuous hydrothermal reactions, feedstocks are introduced into the reaction vessel via a pump. In contrast, one-pot reactions involve pre-loading the feedstocks into the autoclave, which is then heated to a predetermined temperature for a specified duration to produce the final product. Likewise, solvothermal synthesis shares similar reaction mechanisms and equipment with the hydrothermal method but utilizes non-aqueous solvents such as ethanol, methyl ethyl ketone, or isopropyl alcohol. This technique is used for synthesizing metal-organic frameworks (MOFs), sulfides, nitrides, and other complex materials that require precise control over nucleation and growth [81]. For example, BiOCl-G NHS and Ni (OH)₂/Fe₂O₃ photoelectrodes were synthesized via the hydrothermal

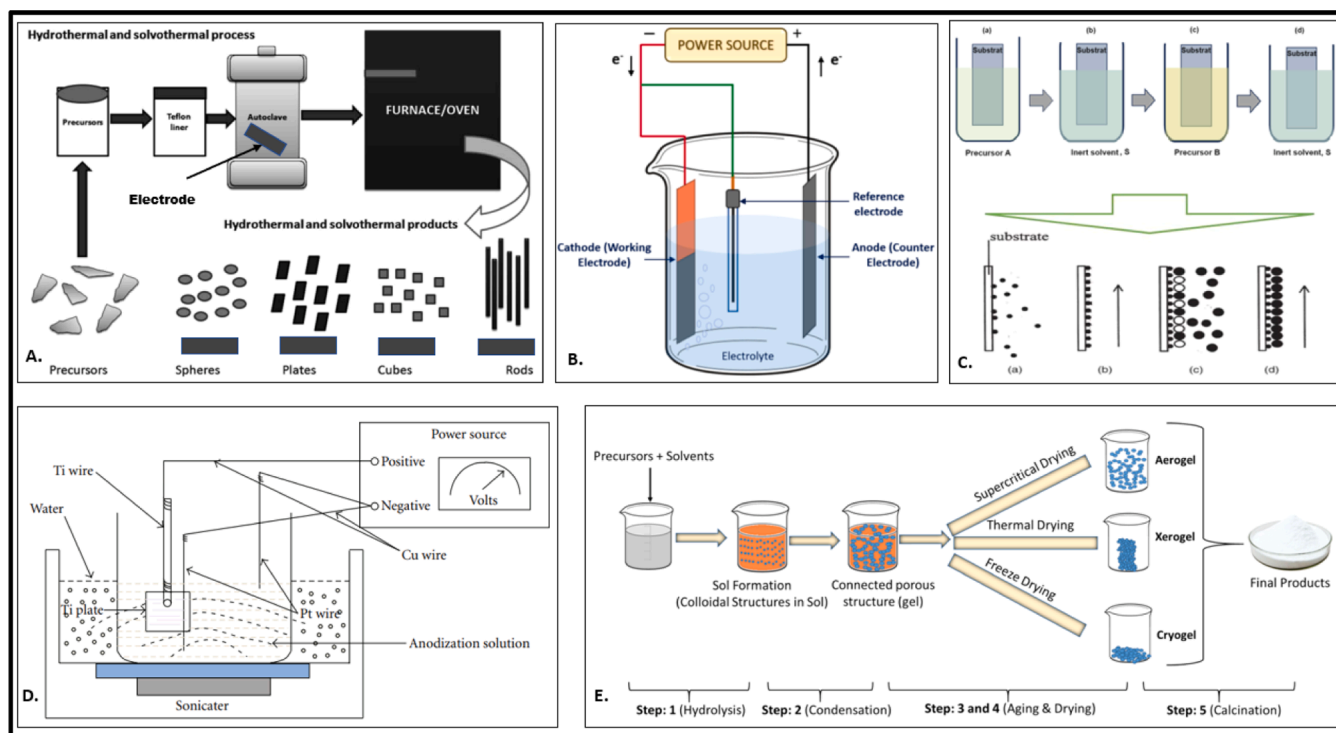


Fig. 7. The Schematic illustrations of the (A) hydrothermal/solvothermal method (Adapted from [82]; with permission from WILEY-VCH), (B) electrodeposition method (Adapted from [83]), (C) SILAR method (Adapted from [84]), (D) anodization method (Adapted from [85]), (E) sol-gel method (Adapted from [86]; with permission from Springer).

method [52,66], while a $\text{Sn}_2\text{O}_4/\text{CdS}$ photoelectrode was prepared using the solvothermal method for glucose detection [34]. Fig. 7A shows the schematic illustration of these methods.

2.3.1.2. Electrodeposition method. Electrodeposition is an electrochemical method for depositing metals from a metal salt solution onto a photoelectrode substrate. It enables the formation of diverse structures, from thin films to nanoscale features like nanotubes, nanorods, and nanoclusters. Typically, it is a cathodic (reductive) process; it can also occur anodically. As shown in Fig. 7B, the setup includes a working (cathode), counter (anode), and reference electrode immersed in an electrolyte containing metal cations. A power source applies a potential that drives cation migration to the cathode and anion movement to the anode [83]. In addition, it is a highly scalable technique that enables the rapid formation of nanomaterials with high porosity and increased surface area. For example, CuO thin films have been electrodeposited onto FTO glass substrates to fabricate photoelectrodes, achieving a remarkable result for PEC glucose sensing [87]. Similarly, Wang et al. developed a nanoporous BiVO_4 -based photoelectrode using this method, which exhibited an excellent PEC performance [88].

2.3.1.3. Successive Ionic Layer Adsorption and Reaction (SILAR) method. Successive Ionic Layer Adsorption and Reaction (SILAR) is a simple and cost-effective wet chemical deposition technique used for the controlled deposition of various thin films and nanoparticles onto substrates. It is particularly practical for large-area applications and is considered a modified variant of atomic layer deposition (ALD) and chemical bath deposition (CBD). As illustrated in Fig. 7C, the step-by-step procedure for thin-film deposition via SILAR includes: (i) adsorption of ionic species onto the substrate by immersion in a cationic precursor solution, (ii) rinsing with deionized water to remove excess adsorbed ions and prevent homogeneous precipitation, (iii) immersion in an anionic precursor solution, where pre-adsorbed cations react with newly adsorbed anions to form the desired material, and (iv) a second rinse with deionized water to eliminate unreacted species and excess ions. These cycles are repeated to gradually form the desired thin film on the substrate. Key parameters affecting the film quality include the concentration and temperature of the cationic and anionic solutions, as well as the duration of each immersion and rinsing step [84,89]. While SILAR offers several advantages, such as ease of implementation, scalability, and cost-effectiveness, it also has limitations, including its time-consuming nature and sensitivity to precursor quality. For example, Pelissari et al. adopted the SILAR method to develop an FTO/ $\text{Fe}_2\text{V}_4\text{O}_{13}$ electrode for non-enzymatic glucose detection, showing remarkable PEC activity [40].

2.3.1.4. Anodization method. Anodization is a simple and cost-effective method for synthesizing nanostructured materials through the electrochemical oxidation of a metal substrate (e.g., titanium or copper) in a suitable electrolyte solution (Fig. 7D). This technique enables the formation of structures with high surface area and well-controlled morphology and is commonly used to prepare TiO_2 -based photoelectrodes for PEC glucose sensors. The process typically involves cleaning the metal substrate (e.g., Ti foil), followed by anodization in a fluoride-containing electrolyte solution (such as NH_4F) under a controlled voltage, usually above 20 V. After anodization, the substrate is rinsed, dried, and may undergo additional chemical modifications to enhance its photoelectrochemical properties. Anodization is highly sensitive to parameters such as voltage, temperature, and electrolyte composition, with small variations significantly influencing the morphology and performance of the resulting nanostructures. Syrek et al. fabricated an anodic TiO_2 -based PEC glucose sensor using this method, which demonstrated promising performance characteristics [90].

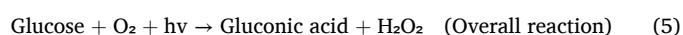
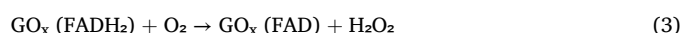
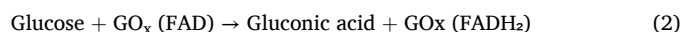
2.3.2. Indirect method

2.3.2.1. Sol-gel method. The sol-gel method is a well-established synthetic approach for preparing high-quality metal oxide nanoparticles, such as ZnO and TiO_2 nanoparticles, before deposition on the substrate for glucose detection. This method offers excellent control over the texture and surface properties of the materials. As described in Fig. 7E, the sol-gel process typically involves several steps: hydrolysis, polycondensation, aging, drying, and thermal decomposition [86]. Notable advantages of the sol-gel method include the high homogeneity and purity of the resulting nanostructures, as well as its low cost and user-friendly reaction conditions. However, one disadvantage is the need for a secondary high-temperature treatment to fabricate suitable nanostructures, which increases the overall processing time [18]. For instance, BiVO_4 thin films were deposited on photoelectrode substrates using this strategy for glucose detection [91].

