Progress in Sensory Devices of Pesticides, Pathogens, Coronavirus, and Chemical Additives and Hazards in Food Assessment: Food Safety Concerns

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Abstract

Food intake gives vitality and supplements to support humans and other living organisms. Food safety and contamination problems associated with food hygiene, storage, chemical additives, enzymes, bacteria, and pesticides are crucial issues because of their direct influence on the health of humans and even animals. New monitoring technologies should be developed for potential food safety and significant environmental benefits. To date, the ultrasensing, early detection, and real- and on-time monitoring of vital reactive species, biomolecules, chemicals, and hazardous agents are important in ensuring food quality. With significant advances in the engineering of sensory devices, the progressive development of accurate quantity screening, early explicit monitoring and assessment, and real-time detection analysis can support the standard food quality through the full control of an extraordinary food safety test. Progress in numerous auto-examining appraisals, sensing protocols, and tools of (i) reactive species and chemical additives associated with human metabolism and various nutritional and industrial processes of foods, including ascorbic acid (AA), H$_2$O$_2$, uric acid (UA), and nitrite and sulfite anions; (ii) extremely organic and inorganic hazards such as heavy metals and bisphenol A; and (iii) food adulteration, pesticides, pathogenic microorganisms are a key challenge for food safety concerns. To date, evidence supporting the possibility of transmitting coronavirus disease 2019 (COVID-19) infection through food products is unavailable. However, in a report on an outbreak in mid-June 2020 in China, food contamination with the causative agent of COVID-19 pandemic, SARS-CoV-2, was discovered. Thus, sensory protocol devices for monitoring the SARS-CoV-2 antigen associated with food products is urgently needed for the future perspective progress in health. As such, we provide details in advanced sensor development in the monitoring, analysis, and evaluation sectors for food safety applications. We also report on next-generation
nano/microscale wearable sensor devices that can wirelessly provide relevant healthy and safety food information data. This review gives evidence that the powerful engineering of mobile food sensor devices is an ongoing acquisition, offering considerable future avenues to the perspective in-home healthcare of aging individuals.

**Keywords:** food safety, monitoring, assessment; food reactive species; bacteria; pesticides; wearable touchscreen sensor devices; COVID-19; food healthcare

1. **Introduction**

Food safety remains one of the most vital issues worldwide because the presence of hazardous species or even urgent additives with risky levels of food poses a real risk to food safety and healthcare. The World Health Organization (WHO) pointed out that foodborne diseases negatively affect the economies of underdeveloped countries. In developing countries, food safety is linked to many different diseases and disorders, such as diarrhea, which leads to about 3 million deaths [1-3]. Therefore, food safety has been recorded as a high priority by the WHO. However, according to a report of the Centers for Disease Control and Prevention, approximately 48 million Americans consume contaminated food and that 3,000 die of foodborne diseases in the United States [2, 3].

With the increasing interest in the food industry, high-quality products with high safety rates have become urgently needed. Therefore, food and quality control systems should be established to meet food safety requirements and maximize the financial profits of food companies. In food markets, many major companies have to ensure the improvement of a quality assurance and controlled production line to avoid losses because of sanctions, product withdrawal, and customer trust loss in brand names [4]. Nanotechnology offers enormous advantages in the growth of food sensor
electronics and identification protocols of different target species and pollutants for food safety and quality evaluation. Various evaluation strategies based on chemical/biological sensors can be used online and are coordinated in the current assembly as quick, basic, and versatile sensors for food species that affect food quality and safety. Commercially available monitors or nano/microsensor devices provide real-time continuous measurement of food ingredients, nutrients, byproducts, pesticide residues, pathogenic microorganisms, chemical food additives, hazards, preservatives, food contaminants, packaging, and other wastes [5]. Current progresses in the design and manufacture of food-based sensors offer great advantages in sensing food contaminants. Moreover, great efforts have been devoted to developing food sensors that feature cost-effectiveness, short response time, and low complexity.

Surveying and estimating the level of definite target molecules in food significantly affect the analysis and prediction of risks to human health [4-6]. Disproportionate qualities and changes in the levels of targeted species, including organic agents, inorganic agents, and biomolecules or their byproducts, threaten human health [7]. Studies have also been performed to improve sensing tools and devices for screening and controlling various targets, including reactive species and additives, such as AA, H₂O₂, UA, and nitrite and sulfite anions. These targeted agents have been produced through transformation in human metabolism or through various nutritional and industrial processes of foods. Moreover, early and real-time monitoring of pathogenic hazardous species, including bacteria, viruses, and pesticide residues, in food have a considerable key factor in food safety assessments [8-10]. Furthermore, global restrictions on tracking and packaging food processes and eating or handling food (including frozen food) via surface touching from individuals have been implemented to avoid the spread of COVID-19 pandemic. Therefore, the development of food-based sensory and screening tools of these targeted agents, hazardous species, and SARS-CoV-2 antigens are vital in food analysis and environmental and health monitoring [8-11]. The early recognition, real-time assessment, and
detection protocols of SARS-CoV-2 antigens, pathogenic microorganisms, toxic species, food additives, and pesticides in food play an essential role to ensure human healthcare. To date, great developments have been achieved to manufacture effective and operative sensing devices for the assessment of food contaminations and risks.

In this context, this review highlights numerous designs of nanomaterial-based sensors associated with food safety concerns. Demands for smart compact nano/microsensor electronics have increased to overcome low temporal/locative settling and conventional techniques associated with analysis problems. Therefore, studies have developed several sensors design and mobile health monitoring technologies that enable the precise management of the levels of various species affecting the food quality. More specifically, this review provides insights into the current state of the COVID-19 pandemic and its relationship with food quality and various industries. In addition, this review enumerates effective strategies and recent progress in numerous analysis techniques, building blocks of sensor designs, and protocols that serve as a basis for improving the controlled management, risk assessment, and early warning detection of food contaminations associated with food hygiene, storage, chemical additives, enzymes, bacteria, and pesticides. Novel conductive materials used in portable sensor devices offer an extraordinary control of a new generation of developed sensing/detection protocols for food safety management.

2. Prospective assessments for food safety

Researchers and industrial partners have recognized the potential of nanotechnology in different food sectors, including agriculture and farming (e.g., pesticides, fertilizers, and vaccines), food processing, food packaging (preservation and protection), and nutrient supplements. The most dynamic assessment reports in food science are based on the advanced fabrication of nanoscale sensor devices that can be utilized in a diverse range of food safety applications. In this manner, assessment strategies
and apparatuses are utilized in food development, creation, handling, and packaging (Scheme 1A) [5, 12]. To date, the rapid increase in the widespread testing and risk assessment of COVID-19 infection is the major concern of worldwide interest. Thus far, the transmission of COVID-19 infection through food products is not yet confirmed. However, the urgent need for monitoring SARS-CoV-2 antigen associated with food products is unquestionable for inexorably global health protection and food safety.

2.1. Key elements for food assessment protocols

For genuine and favorable food safety management board tools, the development of a mature, reliable, and dependable innovation for next-generation nano-future food management devices is crucial. For food safety and management, sustenance examination should focus on the whole food cycle, which includes the final product, nourishment freshness, strengthening, or dangerous ingredients utilized in food or during food processing and microbiological quality. In any case, a device for the fast and accurate response discovery of targeted concentration levels remains to be developed.

For the progress in health integration and food safety, the well-controlled assessment protocol and large-scale manufacturing of food sensors that can be integrated with the exact transmitter into portable devices are required with the following key controls:

(I) To ensure that the duration of targeted and systematic analysis and improve real-time and continuous monitoring

(II) To reduce complications and serious adverse events of other associated problems

(III) To overcome the restrictions of self-revealing techniques

(IV) To offer logical estimations of food species by tracking physiological operations

(V) To improve methodologies for reading out and sorting tool performance and automatically track and distinguish each target
(VI) To determine the effectiveness of long-term food management practices in indoors and outdoors through continuous supply, along with wireless applications.

With regard to the recently created contemporary nanotechnology outskirts and around future proof choices to precise, accurate, reliable, and low-cost individuals, a nanosensory system combined with patch, adaptable, flexible, and intelligent fabrics remains important for managing long-term food safety and healthcare. Such key creative endeavors not only provide a consistent and implantable screening process for target ranges but also permit the complete estimation of controlling food quality or doses and techniques of beneficial food additives.

