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REVIEW ARTICLE

Recent development of antibiotic detection in food and environment: the combination of sensors and nanomaterials

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Abstract

In recent years, the abuse of antibiotics has led to the pollution of soil and water environment, not only poultry husbandry and food manufacturing will be influenced to different degree, but also the human body will produce antibody. The detection of antibiotic content in production and life is imperative. In this review, we provide comprehensive information about chemical sensors and biosensors for antibiotic detection. We classify the currently reported antibiotic detection technologies into chromatography, mass spectrometry, capillary electrophoresis, optical detection, and electrochemistry, introduce some representative examples for each technology, and conclude the advantages and limitations. In particular, the optical and electrochemical methods based on nanomaterials are discussed and evaluated in detail. In addition, the latest research in the detection of antibiotics by photosensitive materials is discussed. Finally, we summarize the pros and cons of various antibiotic detection methods and present a discussion and outlook on the expansion of cross-scientific areas. The synthesis and application of optoelectronic nanomaterials and aptamer screening are discussed and prospected, and the future trends and potential impact of biosensors in antibiotic detection are outlined.

Keywords Biosensor · Antibiotic detection · Aptasensor · Nanoparticles · Food analysis · Environmental samples

Introduction

Antibiotics are a kind of chemical substances produced by some microorganisms in the life process, which can inhibit or kill some other pathogenic microorganisms and have bacteriostatic or bactericidal effects. They can be classified into β-lactams (BLCs), macrolides antibiotics, fluoroquinolones (FQs), aminoglycosides (AGSs), tetracyclines (TCs), phenicols, and sulfonamides (SAs) antibiotics [1], which mainly treat infections caused by micropathogens and are commonly used in the prevention of bacterial growth in plants and livestock [2]. Moreover, they can be used in medical field, such as respiratory infections [3], orthopedics [4, 5], surgery [4], neurological diseases [6], and even cancer treatment [7].

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However, overusing antibiotics in intensive aquaculture become a latent menace to human health due to adverse reactions, including anaphylaxis, impaired liver function, and ototoxicity [8–14]. So the efficient detection of trace antibiotics is important for studying environmental antibiotic residues, metabolism, and environmental effects [15]. Therefore, especially in food and water environment, it is very important to find a suitable method to measure the content of antibiotics to ensure the safety of our production and life.

Recent years, numerous efforts have been made to develop analytical techniques for trace amounts antibiotic detection, such as high-performance liquid chromatography (HPLC) [16], mass spectrum (MS) [17, 18], and capillary electrophoresis (CE) [19] with low detection limit and high accuracy and precision. Pang et al. [20] established an in-tube solid-phase microextraction combined with high-performance liquid chromatography and fluorescence (IT-SPME-HPLC-FLD) for fluoroquinolone (FQs) detection, which were excellent in water and honey. Cui et al. [21] detected trace tetracycline (TC) in water by high-performance liquid chromatography-tandem mass spectrometry (HPLC-MS/MS). This kind of novel diol-based porous organic polymer (NTdiol-POP) was used as a new SPE material with a detection limit of 0.16–4.53 pM.

However, these techniques still have the drawbacks of expensive instruments, complicated pretreatment process, and so on. Therefore, convenient and low-cost detection method with high accuracy and sensitivity is desirable, such as colorimetry [22], fluorescence [23], electrochemistry [24], chemiluminescence [25], electrochemiluminescence [26], and photoelectrochemistry [27]. Among these methods, photoelectrochemical technique has begun to attract attention due to high sensitivity and specificity as well as low background current, in which the nucleic acid aptamers play an integral role. DNA aptamer is a synthetic oligonucleotide sequence, which can bind to target molecules with high specificity and activity. As early as 1990, Ellington et al. established a strategy for artificially obtaining aptamers from nucleic acid libraries [28], and aptamers have become brilliant in many aspects such as cell imaging, new drug development [29, 30], disease treatment [31, 32], and microbial detection [33]. Compared with antibody or receptor proteins, DNA aptamers are inexpensive and easy to synthesize and mix with good thermal stability and lacks immunogenicity and non-toxicity. Zeng et al. [34] designed a zero-backgrounded fluorescence aptasensor by repeatedly reacting chloramphenicol with the probe on the rod, replacing a large number of long DNA strands with the supernatant to fluoresce. Sui et al. [35] measured chloramphenicol content by reducing photocurrent due to the interaction between g-C_3N_4 nanosheets and ferrocene-aptamers via photoelectrochemical method, which reached a low detection limit of 0.22 pM (3σ) and 94.5 to 107.3% recovery rate.

Based on the aptamer detection method, nanomaterials have received extensive attention due to properties of surface and interface effects and small size effects. In the detection of antibiotics, many semiconductor materials, such as TiO_2 [36–40], rGO [41–50], and g-C_3N_4 [35, 51–54], play an important role in fluorescence and electrochemical methods due to their unique electronic effects and optical properties. Wang et al. [38] detected chloramphenicol with a low detection limit of 57.9 pM (S/N = 3) on the use of TiO_2 based nanorod array (NRA) sensitized with Eu(III)-doped CdS quantum dots. Qin et al. [40] used TiO_2 nanotube array-polydopamine-Ag_2S structure to bind aptamer-antibiotics bioaffinity complexes, which obtained a 0.75-pM detection limit (at S/N = 3) of ofloxacin (OFL) in spiked milk samples. Dang et al. [53] used g-C_3N_4 quantum dot–modified reduced graphene oxide to prepare sulfadimethoxyl photoelectric sensor, which has a linear response in the 0.5- to 80-nM SDM concentration range, with a 0.1-nM detection limit (at S/N = 3). Despite the sensitivity and accuracy of these materials, it is an essential requirement to develop materials study in terms of doping modification, film formation, and mechanism of action. The review by Mehlhorn et al. [55] classifies antibiotics and summarizes in detail the aptamer biosensors used to detect antibiotics. Dong et al. [56] reviewed the application of low-dimensional nanomaterials in the detection of antibiotics. Ahmed et al. summarized receptor-based screening assays [57] and immunoassays [58] for the detection of food antibiotic residues in 2017 and 2020, respectively.

Therefore, this review systematically discusses analytical techniques for antibiotic residues detection. At the same time, the optical and electrochemical detections are discussed in detail as well as the advantages and disadvantages of each method. Herein, we give a future perspective including the challenges and opportunities of antibiotic detection.

Classification of antibiotic detection methods

We classify antibiotic detection methods based on different physical signal results, including chromatography, mass spectrometry, capillary electrophoresis, optical detection, and electrical detection. In this section, the principles, progress, advantages and disadvantages, and method evaluation of each test method will be described in detail.

Chromatography

High-performance liquid chromatography is a traditional analytical method that has been widely used for the analysis of antibiotics in different samples. The essence of the chromatographic process is the distribution balance of the molecules to be separated between the stationary phase and the mobile phase. The distribution of different substances in the two phases will be different, which makes it move at different speeds with the mobile phase. The movement of the mobile phase separates the different components in the mixture from each other in the stationary phase. High-performance liquid chromatography fluorescence detection (HPLC-FLD) has high sensitivity, flexibility, and easy operation and is widely used [59]. However, due to the very low concentration of antibiotics in actual samples and complex matrices, proper sample pretreatment must be performed first. The current methods for enriching antibiotics include liquid-liquid extraction (LLE), liquid-liquid microextraction (LLME), solid-phase extraction (SPE), solid-phase microextraction (SPME), magnetic SPE (MSPE), etc. [60–70]. Among them, SPME technique has been widely applied in the field of environmental chemical analysis since it was proposed by Pawliszyn of Waterloo University in Canada and its collaborator Arthur in 1990 [71].

In a recent example, Pang et al. [20] prepared a new metal-organic framework-monolith composite for in-tube solid-phase microextraction phase (IT-SPME) of fluoroquinolones (FQs). Firstly, poly(VA-co-ED)monolith were prepared by in situ polymerization. After ultrasonic treatment of the mixed solution of precursor, the homogeneous solution was injected into the fused silica capillary column modified with γ-MAPS.

After polymerization, the capillary was connected to the high-pressure pump and flushed with methanol. Then, Zn^{2+} and imidazole were used to form the ZIF-8 in the monolith, which was fixed on the polymonomer (VA-co-ED) by coordination bond. After 15 cycles of alternating flow of $Zn(NO_3)_2$ and imidazole methanol, MM-CMC was successfully synthesized. The introduction of ZIF-8 improved the extraction performance of FQs, and the applicability of the method was verified by the ultratrace analysis of FQs in the environmental water and honey samples. The method has the advantages of high sensitivity, convenient operation, partial automation, low sample and solvent consumption.

Negarian, M et al. [72] successfully synthesized core-shell lincomycin-imprinted polymers (Fig. 1). The lincomycin (LIN) in the milk matrix was preconcentrated and purified by the core-shell molecularly imprinted solid-phase extraction (CSMISPE) method and then analyzed by high-performance liquid chromatography equipped with UV detector (HPLC-UV). The test range is on the order of micrograms per milliliter and can be used to measure local pasteurized milk samples.

Using the same method in another analysis, Wang et al. [73] determined 28 veterinary drug residues in pig production wastewater by high-performance liquid chromatography-tandem mass spectrometry. Experiment using orthogonal test method was carried out on the parameters that influence efficiency of d-SPE system research; the limits of detection and limits of quantification for all the analytes ranged from 0.002 to 0.200 ng mL^{-1} and 0.005–0.500 ng mL^{-1}, respectively, which can be used for routine monitoring of pig antibiotic wastewater.

