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Engineering nanomaterials-based biosensors for food safety detection

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ABSTRACT

Food safety always remains a grand global challenge to human health, especially in developing countries. To solve food safety pertained problems, numerous strategies have been developed to detect biological and chemical contaminants in food. Among these approaches, nanomaterials-based biosensors provide opportunity to realize rapid, sensitive, efficient and portable detection, overcoming the restrictions and limitations of traditional methods such as complicated sample pretreatment, long detection time, and relying on expensive instruments and well-trained personnel. In this review article, we provide a cross-disciplinary perspective to review the progress of nanomaterials-based biosensors for the detection of food contaminants. The review article is organized by the category of food contaminants including pathogens/toxins, heavy metals, pesticides, veterinary drugs and illegal additives. In each category of food contaminant, the biosensing strategies are summarized including optical, colorimetric, fluorescent, electrochemical, and immune- biosensors; the relevant analytes, nanomaterials and biosensors are analyzed comprehensively. Future perspectives and challenges are also discussed briefly. We envision that our review could bridge the gap between the fields of food science and nanotechnology, providing implications for the scientists or engineers in both areas to collaborate and promote the development of nanomaterials-based biosensors for food safety detection.

1. Introduction

The World Health Organization (WHO) expressly stated that safety food should be nontoxic and innocuous. Food safety is a multidisciplinary subject regarding food processing, preparation, and storage in ways that prevent foodborne illness. Food safety standards are now extensively adopted in the worldwide food industry, such as BRC, FSSC 22000, IFS, HACCP (Aung and Chang, 2014; Chassy et al., 2004). However, global food safety incidents still frequently occurred, leading to severe healthy, economic and even social problems (Carvalho, 2006; Henson and Caswell, 1999; Li et al., 2015; Pena-Rosas et al., 2012; Wilcock et al., 2004). For instance, beef products from America have been frequently banned to export to other countries due to outbreak of bovine spongiform encephalopathy (Jin and Kim, 2008). A banned chemical, clenbuterol, was found in pork to promote leanness in pigs (Prezelj et al., 2003). In 2008, an unforgettable scandal regarding food safety occurred in China: melamine has been detected in infant formula (milk powder), causing more than 290,000 infants suffered from severe healthy problems such as urinary tract stones (Chen, 2009; Wu et al., 2009). All these events alarm us the importance and urgency of guaranteeing food safety. Except for relevant policies and laws concerning food safety, detection of food contaminants has been attracting remarkable attention in last decade, which ensures the governments and customers to recognize whether the food is safe (Cocolin et al., 2011; Glynn et al., 2006).

A variety of well-developed technologies have been employed for food safety detection such as gas chromatography (GC), high performance liquid chromatography (HPLC), gas chromatography-mass spectrometer (GC-MS), liquid chromatography-mass spectrometer (LC-MS) and enzyme-linked immunosorbent assay (ELISA) (Malik et al., 2010; Patel, 2002; Pico et al., 2006; Rodriguez-Lazaro et al., 2007). Most of these methods have disadvantages such as complicated sample pretreatment and long detection time, especially relying on expensive instruments and well-trained personnel, which limit their applications in some scenarios such as in developing countries and areas with poor equipped facilities and specialists.

Nanotechnology, an emerging developing subject, provides opportunity to address these challenges pertaining to food safety detection. Nanotechnology refers to the science and technology of designing, building, manipulating and understanding materials and systems at

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nanoscale (size ranging from approximately 1 nm to 100 nm in general), which has demonstrated comprehensive impacts on a myriad of applications such as healthcare, environment and energy (Arico et al., 2005; Parak et al., 2015; Sahoo et al., 2007; Whitesides, 2005). In particular, nanotechnologies bring grand benefits to, and impose great impacts on food industries through the whole food chain, including production, processing, safety, packaging, transportation, storage and delivery (Kagan, 2016; Kalpana Sastry et al., 2013; Neethirajan and Jayas, 2011; Rossi et al., 2014; Sozer and Kokini, 2009). Regarding food safety in specific, nanotechnologies exhibit promising potential for detecting food contaminants, in which nanomaterials-based biosensors play important roles. Nanomaterials possess unique physiochemical properties in comparison with their bulk counterparts, providing valuable strategies for food safety detection (Gupta et al., 2016; Rai et al., 2009). Advances in nanomaterials have facilitated the developments of biosensing for food safety detection (Chen and Park, 2016; Sharma et al., 2015; Warriner et al., 2014). Biosensors, coupling biological components with physicochemical detectors, possess enormous benefits for the detection and analysis of food contaminants, because of the high sensitivity and specificity resulting from the precise recognition (Chaudhry et al., 2008; Lan et al., 2017; Rashidi and Khosravi-Darani, 2011). Compared to traditional chromatography methods, nanomaterials-based biosensors have some key advantages including more specific target recognition, improved selectivity and sensitivity, enhanced signal readout, and shorter analysis time (Lim and Kim, 2016; Yang et al., 2016).

We organize this review by the category of food contaminant sources, including pathogens/toxins, heavy metals, pesticides, veterinary drugs and illegal additives. In Fig. 1, we provide an overview of engineering nanomaterials-based biosensors for food safety detection. In this review, we do not intend to summarize all of the nanomaterials ever used in detection of food contaminants. Instead, we selectively focus on some lately important achievements. We further provide our perspectives on the future development.