**Scheme 1**

### 2.2. Sensing techniques for food quality and safety

For food quality and safety, chemical sensor techniques are widely used to measure or distinguish physical parameters and changes in reactive species, agents, and contaminant targets into read-out signals through observation or instrument use. Sensors can permit real-time investigation with high sensitivity and selectivity, thereby revolutionizing examination in the food safety field, including agriculture and farming, food processing, food packaging, and nutrient supplements under basic consideration. Progress on sensors can be identified on the basis of different keys, including (i) intrinsic property evaluation (selectivity, sensitivity, sensory detection range, limitation, and lifetime), (ii) economic manufacturability, (iii) new research methods with a fast response, (iv) simple sensing strategy, and (v) real applicability [13, 14]. For example, the construction and engineering of biosensors primarily depend on incorporations between two components, namely, a bioreceptor and a transducer. Bioreceptors immobilize sensitive biological elements (e.g., enzymes, proteins, antibodies, and whole cells) that act as an active interfacial surface, and transducers convert the physiochemical change resulting from the interaction between food reactive targets and biological...
elements to a quantified signal, whose intensity is associated with target concentrations [15, 16]. Regarding transducing elements, chemical sensors and biosensors can be categorized into electrochemical (potentiometric, amperometric, and conductometric) sensors, optical (including reflectometry or absorptiometry, fluorescence, and surface plasmon resonance [SPR]) and surface-enhanced Raman spectroscopy (SERS) assay sensors, piezoelectric sensors, and thermal sensors, which can convert reactions or binding with target species into a digital signal (Scheme 1B) [13, 15, 17-19].

Studies should focus on the developmental engineering of powerful nanoscale sensor designs, devices, and protocols for a wide scope of food safety assessments. A considerable progress on sensor device manufacturing provides periodical transduction into easy-to-track output signals, fast response analysis, and dead-end-use examination, leading to the precise assessment of food contamination. Therefore, the integral optimization of nanoscale sensor chipsets into robotics, smartphones, and wearable, patches, and touchscreen devices is the key principle for the future steadfast advancement in sensing, analytical, and biotechnological applications. Portable sensor disciplines offer contamination monitoring and environmental food assessment with short time consumption and simple operation.

3. Nanoscale-developed food sensory platform models

Nanoscale sensor platforms have been developed with 0D–3D materials that have hierarchical engineering, well-controlled shape morphology, surface heterogeneity, molecular arrangement structures, and highly physical/optical/mechanical properties, enabling to improve food target diffusivity, electronic surface charge transfer, and sensitive signaling and absorptivity. These platforms serve as precise environments suitable for sensory designs and effective sensor protocols (Scheme 2) [8, 15, 20-27].
Nanoscale sensory techniques are of great interest because they provide basic solutions to many daily problems, especially in environmental, healthcare, and medical sectors. These techniques improve optical and electronic properties, expand the surface of the transducing range, and increase the surface-to-volume ratio for sensitive reading out signals [28-32]. Moreover, nanomaterials with a highly controlled anisotropic composition and surface functionality can be used to generate modern sensors with special features associated with electrical conductivity, power consumption, weight, and diffusion for the precise and highly sensitive real-time detection of targeted food species [33-39].

In general, nanomaterial-based platforms can provide the following advantages for food sensing protocols [11, 33-44]:

i) high surface-to-volume ratio, which stimulates massive and multidirectional target accessibility

ii) changes in the number of surface-active atoms in the correspondence to defects, such as vertices, which hamper unconventional surface reactivity in sensory signaling

iii) aligned geometries in different dimensions provide surface coordinates with electrical association during target detection

iv) surface modification with a highly reactive stationary platform facilitates the selective reaction of target species with active signaling sites

v) production of highly mechanical, conductive, and stretchable/flexible surface use

Scheme 2

To date, nanoscale materials are important for developing nano/microsensor devices and sensing protocols for target species in food. These nanoscalable monitoring designs provide effective assessment efficiency, signal output, amplification, rapid response, real-time analysis, and
analytical diffusivity [8, 11, 29, 30, 35, 38, 39, 41, 42, 45]. To date, nanoscale structure-based sensors show real evidence as alternative devices for the rigorous analysis of the final quality, sensitivity, and other complex features (Scheme 2). Given the great technological advances in material platforms, nanoscale sensory designs provide insights into the following key processing features [15]:

(i) hierarchical superstructures and three-dimensional (3D) architecture geometrics for outstanding electronic and conductive signaling devices

(ii) well-defined atomic-scale configurations and orientation structures, offering actively dense, high-index exposure facets, and versatile binding to ultra-traces of glucose

(iii) surface topographies, interfaces, and vicinities with entirely and diversely interior-space pores as transport gates for the multidiffusive accommodation of glucose targets and nonresistance movements of electrons/charges along surfaces

(iv) multidimensional arrangements and directional alignment around longitudinal and lateral axes for dense electron/charge transfer scales

Nanostructured material platform-based devices with multicomponent metal/nonmetal/metalloid hybrids and composites, interesting morphologies, electronic, optical, electrochemical, and conductive properties, high stabilities and surface activities, and flexible, stretchable, and compact size with low-power consumption can be used to develop diverse sensor devices for monitoring biomolecules, pesticides, pathogens, coronaviruses, chemical additives, and hazardous substances in food products. These nano/microscale sensor devices and protocols may have the following individual basic attributes in the improvement of food assessment:

(i) selective output signal analyses associated with noncomplex readout procedures

(ii) fast and multicultural sensing examination with an easy investigation for early detection purposes
(iii)sensitive operating conditions for the accurate evaluation and specific monitoring of food target dosage levels with >99% accuracy

(iv) real-time and self-interpretation of the examination of pesticides, pathogens, coronaviruses, chemical additives, and hazardous substances in food products to reduce food risks and contaminants

(v) indoor or outdoor online marking and identification, rapid, and convenient with long-range stability of analyses

In general, nanostructure materials are strong sensor candidates that meet the high requirements of the development of smart and compact food sensor electronics because of their optical, electronic, conductive, and magnetic features [20-22, 28, 29, 31, 33-35, 39]. The improvement of nano/microsensor devices and sensing protocols can be identified because of the following: (i) perennial electron transfer, (ii) proper accommodation of target species hosting, (iii) target-to-sensor surface interaction, and (iv) selective binding event and signaling [11, 38-42, 46]. Various nanostructured materials, including conductive polymers, metal hydroxides, oxides, carbon-based materials, quantum dots (QDs), and nanocomposite materials, have been utilized to create active platforms of nano/microsensor devices for food assessment (Scheme 2) [11, 20-22, 28, 40-42, 46-48].

4. COVID-19 pandemic and its relationship with food products

The COVID-19 pandemic is currently a global threat, causing massive loss of life, in addition to economic and commercial losses and disruption of daily activities in most countries. According to the data and last report of WHO on December 20, 2020, the total number of cases worldwide has reached 75 million and more than 1.6 million deaths have been recorded since the beginning of the pandemic [49-54]. Coronaviruses (CoVs) are infectious viruses that attack the human respiratory
system, causing pain, serious problems, and possibly death. Various kinds of CoVs include Middle East respiratory syndrome (MERS), severe acute respiratory syndrome (SARS), and SARS-CoV-2 (2019-nCoV and COVID-19) [55].

At the beginning of the COVID-19 pandemic, many reports related to the relationship between the COVID-19 pandemic and food have indicated that catching COVID-19 through food or its processing is unlikely. However, SARS-CoV-2 infection may be transmitted through direct exposure to respiratory droplets from the sneezing or coughing of an infected patient. Currently, considering the relatively small number of cases, people have some concerns and beliefs that food contamination contributes to the transmission of COVID-19 infection [56]. The first case of the food product infection appeared on June 12, 2020, at Xinfadi Agricultural Products Wholesale Market in Beijing, where SARS-CoV-2 was detected on a cutting board used for salmon. In this regard, frozen foods are considered a potential vector for COVID-19. Subsequent reports have confirmed that many infected cases may be due to food packaging materials. COVID-19 infections have been discovered in many foods, including salmon, shrimp, frozen chicken wings, and inside a shipping container [56, 57].

