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ORIGINAL PAPER

# Enhanced Electrocatalytic Performance of Eco-friendly Nanomaterial-Based Sensors for the Analysis of Pharmaceutically Important Compounds

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**Abstract**
Green synthesis is a sustainable alternative to traditional chemical methods for nanomaterial-based sensors because it is more affordable, scalable, and does not involve any harmful contaminants when using green materials' extracts as stabilizing and reducing agents for nanoparticle synthesis. The green-synthesized nanoparticles are extremely attractive for various pharmaceutical applications. This review article examines the most preferred eco-friendly nanomaterials, their synthesis and characterization, and their pharmaceutical applications based on the selected studies conducted in the last five years. It concludes that the green synthesis methods allow the transformation of metals into nanoparticles or green materials that act as precursors to carbon-based nanomaterials. The nanoscale materials obtained through green synthesis methods contribute to low toxic, environmentally benign, easy, and low-cost sensing and enhanced electrocatalytic performance.

**Keywords** Eco-friendly Nanomaterials · Electrochemistry · Electrocatalytic Effect · Green Synthesis · Pharmaceuticals

## 1 Introduction

The development of pharmaceutical compounds has revolutionized human health; therefore, the pharmaceutical industry is one of the fastest-expanding industries worldwide [1]. Numerous pharmaceutical compounds, including contraceptives, analgesics, non-steroidal anti-inflammatory drugs, antibiotics, lipid regulators, beta-blockers, and neuroactive compounds, are utilized in human medicine [1, 2]. Considering this great diversity, it is very significant to analyze bulk drug compounds, their intermediates, their degradation products, drug formulations, drug products, impurities, and biological samples involving the drugs and their metabolites [3]. Furthermore, pharmaceutical compounds and their metabolites have been considered "pseudo-persistent" because of their frequent usage and release. They are often monitored in environmental mediums such as wastewater, sediment, and biota. Because of their toxic properties and bioaccumulation in living organisms, they can induce perilous impacts on humans, animals, and aquatic organisms [4, 5].

The effectiveness of pharmaceutical compounds is contingent on their purity and proper utilization. To ensure this, various analytical and instrumental techniques have been developed for drug determination [3]. A key consideration in developing these techniques for low concentrations of pharmaceutical compounds is the chemistry of the target analytes, the ability to separate them from interfering substances, and the technique's sensitivity [6]. Common methods for qualifying and quantifying pharmaceutical compounds include gas chromatography (GC) [1, 7], high-performance liquid chromatography (HPLC) [8–11], thin layer chromatography (TLC) [12, 13], spectrofluorimetry [14, 15], colorimetry [16], ultraviolet-visible spectrophotometry

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(UV-Vis) [17, 18], nuclear magnetic resonance (NMR) [19, 20], Fourier Transform Infrared spectroscopy (FTIR) [21], capillary electrophoresis [22], fluorescence spectrophotometry [23], gravimetry [24], Raman spectroscopy [25], and electrochemistry [17, 26, 27].

When the recent literature is examined, it is seen that among the techniques mentioned above, chromatographic methods are frequently applied in determining pharmaceutical compounds. The chromatographic methods could supply the simultaneous separation, qualification, and quantification of various compounds from different samples, integrated with highly selective and sensitive mass spectrometric determination. However, sensitive and accurate chromatographic methods have disadvantages, such as requiring generally complex and high-cost devices, involving laborious sample preparation steps, and necessitating qualified experts, especially in the use of the devices [28–30]. Moreover, the aforementioned analytical methods make real-time monitoring of pharmaceutical compounds impossible and restrict the analysis to the laboratory surroundings since they employ sophisticated and cumbersome bench-top apparatus. Other drawbacks of the former methods are the high analysis cost and the requirement for qualified analysts to utilize the apparatus [31].

The early 21st century witnessed significant developments in electrochemical studies, which made the field truly multidisciplinary, merging chemistry, engineering, materials science, physics, and nanotechnology [32]. Electroanalytical techniques, particularly electrochemical-based sensor systems, are quite promising for drug analysis, offering the potential for on-site monitoring of pharmaceutical compounds. Electrochemical sensors are employed to determine the amounts of various analytes (neutral species, ions, metals, organic, inorganic, and biological molecules) in distinct samples. Today, electrochemical sensor systems are properly employed to determine and monitor many pharmaceutical compounds, antibodies, receptors, peptides, enzymes, lectins, and biomarkers in various biological fluids (serum, blood, saliva, urine, and tissue) [33–35]. Both investigation and development of pharmaceutical compound analysis take seriously innovative processes to merge precision, accuracy, sensitivity, and selectivity with simpleness, quickness, and low cost. The biggest benefit of sensors is that there is no need for pre-enrichment of the sample [36]. Consequently, the attentiveness to sensor systems has increased in recent years because such apparatus combines many of these standards with rapidity and simplicity [37].

Nanomaterials have been widely employed to develop electro-sensing systems. Nanotechnology has been a recognized area of research since Nobel Prize laureate Richard Feynman popularized the term “nanotechnology” in his 1959 lecture “There’s Plenty of Room at the Bottom,” the field has undergone tremendous and revolutionary advances. Nanotechnology has fabricated materials of numerous kinds at the nanoscale level (within a range of 1 to 100 nm) [38, 39]. It investigates matter features at the nanoscale and specifically focuses on solid-state materials’ inimitable, size-dependent features [40–42].

Nanostructured materials, with their wide surface-active area, high conductivity, immense stability, and good selectivity, have always been of interest. However, their conventional synthesis often involves toxic reducing and functionalizing chemicals, posing risks to human health and the environment. Moreover, traditional methods generate harmful by-products that hinder their use in biological and clinical trials. To overcome these limitations, various methods of nanomaterial manufacturing based on natural chemicals have been proposed [43]. In essence, the potential of an environmentally friendly nanomaterial synthesis, where the nanomaterials needed to modify electrodes are made ecologically benign, is on the horizon. This potential is driving the popularity of the so-called “green synthesis” for its simplicity, cost-effectiveness, short analysis time, and environment-friendliness [44].

Green nanomaterial synthesis, with applications in electrochemical (bio)sensing, is a burgeoning research area, allowing for more sensitive electroanalytical detection with less negative environmental impact. Nanomaterials’ exceptional physicochemical qualities combined with green synthesis methods’ eco-friendly and low-cost aspects have produced extremely advantageous outcomes. According to research, natural extracts derived from plant tissues have been successfully used to produce nanomaterials. Bioactive chemicals found in such tissues operate as reducing, capping, and functionalizing agents all at once, allowing nanoparticle formation to be completed more easily. Modifying electrochemical sensors with green-synthesized nanostructures can significantly improve the detection limit for a wide range of important analytes, particularly pharmaceutical drugs [43].

Green synthesis is advantageous in producing nanomaterials with unique morphologies and properties that are not easily achievable through traditional methods. These unique characteristics can significantly enhance the electrocatalytic activity of nanomaterials, leading to improved sensitivity and selectivity in electrochemical sensors. For instance, greenly synthesized nanoparticles often exhibit smaller sizes and more uniform distributions than those produced using traditional methods. This can increase the available surface area for electrochemical reactions, resulting in higher current responses and lower detection limits [45].

Additionally, green synthesis can contribute to developing more sustainable and environmentally friendly electrochemical sensors. By reducing toxic chemicals and

minimizing waste, green synthesis can help mitigate the environmental impact of nanomaterial production and sensor fabrication. This is particularly important in the pharmaceutical industry, with a growing emphasis on sustainability and ethical practices.

This work intends to serve as a worthy source for researchers in eco-friendly nanomaterial-based sensors. It reviews the existing literature on the electrocatalytic effect of nanomaterials and attempts to elaborate on the synthesis and characterization of eco-friendly nanomaterials. Moreover, it focuses on the applications of such sensors in pharmaceutical analysis by referring to the recent five-year studies in the literature. Finally, the review ends with a concluding section that evaluates the overall impact of eco-friendly nanomaterial-based sensors and their applications in pharmaceutical analysis.