Mass spectrometry

Mass spectrometry is a spectroscopic method juxtaposed with the spectrum. Generally speaking, it refers to a special technique widely used in various disciplines to identify compounds by preparing, separating, and detecting gas phase ions. The basic principle is to ionize the components in the sample to generate charged ions with different charge-to-mass ratios. An accelerated electric field forms an ion beam into the mass analyzer. In the mass analyzer, the electric and the magnetic field are reused to cause the opposite velocity dispersion, and they are respectively focused to obtain a mass spectrum for determining the mass [74].

Organic mass spectrometers are classified into gas chromatography-mass spectrometry (GC-MS), liquid chromatography-mass spectrometry (LC-MS), matrix assisted laser desorption time-of-flight mass spectrometry (MALDI-TOFMS), Fourier transform mass spectrometry (FT-MS), and so on due to different application characteristics. Inorganic mass spectrometers include spark source dual focus mass spectrometer, inductively coupled plasma mass spectrometer (ICP-MS), and secondary ion mass spectrometer (SIMS). GC-MS subdivision includes gas chromatography-quadrupole mass spectrometry (GC-QMS), gas chromatography-time-of-flight mass spectrometry (GC-

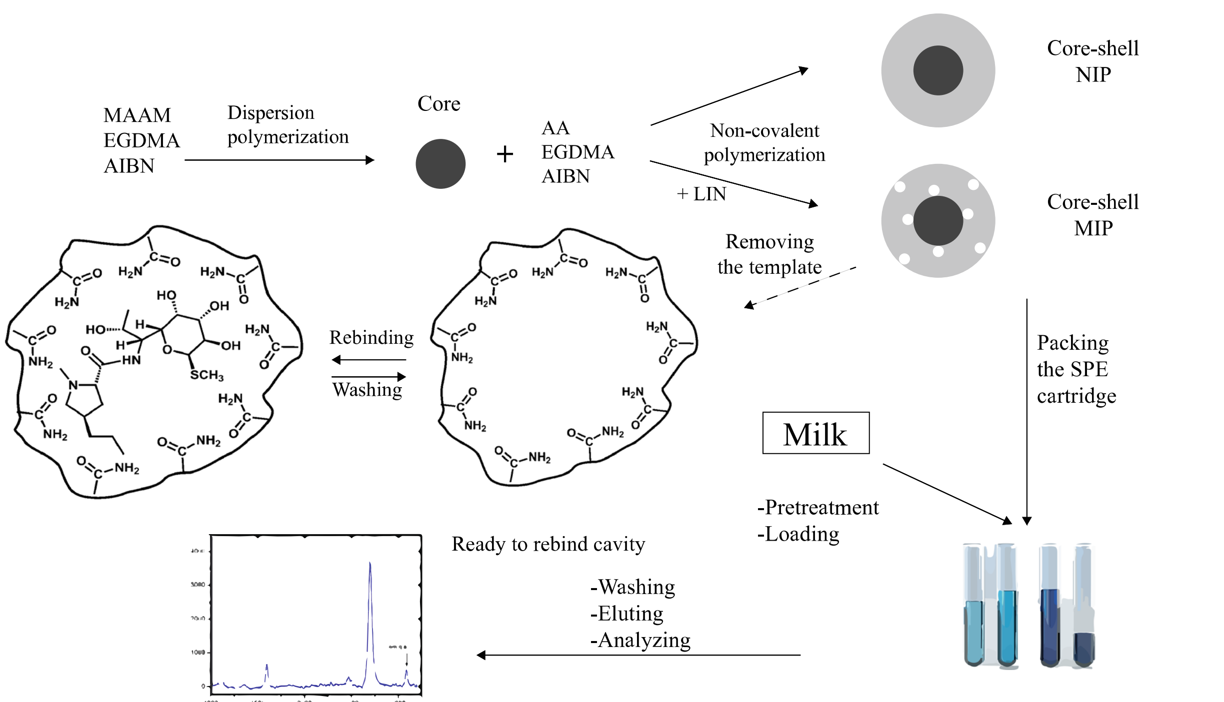

Fig. 1 Schematic illustration of core-shell polymers synthesis and process of CSMISPE-HPLC-UV for LIN residue checking in pasteurized milk sample. Reproduced from Negarian et al. (2019) with permission from Elsevier [72]

TOFMS), gas chromatography-ion trap mass spectrometry (GC-ITMS), etc. LC-MS subdivision has liquid chromatography-quadrupole mass spectrometry (LC-QMS), liquid chromatography-ion trap mass spectrometry (LC-ITMS), liquid chromatography-time of flight mass spectrometry (GC-TOFMS), a variety of liquid chromatography-mass spectrometry-mass spectrometry (LC-MS/MS), and so on.

He et al. [75] proposed a rapid and direct analysis method for seawater samples detection based on droplet spray ionization mass spectrometry. As shown in Fig. 2, the target analyte can be separated from the salt by spontaneous desalination (SD) of the droplet seawater covering the slip angle, and then, the separated target can be dissolved in the spray solvent for MS analysis, so as to measure antibiotics like trimethoprim and clarithromycin in the seawater. They solved the problem that non-volatile salts such as NaCl competed with analyte ions in the process of forming droplets in the outer surface space. Furthermore, it helps to remove the number of ions reaching the mass spectrometer and causing negative effects and serious ion inhibition.

Among these methods, LC-MS/MS is a commonly used method for the analysis of antibiotic residues [61, 64, 76–80]. Cas et al. [81] measured the daptomycin content in human plasma and breast milk by UPLC-MS/MS, which provided a possibility for the study on the possible toxicity of antibiotic drug residues in breast milk to infants during lactation. As the adsorbent has a large specific surface, suitable pore structure and surface structure, it has a strong adsorption capacity for adsorbates. In order to better enrich the sample, Xu et al. [82] simultaneously tested mycotoxin and veterinary drugs and pesticides in egg samples by ultrahigh-pressure liquid chromatography-tandem mass spectrometry (UPLC-MS/MS). Magnetic multi-walled carbon nanotubes (Fe_3O_4MWCNTs) were used as adsorbents in the purification process to achieve faster adsorbent separation using external magnets. Using the same method, Zhao et al. [65] used core-shell nanocomposite Fe_3O_4@MoS_2 as an adsorbent to detect sulfonamide antibiotics (SAs) by HPLC-MS/MS (Fig. 3). The electrostatic interaction between positively charged SAs and negatively charged MoS_2 nanoparticles can enhance the adsorption of SAs on the surface of the adsorbent, which is more conducive to the extraction and detection of real samples (Fig. 4).

Capillary electrophoresis

Capillary electrophoresis (CE) is a new type of liquid phase separation technology that uses a capillary as a separation channel and a high-voltage DC electric field as a driving force [83–85]. The microfluidic analysis system combines the principles and techniques of conventional CE and flow injection sampling technology, so CE has also been well applied in the micrototal analysis system [86, 87]. CE actually contains electrophoresis, chromatography, and its cross-over content. It enables analytical chemistry to go from microliter to nanoliter level and makes single cell analysis and even single molecule analysis possible. The microfluidic analysis system can increase detecting speed and dramatically reduce the cost. The microfluidic CE analysis system is also commonly referred to as integrated capillary electrophoresis (ICE). This system has opened a new chapter in the history of analytical science and brought analytical science into a new world of miniaturization, integration and automation [88–90].

Microchip electrophoresis is an efficient and rapid method to detect antibiotics in environment or food, and CE is a good choice for multiple antibiotic testing [87, 89, 91, 92]. Zhang et al. [87] amplified signal transduction on the basis of a novel stir-bar assisted DNA multi-arm junctions recycling (MAJR) strategy based on a universal dual-t microchip to perform rapid antibiotic detection, as well as chloramphenicol (CAP) and kanamycin (Kana), as is shown in Fig. 5. The detection limits were 1.61 aM for CAP and 0.7 aM for Kana (S/N = 3).

Ma et al. [69] combined offline dispersive liquid-liquid microextraction (DLLME) and online field-amplified sample injection (FASI) technology for CE separation, which can simultaneously detect 4 antibiotics (Fig. 6). And lower detection limits for sulfamethazine (SMZ), sulfamerazine (SMR), sulfadiazine (SDZ), and sulfacetamide (SFA) were obtained, which ranged from 7.2 to 82.6, 7.9 to 93.4, and 15.4 to 226.3 nM in tap water, lake water, and seawater, respectively.