Therefore, the integrity of the global food safety monitoring system, international trade support, and food supply chain must be ensured. With the new generation of SARS-CoV-2, tracking the COVID-19 pandemic and its relationship with food should be prioritized to reduce the global risks that can threaten the food safety supply. A critical goal for food safety is growing fast to provide effective identification and on-time monitoring protocols of COVID-19 in food sources, ingredients, products, and infected food workers [58].

Prior to the outbreak of the COVID-19 epidemic, some speculations have indicated the possibility of the spread of the new coronavirus through food products or packaging as a potential risk factor [59]. Therefore, some precautionary measures have been taken, especially to employees who are working in the field of food retail, manufacturing, and distribution [60, 61]. SARS-CoV-2 can reach and
contaminate food products and then transmit COVID-19 infection in several ways: (i) touching of contaminated food package surfaces, (ii) contacting food products, fruits, and vegetables with an infected person, and (iii) consuming food products infected by animal viruses. However, several studies have confirmed that the real transmission of foodborne SARS-CoV-2 needs approval [62]. In a hypothesis, transmission may occur via frozen food or packaging, although its effective function may terminate within days after shipment [62, 63]. In general, although no logical explanation has confirmed the relationship between COVID-19 and food, the rapid evolution of the current situation regarding COVID-19 and the risks associated with its transmission and the detection of SARS-CoV-2 on frozen foods have contributed to the development of an updated strategy and provided insights into the assessment of the transmission risk through food and a review of the relationship between COVID-19 and food [59-65].

Several optical and electrochemical sensor techniques have been developed for COVID-19 detection through real-time polymerase chain reaction (PCR) to decrease direct contact among patients. However, the on-site, rapid, and accurate detection of COVID-19 by using wearable optical and electrochemical sensor devices remains challenging [66-68]. Currently, reverse transcription (RT) PCR examination is the most important laboratory technique for analyzing SARS-CoV-2. However, RT-PCR has several difficulties and limitations, such as (i) long processing times ranging from 2 h to 3 h to obtain the results, (ii) difficulty in operating, (iii) the necessity for well-trained technicians and qualified laboratories, (iv) expensive equipment, and (v) some false negative results obtained for infection [66-68]. In this regard, several studies on the development of COVID-19 screening have been performed to overcome these limitations [66-68]. Thus, the indoor/outdoor identification assay of SARS-CoV-2 antigen associated with food products and its industrial processing are urgently needed to control the infectious effect of the spread of COVID-19 pandemic.
5. Sensor protocols for the assessment of food ingredients, nutrients, additives, and byproducts

5.1 Scalable sensor devices of AA in the food industry

Vitamin C or AA is a vital nutrient of the monosaccharide family with wide functions. The biological activities of AA are conferred by its strong antioxidant property, and AA enhances the immune system’s efficiency against various diseases, including scurvy and colds [69-71]. It is soluble in water and a distinct ingredient of the Mediterranean diet. In addition, AA and its derivatives are widely utilized in the food industry as an antioxidant material and stabilizer [72, 73]. The consumption of AA in foods such as fruits and vegetables is essential for various metabolic processes because of the absence of L-gulonolactone oxidase that produces vitamin C from glucose [74]. Upon intake, AA exerts low toxicity to the human body. The most common complaints are diarrhea, nausea, stomach issues, and other gastrointestinal disorders because of the lack of a decreased effect of vitamin C on gastrointestinal nutrients. In addition, high AA intake is related with an expanded danger of cardiovascular disease, increased kidney stones, and extensive iron expansion that affect people with a hereditary plaque. The chronic consumption of AA can damage tissues, chromosomes, and DNA; cause cancer development; reduce B12 nutrient and copper levels; accelerate digestion or AA secretion; and promote tooth enamel erosion [75-78]. The permissible and recommended dietary level of AA has been estimated to be 100–120 mg per day. Vitamin C can be obtained from various food products, including citrus fruits, hips, tomatoes, cabbage, strawberries, spinach, and others. Therefore, scalable sensor devices for the accurate detection of AA in the food industry are important to ensure food safety [79-83].

AA detection is critical for the food industry and diagnostic application. The absence or lack of AA concentration leads to scurvy, whereas an increase in AA has many health effects. In this regard, significant endeavors have been given to build up an accurate, portable, straightforward, convenient,
and cost-effective sensor for AA detection (Table 1). Therefore, progressed sensory devices have accomplished genuine headway in daily checking and early warning requirements in relevant fields, such as food safety, military, life security, environment control, and medicinal observation [3, 5, 8, 15, 84-86]. For instance, numerous optical sensor techniques are utilized to detect AA levels [87-89]. In this respect, advancements in nano/micromaterial-based optical sensor devices for quick, ultrasensitive, and specific AA species are remarkable challenges that meet the necessities of daily monitoring assays of AA in the food industry.

Noble metal nanoparticles (NPs), such as Ag NPs and Au NPs, are widely used to design good SERS-based sensors for the visual detection of AA samples in foods [87, 90-93]. Fluorescence-based sensors are generally utilized in numerous applications, such as pharmaceutical enterprises and food and clinical analyses. Basically, a fluorescence approach to construct noninvasive AA sensors has numerous favorable benefits, including high sensitivity, host system protection, low cost, simplified procedure, and analysis, can be completed with fluorescence decay times [94-97]. Diverse analysis techniques based on fluorometric estimations, including chemiluminescence (CL), electrochemical chemiluminescence (ECL), surface-enhanced fluorescence, and surface-enhanced infrared absorbance, are used for AA detection in food, fruit juice and pharmaceutical products, and human samples [94]. For example, Ferreira et al. [98] described the design of optical paper-based sensors for the quantitative analysis of AA by using AgNPs and Ag ions as a chromogenic substance. A clear change in the color of a paper sensor strip indicates the simple detection of AA concentrations in real samples. Rostami et al. [99] fabricated AgNPs through seed-mediated growth on a substrate for a wide range of detection (ROD) of AA (0.25–25 μM) and LOD of 0.054 μM in commercial lemonade and pharmaceutical tablets (Scheme 3A). An Eu (III) luminescent complex is prepared by conjugating two nitroxide radicals as a sensory probe for AA visual imaging in living cells with a LOD of 9.1 nM (Scheme 3B) [100].
Variable carbon-based nanomaterials, including carbon dots (CDs), QDs, graphene QDs, N-, S-, and P-doped CDs, silicon QDs, and polyallylamine-AuNNCs@MnO₂, are used to fabricate the fluorescence sensors of AA in vitamin C tablets, human serum, fresh fruits, and commercial fruit juices [92-97] [95, 101-109]. Furthermore, a fluorescent switchable sensor is constructed by using carbon dots (CDs) doped with heteroatoms, such as sulfur and nitrogen (N, S-CDs), for AA detection in some fruits, including tomato, apple, grape, and kiwi (Scheme 3C) [110]. The designed sensor permits the detection of AA in the ROD of 10–200 μM and LOD of ~4.69 μM. An integrated sensing protocol based on the quenching mechanism of probe by Fe³⁺ is designed by using a dual fluorescent sensor to enhance a linear wide ROD of AA (Scheme 3D).

Along with optical sensors, CL-based sensors have lower emission background that avoids light scattering noise and shortens lifetime compared with fluorescence-based sensors. In general, the selection of substrates, reagents, and immobilization conditions is important for the development of efficient CL sensors. The advantage of CL sensor signal clearance is related to chemical reaction changes; consequently, it can provide an appropriate approach for the quantitative measurement of AA concentrations in the real samples of juice and food [111-118]. In this regard, AuNPs with different sizes are used to catalyze and improve the function of a luminol-K₃Fe(CN)₆ CL sensor, enabling the detection of AA in vitamin C tablets and fruit juice with ROD and LOD of 0.1 nM–1.0 μM and 0.2 nM, respectively [115]. CL analysis shows the improved properties of AA and amino acids on the same system. Cu²⁺ is used to enhance the CL efficiency of a luminescent probe, i.e., graphene QDs doped with nitrogen, during the detection of AA species in fruits [117]. This luminescent probe produces CL through oxidation with H₂O₂. In addition, the presence of Cu²⁺ expands the CL intensity by two orders of magnitude. This enhancement in CL intensity can be
attributed to the Fenton-like catalytic reaction of Cu$^{2+}$. This sensor determines the AA concentration with a LOD of 0.5 $\mu$M in fruits.