3. Photoelectrochemical (PEC) Glucose Sensors (PGSSs)

3.1. Enzymatic PEC Glucose Sensors (EPGSs)

EPGSs have attracted considerable attention due to their high sensitivity, excellent biocompatibility, and strong substrate specificity. These sensors typically employ glucose oxidase (GO_x) as the bio-recognition element, harnessing the synergistic interaction between enzymatic catalysis and the photocatalytic activity of photoelectrode materials under light irradiation. The sensing mechanism is governed by light-induced charge generation in the photoelectrode and enzyme-mediated glucose oxidation. Upon illumination, the photoelectrode generates electron-hole (e^-/h^+) pairs. GO_x catalyzes the oxidation of glucose to gluconic acid and hydrogen peroxide (H_2O_2). Subsequently, H_2O_2 reacts with photogenerated holes to form molecular oxygen (O_2), promoting electron transfer between the redox-active FAD cofactor in GO_x and the electrode surface, thereby amplifying the photocurrent. During this process, flavin adenine dinucleotide (FAD) cofactor is reduced to FADH_2 and subsequently reoxidized by dissolved oxygen, regenerating H_2O_2 and further enhancing signal output [92]. The resulting photocurrent exhibits a linear relationship with glucose concentration, enabling quantitative detection. The fundamental reactions are summarized as follows:



Despite these advantages, EPGSs often suffer from limited photo-conversion efficiency, poor charge carrier separation, narrow light absorption, low selectivity, and the intrinsic instability of enzymes, factors that constrain their implementation in device-level sensors and keep them at the prototype stage. Nonetheless, various studies, as highlighted in Table 3, offer valuable and innovative strategies to mitigate these issues.

For instance, Yang et al. engineered a $\text{TiO}_2(001)/\text{Au}/\text{GO}_x$ composite via a hydrothermal method to overcome the poor photoelectric efficiency of TiO_2 nanomaterials stemming from a mismatch in electron-hole diffusion distances and migration rates through the suppression of hole migration. The incorporation of Au nanoparticles onto the (001) crystal facets of TiO_2 , coupled with their strong affinity for GO_x , significantly enhanced charge separation and enzyme immobilization, as depicted in Fig. 8A. The resulting system exhibited a sensitivity of $16.86 \mu\text{A mM}^{-1}\text{cm}^{-2}$, a linear detection range of 0.01–3 mM, and a low detection limit of 0.83 μM , underscoring its potential for sensitive

Table 3

Performance Comparison of Notable EPGSs.

Photoelectrode Material	Sensitivity ($\mu\text{A cm}^{-2}\text{mM}^{-1}$)	Linear Range (mM)	Detection Limit (μM)	Photocurrent Retention (%), at 4 °C	Reproducibility (%)	Reference
TiO ₂ /Au/GO _x /Ti	16.86	0.01–3.00	0.830	n/a	n/a	[93]
GOD _x /Au@C/TiO ₂ /FTO	29.76	0.10–1.60	49.00	n/a	n/a	[94]
ZnO–Au–Cu ₂ O/GOD	2.880	1–19.0	80.00	n/a	n/a	[95]
PPhECu/GO _x /GCE	n/a	0.0005–5.00	0.160	n/a	n/a	[56]
TiO ₂ /PDA/GO _x /FTO	57.72	0.2–1.00	28.50	n/a	n/a	[96]
SrTiO ₃ /TiO ₂ /PDA/GO _x /FTO	5.370	0–32.0	25.68	88.70 (30 days)	3.10	[44]
PNR/ZnIn ₂ S ₄ NF/FTO	n/a	0.08–30.0	27.00	99.38 (30 days)	0.08	[45]
SrTiO ₃ /Au/PDA/GO _x /Ti	23.70	1.0–20.0	12.00	99.44 (10 days)	n/a	[97]
TiO ₂ NTs/Au/Pt/GO _x /Ti	81.93	0–0.80	1.390	n/a	n/a	[98]
TiO ₂ /rGO/PDA/GO _x /Ti	13.82	0–3.00	0.034	n/a	n/a	[57]
CDs/B-TiO ₂ /GO _x /FTO	1.648	0–9.00	43.0	n/a	n/a	[99]
Bi ₂ WO ₆ /TiO ₂ /GOD/ITO	n/a	0.008–8.00	3.800	95.50 (14 days)	2.30	[39]

*** n/a- not available, PPhECu – polymer phenylethynylcopper, CDs – carbon dots, PDA- polydopamine, GCE – glassy carbon electrode, PNR – poly-(neutral red), GOx/ GOD – glucose oxidase, NTS – nanotubes.

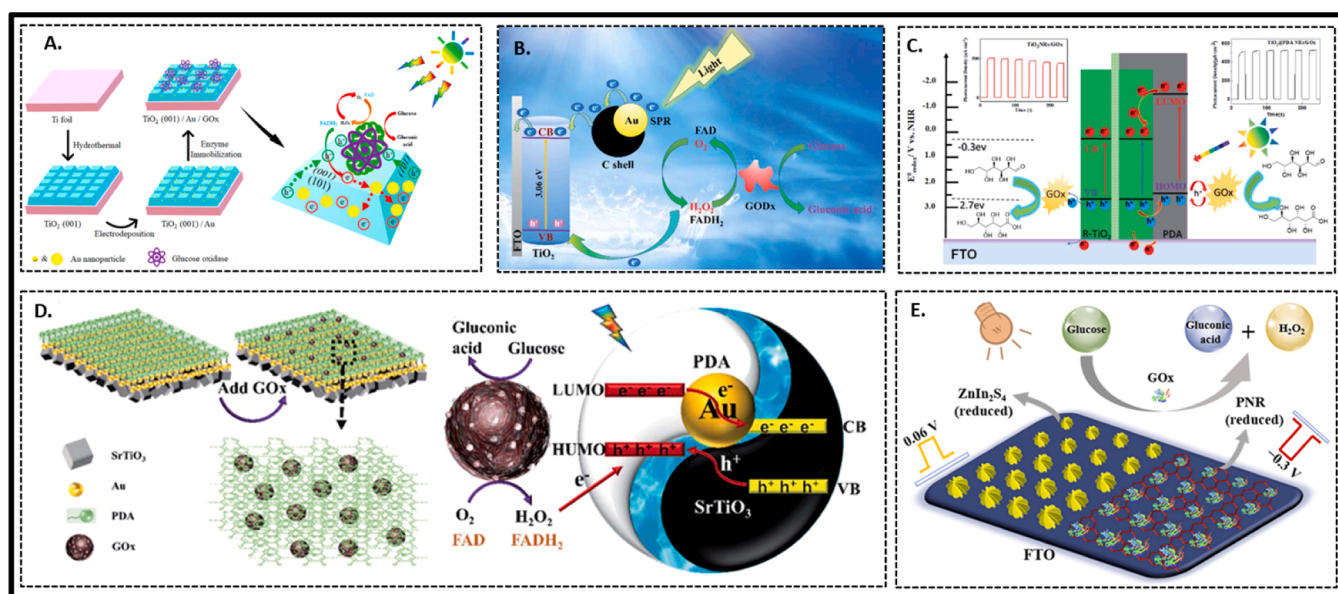


Fig. 8. The Schematic illustrations of the mechanisms of EPGSs based on (A) TiO₂ (001)/Au/GO_x (Adapted from [93]; with permission from American Chemical Society), (B) GOD_x/Au@C/TiO₂ (Adapted from [94]; with permission from the Royal Society of Chemistry), (C) TiO₂/PDA/GO_x (Adapted from [96]; with permission from the Royal Society of Chemistry), (D) SrTiO₃/Au/PDA/GO_x (Adapted from [97]; with permission from the Royal Society of Chemistry), (E) PNR/ZnIn₂S₄ NF/GO_x (Adapted from [45]; with permission from Elsevier).

glucose monitoring [93]. These enhancements were attributed to the synergistic interplay between the engineered crystal facets, the catalytic properties of Au, and the bio-affinity of the (001) surface for enzyme immobilization. Despite these promising outcomes, future optimization of Au nanoparticle deposition and annealing procedures, as well as advanced interfacial characterization, is required.

Likewise, Ge et al. developed an Au-C/TiO₂ NRA/GOD_x composite to address the rapid charge recombination rate and the wide bandgap of TiO₂, which restricts its visible light absorption. The synergistic effects of surface plasmon resonance (SPR) from Au nanoparticles and the carbon shell deposition on the surface of TiO₂ NRAs significantly accelerated the electron transport, decreased the recombination rate, and enhanced overall PEC performance, as illustrated in Fig. 8B. The obtained platform showed outstanding stability and satisfactory reproducibility after 10 cycles of light on/off irradiation, as well as a sensitivity of 29.76 $\mu\text{A mM}^{-1}\text{cm}^{-2}$ in the linear detection range of 0.1–1.6 mM, and a low LOD of 0.049 mM in detecting glucose [94]. However, further validation in complex biological samples and exploration of long-term operational stability remain critical for practical

deployment.

In another study, Xu et al. synthesized a TiO₂/PDA/GO_x composite to mitigate enzyme degradation caused by the oxidative activity of photogenerated holes. The TiO₂ nanorod core, hydrothermally grown on FTO glass, provided efficient charge transport and light absorption due to its one-dimensional (1D) single-crystal arrays, while the PDA shell enhanced interfacial charge transfer and protected the enzyme from oxidative degradation, as described in Fig. 8C. The resulting configuration achieved a sensitivity of 57.72 $\mu\text{A mM}^{-1}\text{cm}^{-2}$, a concentration range of 0.2–1.0 mM, a low detection limit of 0.0285 mM, excellent selectivity, and stable performance under repeated illumination for glucose detection [96]. Nevertheless, broader dynamic range testing, long-term stability assessments, and real-sample validation remain essential.