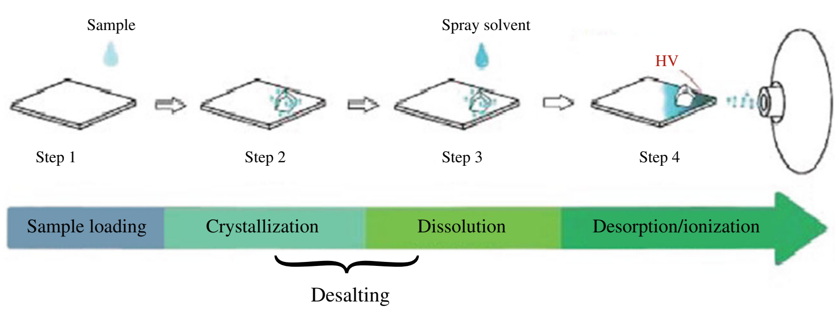

Fig. 2 Schematic setup of SD-DSI. The workflow for sample analysis includes four steps: sample loading, crystallization, dissolution, and desorption/ionization, namely, step 1 to step 4. Reproduced from He et al. (2019) with permission from Royal Society of Chemistry [75]

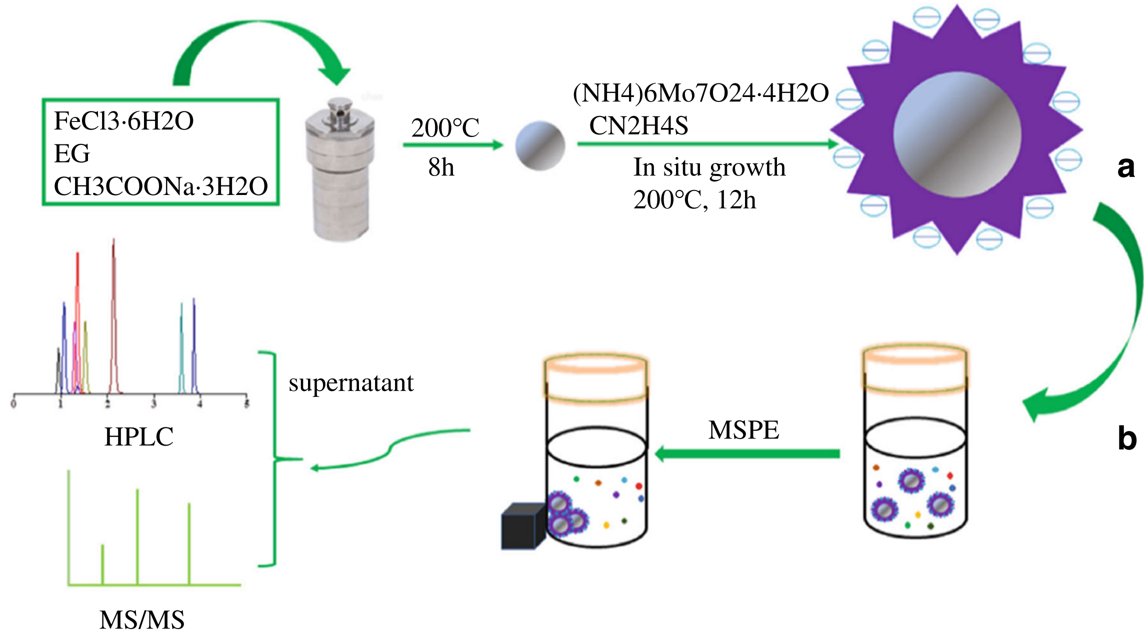

Fig. 3 Schematic diagram for the preparation of the core-shell Fe_3O_4@MoS_2 nanocomposites (a) and their application for enriching SAs (b). Reproduced from Zhao et al. (2019) with permission from Elsevier [65]

Optical detection

Optical inspection can be divided into colorimetry, fluorescence, surface plasmon resonance, chemiluminescence, and surface-enhanced Raman scattering. The advantage of optical detection lies in the fact that even small products can be easily zoomed in to complete the detection of small molecule concentrations like antibiotics or proteins with high accuracy, which are unimaginable in the past.

Colorimetry

Colorimetry is a method to determine the content of components by comparing the color depth of a substance solution.

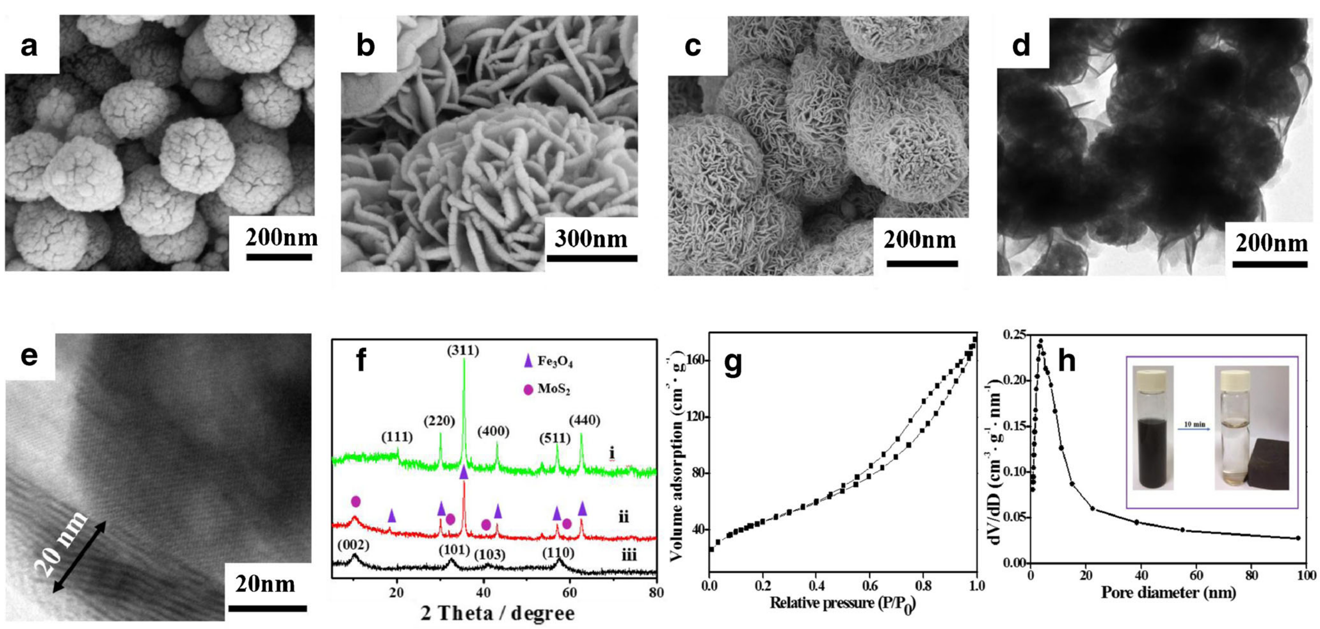

Fig. 4 SEM of Fe_3O_4 (**a**), MoS_2 (**b**), core-shell Fe_3O_4@MoS_2 nanocomposite (**c**), TEM image of core-shell Fe_3O_4@MoS_2 nanocomposite (**d**), and HRTEM of the core-shell Fe_3O_4@MoS_2 nanocomposite (**e**). XRD patterns of Fe_3O_4 (i), core-shell Fe_3O_4@MoS_2 nanocomposite (ii), and MoS_2 (iii) (**f**). Nitrogen adsorption/desorption isotherms of core-shell Fe_3O_4@MoS_2 nanocomposite (**g**). Pore size distribution curves of core-shell Fe_3O_4@MoS_2 nanocomposite (**h**), photograph of separation of core-shell Fe_3O_4@MoS_2 nanocomposites from sample by a magnet (insert of **h**). Reproduced from Zhao et al. (2019) with permission from Elsevier [65]

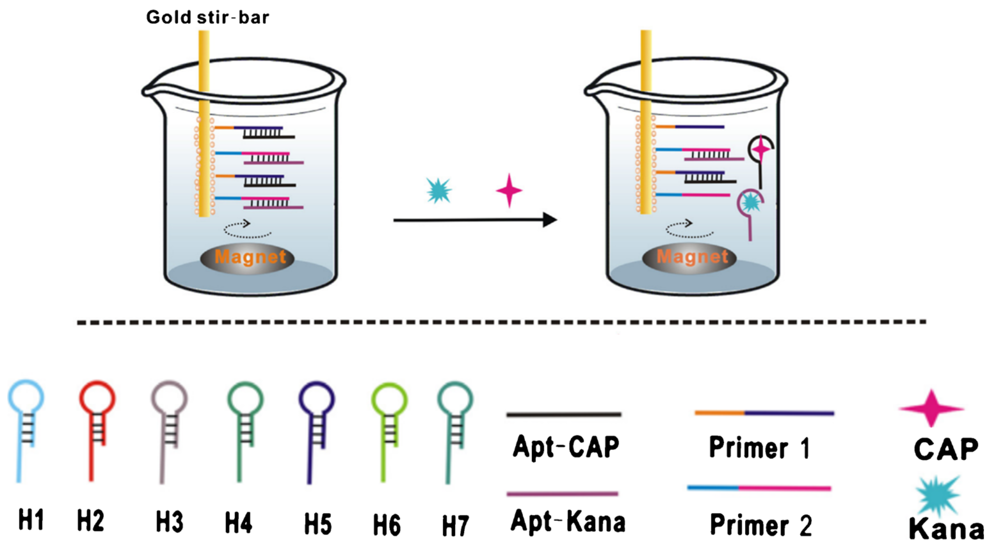

Fig. 5 Schematic illustration of the multiplexed aptasensor for antibiotic detection based on microchip electrophoresis and stir-bar assisted enzyme-free amplification. Reproduced from Zhang et al. (2019) with permission from Elsevier [87]

There are two commonly used colorimetric methods: visual colorimetry and photoelectric colorimetry, both of which are based on Lambert-Beer's law ($A = \varepsilon bc$). The commonly used visual colorimetry method is the standard series method, that is, use different amounts of standard solutions of the test substance in the same set of colorimetric tubes, first develop the color according to the analysis steps, and match the standard color gradation with gradually changing colors. Compared with the visual colorimetry method, the photoelectric colorimetry method eliminates subjective errors and improves the measurement accuracy and can eliminate interference by selecting filters, thereby improving selectivity [93, 94].

