Construction of Electrochemical Sensors for Antibiotic Detection Based on Carbon Nanocomposites

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Abstract: Excessive antibiotic residues in food can cause detrimental effects on human health. The establishment of rapid, sensitive, selective, and reliable methods for the detection of antibiotics is highly in demand. With the inherent advantages of high sensitivity, rapid analysis time, and facile miniaturization, the electrochemical sensors have great potential in the detection of antibiotics. The electrochemical platforms comprising carbon nanomaterials (CNMs) have been proposed to detect antibiotic residues. Notably, with the introduction of functional CNMs, the performance of electrochemical sensors can be bolstered. This review first presents the significance of functional CNMs in the detection of antibiotics. Subsequently, we provide an overview of the applications for detection by enhancing the electrochemical behaviour of the antibiotic, as well as a brief overview of the application of recognition elements to detect antibiotics. Finally, the trend and the current challenges of electrochemical sensors based on CNMs in the detection of antibiotics is outlined.

Keywords: antibiotics; graphene; carbon nanotubes; functionalization; electrochemical activity; aptamers; molecular imprinting

1. Introduction

Antibiotics are chemicals that inhibit the growth of bacteria and even kill them. Since their discovery, antibiotics have considerably improved human and animal health. However, the abuse of antibiotics favours their presence in animals and food, leading to allergies, antibiotic resistance, and superinfections in humans [1–9]. Therefore, sensitive and selective methods are required to detect antibiotics. At present, numerous analytical methods have been constructed for the detection of antibiotics, such as high-performance liquid chromatography (HPLC), enzyme-linked immunosorbent assay (ELISA), capillary electrophoresis (CE), and liquid chromatography–mass spectrometry (LC–MS) [10–16]. Although these analytical methods exhibit high sensitivity and a low detection limit, they still have the disadvantages of expensive instruments and complex pretreatment processes.

Nowadays, rapid methods for the detection of antibiotics include colorimetric, fluorescent, and electrochemical methods [17–20]. Compared to these, electrochemical methods are attracting attention for their high sensitivity, high accuracy, and trace detection. However, traditional electrode materials have obvious drawbacks, such as the high potential value required for the electro-oxidation of these compounds [21–24]. Due to their excellent electrical conductivity, large surface area, and high electronic transport, the use of carbon nanomaterials (CNMs), such as graphene (Gr) and its derivatives, including graphene oxide (GO) and reduced graphene oxide (rGO), as well as carbon nanotube (CNT), including multi-walled carbon nanotube (MWCNT) and single-walled carbon nanotube (SWCNT), has been an effective strategy that has been gaining attention [25–32].
This paper presents a comprehensive overview of reported progress, in recent years, in the electrochemical application of CNMs to detect antibiotics (Figure 1). First, we provide a brief review of the modifications of CNMs. Then, the electrochemical signal amplification of antibiotics is summarized and the application of recognition elements for the detection of antibiotics is outlined. Finally, the challenge of CNMs in antibiotic development is discussed.

Figure 1. An overview of functional CNMs and their application in the detection of antibiotics.

2. Carbon-Based Nanomaterials

CNMs are considered a promising candidate for sensing antibiotic application. In order to endow CNMs with sensing capabilities, it is necessary to functionalize them with various strategies that boost the signal transmission and bring the detection targets to the surface through specific interactions [33–35]. Therefore, functionalized CNMs play a vital role in detecting antibiotics by electrochemical sensors. Different molecules used to modify CNMs have been selected and summarized in Table 1.

Table 1. Different molecules are used to modify CNMs, their processing methods, and their properties.