**Scheme 4**

Several fabrications of electrochemical sensor-based devices with carbon nanomaterials, metal nanoparticles, metal oxide nanostructures, polymers, and their composites have been reported for the promising detection of AA in food products, such as vitamin C tablets, orange juices, and other beverages [119, 120]. As such, an electrochemical sensor with a sulfur-doped microporous carbon microsphere (S-MCMS) electrode is used to detect AA in different resources, such as food and pharmaceutical products (Scheme 4) [11]. The annealing temperature under the N$_2$ flow is a key factor for producing a highly electroactive material with good characteristics of a porous surface, %S content, and microspherical orientation. The designed S-MCMS-modified electrode, which has a LOD of 1 $\mu$M and high stability and reproducibility, is utilized to detect AA in commercial juice products. A NiO/graphene nanocomposite film is employed for the detection of AA and folic acid in the real sample of vitamin C tablets with high sensitivity, selectivity, stability, and reliability [120]. Nanoporous carbon/cobalt oxide (NPC/Co$_3$O$_4$) nanocomposites for AA sensor electrodes in vitamin C tablets are synthesized through the dispersion of Co$_3$O$_4$ molecules (10 ± 2 nm) in the matrix of NPC through the direct thermal treatment of a ZIF-67 precursor (Scheme 5A) [121]. A reliable electrochemical sensor is obtained with high sensitivity (LOD = 20 nM), fast response, reproducibility, good stability, and capacity for the real-time monitoring of vitamin C tablets with 98%–102% recovery. In general, AA electrochemical biosensor electrodes constructed by using carbon materials alone and in combination with inorganic nanostructured materials have attracted great attention because of their promising properties [122, 123]. A glassy carbon electrode modified with Fe$_3$O$_4$/graphene oxide is fabricated using a green methodology for AA detection in vitamin C tablets [123]. Liu et al. [122] applied a plasma-assisted approach to prepare carbon fiber/ZnO core–
shell hybrids and fabricate microelectrodes that demonstrate a robust electrocatalytic biosensing performance to AA and DA with a linear ROD of 600–2000 and 20–200 μM, respectively (Scheme 5B). Metal nanoparticles, such as AgNPs, are employed for AA detection. In this manner, AgNPs are synthesized via microwave technique by utilizing Psidium guajava leaves as a source of capping and reducing agents [124]. The modified electrode with AgNPs exhibits a single electron reaction of AA in the control range of 150 μM ≤ AA ≤ 2000 μM.

**Scheme 5**

Scalable electrochemical organic transistors, such as poly(3,4-ethylenedioxythiophene) polystyrene sulfonate, are used to produce an electrochemical sensor electrode for AA detection in commercial juices, such as orange, and foods rich in vitamin C with a low LOD of 80 μM [125]. Li et al. [126] studied and constructed an electrochemical electrode for AA monitoring through the modification of a glassy carbon electrode (GCE) with AuNPs, MWNTs, and polyethyleneimine (PEI), producing AuNPs/MWNTs/PEI/GCE for stripping voltammetry detection. This electrode provides a high sensitivity toward a wide ROD of AA (0.5 pM–0.1 nM) and LOD of 15 fM. A highly sensitive copolymer of pyrrole and o-phenylenediamine layer is designed by modifying a pencil graphite electrode (PGE) through cyclic voltammetry copolymerization for the electrochemical detection of AA with a LOD of 0.263 μM [127]. Kong et al. [128] prepared poly(o-phenylenediamine-co-o-aminophenol) as a molecularly imprinted copolymer to detect AA electrochemically in orange juices and vitamin C tablets. During copolymerization, AA as a template molecule is incorporated, and electrochemical reduction is achieved by using an ammonium solution. Differential pulse voltammogram analysis reveals a ROD of 0.1–10 mM and a LOD of 36.4 μM. Nanocomposite-based electrochemical sensors are used effectively to detect AA in food products. For example, nanometal/metal oxides and carbon materials, such as graphene, and MWCNTs have
been widely used as active nanosensors for the selective monitoring of AA. A sensitive, fast-response, and reproducible sensor for AA detection in a vitamin C supplement is constructed using TiO$_2$/rGO nanocomposites [129]. The designed sensor yields the fast response detection of AA (<5 s), a LOD of 1.19 μM, a ROD of 25–725 μM, and a sensitivity of 1.061 μA μM$^{-1}$ cm$^{-2}$. Many researchers attempted to construct exceptionally delicate sensing platforms by using graphene-based compounds. Liu et al. [130] designed a nanocomposite of copolymerizing neutral red and RGO for AA detection in real samples, including vitamin C beverage. In addition, graphene–inorganic metal oxides and graphene–inorganic metal oxide/organic polymer nanocomposites have a wide range of applications in this field [131]. Das et al. [132] utilized graphene–iron oxide–polyvinyl alcohol (PIG) to modify a PGE for AA detection with a ROD of 0.04–4.1 mM and a LOD of 0.234 μM (Scheme 5C). Table 1 highlights AA sensory-based protocols fabricated by variable types of materials such as carbon, metal nanostructures, metal oxides, polymers, and composites. These novel sensory systems, sensing protocols, and analytical techniques enable the sensitive detection of AA in various food sources, dietary supplements, and human fluids.

**Table 1**

### 5.2. Sensing designs of reactive oxygen species for a safe food grade

Reactive oxygen has been used in food processing, industry, and medical applications to regulate physiological balance in living cells. H$_2$O$_2$ is the most recognized and stable oxygen reactive molecules. The presence of H$_2$O$_2$ accompanies the degradation and formation of free radicals, including O$_2^-$ and OH$^-$. H$_2$O$_2$ can destroy the walls of bacterial and viral cells through the conversion of the harmful byproduct H$_2$O$_2$ to O$_2$ and H$_2$O. Therefore, it is used as an antibacterial agent in foods, such as wheat flour, egg white, and milk, and as a sterilizing agent in food packaging. Moreover, enough H$_2$O$_2$ dosage should be used for food-grade purposes. Unfortunately, undesired H$_2$O$_2$ levels are considered the main causes of (i) unsafe food grade and (ii) various pathological disorders,
including Parkinson’s and Alzheimer’s diseases [133-135]. Although the use of H$_2$O$_2$ in the food industry is important to avoid food spoilage and increase its shelf life, but H$_2$O$_2$ is harmful because of lipid oxidation, caustic spoilage, reactive oxygen species formation, corrosive properties, and tissue damage [136, 137]. Thus, sensory H$_2$O$_2$ designed with high economic values, sensitivity, fast response, and selective signals in food safety has been widely explored in industrial and nutrition fields (Table 2).

Optical H$_2$O$_2$ sensors based on optical fiber probes are a technological development in chemical and molecular biochemical detection for many sensor applications [138, 139]. A H$_2$O$_2$ sensor based on optical fibers is constructed by coating fiber optics with polyvinyl alcohol decorated with AgNPs connected to SPR (Schemes 6A and 6B) [140]. This sensing system can detect H$_2$O$_2$ in the ROD of 0.01 μM–0.1 M. Potassium ferricyanide has been used as a precursor for the chemical deposition of a PB-nanostructured film into a multimode fiber optical tip, which can serve as a sensitive probe for H$_2$O$_2$ molecules [141]. The sensing mechanism is based on the possibility of using AA to reduce Prussian blue (PB) and produce Prussian white (PW), followed by re-oxidation via H$_2$O$_2$ (Schemes 6C and 6D) [142]. In addition, fiber optics based on HRP immobilization on a bovine albumin/glutaraldehyde matrix is enzymatically modified to improve the selectivity of H$_2$O$_2$ detection with a LOD of 0.5 μM [143, 144].