## 2 Electrocatalytic Effect of Nanomaterials

Electrocatalysis, a significant process in various applications, including electrochemical processes, energy transformation technologies, and environmental remediation, is a topic of practical interest. This entails using nano-electrocatalysts to expedite electrochemical reactions, detect diverse analytes, permit effective energy conversion, and enable the breakdown of pollutants [46]. The role of electrocatalysis in identifying the extent and rate of electrochemical reactions in the presence of an electrocatalyst is a fascinating area of study. It reduces the electrode overpotential that connects electrode material, products, reactants and intermediates at the inter-phase electrolyte-electrode [47, 48].

Electrochemical reactions involve at least one stage where an electron is transferred between species from the electrode surface or the solution side. These reactions, forming at the interfaces, are managed by the interplay of the reactant with the electrode and the solvent. Electrocatalytic reactions mostly occur in the electric double layer that forms between an ionic conductor, such as an electrolyte solution, and an electronic conductor, typically a metal. In electrocatalytic reactions such as oxygen evolution/reduction, hydrogen evolution/oxidation, and $CO_2$ reduction, the structures of interfacial electric double layers play a major role in determining the kinetics, thermodynamics, and selectivity of the product. These reactions have been collectively referred to as the effects of the electric double layer [49–51]. Therefore, electrocatalysts, defined as catalysts participating in electrochemical processes, are significant for these reactions, working on the surface of electrodes or the electrode surface itself in some circumstances [52].

Research for extremely efficient electrocatalysts has been unusually focused on nanoparticles for the last two decades. Due to their finite-size influences, nanomaterials exhibit a variety of intriguing properties. Specifically, among other advantageous electrochemical properties, nano electrocatalytic materials often exhibit enhanced electrical conductivity, highly effective electrochemically active surface area, and elastic strength of intermediate/substrate bonding. Therefore, the selectivity and sensitivity of electrochemical sensors have been greatly enhanced by electrocatalytic signal magnification based on nanomaterials [32].

Thanks to the remarkable physicochemical characteristics of nanomaterials, sensors with larger surface areas, higher sensitivity, and faster response times may now be built. Comparing nanomaterial-based sensors to macro-scale materials, their high surface-to-volume ratio, huge active surface area, and extremely small particle size enhanced the sensing response and increased electrical, optical, and magnetic capabilities [53, 54].

Furthermore, it is desirable to lead the catalytic constituent on a suitable variety of supporting materials to further enhance the stability and electrocatalytic performance of the nanomaterials-based catalysts [55]. Certain conditions must be met by these catalysts, including (i) a wide surface area to guarantee efficient dispersion of catalyst particles, (ii) strong electrical conductivity to facilitate electron transfer, and (iii) exceptional stability to maintain a stable catalytic structure [55–57].

Nanomaterials synthesized via green methods frequently demonstrate distinct surface properties, including increased surface area and enhanced functionalization. This may result in enhanced interaction with target analytes, thereby improving sensitivity and selectivity in sensors. Green synthesis facilitates enhanced control over the morphology and size of nanoparticles. This may result in enhanced electrocatalytic sites, thereby improving the efficiency of electron transfer processes essential for electrochemical reactions [58].

### 2.1 Electrochemical Effect of Nanomaterials

The first consideration in nanomaterials used in electrochemical analysis is the electrical properties of nanomaterials. Nanomaterials are divided into conductors, semiconductors and insulators according to their ability to conduct electricity. Conductive nanomaterials stand out with their high electrical conductivity and catalytic effects and exhibit high electrochemical performance. In addition to providing high catalytic activity, these materials increase the sensitivity and selectivity of sensing elements. Quantum dots and carbon nanotubes are examples of conductive nanomaterials. The electrical conductivity of metal oxides and sulfides, which have moderate electrical conductivity, can be increased by changing the amount and size. These semiconducting

materials are actively used for energy conversion. Insulating nanomaterials offer low electrical conductivity but provide large surface area and large pore volumes, making them suitable for use in electrochemical analysis. The surface area and porosity of materials significantly affect their electrochemical performance. Materials with high surface area provide more active sites for electrochemical reactions, leading to improved charge storage capacity and faster reaction kinetics. The porosity of materials influences ion diffusion and electrolyte accessibility, affecting overall device performance. The most commonly used insulating nanomaterials are metal-organic frameworks (MOFs) and covalent organic frameworks (COFs).

According to their structure, nanomaterials can be classified as carbon-based nanomaterials, metal/metal oxide, conductive polymers and hybrid materials. Metal/metal oxide nanomaterials are promising in electrochemical devices due to their high surface area and redox properties. According to the literature, electrodes and electrocatalysts are typically made of palladium, gold, platinum, and silver in various electrochemical processes. Although these materials have good qualities, their commercial applications are limited by their high cost, limited sources, weak firmness, and specificity. On the other hand, transition metals, such as iron, nickel, and cobalt, are abundant and have favorable electroactive characteristics, which makes them interesting materials for electrocatalytic sensing platforms. Additionally, metal-organic framework materials have garnered increased interest and have been used as electrochemical sensors [59]. Carbon-based nanomaterials have received sustained notice as an ideal electrode material group. Manufacturing carbon-based nanoskeletons with inimitable structures has become a crucial field of investigation in electrocatalysis. These carbon-based nanomaterials can be used as a prominent carrier and active substrate, having a significant role in different electrocatalytic procedures because of their elastic features, including wide surface area, enhanced electrical conductivity, mechanical robustness, acid and alkali survivability, porous structure, and surface functionalization [60, 61]. Carbonaceous substrates such as carbon nanotubes, graphene oxide, and graphitic carbon nitride are generally utilized to connect the nanomaterials and facilitate electron transfer in electrochemical reactions [62]. Conducting polymers such as polyaniline and polypyrrole, with their high conductivity and tunable redox potential, provide versatility in electrochemistry. Hybrid materials are nanomaterials developed by combining two or more classes of nanomaterials. Thanks to synergistic effects; they offer the potential for improved performance and functionality.

The most significant element in defining electrocatalytic features of a catalyst is the structure of the surface (for example, a specific style of faceting process or the dominant crystallographic planes and directions in the catalytic matter, also well known as the Miller indices), not certainly the shape of the nanocatalyst [63]. The size of catalysts has been lowered to nanometers or even single atoms for desired activities to increase the efficiency of atom consumption [61]. One parameter that is especially indispensable for electrochemical implementations of nanomaterials is the specific surface area of the electrode [64]. Several approaches have been put up recently for the design and synthesis of nanomaterial-based electrocatalyst materials for electrochemical uses. With an increase in the electroactive field, more active sites, and an arranged structure that permits preferred diffusion of the target analyte, nanostructured materials provide additional advantages and lead to more effective determination [59].

Various electrochemical-based sensing platforms have several physical and electrochemical features, which directly impact the sensitivity, selectivity, and limit of detection and restrict the feasibility of the final electrochemical sensor system [65]. Electrochemical-based sensors are appliances capable of determining analytes in real-time. The focus has recently shifted to quickly preparing extremely sensitive and selective sensors using environmentally benign nanomaterials and a one-pot synthesis method. The successful use of nanomaterials opens the door to the broad application of various methods for creating innovative, ultra-sensitive platforms for analyte detection [66, 67]. The stability of electrochemical sensors is essential for their long-term performance, resulting in more reliable and consistent readings in pharmaceutical applications. The utilization of natural precursors and simplified processing methods frequently leads to reduced production expenses. This economic advantage may promote the advancement of more accessible and widespread electrochemical sensing technologies [68].

Plant extracts offer a revolutionary approach to producing nanoparticles because of their advantages over chemical and physical processes in mass production, ease of use, economic viability, and environmental benefits. Additionally, they provide biocompatible nanomaterials free of harmful chemicals during synthesis and have a wide range of applications in biomedical disciplines. The abundance of active biomolecular compounds found in plant extracts, such as phenols, alkaloids, flavonoids, proteins, nitrogenous bases, reducing sugars, amino acids, and other oxygenated phytochemical compounds that can function as reducing and stabilizing agents, causes the reduction of metal ions, contributing to an enhancement in their electrocatalytic activity [69]. Considering the increasing significance of green chemistry in nanomaterial production, the next chapter deals with these environmentally friendly substances.