| Carbon Nanomaterials | Modification | Processing Method | Morphology | Properties | Ref. |
|----------------------|--------------|-------------------|------------|------------|------|
| CNTs                 | Ag NWs       | Van der Waals interaction | Network structure | Facilitate the electron transfer. Good sensitivity, electron-transfer rate, and surface area. | [36] |
| MWCNTs               | Au NPs       | In situ growth    | Network structure | Good sensitivity, electron-transfer rate, and surface area. | [37] |
| MWCNTs               | Au NPs (10 nm) and cDNA | In situ growth | Tube with dark dots | High biological binding sites and good conductivity. | [38] |
| GO                   | Au NRs       | Drop-casting      | Rod-shaped particles, dropped-plate pieces | Good surface conductivity and conductivity. | [39] |
| Gr                   | Au NP (15 nm) and PDDA | In situ growth and electrochemical polymerization | Wrinkled flakes with bright dots | Good sensitivity and high selectivity. | [40] |
At present, various strategies have been put in place to make the CNMs functional. The intentional introduction of noble metal nanoparticles (NM NPs) is an effective method and provides great performance. Due to the more significant number of active sites on the surface of NM NPs, the conductivity can be significantly increased, the redox potential of antibiotics can be reduced, and the catalytic efficiency can be increased [36–41,54]. Ayat Mohammad-Razdari et al. reported that gold nanoparticle (Au NP)/rGO nanocomposites were prepared by electrodeposition [55]. The combination of rGO and Au NPs as a nanocomposite provides higher electrochemical activity and high electron transfer capability. In addition, NM NPs can endow excellent biocompatibility with CNMs. Mahmoud Roushani et al. reported that nanocomposite prepared from GO and functionalized with (3-aminopropyl) triethoxysilane/silver nanoparticles (Ag NPs) [56]. This nanocomposite can successfully bind the aptamer by forming a Ag–NH₂ bond between Ag NPs and the NH₂ group of aptamers. In addition, building bimetallic nanohybrid components that modify CNMs to detect antibiotics is of interest. Neeraj Kumar and his colleagues observed that, electrochemically, rGO modified with gold–palladium nanoparticles (Au NPs–Pd NPs–ErGO) is highly resistant to interference [42]. In their study, Au NPs could enhance electronic conductivity and Pd NPs required stability for the fabrication of a stable, rapid, and efficient voltammetric sensor. The complexes indicate high sensitivity and the efficient electrocatalytic activity that facilitated the electron transfer.
Additionally, metal oxides can endow CNMs with remarkable catalytic capabilities and fast electron transfer [43–47,57]. As shown in Figure 2A,B, Gr and zinc oxide nanorods (Gr-ZnO) for the detection of sulfamethoxazole were successfully synthesized [48]. The composite of Gr and ZnO can exhibit high electric conductivity and significantly improve the electron transfer rate compared to the single materials due to the synergistic effect of using them together. In addition, the decoration of CNMs by magnetic spinel ferrite nanoparticles with the chemical formula MFe$_2$O$_4$ (M can be an element such as Ni, Cr, and so on) can enhance electron transfer and increase sensitivity. For example, the NiFe$_2$O$_4$ decorated rGO was successfully synthesized via a solvothermal method because of its synergistic effect. It can enhance electron transfer, speeding up the electron transfer rate and increasing sensitivity [49].

The combination of CNMs and polymers also led to satisfactory results in this field [49–51,58–60]. The polymelamine film on ErGO was designed using a potentiodynamic method for the determination of tetracycline [61]. The three amine groups with a triazine ring make the polymelamine useful for polymerization, and the nitrogen-rich matrix with many π-electrons can interact with the target molecules. The synergistic effect of melamine and ErGO can bolster electrochemical activity.

Thanks to the sustainable development of CNMs, researchers have been devoting themselves to developing new carbon nanocomposites to improve the detection limit and performance of electrochemical sensors for antibiotics. Xian Fang et al. reported that NH$_2$-UiO-66 and rGO composites were successfully synthesized by a solvothermal method (see Figure 2C,D) [52]. In this strategy, the NH$_2$-UiO-66 possesses a large surface area, and the introduction of rGO further increases the surface area of the nanocomposite and facilitates the adsorption of antibiotics. Moreover, ionic liquids are commonly used as surface modifiers because of their high conductivity, wide potential window, high viscosity, and low volatility [53]. The use of electrodes with ionic liquid associated with GO improves absorption.

3. Recent Reports on Antibiotic Detection by the Sensor

3.1. Electrochemical Behaviour of Antibiotics

The electrochemical behaviour of antibiotics is determined by the fundamental skeleton and the derived functional groups. Given the limitations of antibiotic redox behaviour, adding CNMs to electrodes could be an effective way to address this problem. A brief
list of the electrochemical behaviour of commonly used antibiotics has been selected and summarized in Table 2.