Advancingly, Wang et al. constructed a SrTiO₃/Au/PDA/GO_x composite via microwave-assisted hydrothermal synthesis. The authors combined SrTiO₃ for its high electron mobility and photocatalytic activity, PDA for its biocompatibility and electrical conductivity, and Au nanoparticles act as an “electron cloud bridge,” facilitating efficient

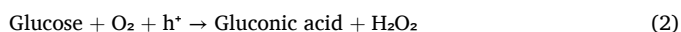
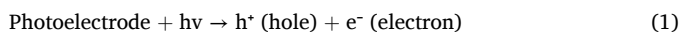
electron transfer at the organic-inorganic interface, as shown in Fig. 8D. The developed system exhibited good light absorption and charge-separation efficiency, resulting in the highest photocurrent value among the tested materials. It delivered a sensitivity of $23.7 \mu\text{A mM}^{-1} \text{cm}^{-2}$, a broad linear range of 1–20 mM, and a detection limit of 0.012 mM for glucose detection, as well as exhibited excellent stability by retaining 99.85 % of its initial photocurrent after nine cycles of intermittent light radiation and showed strong anti-interference ability against common interferents. The biosensor successfully detected glucose concentrations in human serum samples, with results consistent with known values and high recovery rates, confirming its potential for clinical applications [97]. Nevertheless, future studies should focus on expanding the analyte range, evaluating real-time operation, and elucidating the mechanistic role of Au electron bridges.

In addition, Yang et al. designed a dual-signal ratiometric PEC system using poly- (neutral red) (PNR) and ZnIn_2S_4 nanoflowers as cathodic and anodic photoactive materials, respectively, on a single electrode substrate, mitigating common issues like environmental interference and variations in electrode properties. By applying different bias voltages (-0.3 V and 0.06 V), the system effectively separated the responsive (PNR) and reference (ZnIn_2S_4) signals, reducing interference and improving detection accuracy, as represented in Fig. 8E. While the GO_x immobilization on the Au-PNR modified electrode ensured specificity, the set-up achieved a broad linear range of 0.08–30 mM, a LOD of 27 μM , and excellent stability (RSD 0.08 %; retaining 99 % signal over 30 days). It also showed strong selectivity and aligned well with commercial glucose meters in serum testing. Its adaptation for lactate detection confirmed platform versatility [45]. However, the relatively high detection threshold suggests future improvements through alternative photoactive materials and on/off signal strategies.

In this regard, EPGs have demonstrated substantial promise, but critical challenges remain. These include optimizing photoconversion efficiency, ensuring enzyme stability, and broadening the dynamic range in complex matrices. Recent advances in material engineering, nanostructure tuning, and multi-signal strategies are progressively overcoming these barriers, bringing EPGs closer to practical clinical and wearable applications.

3.2. Non-Enzymatic PEC Glucose Sensors (NEPGs)

In response to the limitations of EPGs, recent efforts have focused on developing NEPGs using mimetic nanozymes as photoelectrode materials. These systems preserve the merits of EPGs, such as light-driven operation and high specificity, while offering enhanced stability, lower fabrication costs, and improved robustness under diverse environmental conditions. The sensing mechanism of these systems is based on sequential oxidation and reduction reactions. Upon photon absorption by the photoelectrode, electron-hole pairs (e^-/h^+) are generated. The photogenerated holes oxidize glucose to gluconic acid in the presence of dissolved oxygen, producing hydrogen peroxide (H_2O_2). The H_2O_2 is subsequently reduced to water, facilitating electron transfer and generating a photocurrent signal proportional to the glucose concentration [4]. Thus, the fundamental reactions are summarized as follows:



Despite eliminating the GO_x -related constraints, most NEPGs remain at the proof-of-concept stage and fall short of device-level performance due to challenges such as suboptimal photoconversion efficiency, sluggish charge carrier dynamics, limited light harvesting, and inadequate selectivity in complex biological environments, limiting their practical deployment in clinical or wearable platforms. However,

recent advances, as provided in Table 4, demonstrate significant progress in addressing these bottlenecks through strategic material design, heterojunction engineering, plasmonic enhancement, and microfluidic integration.

For instance, Wang et al. prepared Au- NiO_{1-x} hybrid nanowire arrays (HNAs) through electrodeposition and thermal oxidation process to overcome limitations in electron transfer, nickel instability, and surface poisoning associated with noble metals. The synergistic catalytic interaction between plasmonic Au nanoparticles and partially oxidized NiO_{1-x} enabled efficient glucose oxidation under visible light, as depicted in Fig. 9A. The resulting system achieved a remarkably ultra-high sensitivity of $4061 \mu\text{A cm}^{-2} \text{mM}^{-1}$, an ultralow detection limit of 0.001 mM, and a linear range of 0.005–15 mM, exhibiting a 2.3-fold performance enhancement compared to dark conditions. It demonstrated excellent selectivity toward interfering species, along with good stability (93 % signal retention over 30 days) and high reproducibility. Accurate glucose detection in human serum confirmed its practical viability. This study guides the exploration of metal-semiconductor heterojunctions for surface plasmon-aided glucose sensing [70]. However, precise control of fabrication parameters remains a key area for further optimization.

In another study, a porous BiVO_4 film was synthesized on an FTO substrate via electrochemical deposition and thermal annealing, addressing the poor light absorption and substrate degradation issues associated with TiO_2 -based sensors. The BiVO_4 -based PEC system exhibited dual functionality for glucose detection and solar-to-electricity generation in a photo fuel cell (PFC) configuration, as shown in Fig. 9B. It achieved a sensitivity of $17.38 \mu\text{A cm}^{-2} \text{mM}^{-1}$ within a concentration range of 5–35 mM, and a maximum power density (P_{max}) of $24.9 \mu\text{W cm}^{-2}$. The sensor also demonstrated acceptable selectivity, good stability, and reproducibility [100]. Future improvements should focus on enhancing power output and validating the system's long-term performance for real-world use.

Similarly, Cory et al. electrodeposited a cupric oxide (CuO) thin film on a conductive glass to address the limitations of the low linear range of typical metal oxide (MO)-based sensors. The electrochemical experiments were conducted in a three-electrode set-up, as illustrated in Fig. 9C. The resulting system demonstrated a remarkably wide linear range up to 29 mM, a sensitivity of $263 \mu\text{A cm}^{-2} \text{mM}^{-1}$, and a limit of detection of 0.059 mM. It showed negligible interference from common blood constituents, fast response time (4 s), and excellent reproducibility under both light and dark conditions [87]. Despite these promising results, further investigation into long-term stability, charge transfer mechanisms, and the sensitivity-carrier concentration trade-off is necessary.

In addition, Tao et al. fabricated a $\text{rGO}/\text{NiCo}_2\text{O}_4/\text{ZnO}$ composite grown on nickel foam (NF) through a simple hydrothermal process to address enzyme instability, cost concerns, and limited detection ranges, as described in Fig. 9D. The set-up exhibited dual linear ranges with limited ultra-high sensitivities: $5456.99 \mu\text{A cm}^{-2} \text{mM}^{-1}$ (0.5–483.2 μM) and $4538.2 \mu\text{A cm}^{-2} \text{mM}^{-1}$ (483.2 μM –6.9425 mM), with a low detection limit of 0.5 μM . It showed a fast response time (<3 s), excellent selectivity against common interferents, and strong stability (91.95 % signal retention over 30 days). Reproducibility was also high (RSD = 1.85 % across ten electrodes) [43]. However, further work is required to evaluate clinical applicability, fundamental PEC mechanisms, and scalable manufacturing.

In a more advanced approach, Zhu et al. integrated $\text{CuO-Au}/\text{TiO}_2$ HNTs composite into a custom-designed microfluidic platform, addressing both the environmental sensitivity of enzymatic systems and the performance limitations of existing nonenzymatic sensors. As illustrated in Fig. 9E, the photoelectrode's Z-scheme band alignment enhanced photoelectric conversion, while the microfluidic design improved sample handling and analyte transport. The system achieved a sensitivity of $293.6 \mu\text{A cm}^{-2} \text{mM}^{-1}$, 29 times higher than a non-microfluidic configuration, within the 1–10 mM linear range and a low detection limit of 4.1 μM . It also demonstrated high reproducibility

Table 4

Performance Comparison of Notable NEPGSs.