## 3 Eco-friendly Nanomaterials: General Information

Electroanalytical sensors developed recently utilize metal-metal oxide, carbon-based nanomaterials, and metal-organic framework frameworks [70]. This section discusses the fundamental characteristics of the generally employed environmental-friendly nanoparticles in electroanalytical techniques used for drug detection and the often-utilized procedures for their production and characterization.

### 3.1 Effect of Nanomaterials on Pharmaceutical Component Analysis

Nanotechnology is the scientific field that involves altering the size of substances and modifying their chemical and physical properties in their larger forms [71, 72]. Currently, material engineering is focused on researching various nanomaterial designs for diverse application sectors. Using environmentally sustainable and cost-effective techniques in manufacturing these materials holds significance [73]. Within this framework, the research is mostly centered around the techniques for creating nanomaterials and the specific chemicals utilized in their production [74].

Plant extracts, yeast, fungi, algae, and bacteria are employed as reducing agents in the green synthesis of metal nanoparticles [75–77]. Plant species contain bioactive compounds with very different chemical structures, including flavonoids, alkaloids, steroids, terpenoids, etc., which act as reducing agents in the synthesis of nanoparticles. These plant extracts have considerable advantages over microorganisms (such as bacteria, yeast, fungi etc.) for obtaining ecofriendly nanoparticles since one-steps, nonpathogenic and cost-effective process of it [78]. One of the most important advantages of using plants in nanomaterial synthesis is that it eliminates the cell culture maintenance process required for microorganisms. Since microorganisms are living cells, the detailed cell culture required for each microorganism is essential for maintaining the viability of the microorganism [79]. Nevertheless, plant extracts, specifically leaves, roots, latex, seeds, and stems, are favored for creating metallic nanomaterials due to their advantageous stabilizing and reducing qualities [77]. Plant extracts facilitate fast, cost-effective, and non-hazardous manufacturing of nanoparticles while allowing for synthesis scalability. Furthermore, including several functional groups exclusively present in plant extracts enables the modification of NPs with cupping agents, eliminating the need for chemical substances [75].

The use of plant extract sources obtained from biomass as natural stabilizing and reducing agents can be attributed to the high amount of carboxyl and hydroxyl groups that can easily functionalize various metal nanoparticles, which can increase the distribution of nanostructures and enhance the utilization and efficiency of metal-based electrocatalysts. These environmentally friendly synthesis methods can produce fine particles with high accessible surface area and various metal nanostructures with suitable morphology by controlling the structure and size of active sites on the material [69, 80] Tian et al. have obtained mesoporous zirconium phosphate nanomaterial using yeast biotemplate in a study. As a result of the study, it was stated that the electrocatalytic activity of the nanomaterial synthesized from yeast was at an excellent level. This increase in electrocatalytic activity value was associated with the local current density and it was stated that with the increase in the effective area, the local current density decreased at that level and the reaction rate increased at that rate [81]. Moreover, gold and silver nanoparticles are extensively utilized in biomedical applications, nanobiotechnology, and pharmaceutical electroanalytical sensor approaches [82, 83]. This is because they demonstrate minimal toxicity while enhancing the effectiveness of electrochemical sensors.

Carbon-based eco-friendly nanoparticles are recognized for their superior properties in various biological applications such as sensing and surface coating [84]. Carbon-based eco-friendly nanoparticles are a distinct category of nanomaterials that have gained recognition for their exceptional characteristics in several biological applications, including sensing and surface coating [85]. They substitute metal-based sensors because of their exceptional catalytic characteristics [85–87]. They are also regarded as excellent options for quick diagnostic testing because of the synergistic effect they provide when combined. Thus, researchers serve as a cost-effective substitute for pricey nanoparticles like gold, colored latex, silica, quantum dots, or phosphor nanoparticles in electroanalytical techniques [87, 88]. The predominant carbon structures utilized in drug analysis are multi-walled carbon nanotubes, single-walled carbon nanotubes, graphene, carbon quantum dots, and conductive polymers [89]. Biomass and plant peels are commonly used as starting materials in creating carbon-based nanomaterials [90]. Additionally, many other food and agricultural wastes such as proteins, chitin, lignin, carbohydrates, hemicellulose, and honeycomb have been used to produce carbon-based materials using the green synthesis approach. This approach is an exemplary model for creating distinct, reusable, and valuable materials from garbage to promote a sustainable world [86].

### 3.2 Synthesis of Eco-friendly Nanomaterials

Nowadays, the synthesis design methodologies for nanomaterials utilized in sensor devices must prioritize both

environmental sustainability and cost-effectiveness. In this context, the often employed environmentally friendly synthesis methods are sonochemical synthesis, solvothermal/hydrothermal approach, and microwave-assisted synthesis.

Sonochemical synthesis entails the application of ultrasonic energy to the desired reactants. This approach utilizes a steady ultrasonic pulse as a heat source to promote fast crystallization, forming homogeneous nucleation centers that occur quickly [91]. This process is commonly used to make metal-metal oxide and metal-organic frame materials. When sonication is utilized, the initial formation of bubbles occurs, which subsequently leads to the generation of radicals in the solution due to the collision of these bubbles. This collision-induced process also generates the required heat to facilitate the breaking of chemical bonds [91, 92]. This approach primarily enables the fabrication of metal-metal oxide and metal-organic framework materials.

The microwave approach has gained popularity in recent years as a synthesis technique that facilitates the formation of nanoparticles by rapidly and effectively heating the process [93]. This technology allows for the rapid synthesis of the necessary nanomaterial, unlike traditional techniques that can take several hours. Hence, the utilization of microwave-assisted synthesis techniques enables the reduction of energy consumption and chemical usage in the manufacturing process of a nanomaterial [93, 94]. This approach applies to metal nanoparticles and carbon materials, including carbon quantum dots [95].

The hydrothermal approach involves the reaction of the initial substances and solvent in a hermetically sealed container using gentle circumstances [96]. The procedure is called hydrothermal when water is employed as the solvent and solvothermal when alcohol or another organic solvent is utilized. While the initial substances should dissolve in the solvent fully, it is feasible to accomplish the desired outcome without total dissolution by regulating temperature, pH, and duration [97]. A reduced amount of organic solvent is utilized, particularly in plant extract research [96]. Therefore, a more ecologically sustainable approach is employed. By employing this methodology, it is feasible to generate materials that are composed of carbon as well as ones that are composed of metal.

Today, traditional methods used for the synthesis of nanomaterials have many disadvantages such as the use of high-cost equipment, the use of high amounts of chemicals that are harmful to the environment and human health, high energy consumption, high heat production and low product efficiency. Therefore, it can be said that the main disadvantage of these methods is the use and release of toxic chemicals that cause various environmental problems [98, 99]. In recent years, methods compatible with green chemistry principles have been increasingly preferred for the synthesis of nanoparticles. Because these environmentally friendly methods aim to prevent pollution before it occurs by minimizing the use of hazardous chemicals. In addition, the goals of these methods include “energy efficiency, prevention of waste, reduce derivatives, renewable substances, safer auxiliary substances” [100, 101].

In the synthesis of nanomaterials, the reaction time is one of the important experimental parameters to be considered. This parameter can determine the shape and size of the nanoparticles, as well as the stability and formation rate of the nanoparticles. Green approaches that are environmentally friendly are usually carried out under mild experimental conditions, but this period can be longer to ensure reproducible and efficient nanomaterial synthesis. For example, the time required for the reduction of metal ions, which includes the synthesis of nanomaterials, was found to be approximately 24 to 124 h when bacterial and fungal species were used as biomaterials [102, 103]. In research on the synthesis of nanomaterials using environmentally friendly syntheses, it is also necessary to carefully examine the human health risks associated with nanotechnology. Currently, research on the accumulation and toxicity of nanoparticles is limited, and the small size of these particles makes it easier for them to enter the human body, potentially causing respiratory problems and serious diseases [104]. Meticulous trials and assessments will be essential to address the toxicity and environmental influence of nanomaterials before they can be reliably usage in electrochemical devices.

However, some plant sources that can be used to obtain nanomaterials have a very long growing period and constitute one of the main sources of problems related to time constraints [105]. In addition, some raw materials used for nanomaterial production are secondary products that require further processing during the synthesis process, which makes the synthesis process more complex and costly before they can be used in the environmentally friendly synthesis of nanoscale materials. As a result, the chance of especially economic applicability of these nanomaterials decreases [106].