**Scheme 6**

Fluorescence-based sensors have attracted considerable attention because association with a target species provides a great opportunity for mapping and differentiation [145-153]. Hu et al. [145] reported a selective fluorescence detector for H$_2$O$_2$ based on the aggregation-induced emission manners of tetraphenylethylene interacting with H$_2$O$_2$ (Scheme 7A). Two-photon dyes of 6-hydroxy-2,3,4,4a-tetrahydro-1H-xanthen-1-one fluorescent-based probes are synthesized to detect H$_2$O$_2$ with a fast response in 9 min and can be used in the optical imaging of H$_2$O$_2$ in living organisms and tissues.
Ratiometric fluorescent sensors based on mitochondrion-targetable HBTPB (fluorescent probes) for H$_2$O$_2$ detection with a near-infrared emission present a high selectivity toward H$_2$O$_2$ with a LOD of about 0.59 μM (Scheme 7C) [155]. The performance of these sensors depends on the presence of H$_2$O$_2$ oxidizing HBTPB and the generated free OH group from 1,6-elimination reactions. In addition, a mitochondrion-targeting sensor is developed using a benzothiazole-based radiometric fluorescent chelating agent (HKB) with a wide Stokes shift for H$_2$O$_2$ detection (Scheme 7D) [156]. Shen et al. [157] revealed that a colorimetric and radiometric fluorescence sensor for H$_2$O$_2$ can be developed by altering a mitochondrion target with a coumarin moiety (Scheme 7E). The sensing protocol is dependent on the intermolecular charge transfer (ICT) and has a wide ROD and LOD of 0.1 nM.

A ratiometric near-infrared fluorescent sensor for highly selective H$_2$O$_2$ recognition under alkaline conditions and in vivo imaging application is established using a boric acid derivative to develop a sensitive fluorescence signaling [158]. A water-soluble turn-on fluorescent dipyrrromethene boron difluoride (BODIPY) is designed for selective in vivo H$_2$O$_2$ detection in HepG2/LO2 cells and angelfish (20 days old) (Pterophyllum scalare; Scheme 7F) [159]. As shown in fluorescent images, the probe can penetrate the fish body because of the obtained weak fluorescence; a clear fluorescence is detected after incubation with H$_2$O$_2$ because of the reaction between the probe and H$_2$O$_2$ inside the body.

Scheme 7

H$_2$O$_2$ should be identified and tracked, so simple sensing strategies, including CL and ECL techniques, have been used, and they have high sensing performance and selectivity depending on the control of sensor nanostructure designs. In this regard, various nanostructured materials, such as CuO NPs, CoFe$_2$O$_4$, bovine serum albumin-capped AgNCs, titania nanotubes, and platinum black-
functionalized indium tin oxide (ITO), significantly improve CL signals [160-163]. Other CL sensor designs based on nanostructured materials are potentially efficient candidates for the detection of \( \text{H}_2\text{O}_2 \) in the food sector [164, 165].

Electrochemical sensor designs based on nanostructural electrode surfaces have been widely explored as effective techniques for \( \text{H}_2\text{O}_2 \) species detection in food products [166-168]. In this regard, efforts have been devoted to developing enzymatic-/nonenzymatic-based electrochemical sensors and methods for \( \text{H}_2\text{O}_2 \) monitoring. Moreover, a prospective enzymatic sensor platform for \( \text{H}_2\text{O}_2 \) detection has been designed through enzyme immobilization, such as cytochrome C (Cyt c), into nanostructured materials to improve the electrocatalytic performance and redox efficiency of sensors toward \( \text{H}_2\text{O}_2 \) molecules in terms of sensitivity and selectivity [169-171]. An enzyme-based electrochemical sensor approach has many advantages, including high selectivity, high sensitivity, simplicity, and easy miniaturization (Schemes 8 and 9) [172-174]. Heme proteins, such as myoglobin, hemoglobin, and Cyt c, are used as \( \text{H}_2\text{O}_2 \) biosensors after the immobilization of the heme protein into an electrode surface for simple \( \text{H}_2\text{O}_2 \) detection (Scheme 8) [175]. Hemoglobin is actively immobilized at AgNPs encapsulated in a poly(amidoamine) dendrimer for \( \text{H}_2\text{O}_2 \) biosensing. The sensitivity of the sensor to \( \text{H}_2\text{O}_2 \) is in the ROD of 50–1150 \( \mu \text{M} \) with good reproducibility and stability [176]. However, a drawback of this approach is associated with its environmental sensitivity because of possible degenerate inactivation and aggregation of enzymes, co-enzymes, and proteins after immobilization into an electrode surface. Moreover, the immobilization of active signaling sites used for the electrode fabrication of an enzyme-based electrochemical sensor is more cost-effective than the nonenzymatic approach.

Scheme 8

Scheme 9
Numerous nanostructured materials, including NiO/graphene nanocomposites, cobalt oxide polyhedron-incorporated 3D graphene oxide (3D GO–Co$_3$O$_4$ PH), Co$_3$O$_4$ materials, cobalt(II) dicobalt(II) oxide nanoparticles, MW-CNT nanocomposite, ($\alpha$-Fe$_2$O$_3$/rGO), (NP-Fe$_2$O$_3$/CoO) CoO nanocomposite, and Co$_9$S$_8$ hollow spheres are designed electrochemical electrode sensors for sensitive and selective H$_2$O$_2$ detection [166-168, 177-183]. Nanocomposites of Co$_3$O$_4$ NPs and mesoporous carbon nanofiber (Co$_3$O$_4$-MCNFs), AgNWs, and AgNP-embedded N-doped carbon nanofibers (Ag/NCNFs) are synthesized as a nonenzymatic H$_2$O$_2$ selective sensor in milk samples with high sensitivity, stability, ROD up to 20 mM, and LOD of 0.15 μM [179, 184]. The electrochemically dense and uninformed decoration of PtNPs on a reduced graphene (RGO)/GCE electrode surface is used as a nonenzymatic H$_2$O$_2$ sensor with a LOD of 0.2 μM; this sensor enables the in vitro detection of H$_2$O$_2$ in living cells (Scheme 10A) [31]. Furthermore, graphene-based electrode materials, including Pt NP-decorated nonporous graphene (PG) and graphene (G)/CNT-encapsulated AuNPs, provide a highly active nonenzymatic electrode for H$_2$O$_2$ detection [182-185]. The nanocomposite of MoS$_2$ nanosheets (NS)/N-doped carbon NWs (produced from the sintered polypyrrole NWs) is synthesized for the electrochemical detection of H$_2$O$_2$ with a LOD of 0.73 μM and the possible detection of H$_2$O$_2$ released from A549 cells (Scheme 10B) [186]. Metal organic frameworks, such as ZIF-67, have been used to design electrochemical H$_2$O$_2$ sensors [187, 188]. An encapsulated Au@ZIF-67 is utilized to synthesize a hierarchically porous dodecahedron (Au@C-Co$_3$O$_4$) electrode for the H$_2$O$_2$ detection with a LOD of 19 nM, the recognition of H$_2$O$_2$ released from living organisms, and the identification of cancer cells (Scheme 10C) [187]. The carbonization of cobalt with an MOF (ZIF-67) to produce mesoporous Co-NPs doped with C-N featuring a rhombic dodecahedral morphology for nonenzymatic electrochemical H$_2$O$_2$ detection with a fast response (6 s), a wide ROD (0.001–30 mM), and a LOD of 0.143 μM [188]. A hierarchized Ni(OH)$_2$/RGO/Cu$_2$O sandwich in situ fabricated on Cu foil hydrothermally for H$_2$O$_2$ and glucose
detection exhibits a high amperometric response as a nonenzymatic detector with a LOD of 0.20 μM and a ROD of 0.5–7.5 μM (Scheme 10D) [189].

**Scheme 10**

The modulation of MWCNTs deposited on a tantalum (Ta) sensor electrode by a Pd-doped α-MnO₂ film via a spray pyrolysis approach shows high efficiency and sensitivity to assess the catalytic activity of H₂O₂ in milk samples with a sensitivity of 116.1 μAmM⁻¹ cm⁻², a ROD of 0.0032–22 mM, and a LOD of 0.02 mM [137]. In addition, a hybrid MnO₂/CNT/Ta nanocomposite sensor designed to detect H₂O₂ in milk in the ROD of 3–3025 μM with a LOD of 0.04 μM and a sensitivity of 83.3 μAmM⁻¹ cm⁻² [190]. Easow et al. [191] modified a GC electrode with a bimetallic Ag@Cu nanowire to improve electrocatalytic performance toward H₂O₂ compared with that of the modified GC electrode with Ag seeds and a bare GC electrode. This bimetallic NW sensor shows the successful detection of H₂O₂ in a pasteurized milk sample with a LOD of 3 μM and a wide ROD of 1–10 mM.