Among them, gold nanoparticles (Au NPs) have unique optical properties and high extinction coefficient and are considered to be promising candidate materials for colorimetric signal conversion for the detection of various small molecules and ions [95–98]. Masoudyfar et al. [99] measured the amount of cefixime in solution based on the color change of AuNPs solution via SPR. This detection method combines SPR with colorimetry, which avoids preconcentration or separation steps, shortens inspection time, improves detection efficiency, and has low detection limit. Xu et al. [100] established a colorimetric method for the determination of kanamycin by using gold nanoparticles as signal conversion elements and hybridization chain reaction assisted signal amplification technology. The aptamer hairpin DNA probes (I-Kana) that helps to identify and combine with target kanamycin, which fit body conformation change. This process leads to hairpin I-Kana open hybrid auxiliary signal amplification, triggering a cascade of phosphate group exposed to form dsDNA AuNPs and

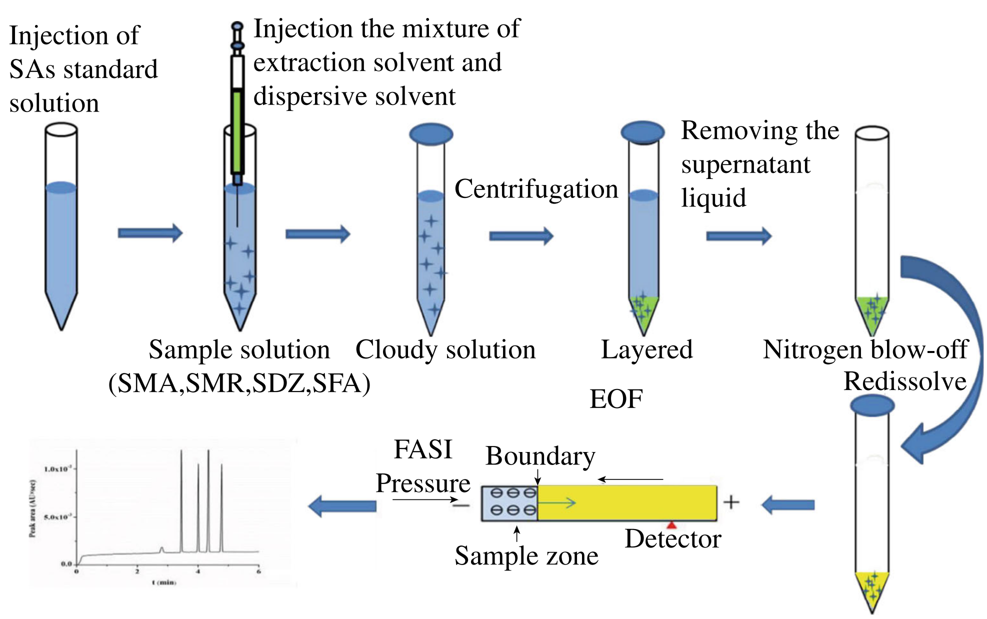

Fig. 6 Schematic of the DLLME-FASI-CE procedure. Reproduced from Ma et al. (2019) with permission from Royal Society of Chemistry [69]

add NaCl after AuNPs between aggregate produced strong repulsion. The color of the solution changes with kanamycin, otherwise solution does not change.

Based on intrinsic peroxidase-like activity of gold nanoclusters (Au NCs), Zhang et al. [101] used tetracycline aptamers to enhance Au NCs to improve the catalytic activity of Au NCs on the oxidation of peroxidase substrates by H_2O_2 and established a colorimetric detection platform for tetracycline (Fig. 7).

Fluorescence

Fluorometric method (FL) for measuring antibiotics is a common method. Fluorescent probes can be detected in the far ultraviolet-near infrared region, and their fluorescent properties can be sensitively changed with their environment. The probes commonly used for fluorescence detection include organic fluorescent dyes (fluorescein, rhodamine, BODIPY, cyanine, etc.), inorganic fluorescent complexes, inorganic quantum dots, and gold nanomaterials. Nanomaterials such as Au NPs [102], Ag NPs [103, 104], carbon QDs [36, 104–107], and carbon nanotubes [108] have excellent luminescence properties, such as broad excitation spectrum, tunable emission wavelength, and excellent light stability [109]. In addition, because of its unique luminous properties, high porosity, and reusability, metal-organic frameworks (MOF) is often used for FL antibiotic detection [110].

In order to improve detection performance, there are many methods of signal enhancement in the fluorescence method. For methods using aptamers, sequence amplification is generally performed, and we generally classify them into enzyme-amplified and enzyme-free amplification. Enzymatic amplification methods include nicking enzyme signal amplification, loop-mediated isothermal amplification, exponential amplification, rolling circle amplification [111], etc. Polymerase chain reaction (PCR), as a common method for exponential amplification in biological analysis, has many applications in biosensors for antibiotic detection [22, 112–115]. Duan et al. [113] combined chloramphenicol specific aptamer and real-time fluorescent quantitative PCR (qRT-PCR) technology. This detection system shows high sensitivity to chloramphenicol, with a detection limit of 0.3 nM and a linear range of 0.1 to 61.9 nM.

No enzyme can overcome the strict experimental conditions of enzyme catalysis, such as temperature, ion concentration, and pH. Hybrid chain reaction (HCR) is a typical enzyme-free amplification, but the amplification of non-specific signals needs to be avoided. Zhang et al. [102] based on SYBR GREENI's fluorescent biosensing, used dual Y-shaped aptamer DNA probes for dual signal amplification to detect kanamycin. The detection limit can reach 0.78 pM. And it obviously overcomes the amplification of non-specific signals by HCR and has good selectivity and sensitivity.

Nadia El Alami El Hassan et al. [116] used a novel immunosensor to detect, which was shown in Figs. 8 and 9. Similarly, the traditional competitive sandwich immunological structure was adopted. Three methods were proposed to immobilize TC-microelectrodes, and the pattern was re-engraved onto PDMS by MEMS process, and the content of tetracycline (TC) was determined by fluorescence change. In the process of functionalization of the immunosensor surface, microcontact printing was performed, and small squares with a side length of about 10 μm were made, and a square array was formed on the glass. This highly sensitive device can detect low-level TC residues in honey samples with a detection limit of 2.5 pM.

In addition, paper-based microfluidic chip is an emerging microfluidic analysis technology platform. Using paper as a substrate to replace materials, such as silicon, glass, and polymers, has a wide range of applications in clinical diagnosis, food quality, environmental monitoring, and other fields, which is also a new method for trace detection of antibiotics. Wang et al. [117] first proposed a dual-functional molecularly imprinted polymer (MIP)–coated paper (MIP-CP) sensor for rapid fluorescence (FL) and visual analysis of norfloxacin (NOR) in aqueous solutions. Lin et al. [118] developed a

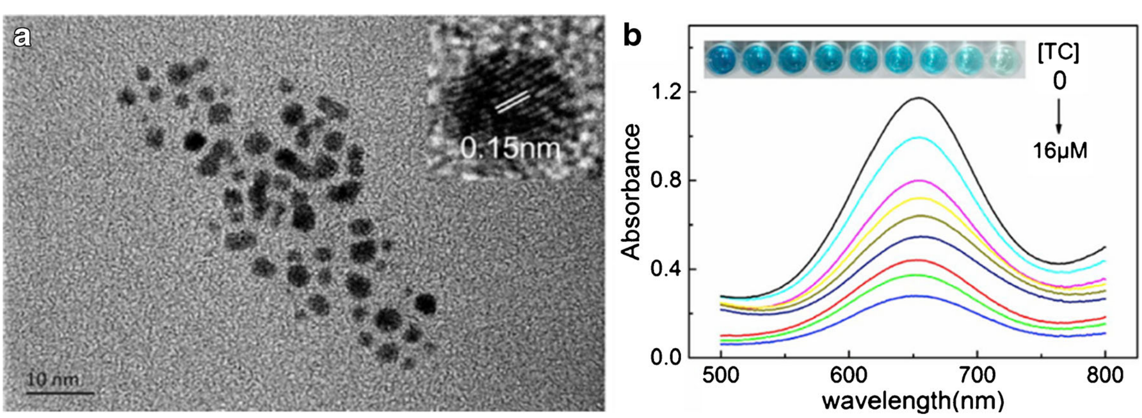

Fig. 7 TEM images of AuNCs-Apt (**a**) and UV-vis absorption spectra of the AuNCs-Apt/TMB/H_2O_2 system in the presence of different concentrations of TC (0, 1, 2, 3, 4, 5, 8, 12, 16 μM) (**b**). Reproduced from Zhang et al. (2020) with permission from Elsevier [101]