### 3.3 Characterization Method for Eco-friendly Nanomaterials in an Electrochemical Sensor

Characterization of eco-friendly nanomaterials is important for revealing their potential applications and for understanding their morphological and topological properties to check their size, shape, dispersity, localized, surface topography, surface area and porosity. There are different methods for characterization of nanomaterials.

#### 3.3.1 Scanning Electron Microscopy (SEM)

SEM is utilized to examine the surface characteristics of nanomaterials, including porosity, irregularity, and size [95]. Accurate particle size measurement is crucial for its effective utilization in electrochemical sensor applications [107]. Furthermore, the distribution of pore sizes is a crucial factor in determining how organic and inorganic molecules are adsorbed onto carbon or metal nanomaterials with different functional groups and cubbing agents [108]. This pore size distribution is vital in explaining the ion transfer mechanism on the material's surface, particularly in electrochemical measurements conducted in a solution medium.

#### 3.3.2 Fourier Transform Infrared Spectroscopy (FTIR)

FTIR is a technique that quantifies the absorption/transmittance of light and transforms it into a wavelength-dependent function [95]. This enables the identification of the functional groups inside a structure. It has a crucial purpose in identifying the functional groups in carbon and metal nanoparticles, particularly those containing cupping agents [109]. Furthermore, these functional groups play a crucial role in defining the electrical conductivity and the material's behavior in water-based environments based on their hydrophobic or hydrophilic nature.

#### 3.3.3 Ultraviolet-Visible Spectroscopy (UV–Vis)

UV-Vis technique is commonly used to analyze the optical absorption properties of nanomaterials and their derivatives. The absorption behavior of eco-friendly metal and carbon-based materials indicates their structural alterations [107–110]. For instance, the absorption pattern of nanostructured graphene is observed at around 300 nm, caused by the n-π* transitions of C–O bonds. Additionally, another absorption peak is observed at 231 nm, which corresponds to the π-π* transitions, showing the presence of aromatic C–C bonds.

#### 3.3.4 X-Ray Diffraction Spectroscopy (XRD)

XRD patterns are useful for elucidating the structure of eco-friendly nanomaterials; however, they are not standalone in providing conclusive information. Nevertheless, functional groups within the structure and their respective concentrations result in the acquisition of distinct angle measurements [95].

#### 3.3.5 X-Ray Photoelectron Spectroscopy (XPS)

XPS method has become an indispensable examination method for surface characterization in various research fields such as material surface characterization, corrosion, adhesion, catalysis, metallurgy, semiconductor materials, medicine, biology, medical applications, textile, geology, energy storage materials, etc [111]. In this method, the elemental composition of biomass (many functional groups exist) [95] is identified by integrating the region (at different binding energies) bounded by the peaks in the spectrum formed when a focused X-ray beam excites the bonding and inner orbital electrons of the target sample. The use of the XPS method together with other characterization methods has many advantages over classical characterization methods because they can simultaneously determine other biomass properties such as morphology and binding states and can significantly reduce the analysis cost [112, 113].

## 4 Applications of Eco-friendly Nanomaterial-Based Sensors for Pharmaceutical Compound Analysis

Expensive, complex, toxic release restrictive methods for synthesizing many nanomaterials, including carbon nanomaterials, have been replaced by low-cost, environmentally friendly methods involving fewer chemical and synthesis steps [110]. Electrochemical sensors developed using environmentally friendly nanomaterials are used for many analytes and are preferred in analyzing active pharmaceutical ingredients from different matrices.

Carbon quantum dots (CQDs) are currently a prominent area of focus in carbon nanomaterial research. CQDs are chemically stable carbon-based nanomaterials with great biocompatibility, low toxicity, and high solubility in organic solvents and water [114]. CQDs, which avoid the semiconductor problem caused by quantum dots, are used in fields such as biosensors, bioimaging, and fluorescent dyes [115]. One of the benefits of these nanoparticles is their capacity to be produced from biomasses and wastes, which makes them particularly appealing for sustainable research [116]. Khasim et al.. synthesized CQDs through the pyrolysis of *Halimeda Opuntia* green algae and doped it with silver nanoparticles (AgNPs) to determine D-glucose and paracetamol simultaneously. The materials employed in surface modification were analyzed for their surface characteristics using SEM, XRD, FTIR, and UV-vis spectroscopy. The SEM analysis revealed that the produced CQDs comprised spherical carbon particles with an agglomerated morphology. Further examination of AgNPs@CQDs indicated that AgNPs were evenly dispersed within the CQD and formed a porous structure, increasing stability. The detection limit for D-glucose and paracetamol were determined to be 0.32 and 0.29 µM, respectively. The method's selectivity, cost-effectiveness, and potential for nanomaterial

manufacture through biosynthesis have enhanced its suitability for future research in determining paracetamol [117].

In the study made by Mathew et al.., carbon dots (C-dots), which also showed fluorescence characteristics, were synthesized from *Simarouba glauca* leaves by hydrothermal method for the determination of doxycycline antibiotic used for respiratory, urinary, and biliary system infections. The homogeneous spherical shape of C-dots was seen using transmission electron microscopy (TEM). XRD, FTIR, XPS, and Raman analyses were also performed to characterize C-dots. It has been stated that the developed sensor increases the analyte response three-fold compared to a bare glassy carbon electrode (GCE). In the study, the selectivity of the developed sensor was also shown using other antibiotic molecules such as amoxicillin, ciprofloxacin, 5-fluorouracil, and other kinds of molecules such as histidine, glucose, or cysteine. The authors compared the sensor's performance with other electrochemical studies to determine doxycycline and found a low detection limit (2.86 nM), and applicability was evaluated in tablets and different water samples [118].

Gold nanoparticles (AuNPs) are commonly utilized nanomaterials in chemical, physical, medical, electrochemical, and biological sciences [119]. Environmentally friendly methods are also important in the synthesis of AuNPs. Nardi et al.. used *Hylocereus undatus* extract (Ext) as a reducing and stabilizing agent to synthesize AuNPs. They also used functionalized multiwalled carbon nanotube (f-MWCNT) to detect hydroxychloroquine (HCQ), traditionally used to treat malaria but has recently demonstrated its efficacy in treating COVID-19. Characterization of AuNP-Ext was performed by UV-Vis and f-MWCNTs/AuNP-Ext nanocomposite by TEM. The electrode surface characterization conducted using electrochemical impedance spectroscopy (EIS) revealed that the electrodes modified with f-MWCNT-Ext, AuNP-Ext, and f-MWCNT/AuNP-Ext exhibited reduced charge transfer resistance in comparison to the unmodified GCE. These results confirm that the nanomaterials employed for modification exhibit a high level of conductivity. The cyclic voltammetry (CV) results showed an increased and more defined HCQ signal using the hybrid material of f-MWCNT-AuNPs-Ext. Square wave voltammetry (SWV) was used to determine the analyte, yielding a detection limit of 0.0093 µM and a limit of quantification of 0.031 µM. It was observed that the proposed method enabled determination within two linear ranges, 0.03–3.5 µM, and 3.5–17.0 µM. The method developed enables the determination of HCQ in pharmaceutical dosage forms and clinical samples [120].