A nanocomposite of PtNPs (2 nm)–TiO₂ (8 nm)-coated RGO is fabricated via a microwave-assisted pathway to design flexible screen-printed sensors for the amperometric detection of H₂O₂ in milk [192]. The modified screen-printed electrodes by TiO₂/RGO, PtNP/RGO, and Pt–TiO₂/RGO can detect H₂O₂ in the ROD from 0 mM to 20 mM and a sensitivity of 40 μAmM⁻¹ cm⁻² [187]. A hybrid magnetic Au/Fe₃O₄-based sensor exhibits an outstanding H₂O₂ detection performance in milk and juice samples with a LOD of 0.108 μM, a ROD of 0.001–1 mM, and a sensitivity of 266 μAmM⁻¹ cm⁻² [193]. The hierarchical structures of N-doped carbon nanofibers comprising CoNPs and hollow mesoporous carbon particles with a good mechanical and electrical conductivity are characterized by excellent sensitivity and selectivity to H₂O₂ in food. A GCE electrode-modulated Co-NC/CNF leads to a high sensitivity to H₂O₂ detection of 300 μAcm⁻² mM⁻¹ in milk. Moreover, the formation of a Co-NC/CNF film on screen-printed electrodes (SPEs) enables the portable sensing of H₂O₂ in juice and milk samples [194]. An amperometric sensor based on the immobilization of the nanocomposite of
MWNTs and [Fe(III)-5,10,15,20-tetakis(\(\alpha,\alpha,\alpha,\alpha\)-2-piv-alamidophenyl)porphyrin] bromide [Fe(TpivPP)Br] on GCE (MWNTs/FeTpivPP/GCE) for \(\text{H}_2\text{O}_2\) detection in commercial beverages with a highly sensitive performance and a LOD of 0.05 \(\mu\text{mol L}^{-1}\) [195]. Challenges in the orientation of nanoscale sensor materials into portable sensing devices for \(\text{H}_2\text{O}_2\) monitoring in food and beverage samples remain sobering scenes.

**Table 2**

### 5.3. Detection protocols of high-purine foods for health risk assessment

High-purine foods, such as alcoholic beverages, some fish, and some kinds of meat are essential and daily food. The metabolism of these kinds of foods produces a uric acid (UA) and appears in different bodily human fluids, such as urine and serum. UA can affect various physiological functions of physical systems [196]. Given its chemical composition, UA serves as an antioxidant agent in physiological human processes. In addition, it acts as a reducing agent in bodily human fluids and accounts for half of the active antioxidants present in blood samples [197]. Protection from some neurological diseases, including Parkinson’s disease, Alzheimer’s disease, and multiple sclerosis, is related to a high UA level. This protection is conferred by the antioxidant activity or other obscure properties of UA [198, 199]. UA levels in bodily human fluids may also be associated with some clinical diseases, such as hypertension, metabolic syndrome, diabetes, gout, hyperuricemia and its related diseases, and erectile dysfunction [196, 197, 200, 201]

Fluorescence biosensors are noninvasive and simple tools for UA detection in human biological samples. Azmi et al. [202] constructed a fluorescence sensor for UA based on a CdS QD–enzyme system encapsulated in a 96-well microplate format. This sensor detects UA based on the quenching properties of \(\text{H}_2\text{O}_2\) produced from the CdS QD–enzyme system toward the QD fluorescence intensity (**Scheme 11A**). The designed sensor has a UA ROD of 0.06–2 mM and a LOD of 50 \(\mu\text{M}\). C dot-
engulfed Fe$_3$O$_4$ nanocomposites are used to design an electrochemical UA detection with a LOD of 6.0 nM in the linear ROD of 0.01–0.145 μM [203].

Tb$^{III}$-dtpa-bis(2,6-diaminopurine) is prepared as an UA fluorescent sensor with a LOD of about 5.80 μM [204]. The quenching feature of H$_2$O$_2$ is used as a base to determine UA concentrations. In this regard, the fluorescence quenching of CdTe NPs capped with various ligands is applied to enzymatically detect UA [205]. A fluorescent sensor containing Co$^{2+}$/H$_2$O$_2$–Amplex® UltraRed (Co$^{2+}$/H$_2$O$_2$–AUR) is designed for UA detection. The mechanism of detection depends on the oxidation of H$_2$O$_2$ and Co$^{2+}$ to provide Co$^{3+}$ and OH radicals that interact with an AUR fluorescence reagent (Scheme 1B) [206]. UA oxidation can produce different species, such as urea, which affects Co$^{2+}$ catalysis and AUR oxidation, leading to a decreased fluorescence intensity. The change in fluorescence intensity is associated with UA concentration. The designed sensor can detect UA in a wide ROD range of (0.05–1.0 μM) and a LOD of 20 nM.

A Cu$^{2+}$-based TMB (3,3,5,5-tetramethylbenzidine) with Fenton-like properties was constructed as a colorimetric-based sensor for UA detection, where H$_2$O$_2$ is a product generated during the enzymatic catalysis of UA [207]. The designed sensing platform can detect UA with a LOD of 0.64 μM. Wang et al. [208] used HAuCl$_4$ to oxidize TMB while inducing a blue color as a function of UA concentration with a ROD of 0.001–80.0 μM and a LOD of 0.67 nM. Various reports have focused on UA detection by using the CL technique [209, 210]. N-hydroxyphthalimide (NHPI) is used as an efficient CL–luminol system to develop a luminol–N-hydroxyphthalimide system. The luminol–NHPI system provides high sensitivity with a LOD of 910 nM [211]. An ITO electrode modified with N-doped graphene is utilized to detect UA electrochemically [212]. Many approaches have been designed for selective UA monitoring in the existence of AA and dopamine (DA) depending on the fabrication of nanostructured metal oxides and carbon materials. For instance, sulfur-doped carbon, NiO, and Fe$_2$O$_3$ show a good selective performance for UA detection with a wide ROD and a low
Various kinds of nanoscale materials of metal/metal oxides and carbon-based materials have been designed as extremely active materials for selective and sensitive UA detection and used for nanodevice fabrication.

**Scheme 11**

6. Nanoscale sensors for the assessment of food packing ingredients (nitrites and sulfites)

Nitrite anion is a well-known inorganic pollutant not only in the environment, water, and soil but also in the human body [214]. The various applications and usage of nitrites in food preservation, corrosion inhibitors, fertilizers and products, dye industries, and antimicrobial activities have led to their spread and harmful impact on public health [214-216]. The hazardous effect of nitrites on the human body is associated with the following reactions:

i) The reaction of nitrites with amines in food leads to the formation of carcinogenic N-nitrosamines (causing stomach cancer).

ii) The combination of nitrites with hemoglobin leads to the transformation of methemoglobin (Fe(II) to Hb-Fe(III)) (causes blue baby syndrome) [215].

Exposure to high nitrite levels may lead to some health risks, such as decreased blood pressure, increased heart rate, decreased ability of tissues to carry oxygen, headaches, abdominal cramps, vomiting, and even death [217]. Thus, developing nitrite sensors, such as optical and electrochemical sensors that feature a fast response, a low cost, high sensitivity and selectivity, stability, and reproducibility have attracted considerable interests (Table 3).

The development of visual detection protocols is one of the greatest approaches used in anion species assessment in foods [218]. Many designs of optical nitrite (NO$_2^-$) sensors based on colorimetric and fluorometric signaling for food analysis have been reported [224-231]. A colorimetric sensor for NO$_2^-$ is designed through the modification and stabilization of AgNPs with $N,N'$-bis(2-
hydroxybenzyl)-1,2-diaminobenzene as a chelating agent [219]. Hu et al. [220] prepared a fluorescent and colorimetric sensor based on CDs through a rapid microwave technique for NO$_2^-$ visual detection with low LODs of 196 and 0.518 nM (Scheme 12A). A design of NO$_2^-$ optosensor is fabricated by incorporating self-adhesive poly(n-butyl acrylate) microspheres with Safranine O reagent, leading to the creation of a visual detection protocol with a concentration ROD of 10–100 ppm in food samples [221]. NO$_2^-$ concentrations are detected in real samples by using strip-typed colorimetric sensors covered by a polyethylene glycol hydrogel to diminish the nonspecific connection of bio-samples and color immobilization-producing probes (Scheme 12B) [222]. The proposed strip sensor modified with a colorimetric sensor based on N-(1-naphthyl) ethylenediamine and sulfanilamide has a ROD of 10–5000 μM and a LOD of 10 μM.