Table 2. CNM-based electrochemical sensor for antibiotics.

| Antibiotics | Detection Principle | Electrode | Linear Range | LOD  | Ref. |
|-------------|---------------------|-----------|--------------|------|------|
| TET         |                     | Ta$_2$O$_5$/ErGO/GCE | 0.2–10 µM | 0.095 µM | [43] |
| Amoxicillin |                     | L-Cys/GR/GCE | 8.0–140 µM | 0.12 µM | [51] |
|             |                     | p-Mel@ErGO/GCE | 5–225 µM | 2.2 µM | [61] |
|             |                     | ErGO/SPE | 20–80 µM | 12 µM | [62] |
|             |                     | Au NPs/Gr/Laccase/GCE | 0.02–310 µM | 0.36 µM | [63] |
| Amoxicillin |                     | Au NPs/en-MWCNTs/SPE | 0.2–10 and 10–30 µM | 0.015 µM | [37] |
| NiFe/O4/MWCNTs/GCE | 0.425–292 µM | 425 nM | [64] |
| NiFe/f-MWCNTs/GCE | 0.6–8.0 and 10.0–80.0 mM | 0.2 µM | [65] |
| β-CD/MWCNT | 10–80 µM | 50 nM | [24] |
| NiO NPs/GO/GCE | 0.04–0.97 µM | 6 nM | [46] |
| NH$_2$-UiO-66/RGO/GCE | 0.02–1 µM | 6.67 nM | [52] |
| Porous-Nafion-MWCNT/BDD | 0.005–10 µM | 5 nM | [66] |
| Poly (L-Cysteine)/Au NPs/rGO | 0.001–100 nM | 3 pM | [49] |
| Levofoxacin  |                     | GO/IL/CPE | 7–700 nM | 0.28 nM | [53] |
| Nitrofurantoin | NiFe$_2$O$_4$/rGO/GCE | 0.09–670 µM | 7.92 pM | [67] |
|            | NiFe/r-MWCNT/SPCE | 0.1–24.8 µM | 0.03 µM | [69] |

3.1.1. Tetracyclines

Tetracycline antibiotics, including tetracycline (TET) and oxytetracycline (OTET), have electroactive groups in their chemical structures that can be electrochemically detected [53,70–72]. Taking TET as an example, as shown in Figure 3A, when the potential is applied to the electrode, part of TET forms a positively charged structure which loses two electrons and a proton directly (the first peak), as the first oxide peak shows. Next, it will be combined with the strong negative oxygen in the hydron, thereby generating a new substance that contains the structure of quinone (the second peak). The two peaks can be clearly shown in Figure 3B [62]. In this way, we can construct an electrochemical sensor with CNM composites for the detection of TET.
Figure 3. (A) Mechanism for the electrochemical oxidation of TET. (B) Adsorptive transfer stripping differential pulse voltammograms corresponding to different mixtures of tetracyclines using ErGO. The black, red, and blue lines are different concentrations of the TET antibiotic mixture. Adapted with permission from Ref. [62].

Due to the synergy between GO and MWCNT, the composite nanomaterial formed by GO and MWCNT increased the electroactive area of the electrode, thereby increasing the electrochemical signal and the stability of the sensor. Ademar Wong’s team reports that an electrochemical sensor based on an electrode modified with a combination of MWCNT and GO was developed for the sensitive and selective determination of TET [63]. Under the optimal conditions, the sensor showed a limit of detection (LOD) of 0.36 µM. The sensor was successfully used to detect tetracycline in river water, artificial urine, and pharmaceutical samples without any sample pretreatment.

Srinivasan Kesavan et al. designed an rGO combined with polymelamine film and modified GCE for electrochemical detection of TET [61]. There are three triazine rings on the three amino groups of melamine so that the molecules can be used for polymerization and the nitrogen-rich matrix with many π-electrons can interact with the target molecules. The results show that the current response increases linearly with the concentration from 5.0 µM to 225 µM, and the LOD is 5 µM. Moreover, the method was also used for the detection of uric acid, which shows the excellent interference resistance of the scheme. Not only polymelamine but also L-Cys can be electrodeposited on CNMs to produce a strongly adsorbed polymer film. A novel electrochemical sensor based on the Gr/L-Cys composite film modified electrode was prepared by Xuemei Sun’s team [51]. The Gr/L-Cys composite film has a uniform morphology with a diameter ranging from 50 to 100 nm, which is beneficial to the charge conduction and the adsorption of tetracycline. Under the optimized conditions, it exhibited a good linear relationship in the concentration range from 8.0 to 140 µM, and the LOD was 0.12 µM. The prepared electrode showed a practical prospect in real samples, recovering from 89.60 to 105.20%.