Oxide nanoparticles exhibit distinct electrical and chemical characteristics compared to their bulk forms [121]. These materials are of significant interest to researchers due to their extensive applications in many domains, including biosensors, tissue engineering, environmental sciences, and catalysis. For instance, strontium oxide nanoparticles (SrONPs), which have many applications in fields including gas sensors, cancer therapy, drug delivery, supercapacitors, and semiconductors thanks to their morphological and surface properties, can be optimized by employing a green and environmentally friendly synthesis process [122]. Abukattab et al.. employed a graphite paste electrode (GPE) coated with SrONPs and synthesized it using green *cardamom* plant extract. The plant extract was employed as a reducing and stabilizing agent in the chemical synthesis of SrONPs, as shown in Fig. 1A. Further, in-situ oxidative polymerization, using $FeCl_3$, was performed to obtain SrONPs-polythiophene (SrO@PTh) nanocomposite (Fig. 1B). In this study, the selection of PTh, a conductive polymer, was based on its mechanical, optical, and electronic durability and the enhanced stability of the sulfur atom in its structure. Cilostazol, a drug used to treat peripheral arterial disease, was determined by employing SrO@PTh/GPE. The SrO@PTh composite was characterized using FTIR, EDX, XRD, and SEM techniques. The FTIR data revealed a notable displacement in the Sr-O vibration bands within the SrO nanoparticle, thereby confirming the interaction between sulfur and the SrONPs within the structure of PTh. The research revealed that cilostazol could be detected using the SW adsorptive anodic stripping voltammetry within the concentration range of $1 \times 10^{-9}$ to $6 \times 10^{-7}$ M, with a detection limit of $1.99 \times 10^{-10}$ M. Real sample applications of the drug were performed on bulk and human plasma samples. The drug was detected in tablet form and exhibited low relative standard deviation values within the recovery range of 99.8–100.33%. The findings indicate that cilostazol can be accurately detected through the environmentally friendly and selective SrO@PTh/GPE. This method can also be effectively employed in diverse applications, including clinical laboratories and tablet dosage forms [123].

In addition to strontium, nickel oxide (NiO) is a noteworthy metal oxide nanoparticle. Due to its distinctive electrical and magnetic properties, NiO has many applications, including in electrochemical supercapacitors, gas sensors, and photoelectrolysis [124–126]. These nanomaterials display an exceptional degree of stability and exhibit notable electrocatalytic activity. Despite the growing demand for using NiO nanoparticles (NiONPs) in various shapes in sensor studies, there are ongoing challenges in developing cost-effective, efficient, and environmentally friendly sensor platforms. The green synthesis method offers an alternative for this purpose. In a study by Likasari et al.., the leaves of *Tagetes erecta L* were extracted and used as a reducer to synthesize NiONPs. The synthesis of NiONPs was achieved by applying microwave irradiation and reflux methodology,

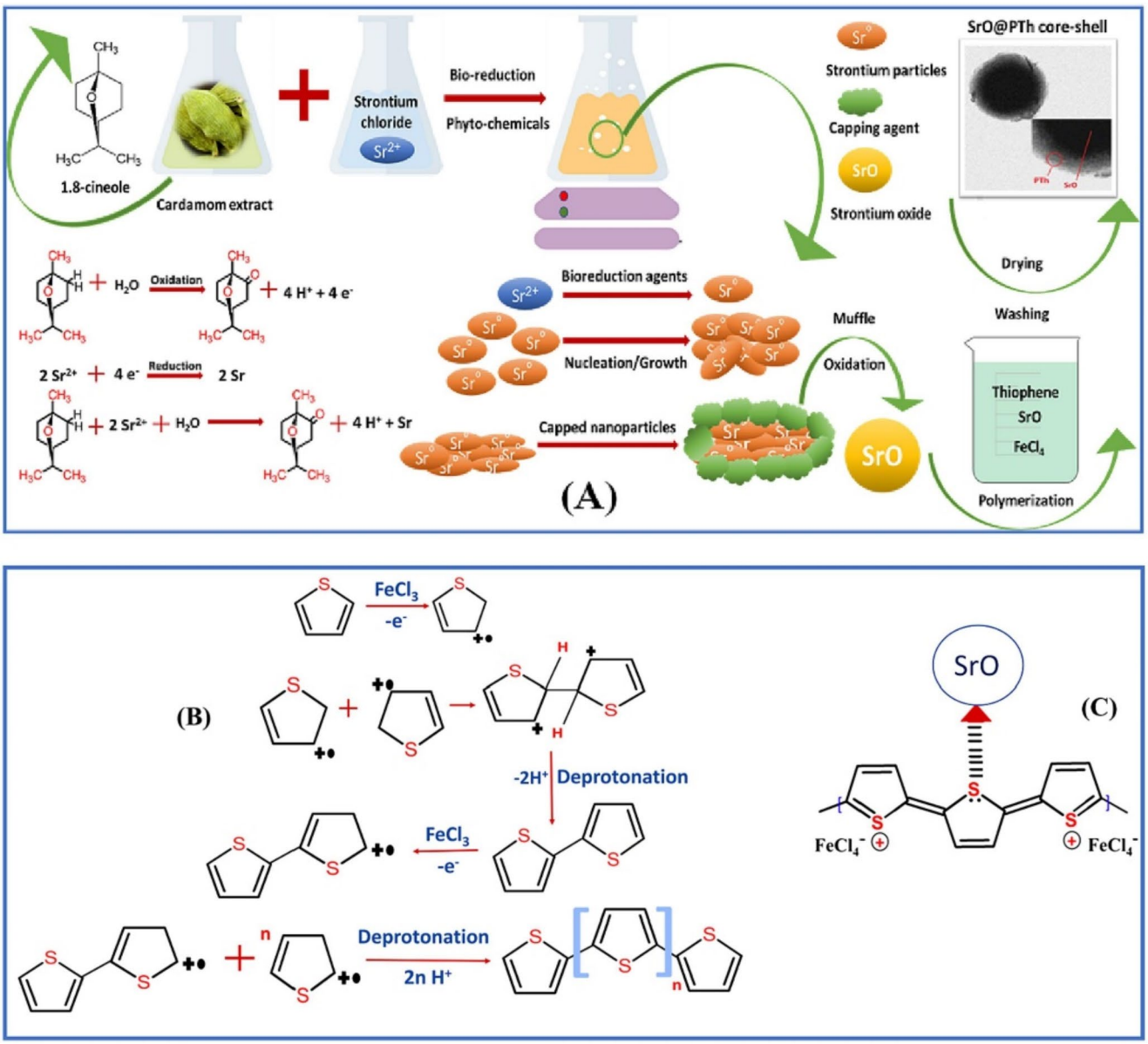


**Fig. 1** The plausible redox reactions mechanism for synthesis of SrO NPs (**A**) and chemical oxidative polymerization of thiophene to form SrO@PTh core-shell nanocomposite (**B**), which leads to the formation of additional polarons and electronic pathways required for enhanced electrical conductivity. Reprinted from Ref [123]. with permission from Elsevier.

as shown in Fig. 2. To conduct the study, the researchers modified the surface of the GPE using NiONPs. They then employed the newly developed sensor to determine glucose. The synthesized nanoparticle was examined using SEM, XPS, TEM, and XRD. The SEM results revealed that the NiONPs synthesized through microwave irradiation exhibited smaller particle sizes and a more homogenous spherical shape than those synthesized through the reflux method. CV was conducted to determine the glucose, which was determined within the range of 0.1 to 1 mM. The sensitivity for determining was to be 2556.41 $\mu A.mM^{-1}.cm^{-2}$, with a detection limit of 83 µM. A study was conducted to assess the stability of the NiONPs/GPE sensor over time. It was found that the sensor maintained a response rate of 93% after 43 days, which was comparable to its initial response on day one. From this data, it can be concluded that the sensor has a high level of stability. In addition, extensive research was carried out to demonstrate the antibacterial properties of the NiONPs synthesized in this study [127].