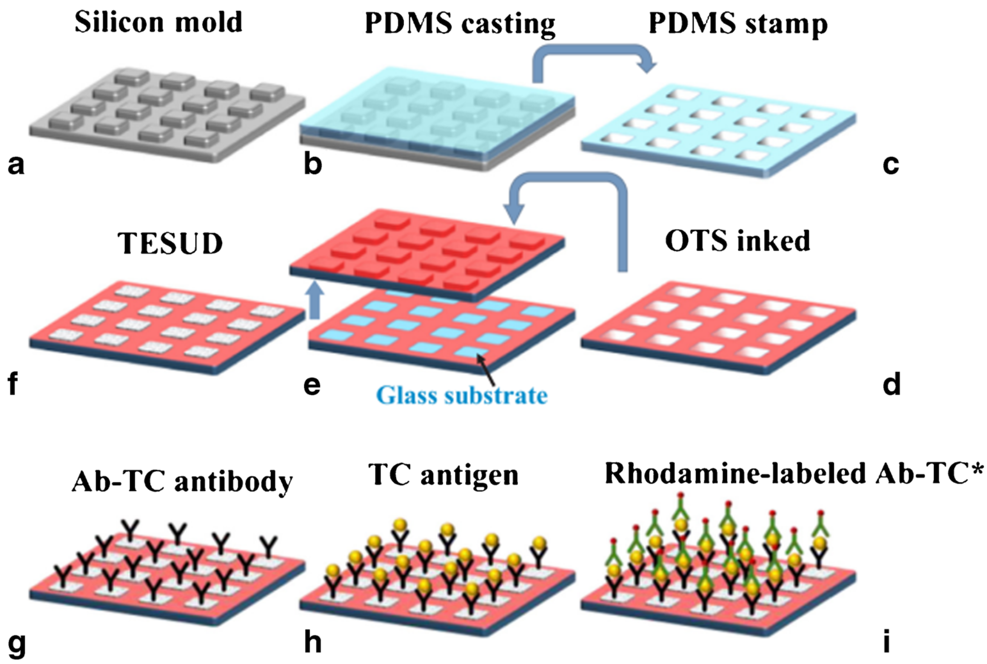

Fig. 8 Microcontact printing process to immobilize Ab-TC onto 1-(triethoxysilyl)undecanal (TESUD) patterns and sandwich immunoassay with TC and rhodamine-labeled Ab-TC. (a)–(c) PDMS and curing agent (10:1 w/w) were poured onto silicon-mold. (d) The stamp was then inked by immersion in heptane solution. (e) PDMS contacts with a freshly activated glass substrate (f) submerged in an ethanol solution containing 1% of TESUD (g)–(i) bonding Ab-TC antibody/TC antigen/Rhodamine-labled Ab-TC. Reproduced from El Hassani NE et al. (2020) with permission from Elsevier [116]

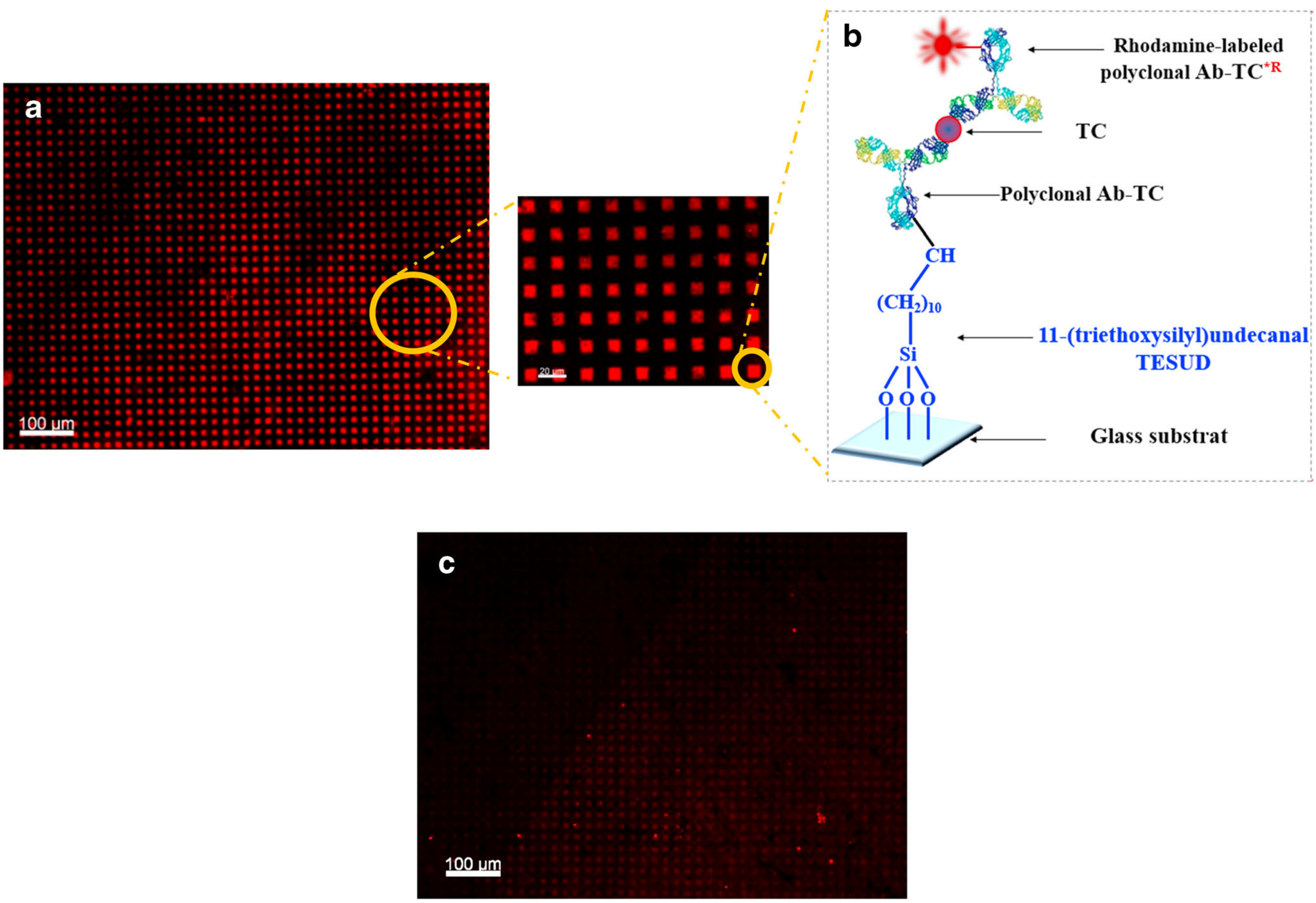

Fig. 9 Fluorescent images of (Ab-TC*R) chemically immobilized on glass substrates after recognition of TC. (a) Positive homogenous pattern with the blackened regions corresponding to the OTS blocked glass surface. (b) Magnification of the positive pattern showing specific detection between Ab-TC*R and TC. (c) Fluorescent image of the biorecognition assay between doxycycline and Ab-TC*R. Reproduced from El Hassani NE et al. (2020) with permission from Elsevier [116]

convenient paper-based chip for labeling functional nucleic acid (H-DNA) and competitive fluorescent side-flow immunoassay (CF-LFI) for ampicillin (AMP) detection. These microfluidic paper chips use wax printing, laser cutting, printing, drawing, inkjet printing, stamp imprinting, and other methods to construct designs on paper and realize various biochemical analyses. The advantages are low cost, easy processing, convenient use, and carrying, making it possible to deeply integrate with smart phones, open source hardware, and 3D printing technology.

Surface plasmon resonance

Surface plasmon resonance (SPR) is an optical phenomenon that can be used to track the interactions in real time between biomolecules in their natural state. Little reflection change between the metal and detected object could induce reflection coefficient change. When light undergoes total reflection on the surface of a prism and a metal film, a fading wave is formed into an optically sparse medium, where a certain plasma wave exists. Resonance may occur when the two waves meet. When the fading wave resonates with the surface plasma wave, the intensity of the detected reflected light is substantially reduced. Energy is transferred from the photon to the surface plasma, and most of the energy of the incident light is absorbed by the surface plasma wave, which drastically reduces the energy of the reflected light. The SPR angle varies with the refractive index of the gold surface, which in turn is proportional to the mass of the molecules bound to the gold surface. Therefore, the specific signal of interactions between biological molecules can be obtained through the dynamic changes of the SPR angle in the process of biological reactions [119]. It can also be used in the sensor to detect the extremely small chemical reaction [99, 120–125]. SPR can detect the change of surface mass concentration of sensor chip in real time [126]. If nanoparticles are condensed to some extent, allowing single particles to pass electron coupling, the surface plasmon oscillation will also change drastically. Agglomeration promotes the coupling of the AuNPs plasma mode, creating a new band containing redshifts and broadening.

Caglayan et al. [120] developed kanamycin and neomycin biosensors based on aptamers using spectroscopic ellipsometer and surface proton resonance enhanced total internal reflection ellipsometer methods as transduction elements, as shown in Fig. 10. The detection time is short, about 10 min, and can be used as an alternative to existing ELISA methods. However, as Si slices are opaque, the spectral ellipsometry analysis used in this study could not simultaneously measure different antibiotics.

Chemiluminescence

Chemiluminescence is a class of molecular luminescence spectroscopy. It is a trace analysis method to determine the content of the substance to be measured based on the principle that there is a linear quantitative relationship between the concentration of the substance to be measured and the chemiluminescence intensity of the system under certain conditions. Nanocomposite materials could enhance the efficiency and intensity of fluorescence conversion, when combined with antibodies or aptamers, they are widely used in antibiotic detection [127–133]. In addition, Li et al. [133] proposed an ultrasensitive chemiluminescence method for fluoroquinolone compounds detection in milk based on the enhancement of the reaction of ruthenium chloride (IV) with methoxylated Cypridina luciferin analogs (MCLA) by fluorescence quantification. Sun et al. [134] prepared nanocomposites (IL/Chit@MGO) through bifunctionalized ionic liquid, chitosan, graphene oxide, and magnetic nanoparticles, which were used in chemiluminescence aptamer sensors for the detection of tetracycline. The new composite material can give aptamers through π stacking, hydrogen bonding, and electrostatic interaction to support more loading sites.

Electrochemiluminescence is a method of converting an electrical signal into optical signal. It is attributed to electrochemical method, which will be described later.