Photoelectrode Material	Sensitivity ($\mu\text{A cm}^{-2} \text{ mM}^{-1}$)	Linear Range (mM)	Detection Limit (μM)	Photocurrent Retention (%), at 4 °C	Reproducibility (RSD%)	Reference
Au–NiO _{1-x} HNAs/ITO	4061.00	0.005–15.0	1.000	93.00 (30 days)	4.50	[70]
BiOBr-TNTA	n/a	0.0005–30.0	0.01	> 95.00 (4 weeks)	5.60	[51]
BiVO ₄ film/FTO	17.38	5.0–35.00	n/a	n/a	7.64	[100]
AuNPs/ZnO/NF	1133.5	0.001–6.917	0.100	90.67 (20 days)	1.24	[27]
Cu/ZnO Nano-Thorns	63.76	0.1–4.500	3.762	n/a	n/a	[101]
CuO film/FTO	263	29.00	59.00	n/a	n/a	[87]
Cu ₂ O/TiO ₂ /ITO	n/a	1–14	14.400	90.00 (30 days)	4.90	[29]
rGO/NiCo ₂ O ₄ /ZnO/NF	5456.99	0.0002–6.9425	0.500	97.63 (14 days)	1.85	[43]
Fe ₂ TiO ₅ nanodisks	2653	0.004–10	0.588	n/a	n/a	[41]
TiO ₂ /CuO	864.0	1.0–9.000	58.60	n/a	n/a	[102]
CuO-Au@TiO ₂ HNTs	293.6	1–10	4.100	n/a	< 7.50	[103]
Cu ₂ O-Cu-ZnO/GCE	1213.0	0–5.0	2.100	n/a	n/a	[68]
Sn ₃ O ₄ /CdS-X/ITO	n/a	0.0005–40.0	0.067	n/a	n/a	[34]
MXene@NiO-ZnO/FTO	7651.2	0.10–5.000	280.0	n/a	5.21	[104]
3D V ₂ O ₅ /NF	82.00	0.3–8.500	1000	n/a	5.56	[37]
rGO/TiO ₂ /Cu-MOFs/NF	1195.13	0.0003–6.0	0.06	93.10 (4 weeks)	1.89	[105]

*** n/a – Not Available, HNAs – Hierarchical Nanostructured Arrays, NF – Nickel Foam, MOFs – Metal-Organic Frameworks, TNTA – Titanium Dioxide Nanotube Arrays, HNTs – Hierarchical Nanotubes.

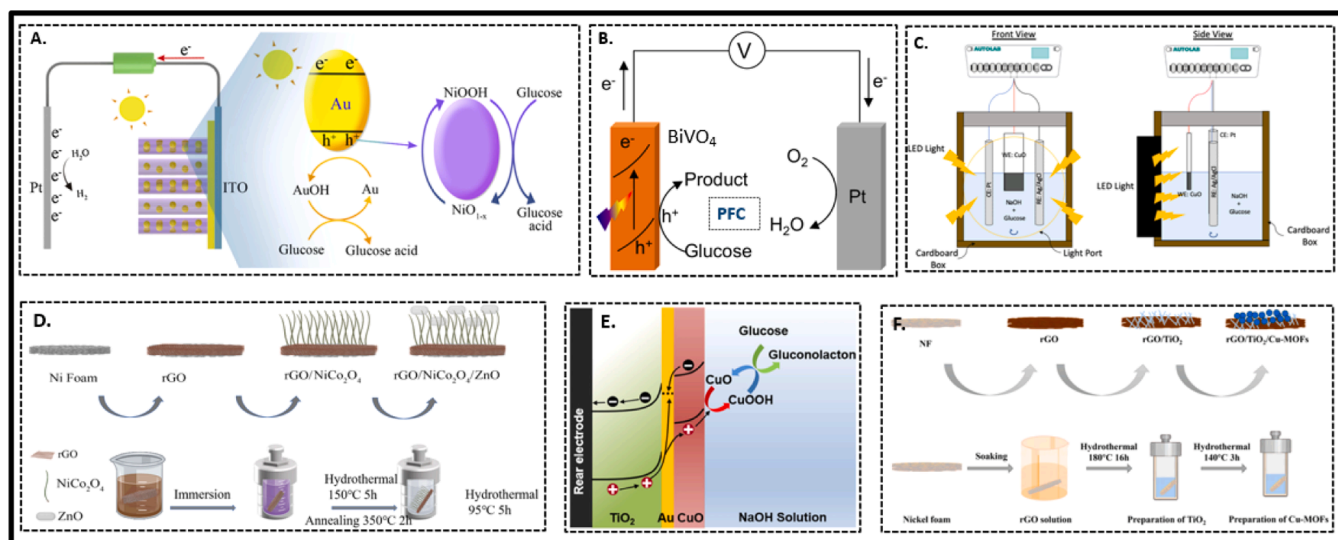


Fig. 9. The schematic illustrations of (A) plasmon-aided glucose oxidation on the Au-NiO_{1-x} HNAs electrode (Adapted from [70]; with permission from Elsevier), (B) PFC constructed by a two-electrode configuration with BiVO₄ photoanode and Pt cathode (Adapted from [100]; with permission from Elsevier), (C) PEC testing procedure of CuO thin film/ITO (Adapted from [87]; with permission from Elsevier), (D) the synthesis of rGO/NiCo₂O₄/ZnO composite (Adapted from [43]; with permission from IEEE), (E) PEC sensing mechanism of the CuO-Au@TiO₂ HNT photoelectrode (Adapted from [103]; with permission from IEEE), (F) synthesis of rGO/TiO₂/Cu-MOFs composite (Adapted from [105]; with permission from Elsevier).

(<7.5 % deviation), strong anti-interference capability (<5 % deviation), and outperformed commercial glucose meters in artificial saliva and sweat [103]. Nonetheless, broader clinical validation and long-term stability testing are needed for practical deployment.

Elsewhere, Tao et al. introduced a dual-analyte detection system using an rGO/TiO₂/Cu-MOFs composite to enable simultaneous sensing of glucose and lactic acid. The combination of conductive rGO, photoactive TiO₂, and redox-active Cu-MOFs allowed for efficient charge transfer and enhanced analyte specificity, as described in Fig. 9F. The system achieved high sensitivities: 1195.13 $\mu\text{A cm}^{-2} \text{ mM}^{-1}$ (glucose) and 1157.77 $\mu\text{A cm}^{-2} \text{ mM}^{-1}$ (lactic acid), with ultralow detection limits of 0.06 μM and 0.03 μM , respectively. It operated within broad linear ranges (0.3–6 mM for glucose; 0.1–12 mM for lactic acid) and maintained strong anti-interference performance against common interferents, along with excellent reproducibility (RSD = 1.89 %) and long-term stability (93.1 % signal retention over one month) [105]. This study represents a significant advancement in multiplexed PEC biosensing.

However, clinical validation with real biological samples and in vivo assessments remains.

Primarily, the development of NEPGSs has advanced significantly through material innovation, device engineering, and integrated sensing platforms. While these systems now demonstrate high sensitivity, broad detection ranges, and improved operational stability, their translation into commercial or clinical devices still requires comprehensive long-term studies, reproducibility across large batches, and rigorous validation with human samples.

3.3. Self-powered Photoelectrochemical Glucose Sensors (SPGSs)

The unveiling of self-powered PEC detection systems has elicited extensive research interest in developing miniaturized, portable, and battery-free visible-light-driven glucose sensors. With the rapid evolution of material science and nanotechnology, in tandem with the Internet of Things (IoT) and artificial intelligence technologies, these

systems are increasingly designed to operate autonomously and wirelessly by harvesting ambient solar light to initiate a PEC reaction for glucose detection. This innovation eliminates over-reliance on external electrical power supplies, facilitating greater integration and extending the applicability of glucose sensors to remote and resource-limited areas. Through the rational design of photoelectrodes, optimization of photocurrent, and integration of supercapacitors, considerable advancements have been made in this area. Similar to EPGs and NEPGs, SPGs remain at the proof-of-concept stage, with reported designs employing either single- or dual-photoelectrode configurations. However, based on the underlying principle of self-generated power, these systems are categorized into two distinct types: photocatalytic fuel cell (PFC)-based and photoelectrochemical (PEC) cell-based glucose sensors.

3.3.1. Photocatalytic Fuel Cell (PFC)-based SPGs

PFC-based SPGs have emerged as dual-functional systems capable of simultaneously harvesting solar and biomass-derived chemical energy while enabling glucose detection without external bias. This integrated functionality represents a sustainable and promising strategy for real-time glucose monitoring. A typical PFC-SPG comprises a photoanode for glucose oxidation, a cathode for oxygen reduction, an electrolyte containing glucose as fuel, and a light source to drive PEC reactions. Although the number of studies in this area remains limited, however, as shown in Table 5, the technological advancements achieved are significant.

In 2016, Zhang and colleagues pioneered the first visible-light-driven, enzyme-free PFC-based SPG, addressing key limitations of enzymatic systems such as poor stability and specificity. As shown in Fig. 10A, the dual-chamber system employed a Ni (OH)₂/CdS/TiO₂ photoanode, capitalizing on the synergistic photocatalytic and electrocatalytic properties of CdS quantum dots and Ni (OH)₂, and a hemin-graphene-modified cathode to enhance H₂O₂ reduction. The system achieved a detection limit of 5.3 μ M, a linear range of 0.01–0.50 mM, and a maximum power density of 10.5 μ W cm⁻². It also demonstrated long-term operational stability (94.5 % photocurrent retention over 40 days), excellent reproducibility, and selectivity against common interferents [106]. Nonetheless, the system faced limitations typical of PFCs, including low power output, limited analyte specificity due to the absence of molecular recognition elements, and sensitivity to pH-induced chemical fluctuations. These challenges underscore the need for improved output efficiency, integration of selective sensing components, and enhanced environmental robustness.

Building on this, Yang et al. developed a dual-function glucose–air PFC using a Ni (OH)₂/TiO₂ photoanode (Fig. 10B) that enabled visible-light-driven glucose sensing and self-powered operation. Ni (OH)₂ enhanced the system's activity by promoting charge separation in TiO₂ and providing redox-active sites for glucose oxidation via the Ni (OH)₂/NiOOH cycle. This system achieved a high sensitivity of 163.5 μ A mM⁻¹ cm⁻², a low detection limit of 0.19 μ M, and a linear range of 0.5–20 μ M. As a PFC-SPG, it generated output proportional to the logarithm of glucose concentrations (5–100 μ M) and retained 93.2 % of its photocurrent over two weeks [107]. However, overloading

Ni (OH)₂ impeded TiO₂ light absorption, indicating the need for optimized cocatalyst deposition. Furthermore, although superior to enzymatic systems in cost and durability, its performance should be more comprehensively benchmarked against other non-enzymatic systems. The device's reliance on alkaline conditions (0.1 M NaOH) and lack of real-world testing limit its immediate clinical applicability.