Another study using metal and metal oxide nanoparticles together is performed by Khalilzadeh et al.., who developed a screen-printed electrochemical sensor based on $Fe_3O_4$@cellulose nanocrystals (CNC)/Cu nanocomposite ($Fe_3O_4$@CNC/Cu) for the sensitive detection of venlafaxine, a

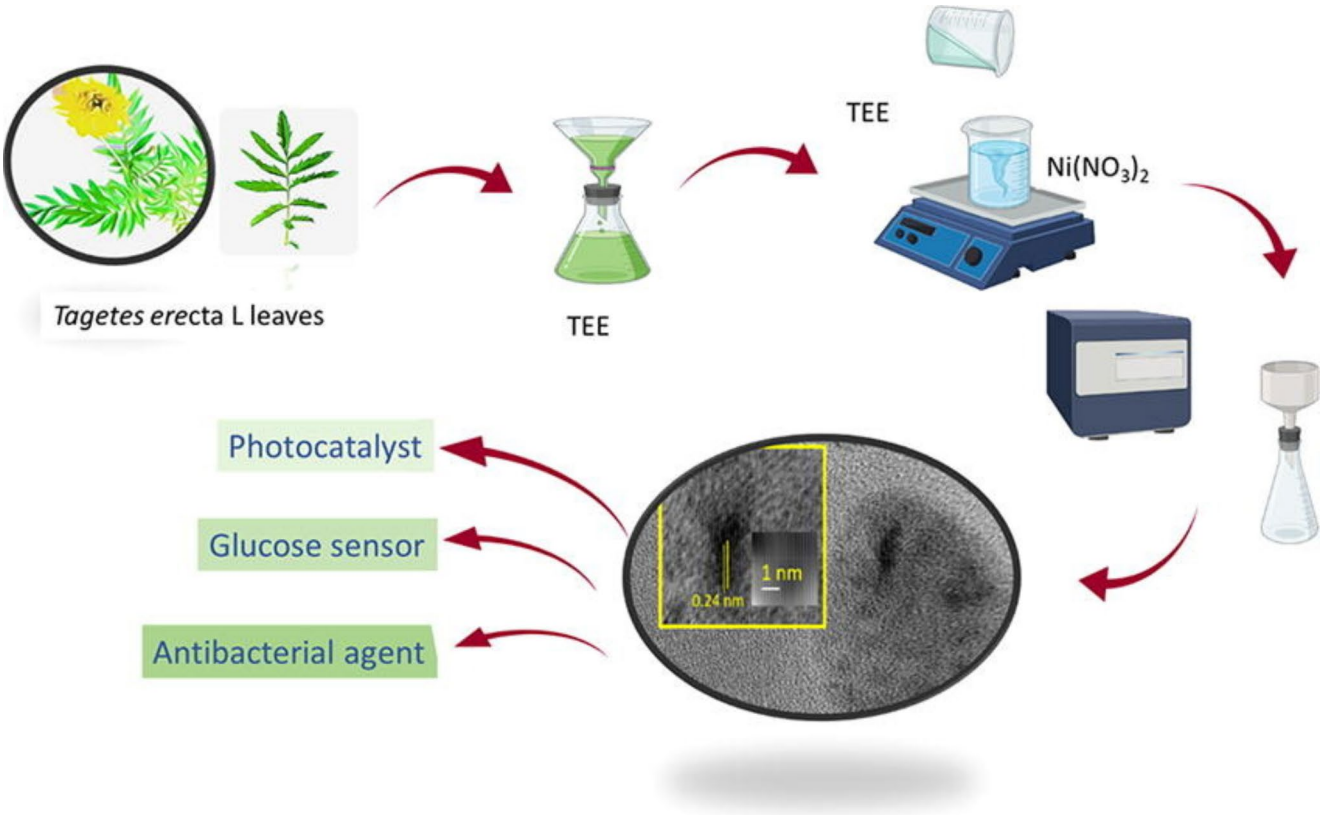


**Fig. 2** Schematic representation of NiONPs preparation. Reprinted from Ref [127]. with permission from Elsevier.

norepinephrine reuptake inhibitor, is widely used in the treatment of depression, anxiety and panic disorders. The modified graphite screen-printed electrode (GSPE) provided high sensitivity and reproducibility to the developed method. In the study, magnetic nanocomposites were obtained using ferrimagnetic iron oxide magnetic nanoparticles. However, to safeguard the alteration, $Fe_3O_4$NPs were covered with a cellulose nanocrystal layer because they are very reactive chemically and oxidize quickly in air. The performance and chemical stability of the nanoparticles were improved by the cellulose nanocrystal structure. Using HBr acid to hydrolyze Whatman filter paper, cellulose nanocrystals were created. Conversely, distilled water was used to suspend dried Petasites hybridus leaves at a temperature of 70º C. The resulting combination was then centrifuged at 6000 rpm. After that, the supernatant was put on filter paper. Lastly, a portion of the water was evaporated and heated to concentrate the green extract. Copper-reducing flavonoids and phenol chemicals are present in the final extract. As a result, $CuCl_2$ and leaf extract were combined as a reducing agent. XRD analysis and TEM images were used to verify the produced cellulose nanocrystals. The sensor's enormous surface area and strong electrical conductivity were provided by the plant-based nanomaterial. Differential pulse voltammetry (DPV), chronoamperometry, and CV were used to examine the electrochemical behavior of venlafaxine on the sensor. Using the modified electrode, the peak potential of venlafaxine was approximately 680 mV, almost 100 mV lower than the value observed for the bare GSPE. The beneficial impact of the alteration was demonstrated by the significantly greater anodic peak current measured with the modified electrode compared to the unmodified GSPE. The developed sensor's linear range for venlafaxine concentrations under ideal circumstances was determined to be 0.05–600.0 µM, with a 0.01 µM detection limit. Utilizing the method established to identify venlafaxine in urine, water, and pharmaceutical formulation samples demonstrates creating a sensitive, easy-to-use, and selective sensor [128].

In addition to the aforementioned studies, Table 1 summarizes some other examples of applications of sensors developed with materials obtained through green synthesis in drug analysis in the last five years. In doing that, the table focuses on the green sources, the electrode-modifying nanomaterials, electrochemical techniques utilized, the analytes explored in various matrices, and linear range and LOD values obtained.

The recent literature summarized in Table 1 reveals that green sources are widely used as reducing agents particularly for the synthesis of metal/metal-oxide nanomaterials

**Table 1** Some drug analysis studies conducted in the last five years on nanosensors synthesized with environmentally friendly methods

| ANALYTE | GREEN SOURCES | METHOD | ELECTRODE SURFACE | LINEAR RANGE | LOD | APPLICATION | REF |
|---|---|---|---|---|---|---|---|
| *Carbon-based green sensors* | | | | | | | |
| Ractopamine | Biochar | DPV | **NBC**/GCE | 0.1 − 1.75 µM | 0.041 µM | Pork sausage sample | [129] |
| 5-hydroxytryptamine | Black phosphorene | SWV | BP-IL-**SWCNH**/GCE | 0.3 − 115 µM | 0.1 µM | Rat blood serum samples | [130] |
| Tetracycline | *R. graveolens* leaves | CV | **N-CQD**/GCE | 5 − 30 nM | 0.80 nM | Water samples | [131] |
| Eslicarbazepine | Banana peels | DPV | **CQDs**/CPE | $3.04\times10^{-8}$–$9.66\times10^{-5}$ M | $7.57\times10^{-9}$ M | - | [132] |
| Ascorbic acid | Garlic green leaf | Chrono-amperom-etry | **$NiCo_2O_4$**/GCE | 0.5 − 8.5 mM | 0.01 mM | Chewable ce-cone tablets samples | [133] |
| Paracetamol | Sugarcane bagasse | DPV | Sugarcane bagasse-acti-vated biochar/GCE | 5–950 µM | 2.5 µM | Waste water samples | [134] |
| *Metal/Metal oxide based green sensors* | | | | | | | |
| Pramipexole | Sucrose | CV | SL-**$Pr_2Ce_2O_7$**/IL/CPE | 0.2–360 µM | 0.04 mM | Tablets, urine, and human blood serum | [135] |
| Capecitabine | Nano-ovalbu-min biocatalyst | DPV | DPB/**Cu**-MCM-41/CPE | 2.0–80.0 µM and 80.0–800.0 µM | 0.144 µM | Blood serum sample | [136] |
| Morphine | Crataegus and walnut leaf | DPV | **$TbFeO_3$/CuO**/SPE | 0.07 − 300.0 mM | 10 nM | Morphine ampoule, serum and urine samples | [137] |
| Promethazine hydrochloride | Pineapple gum | SWV | **AuNPs**/CPE | 4.0–15.7 µM | 1.09 µM | Pharmaceutical sample | [138] |
| Paracetamol | Curcumin | Amperom-etry | **CsNPs**/GCE | $0.59\times10^{-6}$–$342.1\times10^{-6}$ M | 0.29 µM | Water samples | [139] |
| Ornidazole | *Phoenix dacty-lifera* seed | SWV | **AgNPs**/CPE | $1.0\times10^{-5}$–$1.0\times10^{-3}$ M | $7.58\times10^{-7}$ M | Milk and water samples | [140] |
| Chlorambucil | *Vitis vinifera* | DPV | SL-**$Pr_6O_{11}$**/SPE | 8 nM − 220 µM | 1.88 nM | Tablet, human urine and serum samples | [141] |
| Paracetamol | *Scoparia dulsis* | CV | **α-$Fe_2O_3$NPs**/CPE | 1–6 mM | $0.95\times10^{-3}$ M | - | [142] |
| Chloramphenicol | Quercetin | LSV | **AgNPs**/GCE | 0 − 80 µM | 265.6 nM | Water, milk and honey samples | [143] |
| Ruxolitinib | Waste sponges | AdSDPV | SC-**$Co_3O_4$**/GCE | 0.08–20 µM | 6.73 nM | Tablet and serum samples | [144] |
| Nitrofurantoin | Urea | DPV | **α-$Fe_2O_3$**/h-BN/GCE | 0.025 − 22.95 µM | 15 nM | Human urine samples | [145] |
| Pazopanib | Waste masks | AdSSWV | **CuONPs**/GCE | $2.0\times10^{-10}$–$1.0\times10^{-6}$ M | $4.94\times10^{-11}$ M | Pharmaceutical dosage form and human serum samples | [146] |
| Glucose | *Trigonella subenervis* | CV | **NiONPs**/GCE | 10–200 µM | 3.2 µM | - | [147] |
| Acetaminophen | Coffee husks | CV | **ZnONPs**/SBA-15/GCE | 4 − 32 µM | 0.11 µM | Pharmaceutical sample | [148] |
| Lamivudine | *Senna Alata L.* leaf | CV | **ZnONPs**/GCE | 10–300 µM | 1.902 µM | - | [149] |
| Mephedrone | Saffron | SWV | **AgNPs**/CPE | 1.841 − 80.00 ng/L | 0.608 ng/L | Urine samples | [150] |
| Paracetamol | Christ's thorn jujube | LSV | **IONPs**/CPE | 0.8–10 µM | 0.287 µM | Adol tablet samples | [151] |
| Atropine | *Spirulina platensis* | SWV | **Ag**/HMIM $PF_6$/CPE | 0.01 − 800 µM | 5 nM | Atropine sulfate ampoule samples | [152] |

