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Recent advances in nanomaterial-based biosensors for antibiotics detection

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ABSTRACT

Antibiotics are able to be accumulated in human body by food chain and may induce severe influence to human health and safety. Hence, the development of sensitive and simple methods for rapid evaluation of antibiotic levels is highly desirable. Nanomaterials with excellent electronic, optical, mechanical, and thermal properties have been recognized as one of the most promising materials for opening new gates in the development of next-generation biosensors. This review highlights the current advances in the nanomaterial-based biosensors for antibiotics detection. Different kinds of nanomaterials including carbon nanomaterials, metal nanomaterials, magnetic nanoparticles, up-conversion nanoparticles, and quantum dots have been applied to the construction of biosensors with two main signal-transducing mechanisms, *i.e.* optical and electrochemical. Furthermore, the current challenges and future prospects in this field are also included to provide an overview for future research directions.

1. Introduction

1.1. The dangers of antibiotics

It is well known that antibiotics are used for treating diseases and promoting animal growth worldwide. With their capability to enhance growth rates and improve feed efficiency, antibiotics have been extensively applied to human and veterinary medicine (Ur Rehman et al., 2015). Generally, antibiotics can be classified into seven groups, *i.e.* tetracyclines, macrolide antibiotics, aminoglycosides, peptide antibiotic, lincosamides, streptogramins, and β-lactam antibiotics. However, antibiotics are able to be accumulated in human body by food chain, which may produce negative influence on human health even at low concentrations, such as hearing loss, toxicity to organs, and so on (Aghdam et al., 2016). In addition, the abuse of antibiotics has improved the frequency of resistance genes, which may result in a decrease in the efficiency of diseases treatment (Leibovici et al., 2016). Furthermore, the development of antibiotic-resistant bacteria is likely to spread to other microbial populations, posing a potential threat to human and animal health (Sapkota et al., 2007). Hence, the control of antibiotics is extremely important to protect human health and safety.

1.2. Conventional detection methods

In the past decades, numerous efforts have been made to develop analytical methods for qualitative/quantitative determination of antibiotics. Chromatographic methods including thin-layer chromatography (TLC), gas chromatography combined with mass spectrometric (GC-MS) (Posyniak et al., 2003), gas chromatography coupled with electron capture (GC-EC), and high-performance liquid chromatography (HPLC) (Blanchaert et al., 2013), are the most conventional detection methods for antibiotics. However, the inherent disadvantages of chromatographic methods such as the expensive apparatus and time-consuming have limited their wide applications. Except chromatographic methods, capillary electrophoresis (CE), diode array (DA), flame ionization (FI), and enzyme-linked immunosorbent assay (ELISA), are also successfully established for the detection of antibiotic residues with accuracy and precision, but still face some drawbacks such as complicate sample pretreatment process and the requirement of highly trained technical personnel (Chauhan et al., 2016). Therefore, a sensitive, selective, convenient, robust, and rapid detection method for antibiotic residues for human health and safety is highly desirable.

Biosensors appear to be suitable alternative or complementary analytical tools for the detection of antibiotics due to their advantages of high selectivity, rapid detection, and *in-situ* applications. Recently, there is a growing rise in the fabrication of antibiotic biosensors. Here, we summarize the current research literature on nanomaterial-based biosensors for antibiotic residues detection. Our aim is to give the readers a complete concept about the state-of-art of nanomaterial-based biosensors for the detection of antibiotic residues and expect more nanomaterials as well as more biosensing strategies be enrolled into this research field that create really workable biosensing devices serving for human world.

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2. Nanomaterials in antibiotic biosensors

Nanomaterials have been widely used for the construction of biosensors in view of their excellent electronic, optical, mechanical, and thermal properties. They are recognized as one of the most attractive materials for opening new gates in the development of next-generation biosensors. With their high surface area to volume ratio, great electronic conductivity, excellent magnetic and physicochemical properties (Wang et al., 2010), different kinds of nanomaterials (including carbon nanomaterials, metal nanomaterials, magnetic nanoparticles, up-conversion nanoparticles, and quantum dots) have been successfully applied to develop numerous biosensors for antibiotics detection. In this section, the basic properties of these five types of nanomaterials used in antibiotic biosensors are discussed.

2.1. Carbon nanomaterials

Carbon nanomaterials, including carbon nanotubes, carbon nanofibers, fullerene, graphene quantum dots, and graphene, are obtaining a lot of attention for their extraordinary properties (Yang et al., 2015). Among them, carbon nanotubes and graphene are the most commonly used carbon nanomaterials in biosensors for the detection of antibiotics.