Surface-enhanced Raman scattering

In some specially prepared metal conductor surfaces or sols, in the excitation area, the Raman scattering signal of the adsorbed molecule is greatly enhanced than the ordinary Raman scattering (NRS) signal due to the enhancement of the electromagnetic field on or near the sample surface. This phenomenon is called the surface enhanced Raman scattering (SERS) effect. This method overcomes the shortcomings of low sensitivity of Raman spectroscopy and can obtain structural information that is not easily obtained by conventional Raman spectroscopy. It is widely used in surface research, surface state research of adsorption interface, interface orientation and configuration of bio-sized molecules, conformation research, structural analysis, etc. [135–146]. It can effectively analyze the adsorption orientation, change of adsorption state, interface information, etc. [147, 148].

Metal plasma nanoparticles can generate SERS [149, 150]. Developed by Anh h. Nguyen et al. [151], the GO and AuNPs conjugated aptamer tested kanamycin in drinking water, orange juice and milk. As shown in Fig. 11, the nanocomposite has high thermal stability and low thermal radiation, providing plasma resonance for SERS-based sensing in microfluidic detection systems, while reducing blinking effects. In addition, Zhang et al. [152] used a simple self-assembly method to prepare a magnetic Fe_3O_4@polyethyleneimine@Ag

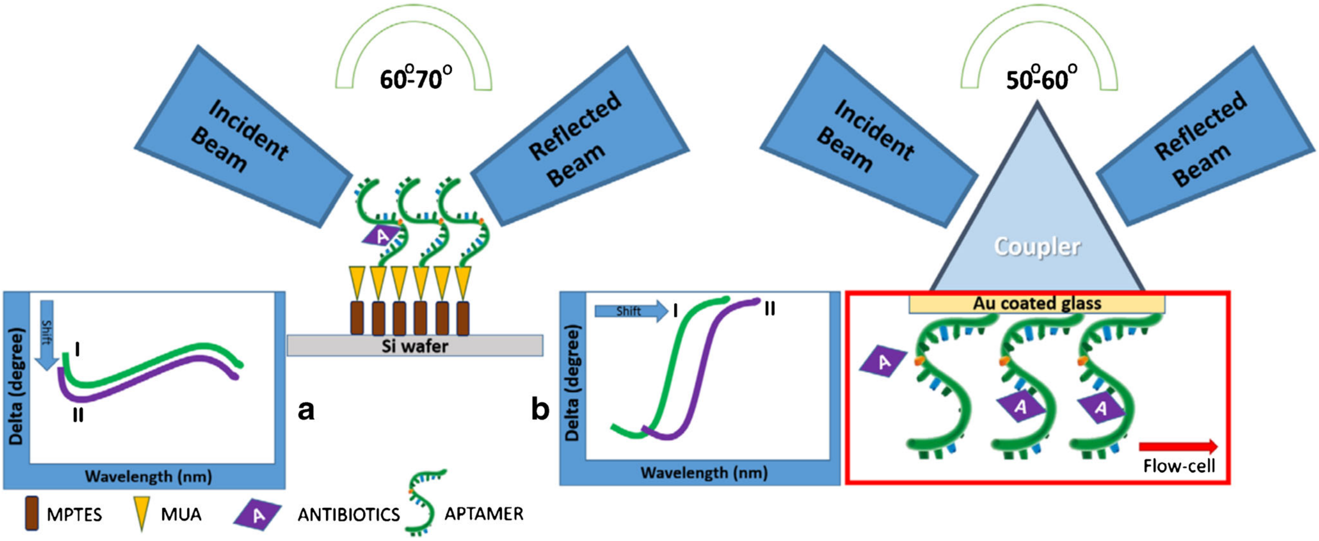

Fig. 10 The ellipsometry and its typical sensor response to (a) SE and (b) SPRe-TIRE. Schematic graphs of sensor response and wavelength represent the usual sensor signal before (I) and after (II) target capture. A "shift" arrow represents the corresponding delta shift after the target is captured. Reproduced from Caglayan et al. (2020) with permission from Springer [120]

composite substrate for tetracycline hydrochloride (TC) detection by SERS (Fig. 12).

Electrochemistry

Electrochemical analysis is a type of instrumental analysis method based on the electrochemical properties of substances in solution. In antibiotic testing, it can be divided into general electrochemistry, electrochemiluminescence, and photoelectrochemistry. The high sensitivity and accuracy of electrochemical analysis makes it popular for trace target analysis.

General electrochemistry

The electrochemical detection method is based on the measurement relationship between the electrical quantity of electric potential, conductance, impedance, current and electric quantity, and the antibiotic content [153–161]. In this way, electrochemical images under different gradient contents can be obtained, so as to obtain more information [159] . In the past decade, electrochemical measurements have been studied to detect antibiotics, and different nanomaterials show different observed electrical signals. Recently, there have been many research results on simultaneous detection of two or more antibiotics [162].

Shen et al. [163] designed an electrochemical sensor for multiple antibiotic detection (e.g., kanamycin and ampicillin) using a metal-ion-encoded apoferritin probes and a double stirring bars-assisted target recycling for signal amplification. The encoding probes were prepared from apoferritin containing Cd^{2+} and Pb^{2+} ions and labeled with duplex DNA, respectively. A signal amplifying device with double agitators assisted target recovery was prepared. Therefore, this method has potential advantages in sensitive screening of antibiotic residues in food.

Besides, Yu et al. [162] detected streptomycin and kanamycin by an all solid-state potentiometric aptasensor array with a dual internal calibration system (Fig. 13). The screen-printed carbon electrode made of reduced graphene oxide and dendritic gold nanostructure is connected to the aptamer of different antibiotics and then specifically binds the antibiotics, resulting in different current values.

Other typical examples of electrochemical methods for measuring antibiotics in 2019–2020 are shown in Table 1.

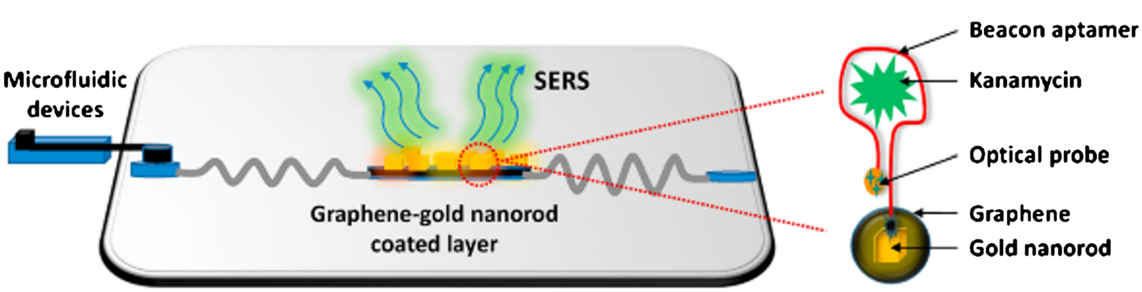

Fig. 11 Scheme of the experimental platform. Reproduced from Nguyen et al. (2019) with permission from Elsevier [151]

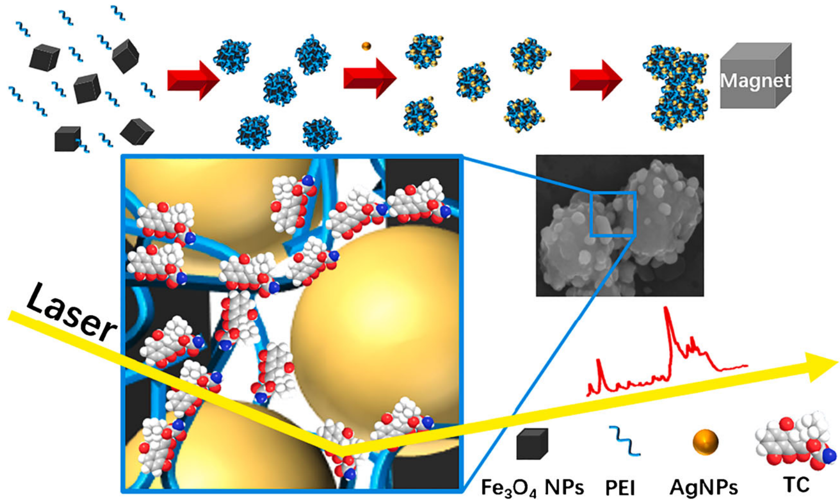

Fig. 12 Schematic illustration of the fabrication of Fe_3O_4@PEI@AgNPs for SERS detection of TC. Reproduced from Akbarzadeh et al. (2020) with permission from Elsevier [152]

Electrochemiluminescence

Electrochemiluminescence (ECL) is a reaction analysis method that combines electrochemical and chemiluminescence methods, resulting in higher detection sensitivity and faster reaction rates. The chemiluminescent agent and the electron donor lose the electron oxidation reaction on the cathode surface at the same time, forming a strong oxidant and cationic radicals. The latter is very unstable and spontaneously loses electrons to form a strong reducing agent. The two highly reactive groups react rapidly on the electrode surface and emit photons. This process takes place over and over again on the electrode surface, producing many photons that enhance the light signal. Recently, there are many research on the use of ECL for trace detection of kanamycin [182], tetracycline [183], chloramphenicol [184], and sulfanilamide [185]. Feng et al. [186] built a dual gears ECL aptasensing platform to simultaneously detect two antibiotics, kanamycin and neomycin, by single ECL luminophore. Taokaenchan et al. [187] proposed that (2,2′-bipyridyl)ruthenium(II) system uses L-cysteine-capped cadmium telluride quantum dots (CdTe-QDs) to achieve resonance energy transfer in aqueous solution, thereby improving the performance of ECL for nitrofuran antibiotic detection. In addition, Hu et al. [188] used Fe_3O_4@SiO_2@ZnO nanocomposites as adsorbents and the carbon nanodots prepared by using urea and ethylenediaminetetraacetic acid as raw materials; also, $Ru(bpy)_3^{2+}$ was used as an electrochemical probe to detect tetracycline.