**Table 1** (continued)

| ANALYTE | GREEN SOURCES | METHOD | ELECTRODE SURFACE | LINEAR RANGE | LOD | APPLICATION | REF |
|---|---|---|---|---|---|---|---|
| Adrenaline | Lactose | SWV | S-CSS/GCE | 0.05–1.45 µM | 0.005 µM | Pharmaceutical drug and human blood serum sample | [153] |
| Ceftriaxone | *Fagonia cretica* | DPV | **$NiFe_2O_4$NPs**/GCE | $1.00\times10^{-8}$–$3.20\times10^{-6}$ M | $1.60\times10^{-9}$ M | Urine and serum samples | [154] |
| 5-Fluorouracil | *Rosa damascena* | DPV | **ZnO**/CPE | 0.05–140.0 µM | 0.0016 M | Fluorouracil ampoule samples | [155] |
| Dopamine | *Salvia officinalis* | DPV | **MgONPs**/CPE | $15.0\times10^{-6}$–$160.0\times10^{-6}$ M | $8.7\times10^{-7}$ M | Dopamine ampule, ascorbic acid tablet samples | [156] |
| Ascorbic acid | | | | $25.0\times10^{-6}$–$2750.0\times10^{-6}$ M | $1.65\times10^{-6}$ M | | |
| Atenolol | *Hibiscus sabdariffa* | CV | **DMZN**/CPE | 0.11 − 125.87 µM | 10.10 nM | Human blood serum samples | [157] |
| Aspirin | *Lycoris aurea* leaf | DPV | **AuNPs**/GCE | 100 µM − 2.6 mM | 11.3 µM | - | [158] |
| Etoposide | *Cuscuta epithymum* | DPV | **CEZLNCs**/CPE | 0.04–120 µM | 2.7 nM | Human serum and urine samples | [159] |
| Isoniazid | Banana peel | DPV | BSA/CYP2E1/GLU/Thioglycolic acid-**AuNPs**/AuE | 0.1683 − 6 µM | 0.0556 µM | Human serum samples | [160] |
| Diclofenac | *Ginkgo biloba* | DPV | **AuNPs**/GCE | 5 − 600 µM | 0.524 µM | - | [161] |
| *Metal/ Metal oxide @ Carbon Combine green sensors* | | | | | | | |
| Ornidazole | *Phoenix dactylifera* | SWV | **Lignin-AgNPs**/CPE | $8.0\times10^{-6}$–$1.0\times10^{-3}$ M | 0.362 µM | Milk, river water, and tap water | [162] |
| Mexiletine | *Citrus Reticulata* | AdSDPV | **CQDs/FeNPs**/GCE | $1.0\times10^{-7}$–$1.0\times10^{-6}$ M | $6.14\times10^{-8}$ M | Human serum samples | [163] |
| Balsalazide | Waste surgical masks | DPV | **CSC-CdO**/GCE | $8.00\times10^{-9}$–$8.00\times10^{-5}$ M | $2.20\times10^{-9}$ M | Human serum and urine sample | [164] |
| Metronidazole | *Sambucus ebulus* L. leaves | DPV | **GRNSs/$Fe_3O_4$NPs**/GCE | 0.05–5 µM and 5–120 µM | 0.23 nM | Real aqueous samples | [165] |
| Hydrocortisone | *Nigella sativa* seeds | DPV | **Nafion-CoONPs**/GCE | 0.001–1 µM and 1–9 µM | 0.49 nM | Pharmaceutical injections and blood serum samples | [166] |
| Paracetamol | *Araucaria angustifolia* | SWV | **AgNP-xGNPs**/GCE | $4.98\times10^{-6}$–$3.38\times10^{-5}$ M | $8.50\times10^{-8}$ M | Paracetamol tablet samples | [167] |
| Tryptophan | Chitosan | DPV | **Cs/Ce-MOF**/GCE | 0.25–331 µM | 0.14 µM | Human serum sample | [168] |
| Furazolidone | Polystyrene | DPV | PS/**β-CD@AuNPs**/GCE | 0.05–715 µM | 2.03 nM | Human blood serum sample | [169] |
| Gatifloxacin | Urea (hydrogen-bond donor) | LSV | **$ZnCo_2O_4$/MWCNT-COOH**/GCE | 0.01–10 µM | 2.0 nM | Water samples | [170] |
| Phenylbutazone | *Azadirachta indica* leaves | SWV | **CTN-$Fe_3O_4$/g-$C_3N_4$@**GCE | 0.6 − 10 µM | 1.28 nM | Water and injection samples | [171] |
| Acetaminophen | *Ocimum tenuiflorum* leaves | DPV | **AgO/rGO**/GCE | 0.001 − 0.3 mM | 0.012 mM | Water samples | [172] |
| Methyldopa | Black tea extract | DPV | **TbFeO$_3$/CuO**/CPE | 0.04 − 300 µM | 0.009 µM | Tablet, urine and human blood serum samples | [173] |