The use of metal peroxides also showed excellent results. An electrochemical sensor for the determination of OTET by GCE modified by Ta$_2$O$_5$-rGO nanocomposites was reported by Felista Magesa et al. [43]. Ta$_2$O$_5$-rGO composites have a large electrochemically active area that amplifies OTET oxidation signals, significantly increasing the electrochemical performance. Detection in milk samples showed that, except for a good recovery rate of 100.1–120.9%.

3.1.2. Amoxicillin

Amoxicillin can present a weak voltammetric signal on electrodes, but CNM-modified electrodes can enhance this signal [37,45,64,65,73]. As shown in Figure 4A, the electrochemical response showed an oxidation peak, which is due to the oxidation reaction of the phenolic (–OH) substituent to the respective carbonyl group (=O) on the side chain of the molecule [71]. In the study by Behzad Rezaei and Sajjad Damiri, amoxicillin yielded a clear oxidation peak on the MWCNT-modified electrode at nearly 1.0 and 0.6 V for pH 3.0 and 7.5, respectively, and the oxidation peak current significantly increased compared with that.
at the bare electrode. Neeraj Kumar et al. [72] reported that AuNP-PdNP-ErGO was used to modify GCE for the determination of amoxicillin [42]. The synergy between Au and Pd nanoparticles on rGO improved sensor sensitivity and performance, thereby improving charge transfer and creating a large number of adsorption sites on the modified GCE surface. As shown in Figure 4B, the peak current of amoxicillin was found to increase with the increasing concentration of amoxicillin. Furthermore, this sensor has been developed for the individual and simultaneous determination of lomefloxacin and amoxicillin, and thereby, this shows its excellent selectivity. The LOD was as low as 81 nM for lomefloxacin and 9 \( \mu M \) for amoxicillin. The method was successfully applied to the detection of the presence of lomefloxacin and amoxicillin in the complex matrix.

3.1.3. Quinolones

The redox behaviour of quinolone antibiotics allows their electrochemical study. Ciprofloxacin is a third-generation quinolone antibiotic widely used in humans and livestock. The reaction pathway of ciprofloxacin is based on the oxidation of the terminal nitrogen atom within the piperazine ring (as shown in Figure 5A) [24]. Jorge M.P.J. Garrido’s team took advantage of cyclodextrin (CD) to chemically identify the guest molecules, resulting in a novel polyaniline (PANI-\( \beta \)-CD/MWCNT) that significantly improved the electron transfer rate [66]. Under optimized conditions, a linear calibration curve was obtained for ciprofloxacin in the concentration range of 10–80 mM with an LOD of 50 nM. In order to achieve rapid determination of trace antibiotics, the behaviour of heavy metals that can form stable complexes with ciprofloxacin attracted attention. Xian Fang and his team reported an electrochemical sensor for detecting ciprofloxacin using NH\( _2 \)-UiO-66 and rGO composites as working electrodes [52]. In the presence of ciprofloxacin, the oxidation current peak of Cu\( ^{2+} \) decreases, apparently due to complex formation. The electrochemical sensor shows a low LOD (6.67 nM) and high sensitivity (10.86 \( \mu A/\mu M \)) based on this principle. Meanwhile, the electrochemical sensor can detect ciprofloxacin in real water samples with satisfactory recovery.

In addition, electrodes modified with nickel oxide nanoparticles (NiO NPs) have a remarkable catalytic capacity. Anderson Martin Santos et al. report a simple and highly selective electrochemical method for simultaneous determination of paracetamol and ciprofloxacin based on the binding advantages of GO and NiO NPs [46]. The sensor shows an LOD of 6.7 and 6.0 nM, respectively (see Figure 5B). The boron-doped diamond (BDD) electrode substrate was modified as the MWCNTs, which are dispersed in a porous Nafion film to develop a synergistically amplified electrochemical sensing platform for the detection of ciprofloxacin. As shown in Figure 5B, ciprofloxacin showed the highest peak at the porous-Nafion-MWCNT/BDD electrode, and DPV studies revealed an LOD of 5 nM [67].
Figure 5. Mechanism for the electrochemical levoxidation of ciprofloxacin (A) and ofloxacin (C). (B) DPV curves of 50 µM ciprofloxacin on BDD, Nafion/BDD, porous-Nafion/BDD, Nafion-MWCNT/BDD, and porous-Nafion-MWCNT/BDD electrodes in 0.1 M KH$_2$PO$_4$ solution (pH = 4.50). Adapted with permission from Ref. [67]. Copyright 2016 ACS. (D) DPV response was obtained for Ag/AgVO$_3$/N-rGO/SPCE by linear addition of levofloxacin in the N$_2$ environment. Adapted with permission from Ref. [74]. Copyright 2021 ACS.