In addition, Cheng et al. [182] produced an ultrasensitive electrochemiluminescence aptasensor for kanamycin detection through silver nanoparticle-catalyzed chemiluminescent reaction of luminol and hydrogen peroxide (Fig. 14). Since the specific interaction between kanamycin and aptamers will significantly reduce the ECL signal, the concentration of kanamycin can be determined. The detection limit was 0.103 nM.

Until now, due to the low quantum yield of chemiluminescence, it is critical to develop sensitizing reagents. And it is necessary to adjust the bandwidth by selecting a suitable luminescent material for synthetic conformational change, new material recombination, and doping to increase luminescence quantum yield and thus improve the sensitivity and accuracy.

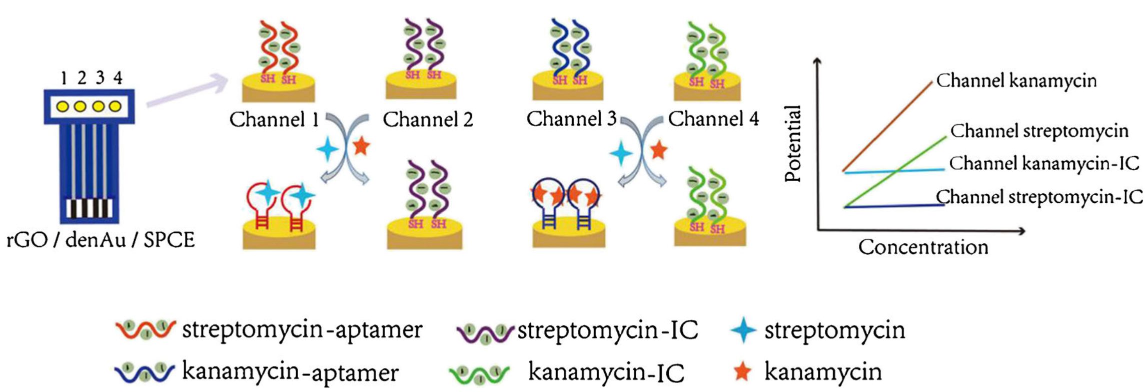

Fig. 13 Schematic illustration of the aptasensor for the simultaneous potentiometric detection of streptomycin and kanamycin with dual-channel internal calibration. Reproduced from Yu et al. (2020) with permission from Elsevier [162]

Table 1 Typical example of measuring antibiotics by electrochemical method in 2019–2020

Detection method	Antibiotics	Materials	Range of detection	Limit detection (LOD)	References
Electrochemical sensor	Chloramphenicol	(RGO@Cu_2S NC)	0.060–1954.5 μM	12.68 nM	[42]
	Oxytetracycline	Ce-MOF@COF	0.22–1.1 nM	37.8 fM	[164]
	Chloramphenicol	GC/Co_3O_4@rGO	1–2000 μM	0.55 μM	[44]
	Sulfadimidine	MIP/$NiCo_2O_4$	0.8–3987 nM	0.674 nM	[165]
	Sulfonamide	2D Cu-TCPP(Fe)	6.927–163.830 nM	2.307 nM	[166]
	Chloramphenicol	GO/ZnO	0.2–7.2 μM	0.01 μM	[167]
	Metronidazole	PrV/SCN	0.001–2444 μM	0.8 nM	[168]
	Ciprofloxacin	NH_2-UiO-66/RGO	0.02–1 μM	6.67 nM	[48]
	Cefixime	GO/AuP	20.0–950.0 nM	7.1 nM	[169]
	Norfloxacin	CH-Y_2O_3/ITO	1 pM-10 μM	3.87 pM	[170]
	Amoxicillin	PGA/3D-GE/GCE	2–60 μM	0.118 μM	[171]
	Nitrofurantoin	beta-CD/CNF	0.004–308 μM	1.8 nM,	[172]
Electrochemical aptasensor	Tobramycin	Ce/Cu-MOF	0.02 pM–21 nM	4 fM	[173]
	Oxytetracycline	OTC-Eu(III)	10–500 nM	1.9 nM	[103]
	Ampicillin	Co-MOF@TPN-COF	2.9 fM–5.7 nM	0.621 fM	[174]
	Azlocillin	EDC-NHS	2.17 pM–216.69 μM	2.60 pM	[175]
	Penicillin	GNP/RGO/PGE	1.0 fM–10 μM	0.8 fM	[46]
Voltammetric sensor	Tetracycline	CNTP-RDE	0.1–2.0 μM	40 nM	[176]
	Levofloxacin	Ag NPs	0.25–100 μM	27 nM	[177]
	Sulfamethazine	poly(NIPAM-co-MAA)	0.01–491 μM	5 nM	[178]
	Azithromycin	MIP-GCE	13.33 nM - 66.67 μM	0.85 nM	[179]
	Ofloxacin	LGCE	0.25–200 μM	75 nM	[180]
	Kanamycin	Au@Pt-CS_2/TH	1 pM–1 μM	0.16 pM	[181]

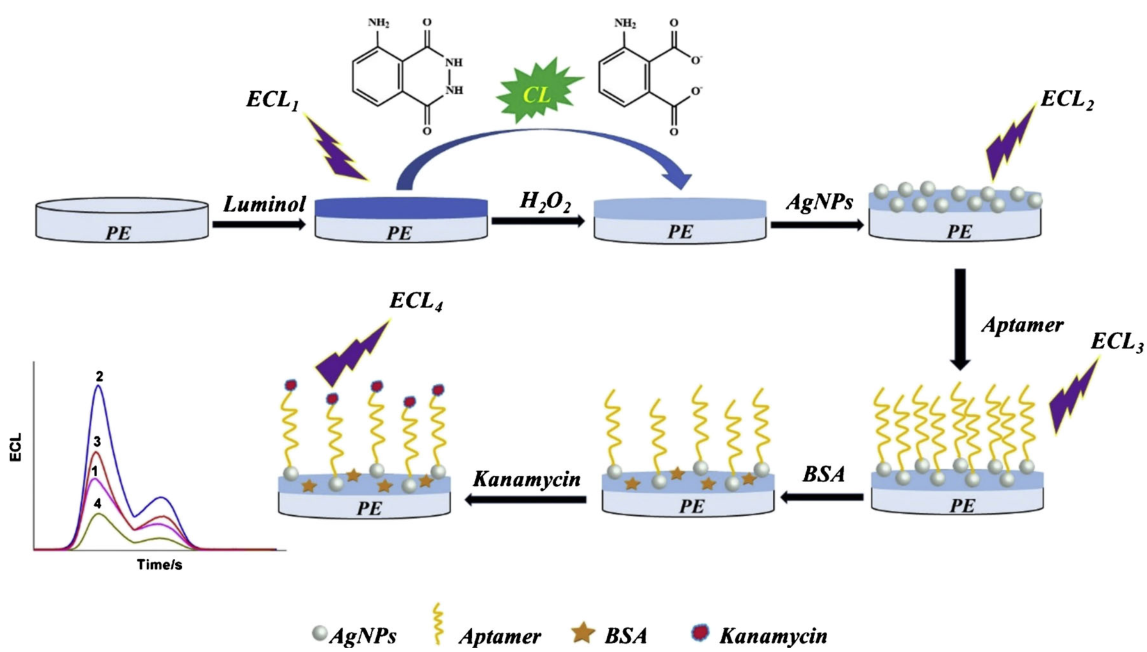

Fig. 14 The schematic illustration of the aptasensor assembled procedure. Reproduced from Cheng et al. (2020) with permission from Elsevier [182]