He et al. later introduced a PFC-SPG based on hematite nanorods (Fe₂O₃NRs), achieving dual functionality in solar energy harvesting and glucose detection (Fig. 10C). The system exhibited a sensitivity of 100.46 μ A cm⁻² mM⁻¹, a linear range of 0.2–2.0 mM, and good selectivity against common interferents [109]. Integrated into a PFC with a platinum cathode, it delivered a maximum power density of 8.0 μ W cm⁻² and a linear output response of 3.53 μ W cm⁻² mM⁻¹ across 0.5–2.5 mM glucose. While Fe₂O₃ is cost-effective and active under visible light, the photoanode suffers from limited sensitivity in neutral media due to photochemical instability. This constrains its usability to alkaline environments and raises concerns about long-term stability, large-scale fabrication, and cost-efficiency in practical applications.

To overcome some of these challenges, He et al. later incorporated Ni (OH)₂ into Fe₂O₃NRs to enhance charge injection and catalytic efficiency. The resulting Ni (OH)₂/Fe₂O₃ photoanode, paired with a Pt cathode, formed a PFC capable of converting solar and chemical energy via glucose oxidation and oxygen reduction (Fig. 10D). This configuration demonstrated a narrow detection range (0.05–0.25 mM), high sensitivity (24.59 μ W cm⁻² mM⁻¹), and a maximum power density of 16.7 μ W cm⁻², markedly superior to that of bare Fe₂O₃ (3.53 μ W cm⁻² mM⁻¹). The system also showed excellent selectivity, high recovery in real-sample analysis, and 95 % signal retention over two weeks [110]. Nonetheless, the narrow detection range, limited long-term data, and performance attenuation at high Ni (OH)₂ loadings highlight the need for further optimization. Moreover, validation in complex biological matrices remains outstanding.

More recently, Li et al. introduced a dual-photoelectrode PFC system comprising a polyaniline (PANI)-coated p-type SiNW photocathode and an In₂S₃-modified n-type SiNW photoanode to form an S-scheme heterojunction (Fig. 10E). This structure improved charge separation and interfacial transfer, yielding an open-circuit voltage of 0.83 V, a high-power density of 163.01 μ W cm⁻², a broad linear detection range of 0–50 mM, and a low detection limit of 0.998 μ M [49]. The system also exhibited good selectivity and stable operation over 10 days, with only a 10 % signal decline. Despite these advances, typical limitations persist, including incomplete fuel oxidation, moderate energy conversion efficiency, and vulnerability of SiNWs to photo-corrosion. While surface modification with PANI and In₂S₃ offers some protection, further strategies are needed to ensure long-term durability under real-world conditions.

In conclusion, PFC-based SPGs represent an innovative platform for sustainable and bias-free glucose sensing by combining energy conversion with biosensing. Continued progress hinges on enhancing power output, expanding detection ranges, improving selectivity through molecular recognition, and ensuring stability in complex environments.

Table 5
Performance Comparison of Notable PFC-based SPGs.

Photoelectrode Material	Sensitivity (μ A cm ⁻² mM ⁻¹)	Linear Range (mM)	Detection Limit (μ M)	Maximum Power Density (P _{max}) (μ W cm ⁻²)	Photocurrent Retention (%), 4 °C	Reference
Ni (OH) ₂ /CdS/TiO ₂ /FTO	n/a	0.01–0.500	5.300	10.500	94.5 (40 days)	[106]
Ni (OH) ₂ /TiO ₂ /FTO	163.50	0.0005–0.020	0.19	3.600	93.2 (2 weeks)	[107]
TiO ₂ -NRA/FTO	201.50	0.01–0.200	n/a	n/a	n/a	[108]
Fe ₂ O ₃ -NR/FTO	100.46	0.20–2.000	5.500	8.000	98.0 (half month)	[109]
Ni (OH) ₂ /Fe ₂ O ₃ -NR/FTO	n/a	0.05–0.250	9.000	16.700	95.0 (half month)	[110]
p-SiNWs/PANI	n/a	0–50.000	0.998	163.010	90.0 (10 days)	[49]
n-SiNWs/In ₂ S ₃						

***n/a – not available, NR-Nanorod, NW- Nano-walls, PANI – Polyaniline, NRA -nanorod array.

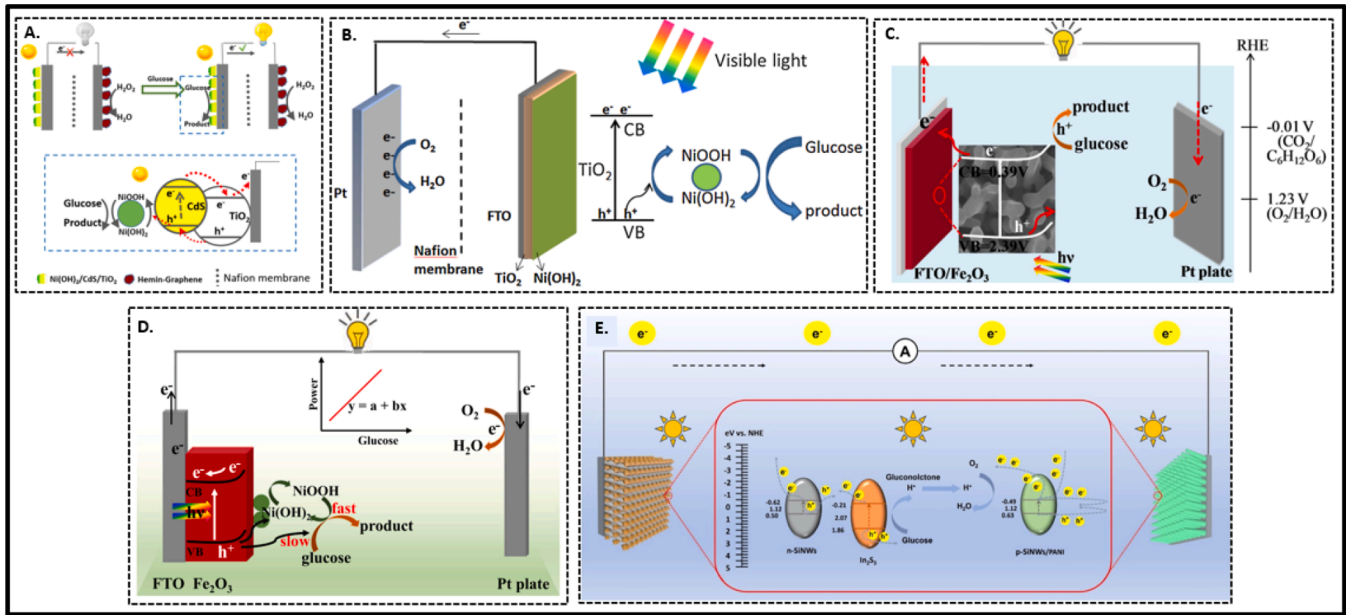


Fig. 10. Schematic illustrations of the mechanisms of PFC-based SPGs based on (A) Ni (OH)₂/Cds/TiO₂ photoanode and hemin-graphene (HG) cathode (Adapted from [106]; with permission from American Chemical Society), (B) Ni(OH)₂/TiO₂ photoanode (Adapted from [107]; with permission from Elsevier), (C) Fe₂O₃NR/FTO photoanode (Adapted from [109]; with permission from Elsevier), (D) Ni (OH)₂/Fe₂O₃/FTO photoanode (Adapted from [110]; with permission from Elsevier), (E) n-SiNWs/In₂S₃ photoanode and p-SiNWs/PANI photocathode (Adapted from [49]; with permission from Elsevier).

Addressing these challenges will be pivotal in transitioning these technologies from laboratory prototypes to practical, deployable biosensors.

3.3.2. PEC cell-based SPGs

Unlike PFC-SPGs, which depend on dual-chamber redox reactions and often require sacrificial reagents or external load resistance, PEC cell-based SPGs operate solely under light illumination. These systems utilize a photoelectrode that drives glucose oxidation upon irradiation, generating a photocurrent proportional to the glucose concentration. This self-sustained approach eliminates the need for an external power source, effectively integrating energy harvesting and glucose sensing into a compact, real-time platform. Encouragingly, recent advances in photoelectrode engineering, nanostructure optimization, and system design have markedly improved the performance of PEC-SPGs; nevertheless, similar to other configurations, they remain predominantly at the laboratory scale, as summarized in Table 6.

For instance, Han et al. introduced a cathodic PGS based on a 0D/2D/3D hybrid heterostructure comprising Au nanoparticles, NiO nanosheet arrays, and BiOI nanoflowers (Fig. 11A). This multi-dimensional architecture synergistically improved light absorption, charge

separation, and surface plasmon resonance effects. Glucose quantification was achieved via photocurrent suppression resulting from oxygen consumption catalyzed by glucose oxidase (GO_x). The device demonstrated a wide detection range (0.0001–50.00 mM), a low detection limit (0.0871 μM), high selectivity, and good short-term reproducibility [111]. Nevertheless, it was only validated in diluted glucose solutions, limiting its relevance for real biological environments. Moreover, enzyme stability, material degradation, and fabrication scalability were not addressed, raising concerns about durability and the feasibility of large-scale applications. Environmental and biosafety issues related to BiOI, NiO, and enzyme immobilization also remain unexplored.