The overall possible explanation for the electron transfer mechanism toward levofloxacin is represented in Figure 5C. Levofloxacin shows an oxidation peak variation, which is recorded for the peak produced by the piperazine moiety by a two-step condensation emitting one electron each (as shown in Figure 5D) [68,70,74]. The use of ionic liquids as modifiers on the surface of electrochemical sensors can improve compounds’ stability, sensitivity, and selectivity for oxidation and/or reduction. An electrochemical sensor based on a carbon paste electrode modified with GO and ionic liquids for the sensitive voltammetric determination of ofloxacin [53]. It shows a low LOD of 0.28 nM.

3.1.4. Nitrofurans

Nitrofurans are synthetic broad-spectrum antibiotics with 5-nitrofuran as their basic structure. The electrochemical activity of furan is related to its irreversible reduction of the nitro (–NO$_2$) to nitroso intermediate, which is rapidly reduced to the corresponding hydroxylamine group (–NHOH), as shown in Figure 6 [48,69,75]. Kuo-Yuan Hwa and Tata Sanjay Kanna Sharma reported using a NiFe/MWCNTs hybrid composite as an electrocatalyst to detect nitrofurantoin. The composite has a high surface area and high electron transport, which reduces the charge-transfer resistance. The results show that the composite has good conductivity and excellent electrocatalytic activity. The LOD is 0.03 µM and the sensitivity is 11.45 µA µM$^{-1}$cm$^{-2}$. The NiFe$_2$O$_4$/rGO nanocomposites were prepared using a hydrothermal method. The high specific surface area and better electrical conductivity of graphene help to improve the signal. In addition, graphene as a carrier modified by NiFe$_2$O$_4$ nanoparticles can prevent any aggregation of these particles [75]. The electrochemical sensor has a linear correlation with the concentration of furazolidone in the linear range of 0.1–10.0 µM and 10.0–150.0 µM, and the LOD is 0.05 µM.
3.2. Application of Recognized Components in the Detection of Antibiotics

Aptamers and molecularly imprinted polymers (MIPs) were also used to detect antibiotics in order to improve binding between antibiotics and electrodes.

3.2.1. Aptamer

Aptamers, due to their remarkable virtues, including a wide variety of specific targets, excellent stability, and cost-effectiveness, have aroused much attention from researchers. The introduction of CNMs can significantly enhance the performance of the sensors, as shown in Table 3 [56,76–82]. An aptamer-based impedance method for the determination of kanamycin was reported by Azadeh Azadbakht and Amir Reza Abbasi [41]. In this report, molybdenum selenide nanoflowers (MoSe$_2$) were synthesized by a hydrothermal method, and then CNTs were modified with MoSe$_2$ and Au NPs as a signal amplifier. The results showed that the calibration curve was linear in the concentration range of 1 pM–0.1 nM kanamycin, and the LOD was 0.28 pM. The recovery of kanamycin in milk ranged from 96.2 to 101.5%, indicating that the method has been successfully applied to the detection of kanamycin in actual samples. Moreover, a sensor for the detection of kanamycin was prepared using the strong physical adsorption between 15-mer of poly-C and GO [82]. As shown in Figure 7, MWCNTs play a role in amplifying electrochemical signals. The designed diblock DNA includes an optimal 15-mer of poly-C and another block aptamer of kanamycin. Therefore, diblock DNA can be immobilized on GO and used for hybridization with targets. The sensor showed high sensitivity and specificity to kanamycin, with a linear range of 0.05 pM to 100 nM and an LOD of 0.0476 pM. Furthermore, the scheme has been successfully applied to the detection of milk. The results show that the scheme has a good application prospect in the detection of antibiotics in food. While indications have significant advantages, they are also limited by environmental variables such as salt concentration and pH value. Those variables also increase the complexity of their development.
Table 3. CNMs-based aptasensors for antibiotics.