**Scheme 12**

Cardoso et al. [223] designed μPADs (microfluidic paper-based analytical devices) for colorimetric NO$_2^-$ detection in environmental, food, and clinical samples with a LOD of 5.6 μM. The mechanistic detection of NO$_2^-$ is achieved through the modified Griess reaction by using a solution of sulfanilamide, hydrochloric acid, and N-(1-naphthyl) ethylenediamine. Zhan et al. [224] reported diazonium salts formed through the reaction of NO$_2^-$, and aminated m-CDs can be used for visual NO$_2^-$ monitoring for food quality evaluation and control. The designed fluorescent sensor successfully detects a low level of NO$_2^-$ species in a ROD of 0.063–2 μM and a LOD of 0.018 μM in food and environmental products. Moreover, an optical fluorescence sensor based on hyperbranched polyethylenimine-capped CdS quantum dots (5 nm diameters) is fabricated for the selective and sensitive monitoring of NO$_2^-$ species in vegetable and water sources in a wide ROD of 0.1–100 μM [225]. The mechanistic TMB oxidation reaction by nitrite species to form yellow ox-TMB leads to the formation a colorimetric sensor assay. NO$_2^-$ reading out signals are detected through digital image colorimetry (DIC) designed to detect nitrite in food samples via a smartphone [226].
The reading out strategy relies on the color contrast of the saturated images and analyzes color by using a red, green, and blue (RGB) model. In addition, this sensing strategy provides the quantitative detection of NO$^{2-}$ in cabbage, pickle, and pork in the ROD of 10–440 μM and a LOQ of 2.34 μM. Studies related to the design of active electrode materials have been devoted to the effective detection of electrochemical NO$^{2-}$ [227-229]. The air annealing treatment of carbon fiber paper (CFP) activates surface properties in terms of increased oxygen containment, higher roughness, enhanced wettability, and produced more defective edge/surface locations [230]. The electrochemical sensors designs have been developed based on nanoengineering of various metal nanoparticles, metal oxides nanostructure and their campsites for sensitive and selective monitoring of nitrites [231-238]. This treated CFP is used to detect NO$^{2-}$ species in food samples in the ROD of 0.1–3838.5 μM, a LOD of 0.07 μM fast current response of 1 s, and a sensitivity of 930.4 μAmM$^{-1}$ cm$^{-2}$. Selective NO$^{2-}$ is detected in meat samples by using a portable Pt-Nafion-based electrochemical sensor [239]. Üzer et al. [240] fabricated an electrochemical sensor for NO$^{2-}$ species by electrochemically modifying a working electrode (Au) by using polyaminothiophenol (PATP) and Au NPs. The designed Au/PATP-AuNP electrode sensor shows high stability and sensitivity for NO$^{2-}$ identification in sausage samples with a LOD of 0.12 mg L$^{-1}$ and a wide ROD of 0.5–50 mg L$^{-1}$. An electrochemical sensor electrode is designed through GCE modulation by using ZnO/PtNP/MWCNT nanocomposites to detect NO$^{2-}$ in pickled food with high sensitivity [241]. This well-designed ZnO/PtNP/MWCNT/GCE sensor shows the detection of NO$^{2-}$ in the ROD of 0.4–200 μM and a LOD of 0.082 μM. Rao et al. [242] designed sensing protocols by using a AuNP/chitosan @N,S co-doped MWCNTS electrode for the selective and sensitive monitoring of NO$^{2-}$ detection in some food products (mustard tuber, ham sausage, and fermented bean curd) within the ROD of 1–7000 μM and a LOD of 0.2 μM. A rGO/MoS$_2$/poly (3,4-ethylene dioxythiophene) (PEDOT) nanocomposite is used to design a nonenzymatic NO$^{2-}$ sensor electrode in water and milk samples [243]. rGO/MoS$_2$/PEDOT
nanocomposite-modified GCE enhances the electrocatalytic performance toward NO$_2^-$ oxidation, leading to a good NO$_2^-$ sensitivity of 874.19 μAμM$^{-1}$ cm$^{-2}$, a LOD of 0.059 μM, and a wide ROD 0.001–1 mM. These reports have reflected the necessity of NO$_2^-$ detection to ensure food quality and healthcare. In addition, these sensor patterns and devices have been used for nanosensor designs and NO$_2^-$ detection in the food chain.

Inorganic anion species, such as sulfites (SO$_3^{2-}$), are important in food evaluation because of their wide use in food processing industries, including food storage and preservative additives for fruit juices, dried fruits, and wines. Sulfate anions are important in improving flavor manifestation through all food processing stages, including preparation, storage, distribution, and color stabilization [244, 245]. Furthermore, SO$_3^{2-}$ is commonly used for antimicrobial growth, bleaching agents, enzyme inhibitors, oxygen scavengers, and antioxidants [246, 247]. In this regard, great endeavors have been made for designing sensitive and selective SO$_3^{2-}$ sensors, especially in the food industry [248-250].

Several sensing methods, including colorimetry, electrochemistry, fluorimetry sensors, are designed on the basis of a nanosensor platform to detect SO$_3^{2-}$ with satisfactory selectivity and sensitivity. For example, the capability of SO$_3^{2-}$ to inhibit Ag$_2$O and Ag$_2$O nanoparticles enables the colorimetric detection of SO$_3^{2-}$ in the ROD of 0–300 μM and a LOD of 1.74 μM [251, 252]. Ag$_2$O acts as an oxidase that catalyzes the oxidation of peroxidase substances. SO$_3^{2-}$ is detected using colorimetric and near infrared fluorescent probes. Fluorescent organic nanoparticles (FONs) are designed via the self-assembly of tetraphenyl imidazole (TIBM) for ultrafast SO$_3^{2-}$ detection (15 s) and a LOD of 7.4 nM [253]. A red-emitting ratiometric fluorescence probe of 6-(dimethylamino) quinoline-2-carbaldehyde) acetonitrile and 2-(1-H-benzo[d]imidazol-3-ethyl-2-yl (BIQ) is fabricated for the sensitive and selective detection of HSO$_3^-$ in food chains of white wine and sugar [254]. A BIQ probe displays fast fluorescence and color response (<2 min) with a LOD of 0.29 μM. Fang et al. [255]
considered the quenching properties of fluorescent carbon quantum dots (C-dots) with SO$_3^{2-}$ and Cr(VI) ions to design an effective fluorescence probe for SO$_3^{2-}$ monitoring with a LOD of 0.35 μmol/L.

The modification of carbon paste electrode (CPE) with 3-n-propyl(4-methylpyridinium) silsesquioxane chloride (Si4Pic$^+$Cl$^-$)/AuNPs(45nm) is designed an electrochemical SO$_3^{2-}$ sensor in coconut water and white wine samples [256]. The designed (Si4Pic$^+$Cl$^-$)/AuNPs/CPE sensor shows good performance in the ROD of 2.54–48.6 mg/L with a LOQ of 0.88 mg/L and a LOD of 2.68 mg/L. Devaramani and Malingappa [257] modified CPE by using cobalt nitroprusside (CoNP) NPs and showed high sensitivity to detect SO$_3^{2-}$ in food samples, including sugar, dry grapes, and wine with a LOD of 0.04 μM and a LOQ of 0.229 μM in the ROD of 1–8 μM. Devadas et al. [258] used a praseodymium hexacyanoferrate electrode for the electrochemical detection of SO$_3^{2-}$ in red wine samples with good high sensitivity of 0.036 mA mM$^{-1}$ cm$^{-2}$ and a LOD of 2.15 mM. The modification of graphitic carbon electrode by using cobalt hexacyanoferrate is designed for the amperometry detection of SO$_3^{2-}$ in various food samples of tomato ketchup, wine, jam, sugar, and dry grapes with a LOD of 1.7 mM in the ROD of 4–128 mM [259]. The immobilization of synthesized phenothiazine imidazolium and hexafluorophosphate counter anion (PTZ-IL) into the modified GCE with MWCNTs to produce a PTZ-IL/MWCNT/GCE sensor utilized to detect SO$_3^{2-}$ in the ROD of 30–1177 μM with a LOD of 9.3 μM and a sensitivity of 282.2 μA mM$^{-1}$ cm$^{-2}$ [260]. Moreover, the modification of CNTs with active organic materials, such as chitosan and graphene transistors, are used for sensitive sulfite anion detection in food chains [261, 262]. Currently, various sensing strategies and nanomaterial-based sensors are used to efficiently screen sulfites in food samples (Table 3). However, the development of portable nanosensor devices for the selective and sensitive assays of nitrite and sulfate species in food products and industry has attracted increasing interests.