Wang et al. developed a light-driven, non-enzymatic PGS based on AuNi_{0.7} nanodendrites (Fig. 11B). The nanodendritic morphology offered a large active surface area and efficient charge separation under visible light, supported by the plasmonic properties of Au and the catalytic activity of Ni. Operating without external power, the system achieved a high sensitivity (~183 μA cm⁻² mM⁻¹), a wide linear range (0.1–18 mM), and a low detection limit (0.01 mM), with 88 % of the photocurrent retained after 30 days [115]. Despite these strengths, its performance under dark conditions was limited (linear range:

Table 6

Performance Comparison of Notable PEC-SPGs.

Photoelectrode Material	Sensor Type	Sensitivity (μA cm ⁻² mM ⁻¹)	Linear Range (mM)	Detection Limit (μM)	Photocurrent Retention (%)	Reproducibility (RSD %)	Reference
Au NPs@BiOI NFs/NiO NSAs	Enzymatic	n/a	0.0001–50.00	0.0871	n/a	n/a	[111]
NiO NSS/RGO/BiOI/ITO	Enzymatic	n/a	0.0001–100.0	0.047	n/a	4.1	[112]
Al/TiO ₂ /Au NPs/GO _x	Enzymatic	3780	0.0003–0.001	0.050	92 (30 days)	5.0	[113]
CuO@TiO ₂ HNT	Non-enzymatic	138.9	0.001–0.20	0.700	n/a	n/a	[114]
AuNi/ITO	Non-enzymatic	183.0	0.10–18.00	10.00	88 (30 days)	2.8	[115]
n-BiVO ₄ /FTO	Enzymatic	n/a	0.500–20.00	n/a	n/a	n/a	[116]
p-Cu ₂ O@TiO ₂ /Cu	Enzymatic	n/a	0.500–20.00	n/a	n/a	n/a	[116]
ERGO/ZnONWs/CdS QDs/FTO	Non-enzymatic	430.06	0.01–1.00	1.100	95.23 (10 days)	0.467	[117]
p-Ti ₃ C ₂ /Cu ₂ O/FTO	Enzymatic	n/a	0.5–4.50	0.079	n/a	n/a	[50]
n-Ti ₃ C ₂ /Bi ₂ S ₃ /FTO	Enzymatic	n/a	0.5–4.50	0.079	n/a	n/a	[50]

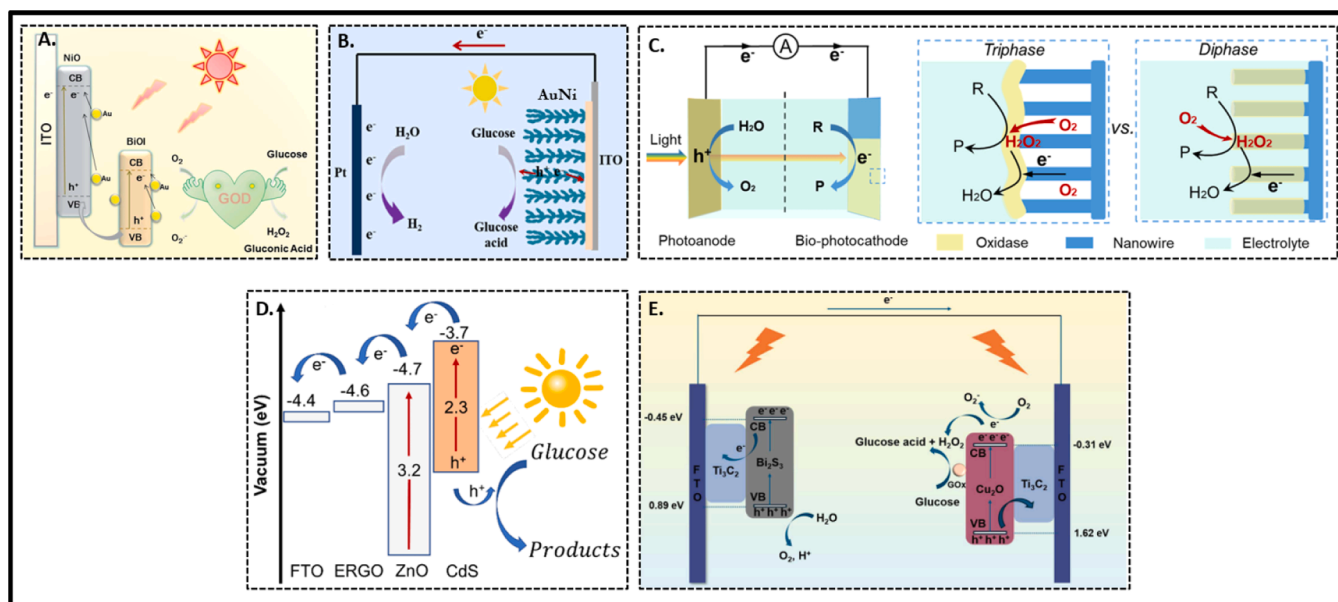


Fig. 11. Schematic illustrations of the mechanism of PEC-SPGS based on (A) Au NPs/BiOI/NiO electrode (Adapted from [111]; with permission from the Royal Society of Chemistry), (B) AuNi nanodendrites (Adapted from [115]; with permission from the Royal Society of Chemistry), (C) BiVO₄ photoanode and Cu₂O@TiO₂ nanowire arrays biophotocathode (Adapted from [116]; with permission from Elsevier), (D) ERGO/ZnONWs/CdS photoanode (Adapted from [117]; with permission from Elsevier), (E) Ti₃C₂/Cu₂O (photocathode) and Ti₃C₂/Bi₂S₃ (photoanode) dual-photoelectrode system (Adapted from [50]; with permission from the American Chemical Society).

0.1–13 mM), and its overall analytical capability, although strong, did not surpass that of the best-reported sensors. Furthermore, the lack of testing in real biological samples limits insight into its practical deployment. Robustness under variable environmental conditions also requires further investigation.

Another group introduced a self-powered PEC bioassay integrating a nanoporous BiVO₄ photoanode and a Cu₂O@TiO₂ nanowire biophotocathode featuring a solid–liquid–air triphase interface (Fig. 11C). This interface significantly enhanced oxygen transport to the enzyme-active sites, improving oxidase kinetics and extending the glucose detection range to 0.5–20 mM, twentyfold broader than traditional diphasic systems. Efficient band alignment between BiVO₄ and Cu₂O facilitated charge separation and improved photocurrent generation. Furthermore, localized enzymatic reactions at the cathode improved anti-interference performance [116]. Despite its innovation, the sensor's sensitivity at low glucose concentrations remains suboptimal for clinical diagnostics. In addition, long-term stability, performance in complex matrices, and scalability of the tri-phase architecture were not evaluated, limiting the current system's readiness for translational applications.

Furthermore, Gur et al. reported a cost-effective, environmentally conscious non-enzymatic PGS using a 3D ERGO/ZnONW/CdS QD photoanode (Fig. 11D). The hierarchical structure offered high surface area, enhanced light absorption, and improved charge transport. Under simulated sunlight, the device delivered strong PEC performance, with high photocurrent density (1150 $\mu\text{A cm}^{-2}$), a wide detection range (0.01–15.00 mM), high sensitivity, and a low detection limit (1.1 μM). It also exhibited good selectivity, reproducibility, and stability, indicating potential for practical, enzyme-free glucose sensing [117]. However, the use of cadmium-based QDs introduces toxicity and environmental concerns, restricting biocompatibility. Moreover, the system's long-term stability under physiological conditions and fabrication reproducibility across batches remain to be verified. Future studies should focus on replacing toxic materials with eco-friendly alternatives and validating performance in real biological fluids.

Recently, Gao et al. developed a SPGS employing a dual-photoelectrode configuration, Ti₃C₂/Bi₂S₃ as the photoanode and

Ti₃C₂/Cu₂O as the photocathode (Fig. 11E). This design exploits the Fermi level difference between electrodes for bias-free charge transport. Ti₃C₂ facilitated both electron and hole transport, while promoting efficient charge separation. Upon illumination, photoinduced carriers were transferred through an external circuit, enhancing photocurrent generation. Glucose detection was enabled via GO_x functionalization, yielding a linear detection range of 0.5–4.5 mM and a detection limit of 0.0790 mM, along with good selectivity and operational stability [50]. Nonetheless, the relatively narrow linear range may limit adaptability in fluctuating glucose environments. Further improvements in sensitivity, replacement of Cu₂O with more efficient photocathode materials, and enhanced photoanode stability are needed. Additionally, real-sample validation is essential for clinical translation.

Generally, PEC cell-based SPGSs offer a compelling route to self-powered, miniaturized glucose sensors, merging light harvesting and biosensing into a unified platform. While recent studies have made notable strides in sensitivity, stability, and device architecture, practical deployment still faces hurdles, including biological matrix validation, operational robustness, material safety, and scalable fabrication. Future research should prioritize eco-friendly materials, real-world testing, and modular system integration to fully realize the clinical and portable potential of PEC-SPGSs.

n/a – not available, NP-Nanoparticles, ERGO- electrochemically reduced graphene oxide, QDs – Quantum dots, NW- Nano-walls, NFs – Nanoflakes, NRA -nanorod array, NSA- nanosheet arrays, HNT - Hierarchical nanotube.