| Antibiotics   | Aptamer Sequence                                                                 | Electrode                        | Linear Range         | LOD             | Ref.   |
|---------------|----------------------------------------------------------------------------------|----------------------------------|----------------------|-----------------|--------|
| TET           | CGTACGGAAATTCGCTAGCCCCCCCCGCAAGCCACGGCTT GGGTTGTCACCACGGGCGGATCCAGCTCCACGT-3' | Apt/MWCNTs/GCE                   | 10 nM–50 µM          | 5 nM            | [78]   |
|               | 5'-SH-GGAATTCGCTAGCGCTGGGCTGGCCGGTT GGGTTGTCAGCTCCACGT-3'                      | cMWCNTs/Au NPs/cDNA/Apt/TFGE    | 1 × 10^{-13}–1 × 10^{-5} g/mL | 3.1 × 10^{-14} g/mL | [38]   |
| OTET          | 5'-NH2-CGTACGGAAATTCGCTAGCCCCCCCCGCAAGCCACGGCTT GGGTTGTCACCACGTCCACGT-3'       | Apt/GO/GCE                       | 0.1 pM–10 µM         | 29 fM           | [79]   |
|               | 5'-SH-CGACGCACAGTCTGTGGATCCTGGTGTGACGT-3'                                     | Apt/Au NPs/HRP&Au NPs/Gr/GCE     | 5 × 10^{-10}–2× 10^{-3} g/L | 4.98 × 10^{-10} g/L | [81]   |
| Kanamycin     | 5'-NH2-(CH2)6-TGGGGTGGTGATAGTTAACAGCCGAC-3'                                    | Apt/Au NP/CNT/MoSe2/GCE          | 1 pM=0.1 nM and 100 nM–10 µM | 0.28 pM        | [41]   |
| Streptomycin  | 5'-NH2-(CH2)6-TAGGGAAATTCGCTAGCGCTGGGCTGGATAGTTAACAGCCGAC-3'                   | Apt/CNT/Chi/Pd NPs/GCE          | 0.1–1500 nM          | 18 pM           | [76]   |
|               | 5'-NH2 -AGATGGGGTTGAGGCTAAGCCGAC-3'                                           | Apt/HNP-PtCu/Gr-Thionine/GCE     | 5 × 10^{-7}–5 × 10^{-2} µg/mL | 0.42 pg/mL     | [77]   |
| Chloramphenicol | 5'-NH2 -ACTTCAGAGGTTTCACCAGGTGAGTGGTGGTGGTGGTAG-3'                              | Apt/Ag NPs/[NH2-Si]-f-GO/GCE     | 10 pM–0.2 µM         | 3.3 pM          | [56]   |
3.2.2. Molecular Imprinting Polymers

Molecularly imprinted polymers (MIPs) are composites of functional monomers, crosslinkers, and template molecules that can withstand a more comprehensive pH and temperature range than biomaterials. Therefore, MIPs have been widely used in the preparation of electrochemical sensors for the detection of antibiotics [40,82,83]. CNTs have been studied more extensively as carriers of MIPs. The scheme of acrylamide functionalized MWCNTs used to synthesize molecularly imprinted polymers to detect kanamycin was reported by Benzhi Liu et al. for the first time [84]. The results show that the detection range is 4 to 50 µM and the LOD is 0.68 µM. Moreover, the sensor has a good recovery rate of about 94.0–95.3% in milk and milk powder. Bimetallic particles are also used to increase their pore size to enhance the response signal. A new electrochemical sensor was composed of MWCNTs-MIP, SWCNTs, and dendritic platinum and palladium nanoparticles [85]. The ion exchange strategy was used for the first time to branch ionic liquids onto the surface of MWCNTs to prepare MIP. The function of MWCNTs is to increase the specific surface area of the electrode and increase the electrocatalytic activity of amoxicillin. The results show that the sensor obtained under the optimal conditions has a linear response to amoxicillin with a low LOD of 8.9 nM, in the range of 1.0–1000 nM. The prepared electrode was successfully applied to the determination of amoxicillin, with the recovery in honey and milk samples showing a practical prospect in real samples.

4. Conclusions and Prospect

This paper reviewed the latest progress in the electrochemical detection of antibiotics based on CNMs. CNM-based composites have electrocatalytic activity, good electrical conductivity, and a large specific surface area. They not only catalyze the redox reaction of antibiotics, but are also compatible with existing biometrics. Furthermore, carbon-based electrochemical sensors have been successfully developed as a rapid and sensitive method for accurate monitoring of antibiotics in food or other substances. With the rapid development of interdisciplinary cooperation, carbon-based sensors will shine in many fields, such as food, biomedicine, the environment, and so on. Recently, portable detection devices have gained popularity among researchers. Based on the excellent malleability of CNMs, the development of portable detection devices based on CNMs for the detection of antibiotics by electrochemical detection methods is also a hot topic for the future.
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