2. Pathogens and toxins

Although advanced food-packaging technologies such as vacuum packaging and pasteurization treatment allowed food to be stored for a relatively long time, the emergences of foodborne pathogens infecting food have arisen inevitably, resulting in severe diseases of consumers, such as acute emesis and acute abdominalgia (Oliver et al., 2005). The most common foodborne pathogens are *Escherichia coli* (*E. coli*), *Salmonella* and *Listeria*. In last decades, events of foodborne pathogen infection occasionally occurred worldwide (Crim et al., 2014). These events gave rise alarm that preventive action should be taken before consuming, such as detecting pathogens in food. On the other hand, in many cases of foodborne pathogen infection, toxins produced by

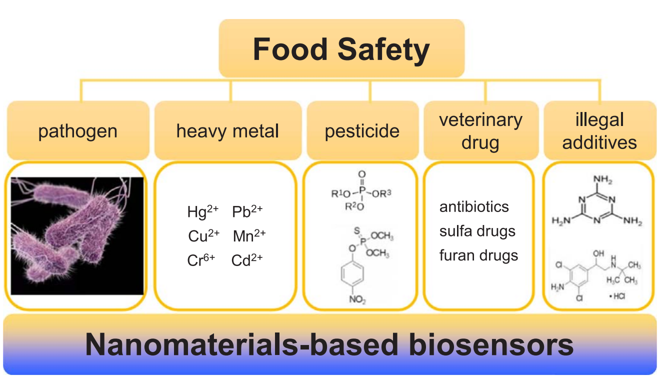

Fig. 1. Overview of nanomaterials-based biosensors for food safety detection. This review is organized by the category of food contaminant sources including pathogens/toxins, heavy metals, pesticides, veterinary drugs and illegal additives.

pathogens were subsequently released into food.

A variety of methods have been proposed to detect and quantify pathogens and toxins in food. Meanwhile some challenges still existed, including low specificity, low sensitivity and long time for identification (Lazcka et al., 2007). Nanomaterials-based biosensors provide alternative strategy to address these challenges (Burris and Stewart, 2012). Nanoparticles (NPs), quantum dots (QDs), nanorods and carbon nanotubes (CNTs) based biosensors have been developed to realize the detection of foodborne pathogens and toxins (Cho et al., 2014; Kaittanis et al., 2010). Various pathogens and toxins have been effectively detected by nanomaterials-based biosensors with numerous biosensing strategies (Table 1).

Functional noble NPs have been widely applied in pathogen detection, realizing colorimetric sensing in specific. It is notable that NPs such as gold and silver possessed excellent plasmon absorption, thus enhancing signal of detection. The colorimetric sensing was realized by selecting appropriate organic ligands to modify NPs. Phillips et al. used poly(paraphenyleneethynylene) (PPE) conjugated gold nanoparticles (AuNPs) to achieve fast and efficient identification of pathogens (Fig. 2a) (Kaittanis et al., 2010; Phillips et al., 2008), in which the interaction between anionic PPE and cationic AuNPs quenched the fluorescence of PPE. Specific pathogens were associated with AuNPs, resulting in the disassociation between AuNPs and PPE. Consequently, fluorescence PPE was released into solution and the fluorescence of PPE was turned on, which exhibited enhanced fluorescence emission. Similarly, Cheng and coworkers demonstrated a fast detection (in 4 h) of *E.coli* by using glassy carbon electrode modified with platinum NPs (PtNPs), which had high surface area ratio, thus improving the detection sensitivity to *E.coli* due to the electro-catalytic ability of PtNPs (Cheng et al., 2008). Hosseini et al. developed a colorimetric and chemiluminescence method based on aptamers conjugated AuNPs to detect aflatoxin B1 (AFB1), in which aptamers acted as specific recognition ligand and AuNPs exhibited high absorption coefficient. The desorption of the AFB1 binding aptamer from the surface of AuNPs induced the aggregation of AuNPs, leading to the apparent color change of the solution (Hosseini et al., 2015).

In comparison with normal NPs, magnetic NPs (MNPs) possess special properties, allowing for rapid separation and enrichment of target analytes. Joo et al. (2012) utilized antibody conjugated MNPs to capture *Salmonella* from milk. Once an external magnetic field was applied, the superparamagnetic MNPs enabled selective separation of target bacteria. The enrichment effect of MNPs rendered this method with high sensitivity (100 cfu/mL) (Fig. 2b). Similarly, Cheng and coworkers employed bio-functionalized MNPs (BMNPs) to detect *E. coli* in pasteurized milk. BMNPs were fabricated by immobilizing a specific anti-*E.coli* antibody on the surface of amine-functionalized MNPs, which showed high capture efficiency to *E. coli* (Cheng et al., 2009). Ma and coworkers demonstrated a novel colorimetric biosensor for detection of *Salmonella typhimurium (S. typhimurium)* by coupling MNPs and AuNPs. The MNPs and AuNPs were combined respectively with capture DNA and probe DNA which were partially complementary to the sequences of the *S. typhimurium* target DNA, which were fabricated as the capture probes and signal probes. In the presence of *S. typhimurium* target DNA sequences, complementary base pairing of DNA induced the probes and target to form sandwich-like structures, thus AuNPs aggregated and the color changed from red to blue. The absorbance spectra of AuNPs red shifted as the intensity ratio of A700/A521 changed, and the intensity ratio of A700/A521 had a linear correlation with the amount of *S. typhimurium* target DNA, therefore realizing the quantification of *S. typhimurium* (Ma et al., 2017).

Gold nanorods had higher sensitivity to the local dielectric environment, and demonstrated distinct optical properties due to their specific shape (Jana et al., 2001). Wang and Irudayaraj constructed gold nanorod bioprobes by functionalizing the amino-terminated gold nanorods with antibodies. Two major species of foodborne bacteria (*E. coli* and *S. typhimurium*) were simultaneously detected in less than