**Table 1** (continued)

| ANALYTE | GREEN SOURCES | METHOD | ELECTRODE SURFACE | LINEAR RANGE | LOD | APPLICATION | REF |
|---|---|---|---|---|---|---|---|
| Glutathione | *Ananas Comosus* | DPV | **CQDs**/GCE | 0–250 nM | 8.8 nM | Bakhra river, Patiala and tap water sample | [174] |
| Lapatinib | *Azadirachta indica* | SWV | **β-CD@CQDs**/GCE | $0.99\times10^{-8}$–$6.17\times10^{-6}$ M | $1.81\times10^{-10}$ M | Urine samples | [175] |
| | | DPV | | $1.99\times10^{-8}$–$8.59\times10^{-6}$ M | $8.56\times10^{-10}$ M | | |
| | | LSV | | $4.99\times10^{-8}$–$6.17\times10^{-6}$ M | $2.51\times10^{-9}$ M | | |
| Sulfaguanidine | Thiourea | Amperometry | **$Ce_2S_3$@CNS**/GCE | 0.01–1131.5 µM | 0.0053 µM | Blood serum samples | [176] |
| Vortioxetine | *Hyssopus officinalis* | DPV | **Au@g-$C_3N_4$**/GCE | 0.01–9.36 µM | 3.2 nM | Fonksera tablet and human plasma samples | [177] |
| Alpha lipoic acid | *Actinidia deliciosa* (Kiwi) peel | DPV | **$SnO_2$-β-CD-GO**/GCE | $6.5\times10^{-8}$–$7.22\times10^{-5}$ M | $1.42\times10^{-9}$ M | Tablet samples | [178] |
| | | SWV | | $3.12\times10^{-8}$–$4.99\times10^{-5}$ M | | | |
| Deferiprone | Natural asphalt | DPV | **NA@NiCo**-LDH NSs/GCE | 0.5–2500 µM | 0.19 µM | Human blood serum sample | [179] |
| Tryptophan | *Ziziphus mucronata* | SWV | **Cu-PANI**/GCE | 5.11 − 25.85 µM | 3.9 µM | Pineapple fruit | [180] |
| L-tryptophan | *E. tereticornis* | DPV | **AuNPs/rGO**/CE | 0.5–500 µM | 0.39 µM | Serum and salvia samples | [181] |
| Riboflavin | *Arum italicum* leaves | DPV | **Ag/rGO**/GCE | 0.002 − 2.2 µM | 0.6 nM | Riboflavin tablet samples | [182] |
| Repaglinide | Agro-waste cotton peels | SWV | **$SnO_2$@p-rGO**/GCE | $1.99\times10^{-8}$–$1.45\times10^{-5}$ M and $4.99\times10^{-8}$–$1.83\times10^{-5}$ M | $0.85\times10^{-9}$ M | Urine and pharmaceutical formulation samples | [183] |
| Nevirapine | Banana peel | DPV | **Au/$NiSe_2$QDs/ Nafion**/AuE | 0–1.21 pM | 0.024 pM | Drug and wastewater samples | [184] |
| Levodopa | Orange peel | CV | **Boroncarbonitride**/CPE | 0.2–160 µM | 0.14 µM | Commercial tablets and human urine samples | [185] |
| Metronidazole | *Goniothalamus wightii* | Amperometry | **GO@AgNPs**/GCE | 0.09 µM − 4.594 mM | 69 nM | Commercial tablets | [186] |
| Dopamine | Orange peels | DPV | **Ag-$Fe_3O_4$/NC**/GCE | 0.01–10.0 µM | 6.0 nM | Dopamine HCl injection, human serum, and urine samples | [187] |
| Acetaminophen | | | | 0.05–15.0 µM | 8.0 nM | | |

**Table 1** (continued)

| ANALYTE | GREEN SOURCES | METHOD | ELECTRODE SURFACE | LINEAR RANGE | LOD | APPLICATION | REF |
|---|---|---|---|---|---|---|---|
| Clenbuterol | Paper waste pulp | DPV | **$NiFe_2O_4$–NPCs**/GCE | $10^{-11}$–$10^{-5}$ M | $3.03 \times 10^{-12}$ M | Human urine samples | [188] |
| Venlafaxine | *Petasites hybridus* leaf | DPV | **$Fe_3O_4$@CNC/Cu**/SPE | 0.05 − 600.0 µM | 0.01 µM | Venlafaxine tablet samples | [128] |

ADSDPV: Adsorptive stripping differential pulse voltammetry, AgNPs: Silver nanoparticles, AuNPs: Gold nanoparticles, AuE: Gold electrode, BP: Black phosphorene, BSA: Bovine serum albumin, C: Carbon, CD: Cyclodextrin, C-dots: Carbon dots, CNC: Cellulose nanocrystals, CNS: Carbon nitrides sheets, CoO NPs: Cobalt oxide nanoparticles, CPE: Carbon paste electrode, CQDs: Carbon quantum dots, CSC: Carbon-based core–shell structures, cSNPs: Capped silver nanoparticles, CV: Cyclic voltammetry, CYP2E1: Cytochrome P450-2E1, DCF-PM: Diclofenac phosphomolybdate, DMZN: $DyMnO_3$-ZnO nanocomposites, DPB: Tetrahydrodipyrazolo pyridine, DPV: Differential pulse voltammetry, $Fe_3O_4$: Ferrimagnetic iron oxide, FeNPs: Iron nanoparticles, GCE: Glassy carbon electrode, GLU: Glutaraldehyde, GPE: Graphite paste electrode, GO: Graphene oxide, GrNSs: Graphene nanosheets, GT: gadoliniumtitanate ($Gd_2Ti_2O_7$), $HMIMPF_6$: 1-Hexyl-3 methylimidazolium Hexafluorophosphate, h-BN: Hexagonal boron nitride, IL: Ionic liquid, IONPs: Iron oxide nanoparticles, LIGE: Laser-induced graphene electrode, LOD: Limit of detection, LSW: Linear sweep voltammetry, MOF: Metal-organic frameworks, MS $Pr_6O_{11}$: Mesoporous $Pr_6O_{11}$, MS-$Pr_2Ce_2O_7$ mesoporous praseodymium, MWCNT: Multi-walled carbon nanotubes, NA: Natural asphalt, NBC: Nafion-biochar-supported, NC: Nanocellulose, N-CQDs: Nitrogen-doped carbon dots, N-GQD: Nitrogen-doped graphene quantum dots, $NiCo_2O_4$: Nickel–cobalt oxides, NiCo-LDH, NSs: Nickel–cobalt double hydroxide nanosheets, $NiFe_2O_4$-NPs: Nickel ferrite nanoparticles, $NiSe_2$QDs: Nickel selenide quantum dots, NPCs: Nanocarbon-supported, PAM: Pralidoxime iodide injection, PANI: Polyaniline, PNZ-PT: Pantoprazole phosphotungstate, PS: polystyrene microspheres, PTh: Polythiophene, $Pr_2Ce_2O_7$: Praseodymium cerate, p-rGO: Porous reduced graphene oxide nanostructure, rGO: Reduced graphene oxide, SBA-15: Santa Barbara-15, SC: Waste sponges, S-CSS: Irregular-edge carbon stacked sheet, SL-$Pr_6O_{11}$ NPs: Sponge-like praseodymium oxide nanoparticles, SL: Sponge-like, SPE: Screen-printed electrode, SWCNH: Single walled carbon nanohorn, SWV: Square wave voltammetry, TB: Terbium, xGnPs: Graphite nanoplatelets, β-CD: β-cyclodextrin

used for electrode modification in nanosensor design. Besides, these sources are also preferred as a carbon source for construction of carbon nanomaterials used as modifiers. The electrosensors developed through the contribution of green chemistry helped detection of pharmaceutical compounds from various matrices effectively.

## 5 Conclusion

Nanotechnology has revolutionized various fields, including environmental monitoring, healthcare, and energy storage. This advancement is very much related to the use of nanomaterials in electrochemical sensors, whose performance is significantly influenced by the properties of the nanomaterials used in their fabrication. While conventional nanomaterial synthesis methods often involve harsh chemicals and high energy consumption and thereby raise concerns about environmental sustainability, more recently, green synthesis has emerged as a promising alternative, utilizing environmentally friendly techniques to produce nanomaterials.

This study reviewed the selected literature produced in the last five years on the role of green synthesized materials in electrochemical sensors, their pharmaceutical applications, and the electrocatalytic impact. The literature revealed that biological agents such as plants, fungi, yeast, or enzymes obtained through environmentally safe methods could either be employed as reducing agents, transforming particularly metals into nanoparticles, hence enhancing their electrocatalytic activity, or as precursor materials used to create carbon nanoparticles in a variety of shapes, including nanofibers, nanotubes or nanoporous topologies. These nanomaterials obtained via less toxic, cheaper and simpler green techniques contribute not only to environmental sustainability but also to the development of quicker, easier, and more sensitive detection of pharmaceuticals in various matrices.

In conclusion, green synthesis of nanomaterials offers a sustainable and environmentally friendly approach to fabricating electrochemical sensors. By utilizing biological agents and avoiding harsh chemicals, green synthesis can produce nanomaterials with unique properties that enhance the performance of these sensors. The ability of nanomaterials derived from green synthesis to act as reducing agents and to be functionalized with specific groups further expands their potential applications. As research advances, green synthesis is poised to be crucial in developing innovative and sustainable electrochemical sensor technologies.

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