4. High-ranking PEC Glucose Sensors (PGSs)

As detailed in the preceding sections, the field of PGSs, comprising enzymatic, non-enzymatic, and self-powered systems, has undergone significant evolution, driven by innovations in material science, nanostructure engineering, and device architecture. While each category presents distinct operational principles and design considerations, comparative analysis across key analytical performance metrics, namely sensitivity, linear detection range, and detection limit, offers critical insights into their relative strengths and potential for clinical or

environmental deployment. Equally important are attributes such as selectivity, response time, reproducibility, and long-term operational stability, which determine their practical viability in real-world bio-sensing applications. This section highlights material designs that demonstrate superior performance within each category, based on key analytical metrics, including their underlying mechanisms, schematically illustrated in Fig. 12.

Within the EPGs category presented in Table 3, three systems stand out for their strategic use of photoelectrode materials and tailored nanostructures to enhance analytical performance. Yang et al. engineered TiO_2 nanotubes (NTs) functionalized with Au and Pt nanoparticles (Fig. 12A), leveraging plasmonic and catalytic synergies to achieve an impressive sensitivity of $81.93 \mu\text{A mM}^{-1} \text{cm}^{-2}$ and a low detection limit of $1.39 \mu\text{M}$. Although its linear range (0–0.8 mM) is narrow, it offers exceptional selectivity and operational stability, making it ideal for low-concentration glucose detection [98]. In contrast, Wang et al. addressed clinical applicability through a $\text{TiO}_2/\text{SrTiO}_3/\text{PDA}$ heterostructure (Fig. 12B) that delivered a remarkably broad linear range of 0–32 mM, suitable for both normoglycemic and hyperglycemic monitoring. While the sensitivity ($5.37 \mu\text{A mM}^{-1} \text{cm}^{-2}$) is low, the system's robustness, anti-interference capacity, and serum compatibility underline its practical promise [44]. Complementing these advances, Li et al. proposed a $\text{TiO}_2/\text{rGO}/\text{PDA}$ heterostructure (Fig. 12C), incorporating a Z-scheme mechanism and co-exposed crystal facets, which significantly boosted charge separation and enzyme retention. This design achieved an ultra-low detection limit of $0.034 \mu\text{M}$ and a sensitivity of $13.82 \mu\text{A mM}^{-1} \text{cm}^{-2}$ across a 0–3 mM range [57]. Together, these three systems highlight the evolving design strategies in EPGs,

from plasmonic enhancement and heterojunction formation to Z-scheme integration, each optimizing different facets of sensor performance. Yet, they also share common limitations, such as narrow detection ranges or concerns about long-term stability, pointing to future directions in material optimization, enzyme-free systems, and real-world validation.

Likewise, in the NEPGs category as provided in Table 4, a clear progression in material engineering and sensor architecture has enabled outstanding analytical performance, with each design uniquely optimizing sensitivity, linear range, and detection limit. Hilal et al. introduced a breakthrough MXene@NiO-ZnO (NMZ) composite (Fig. 12D), achieving an exceptional sensitivity of $7651.2 \mu\text{A mM}^{-1} \text{cm}^{-2}$ and a rapid 4-second response time. The synergy between MXene's high conductivity and the NiO/ZnO heterojunction's internal electric field enhanced charge separation and catalytic efficiency, positioning this architecture as a frontrunner despite pending investigations into its long-term stability and performance in biological media [104]. Similarly, Li et al. developed a $\text{Sn}_3\text{O}_4/\text{CdS}$ composite (Fig. 12E) with an ultra-broad linear range (0.5 μM –40 mM) and a low detection limit of $0.067 \mu\text{M}$. Its broad visible-light absorption and efficient charge separation allowed for high selectivity and reproducibility in real sample analysis, although concerns around CdS toxicity and environmental robustness remain [34]. Complementing these, Wu et al. designed a BiOBr-modified TiO_2 nanotube array (BiOBr-TNTA) composite (Fig. 12F), achieving a remarkably low detection limit of 10 nM and a broad linear range up to 30 mM. Its enhanced charge transport and stability in serum samples demonstrated strong clinical potential, though its reliance on highly alkaline conditions may restrict biocompatibility [51]. Collectively, these NEPGs reflect a dynamic evolution in photoelectrode material design, ranging

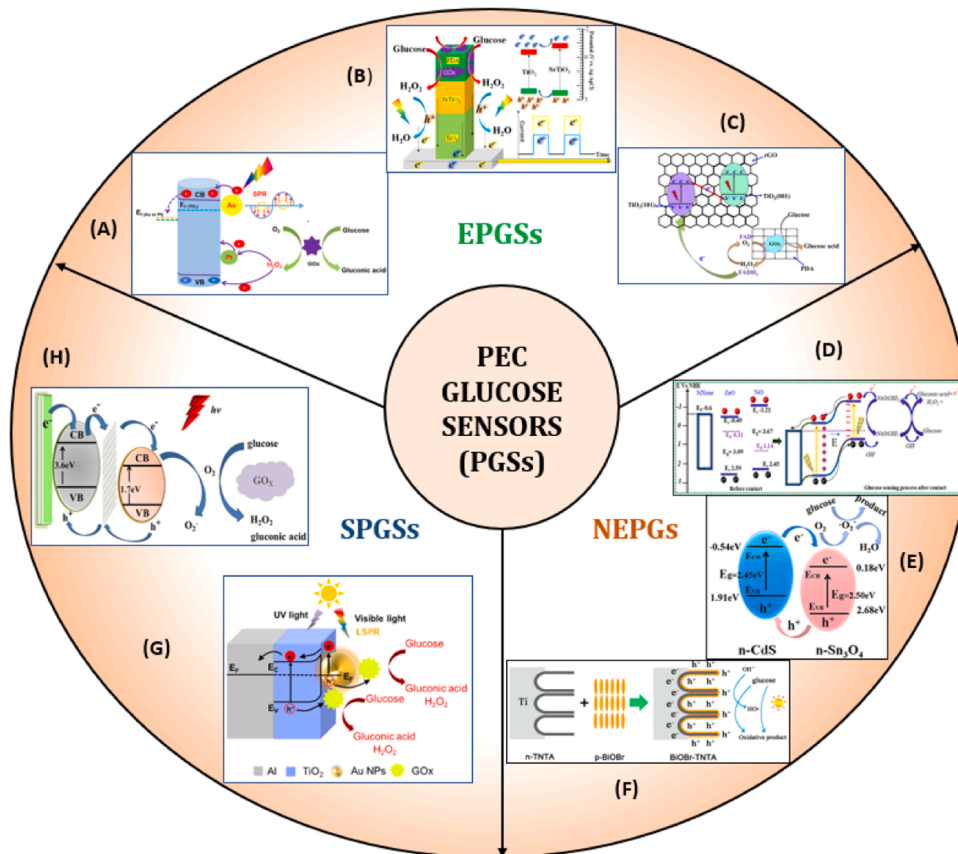


Fig. 12. Schematic illustrations of the mechanisms of the high-ranking PGSSs based on (A) TiO_2 NTs/Au/Pt/ GO_x (Adapted from [98]; with permission from the American Chemical Society), (B) $\text{TiO}_2/\text{SrTiO}_3/\text{PDA}/\text{GO}_x$ (Adapted from [44]; with permission from Elsevier), (C) $\text{TiO}_2/\text{rGO}/\text{PDA}/\text{GO}_x$ (Adapted from [57]; with permission from the American Institute of Physics (AIP)), (D) MXene@NiO-ZnO (Adapted from [104]; with permission from Elsevier), (E) $\text{Sn}_3\text{O}_4/\text{CdS}$ -X (Adapted from [34]; with permission from Elsevier), (F) BiOBr-TNTA (Adapted from [51]; with permission from Elsevier), (G) Al/ TiO_2 /AuNPs/ GO_x (Adapted from [113]; with permission from the American Chemical Society), (H) NiO/RGO/BiOI (Adapted from [112]; with permission from Elsevier).

from 2D conductive substrates and Z-scheme composites to heterojunction-enhanced nanotube arrays, each tailored to amplify photoelectrochemical activity while confronting the ongoing challenges of biocompatibility, operational durability, and real-world adaptability.

The most advanced SPGSs showcased in Table 6 exemplify the strategic integration of photoelectrode materials and catalytic interfaces to deliver high-performance, bias-free glucose detection. Yu et al. introduced a GO_x-functionalized Schottky junction built on Au-decorated TiO₂ films supported by Al substrates (Fig. 12G), leveraging an optical resonance cavity effect and slight Al doping to enhance light harvesting and charge separation. The inclusion of Au nanoparticles extended spectral responsiveness into the visible–NIR region, while the immobilized GO_x ensured enzymatic specificity. This elegant configuration yielded a high sensitivity of 3780 $\mu\text{A mM}^{-1} \text{cm}^{-2}$, a broad linear range of 0.3 μM to 10 mM, and an ultralow detection limit of 0.05 μM , all achieved at or near zero bias [113]. In parallel, Wang et al. developed an ITO/NiO/RGO/BiOI heterostructure featuring a multilayered architecture with dual catalytic behavior: BiOI's catalase-like activity coupled with GO_x enzymatic action (Fig. 12H). This synergy enabled the system to achieve an ultra-wide linear detection range (100 nM to 100 mM) and an exceptionally low detection limit of 47 nM, along with high reproducibility and successful application in injectable glucose solutions [112]. Both systems demonstrate excellent analytical capabilities, with clear advantages in sensitivity, stability, and anti-interference characteristics. However, their dependence on enzymatic catalysis raises concerns about long-term durability under physiological stress, while complex electrode designs may challenge scalability. Future directions should prioritize replacing enzymatic components with robust non-enzymatic catalysts, enhancing material biocompatibility, and simplifying architectures for integration into portable, wearable, or implantable diagnostic platforms.