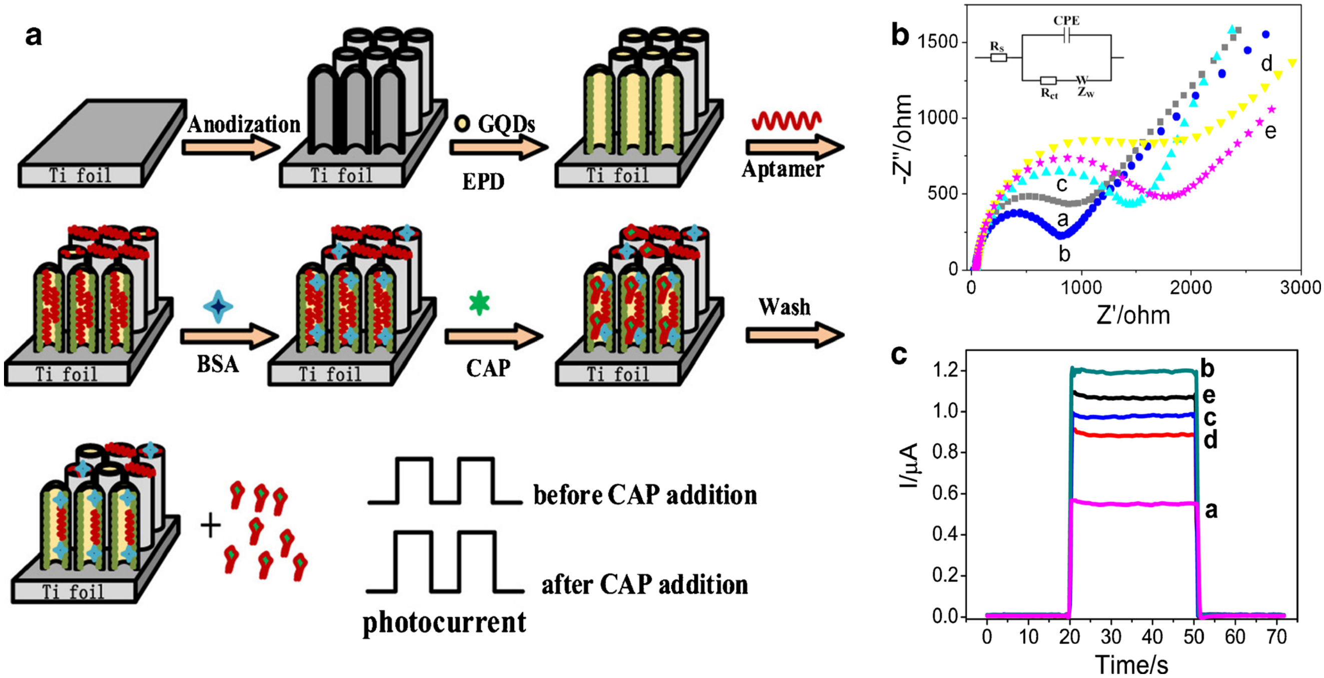

Fig. 15 **a** Fabrication process of PEC aptasensor for CAP detection. **b** EIS spectra of the designed aptasensor at different modification stage. The inset shows the equivalent circuit. **c** The photocurrent response of different modified electrodes. a, TiO_2 NTs; b, GQDs/TiO_2 NTs; c, aptamer/GQDs/TiO_2 NTs; d, BSA/aptamer/GQDs/TiO_2 NTs; e, CAP/BSA/aptamer/GQDs/TiO_2 NTs. The PEC tests were performed in 0.1 M PBS(pH 7.0) containing 0.01 M AA with 0 V applied potential under visible-light irradiation. Reproduced from Qin et al. (2020) with permission from Elsevier [39]

Photoelectrochemistry

As a novel analytical method, photoelectrochemical (PEC) technology has attracted more attention due to its advantages of low background current, fast response, time-saving, and high sensitivity. Photoelectric materials are an essential part of PEC detection [189]. Photosensitive materials have been widely used in energy and environmental fields due to their unique and excellent optical and electrical properties [190–195]. Qin et al. [39] used graphene quantum dot–sensitized TiO_2 nanotube arrays (GQDs/TiO_2 NTs) as photoelectric materials. The aptamer-CAP structure and ascorbic acid removed photogenerated holes to form electron donors to increase photocurrent, thereby detecting chloramphenicol content. The PEC aptasensor production steps are shown in Fig. 15; the aptamer of CAP was immobilized on GQDs/TiO_2 NTs photoelectrode by π-π stacking interaction between GQD and aptamer nucleobases. Aptasensor detects the label-free analysis of CAP by monitoring the photocurrent increase caused by the specific recognition reaction between CAP and aptamer. The aptamer sensor showed a wide linear range from 0.5 to 100 nM in CAP detection, with a low detection limit of 57.9 pM.

In addition, in order to establish a photoelectrochemical platform for the detection of kanamycin, Zeng et al. [50] improved the photoactivity of the matrix by reducing rGO and Au NPs, and using t-bHCR for efficient signal amplification and establishment. In particular, HCR can be exponentially amplified in a short time, which is a very developmental approach in antibiotic detection.

Other literature will not go into details here, and a summary of the antibiotic content detected by the PEC method in recent years is shown in Table 2.

Conclusions and perspectives

This article reviews currently antibiotic detection technologies and the latest research. And the pros and cons of each technology are also analyzed. What all methods have in common is the direction of cross-science: integration and linkage of existing methods, composite and doping of materials, physical and chemical analysis of bio-specific grasping of small molecules, etc.

In this article, according to the principle of light absorption and emission, optical detection methods are divided into colorimetry, fluorescence, surface plasmon resonance, chemiluminescence, and surface enhanced Raman scattering. The advantage of this non-contact measurement is that it does not cause any damage to the observer or the small molecule being detected, thus improving the reliability of the system. In addition, optical detection can obtain physical and chemical information of small particles such as molecules or ions. For example, SPR is the local vibration and resonance wavelength of metal nanostructures that

Table 2 Summary of the detection of antibiotics by the PEC method in recent years

Year	Antibiotics	Materials	Range of detection	Limit of detection	Recovery rate	Reference
2020	Chloramphenicol	Flower-like BiOBr	0.03–319 nM	9.35 pM		[193]
2020	Ciprofloxacin	Nitrogen-deficient graphitic carbon nitride	0.181–57.6 nM	0.060 nM		[191]
2020	Doxycycline	BiOBr nanosheets	0.50 μM −1 mM	0.14 mu M		[190]
2019	Tetracycline	MIL-68(In)-NH2/MWCNT/CdS	0.1 nM–1 μM	0.015 nM		[196]
2019	Kanamycin	rGO-Bi_2WO_6-Au	1 pM–5 nM	0.78 pM		[50]
2019	Ampicillin	aptamer-antibiotic conjugate	0.1–100 nM	32 pM	94–105.4%	[197]
2019	Chloramphenicol	GQDs/TiO_2 NTs	0.5 nM–100 nM	57.9 pM	96.0–102.1%	[39]
2019	Ofloxacin	TiO_2 NTA	5.0 pM–100 nM	0.75 pM	97.6–99.0%	[40]
2019	Tetracycline	Fe_2O_3 on Bi_2WO_6		0.3 μM		[198]
2018	Chloramphenicol	WS_2 nanosheets	10 pM–10 nM	3.6 pM	94.8–110%	[199]
2018	Tetracycline	CN/BiOBr	16.6–10.8 × 10^3 nM	7.9 nM	98.30–103.67%	[200]
2018	Chloramphenicol	TiO_2 NRA@Eu(III)-CdS QD	1.0 pM–3.0 nM	0.36 pM	95.4–101.2%	[38]
2018	Chloramphenicol	g-C_3N_4	1 pM–100 nM	0.22 pM	94.5–107.3%	[35]
2018	Erythromycin	NiTAPc-Gr	0.40–20.00 μM	0.08 μM		[201]
2018	Kanamycin streptomycin	CNFs and OMC-AuNPs.	0.1 to 1000 nM	87.3 and 45.0 pM	93.43–109.74%	[202]
2018	Sulfadimethoxine	g-C_3N_4 QDs & rGO	0.5 nM–80 nM	0.1 nM	95.3–103.2%	[53]
2017	Oxytetracycline	ITO/TiO_2/H-DNA@QD/aptamer	2–300 nM	0.19 nM	89.4–109.2%	[203]
2017	Tetracycline	Au/TiO_2-quercetin	0.3–1600 nM	0.1 nM	93.7–105.3%	[204]
2017	Tetracycline	ZnO-CdS@Au nanocomposite	50–200 nM	4.5 nM	99.75–102.1%	[205]
2016	Ciprofloxacin	g-C_3N_4/BiOCl	1.51–5553 nM	0.6 nM	90.5–112.6%	[52]
2015	Oxytetracycline	Aptamer/BiOI-G/FTO	4.0–150 nM	0.9 nM		[206]

can obtain their free electrons through composition, size, shape, and surrounding dielectric refractive index (RI). For electrochemical detection, the main advantage lies in its simplicity and ease of operation. The chemical parameters which are difficult to be obtained directly in the experiment can be converted into the electrical parameters which are easy to be detected. Since electrochemical reactions are performed according to Faraday's law, even small changes in matter can be measured by easily measurable currents or charges. Moreover, electrochemical measurement is real-time and convenient for monitoring and control. In addition, photoelectrochemical detection has a huge application prospect in the analysis field. In the PEC detection process, electrons excited by light from the semiconductor material are transferred and a photocurrent is generated as a detection signal. By effectively separating the excitation source and the detection signal, a large number of background signals are greatly reduced. PEC detection method has been widely concerned by researchers due to its advantages of low cost, simple operation, high sensitivity and fast response time. For the required photoelectric conversion materials, the same material or two different semiconductor materials can be made into a PN junction cell structure by appropriate compounding or doping methods to improve the photoelectric conversion efficiency and device characteristics.

Compared with immunosensors, the application and development of aptamers have opened up a new way for antibiotic detection, making the detection cost lower, and the process faster and easier. At the same time, the aptamer is a single-stranded sequence, so it has excellent thermal stability, sensitivity and specificity. In addition, it is a promising research direction to associate functional DNA or RNA short chain with antibiotic detection [207]. Binding of nucleic acid layer needs to reflect the measurement results through some amplification method. PCR and HCR can easily conduct tens of thousands of times signal amplification in a short time, which is efficient and labor-saving. But there are also challenges: First, does SELEX screen for aptamers with the desired rate of capture? The higher the accuracy of target acquisition, the lower the detection limit of antibiotic detection. Secondly, false positive is the most likely occurrence of fluorescence detection. Further studies are needed to improve the accuracy of detection. Therefore, for the future development trend, the first step is to innovate the method and hope to be more capable of real-time detection of multiple antibiotics. In addition, the correct selection of optoelectronic materials ensures the accuracy and efficiency of the test. Moreover, the visualization of signals is also very important, which can form the Internet of things through the interconnection application with artificial intelligence. At the same time, we can use micromechanical systems to build functional platforms to miniaturize the entire experiment and testing process [208, 209]. The portability and integrated nature of the chip can play a significant role in the biosensor used for antibiotic or other small molecule detection.

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Compliance with ethical standards

Conflict of interest The authors declare that there are no conflicts of interest.

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