2.1.1. Carbon nanotubes

Carbon nanotubes (CNTs), one-dimensional (1D) carbon nanomaterials, are sp^2 hybridized carbon atom rolled graphene sheets that were discovered by Iijima in 1991 (Iijima, 1991). Since their discovery, CNTs have gained enormous attention due to their unique mechanical, thermal, and electronic properties. For example, their great mechanical flexibility, fast electron transportation, excellent electrochemical stability, and unique thermal conductivity make them very attractive materials in the application of electrochemical biosensors (Lawal, 2016). Moreover, CNTs can be easily functionalized with various chemical groups which are able to connect the biomolecules (*e.g.* protein and nucleic acid) or organic molecules (Katz and Willner, 2004). In the past decades, CNTs-based biosensors have been extensively used for the detection of antibiotics.

2.1.2. Graphene

Graphene, two-dimensional (2D) carbon nanomaterials, is a sheet of sp^2 bonded carbon atoms that are arranged into a rigid honey comb lattice, exhibiting the highest mechanical strength among the known materials, extraordinary electron transfer capabilities, excellent electrical conductivity, ultra-large specific surface area, unprecedented pliability and impermeability, and favorable biocompatibility (Ping et al., 2015). Since its discovery in 2004, graphene has been successfully applied into various fields, such as catalysis, energy storage, sensor, and electronic devices. In addition, graphene can be easily oxidized into graphene oxide (GO), which contains many hydrophilic groups such as hydroxyl, epoxy, carbonyl, and carboxyl groups (Loh et al., 2010). These hydrophilic groups make GO aqueous dispersibility and easy to be functionalized with biomolecules, which are highly important features in biosensor applications.

2.2. Metal nanomaterials

Metal nanomaterials are a class of functional materials with unique physical and chemical properties, which are closely related to their size, composition, shape, and structure. Great progress has been made in the synthesis of novel metal nanocrystals and their potential applications in diverse fields such as in catalysis, electronics, sensors, and medicine. With their high specific surface area, excellent electron transfer kinetics, and plenty absorption sites to antibodies, enzymes and antigen, metal nanomaterials have attracted extensive attention in the development of electrochemical biosensors for antibiotic detection (Sozer and Kokini, 2009). In addition, metal nanomaterials, especially gold nanoparticles (AuNPs), possess a distinctive phenomenon termed as surface plasmon resonance, in which any change/alteration in the size, shape, or geometry of particles alters the local electron confinement that is thereby reflected in the absorption maxima and color of colloidal solution (Saha et al., 2012). In the past decades, optical biosensors based on AuNPs have been successfully developed for antibiotics detection.

2.3. Magnetic nanoparticles

Magnetic nanoparticles (MNPs) have gained an increasing attention in the development and applications of biosensors recent years. Enormous efforts have been made to develop MNPs because of their particular characteristics such as large surface area, high mass transference, unique physicochemical properties, biocompatibility with biomolecules, and easy production (Akbarzadeh et al., 2012; Reddy et al., 2012). MNPs display superparamagnetic property below 50 nm size and perform best at 10–20 nm, making them a suitable choice in magnetic fields when quick response is required (Netto et al., 2013). Moreover, MNPs are able to be integrated into the transducer materials, attracting analytes in the samples by an external magnetic field (Rocha-Santos, 2014). Compared to non-MNPs-based strategy, biosensing strategy based on MNPs has many merits including improved sensitivity, lower detection limit, less noise, and quicker analysis (Justino et al., 2013).

2.4. Up-conversion nanoparticles

Up-conversion nanoparticles (UCNPs) are a novel class of luminescent materials that transform the near infrared radiations (lower energy) into visible radiation (higher energy) (Wang et al., 2011). It has been paid significant attention in various fields due to their low background noise, high photo stability, low toxicity, and great penetration of signals in tissues with low absorption (Zhang et al., 2016). These advantages make them a better choice in comparison to down-conversion fluorescent materials, such as organic fluorescent dyes and inorganic quantum dots. Generally, they are used as fluorescent labels combined with biorecognition molecules for antibiotics detection in the optical biosensing systems (Wang et al., 2005).

2.5. Quantum dots

As semiconductor nanocrystals, quantum dots (QDs), including CdS, ZnS, and CdTe, display unique optical and electronic properties, such as high luminescence, long-term photo stability, resistance to photo-bleaching, broad absorption bands, and size-tunable, narrow, symmetric emission (Bonilla et al., 2016), which make QDs very attractive in analytical chemistry. Moreover, QDs provide the convenience of conjugation to aptamer/antibody without affecting either their emission properties or aptamer/antibody specificity (Stanisavljevic et al., 2015).

3. Nanomaterial-based biosensors for antibiotics detection

3.1. Optical biosensors

Optical detection comprises of transducers that can capture signals produced by the interactions of biorecognition element with target analyte and can transform them into optical signals (Yoo and Lee, 2016). In the past years, optical biosensors have been extensively used in antibiotics detection owing to their advantages such as simplicity of operation, convenience, and sensitivity (Adrian et al., 2009). The introduction of nanomaterials into optical biosensors have enabled the ultrasensitive and label-free strategies in antibiotics detection. According to the optical signal transducing mechanism, the nanoma-