Interestingly, the current landscape of PGSSs showcases a diverse range of high-performing systems, each excelling in specific domains, be it sensitivity, detection range, or detection limit. EPGSSs benefit from enzymatic selectivity and tailored nanostructures, NEPGSSs capitalize on material-driven catalysis for enhanced stability and broader applicability, and SPGSs offer low-power solutions suitable for portable diagnostics. However, no single material design meets all the ideal criteria. Future progress lies in combining the strengths of each system, integrating robust, earth-abundant materials, non-enzymatic catalysis, simplified architectures, and real-sample validation, to develop PGSSs that are not only analytically superior but also clinically and commercially viable.

5. Application of computational tools and Machine Learning (ML) models

While experimental methods have significantly driven the development of PGSSs, the integration of computational tools such as density functional theory (DFT) and machine learning (ML) models is becoming increasingly important for rational material and device design, as shown in Fig. 13. In particular, DFT calculations, conducted using software like the Vienna Ab initio Simulation Package (VASP) and Quantum ESPRESSO, provide atomic-scale insights into key material properties, including band alignment, charge carrier dynamics, adsorption energies, and surface reactivity. Although direct applications of DFT in PGS remain limited, however, related studies offer valuable insights that can inform sensor design. For instance, DFT simulations have elucidated the charge transfer mechanisms in CdSe/TiO₂ NTs heterojunctions, revealing efficient electron migration from CdSe to TiO₂ and hole-driven glucose oxidation, which correlates with a low detection limit of 3.1 μM and an enhanced PEC response under visible light [118]. Similarly, DFT-based studies of a 3D CuO–TiO₂ heterojunction revealed strong interfacial adsorption energies (−8.34 eV) and favorable band alignments, resulting in effective electron–hole separation and enhanced photocatalytic activity [28]. In PEC water splitting, DFT analysis demonstrated that CuO coupling lowers the overpotential of WO₃ from 0.82 V to 0.63 V, enhancing oxygen evolution kinetics through interface-induced charge redistribution [30]. Moreover, co-doping strategies such as La–Fe or La–Ni in SrTiO₃ systems have been computationally shown to fine-tune the band structure for optimal redox alignment, extend light absorption, and increase carrier lifetimes, offering valuable insights for photocatalyst engineering [119]. In parallel, the rapid emergence of data-driven technologies has positioned ML as an indispensable tool in sensor development. Recent studies show that ML algorithms can significantly enhance the physicochemical properties of carbon dots (CDs), such as fluorescence, stability, and electrocatalytic activity, while optimizing synthesis routes. Furthermore, ML facilitates the screening of carbon dot precursors and the prediction of their resulting properties, thereby accelerating the development of high-performance CQDs tailored for specific applications. This approach not only improves material efficiency but also holds significant promise for broadening the scope of PEC biosensors in environmental and biomedical diagnostics [120]. Complementarily, Giordano et al. highlighted the transformative potential of supervised ML models trained on large datasets generated from electrochemical biosensors. These models have been shown to improve sensitivity, reproducibility, and accuracy, and enable simultaneous multi-analyte detection via multi-output regression, despite challenges such as electrode fouling, low signal-to-noise ratios, chemical interferences, and complex matrix effects. By minimizing the reliance on extensive experimental protocols,

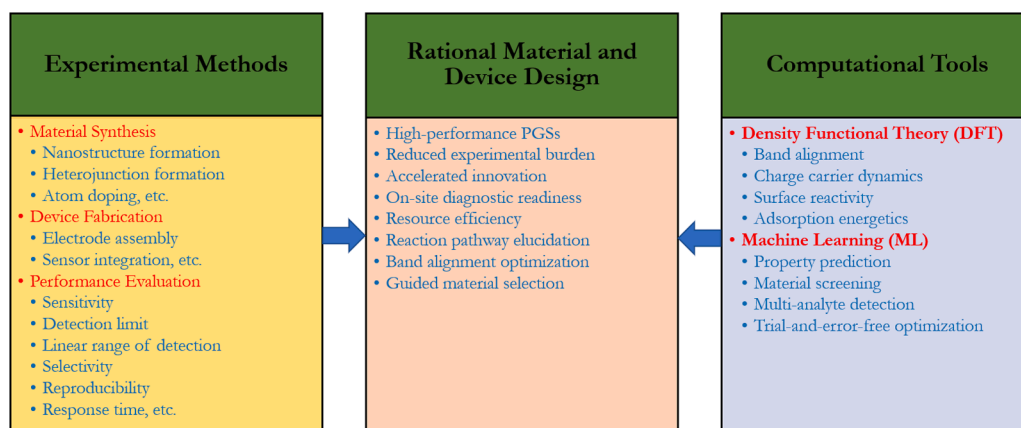


Fig. 13. Experimental-computational synergy for high-performance PGSSs development.

ML facilitates more efficient, reliable, and practical sensor platforms, thereby accelerating the translation of sensing technologies into real-world, on-site applications [121]. Collectively, these applications demonstrate the untapped potential of DFT and ML in guiding material selection and system-level optimization for high-performance PEC glucose sensing.

6. Conclusion, challenges, and future perspectives

The field of PGSSs has significantly evolved over the past decade, driven by continuous advancements in materials science and nanotechnology. EPGSSs have benefited from the high selectivity of GO_x immobilized on tailored photoelectrodes, enabling sensitive glucose detection under light illumination. However, their practical deployment is hindered by issues related to enzyme instability and limited operational lifespan. To address these limitations, NEPGSSs have emerged, leveraging the photocatalytic properties of mimetic nanomaterials and engineered heterostructures to achieve enhanced stability, simplified design, and improved reproducibility. In parallel, SPGSSs, particularly those based on PEC and PFC configurations, represent a promising paradigm shift, integrating energy harvesting with biosensing for autonomous, bias-free operation. Despite these promising developments, several critical challenges remain unresolved. Many current systems still suffer from limited sensitivity, narrow linear detection ranges, high detection limits, inadequate selectivity in complex biological environments, and low power output. In enzyme-based systems, preserving enzymatic activity and immobilization stability over time continues to pose significant obstacles. In non-enzymatic platforms, achieving enzyme-like selectivity without sacrificing sensitivity remains a major hurdle. Moreover, complex fabrication techniques, poor scalability, and concerns over material toxicity further limit clinical translation and environmental sustainability. The scalability and reproducibility of advanced photoelectrode architectures, especially dual-photoelectrode configurations, are also critical concerns. These architectures, while capable of significantly enhancing photocurrent responses and selectivity, often require precise nanofabrication methods that are not yet feasible at an industrial scale. Additionally, the real-world applicability of PGSSs remains under-explored, with limited studies validating sensor performance in physiological fluids, continuous monitoring conditions, or real-time operation. Importantly, certain relevant data remain underreported, such as quantitative kinetic parameters (reaction rate constants and apparent activation energies) for enzyme versus non-enzyme systems, and energy output metrics, particularly photoelectric conversion efficiency (PCE) and maximum power density, which are available mainly for PFC-based SPGSSs but are scarce for purely PEC-driven designs. Systematic inclusion of these metrics would enable more rigorous benchmarking and better assessment of device-level feasibility. Looking ahead, a more integrated and interdisciplinary approach is essential. The rational design of high-performance photoelectrodes should prioritize earth-abundant, environmentally benign materials that simultaneously offer broad-spectrum absorption, efficient charge separation, and robust catalytic activity. Strategies such as single-atom doping, heterojunction engineering, and the development of biomimetic catalytic sites show significant promise in this regard. Also, future studies should place more emphasis on quantitatively characterizing surface reaction kinetics. Moreover, dual-photoelectrode systems should continue to be explored as a means of overcoming the inherent limitations of single-photoelectrode configurations. The convergence of experimental methods with computational tools, particularly density functional theory (DFT) and machine learning (ML), presents a transformative opportunity for the PEC sensing field. DFT simulations can provide atomistic insights into band alignment, charge transfer dynamics, and surface-analyte interactions, guiding the intelligent design of materials with tailored functionalities. ML algorithms, on the other hand, can accelerate material screening, optimize synthesis conditions, and enable advanced data interpretation for real-

time biosensing. Furthermore, integrating PGSSs with emerging technologies such as microfluidics, wireless data transmission, 3D printing, and artificial intelligence can lead to the development of portable, smart, and user-friendly sensing platforms suitable for personalized healthcare. Emphasis should also be placed on establishing standardized protocols for sensor evaluation in biological matrices and conducting long-term operational stability assessments under physiologically relevant conditions. In conclusion, while current PGSSs fall short of fulfilling all ideal performance criteria, the collective advancements in material innovation, device architecture, and data-driven design chart a clear path toward their future realization. Continued interdisciplinary collaboration will be pivotal in translating PEC sensing technologies from laboratory prototypes into reliable, commercially viable, and clinically impactful tools for diabetes monitoring and broader biomedical applications.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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