Table 3
7. Nanosensor designs for the detection of organic and inorganic toxins in foods

7.1 Sensing protocols of toxic heavy metals in food products

Environmental pollution with heavy metals and their direct or indirect association with food processing threatens the quality of food and agricultural products and human health. In general, food can become contaminated with heavy metals from environmental/industrial tools, agricultural water, marine, and beverage chemicals and additives. Food delivery associated with labeling materials and package contents (text/ink patterns, and packaging bottles and papers) may lead to polluted food [263-265]. Adverse health effects are unquestionable with the consumption or intake of large/low amounts of contaminated food sources.

For instance, marine environmental pollutants by naturally occurring amounts of heavy metals pollute marine organisms, fish, and seafood products (fish and shellfish). The accumulation of nondegradable and harmful heavy metal contaminants in marine food chains above the legal limits potentially threaten human health. Several studies have been conducted in Bangladesh (Saint Martin Island) and showed evidence on the presence of high level of toxic heavy metals such as Pb²⁺, Hg²⁺, As³⁺, Cr⁶⁺, Cd²⁺, Fe³⁺, Cu²⁺, Zn²⁺, and Mn²⁺ in the muscles/armor/exoskeleton of shrimp, lobsters, and crabs [266]. Several sensing techniques, such as atomic absorption/emission spectroscopy (AAS/AES) and inductively interconnected plasma mass spectroscopy (ICPMS), are used to detect the traces of heavy metal contents in food and environmental samples [263, 266, 267]. These sensing techniques are used to detect sensitively toxicant metals, but some challenges are associated with the high cost, sample preparation, and complexity of analysis and users’ technical expertise. In this regard, great efforts have been devoted to developing a simple, accurate, and cost-effective approach for the fast tracking and indoor/outdoor field trials of heavy metals to ensure food quality (Table 4) [33, 84, 85, 268-275].
Optical and electrochemical sensors have been widely used for metal ion monitoring and the efficiency of remediation/treatment with good selectivity and sensitivity without requiring further treatment [271, 276-291]. In this context, different chelating agents, such as N,N’-disalicylidene-4,5-diamino-6-hydroxy-2-mercaptopyrimidine (DSAHMP), (4,5-diamino-6-hydroxy-2-mercaptopyrimidine and diphenylthiocarbazone), and dicarboxylate 1,5-diphenyl-3-thiocarbazone (III) are used as colorimetric probes for the visual detection and extraction of Zn\(^{2+}\), Pd\(^{2+}\), Cd\(^{2+}\), Cu\(^{2+}\), Co\(^{2+}\), and Hg\(^{2+}\) [85, 279, 280, 292]. These visual sensors provide fast and easy-to-use on-site detection of target metal ions in water, food products, marine organisms, and other environments. Fluorometric sensors have been widely used to monitor heavy metals in various samples, including water, food, and pharmaceuticals [293-296]. A sensitive and selective fluorescent probe, namely, 2,2’-((1Z,12Z)-5,9-dithia-2,12-diazatrideca-1,12-diene-1,13 diyl) diphenol, is synthesized and loaded on Zr-MOFs for the detection of Zn\(^{2+}\) and Bi\(^{3+}\) [294, 297]. Tumay et al. [298] fabricated an optical sensor based on pyrene-modified nanocrystalline cellulose to detect Cd(II) ions in food plant samples in a linear ROD of 0.1–60 \(\mu\text{M}\) with a LOD of 0.09 \(\mu\text{M}\).

Nanostructure-based materials for electrochemical sensor designs are widely used to improve the performance of detection sensitivity [299, 300]. For the electrochemical monitoring of heavy metals, a number of identified compounds, such as aptamers [301] and anticell and enzymes [302, 303], are used with nanostructured carbon-based materials. An imprinted chitosan/GR/GCE electrode is designed for Cd\(^{2+}\) detection in milk and drinking water in the ROD of 0.1–0.9 \(\mu\text{M}\) and a LOD of 0.162 nM [304]. Wang et al. [305] designed an electrochemical aptasensor-based sensor. This sensor can detect Hg\(^{2+}\), Pb\(^{2+}\), Cd\(^{2+}\), and arsenic in food samples. A double-sided conductive carbon tape modified by a thin Au layer enhances the electrochemical detection of Cd\(^{2+}\) in rice based on a paper-based electrode with a LOD of 0.1 \(\mu\text{g/L}\) and a linear ROD of 2–50 \(\mu\text{g/L}\) [306]. Wu et al. [307] fabricated Fe\(_3\)O\(_4\)/F-MWCNT nanocomposites to detect heavy metals in food chains. The designed
Fe\textsubscript{3}O\textsubscript{4}/F-MWCNT platform is effectively used for Cd\textsuperscript{2+}, Pb\textsuperscript{2+}, Cu\textsuperscript{2+}, and Hg\textsuperscript{2+} detection in food with good sensitivity of 108.79, 125.91, 160.85, and 312.65 μA mM\textsuperscript{-1} cm\textsuperscript{-2}; a LOD of 0.05, 0.08, 0.02, and 0.05 nM; and a wide ROD of 0.5–30, 0.5–30, 0.5–30, and 0.5–20 μM, respectively. Moreover, a modification of working electrode by MOF materials has been effectively used to detect multiple heavy metal actions, such as Pb\textsuperscript{2+}, Cd\textsuperscript{2+}, Cu\textsuperscript{2+}, and Fe\textsuperscript{3+} with high sensitivity and selectivity [308-310]. Indeed, current reports indicate the key roles of nanostructured materials in designing extremely selective and sensitive sensors for the critical assessment of heavy metals in food samples. The development of smart and compact nanosensor devices for toxic metal detections in food remains a practical challenge. Nanosensors integrated in portable devices enable on-time usage in the basic laboratory assessment and evaluation of food quality in-home and commercial markets.

7.2 Sensing protocols and designs of bisphenol A toxicants

Bisphenols, such as bisphenol A (BPA), are a group of organic compounds that containing two hydroxyphenyl bridged by one carbon atom. From the 1960s, BPA has been widely used in the production of polycarbonate plastics and resin industries [311]. It has been magnified as a serious toxicity to food and human health. It can be released to food through the packaging and contact process (food packaging, tableware, and cooking utensils), which poses a health threat associated with an endocrine disrupter with a reduced estrogenic behavior [312-314]. Recently, the European commission decreased the permissible contents of BPA in plastic materials from 0.6 mg/kg to 0.05 mg/kg [315]. In Catalonia, Spain, traces of PBs corresponding to 93% and 36% are present in canned and noncanned foods, respectively [316]. Many techniques, including optical, liquid chromatography-tandem mass spectrometry, electrochemical, and other methods, have been used to evaluate the risk and detect BPA with good sensitive and selective screening in various samples (Table 4) [311, 317-320].
This critical report aims to assess and recognize the BPA in fish and water samples. For example, a colorimetric sensor is designed using Fe$^{3+}$ nanocomposite and fabricated BTSIXO (3',6'-bis(diethylamino)-2-((3,4,5-trimethyl benzylidene) amino) spiro[isoindoline-1,9'-xanthen]-3-one) [321]. This optical sensor performs a high sensitivity with a LOD of 0.02 ppm and a wide ROD (0.1–150 ppm). Several studies have revealed the importance of AuNPs for the potential design of aptasensors in the colorimetric detection of BPA [322-324]. Jia et al. [322] reported a label-free aptasensor with the truncated 38- and 12-mer aptamers conjugated to AuNPs, providing a LOD of 7.60 and 14.41 pM, respectively. Color changes from red to blue associated with AuNP aggregation in the presence of low concentration BPA levels. This colorimetric sensor is used for BPA detection in grain rice with a LOD of 0.004 nM [323]. In addition, AuNP aggregation induced by the BPA–aptamer and cationic polymer is used to detect BPA with a LOD of 1.5 nM [324].

A visual microfluidic paper detector is designed through the integration of ZnFe$_2$O$_4$, molecularly imprinted polymer (MIP) and cellulose fibers to detect BPA with a LOD of 6.18 nM in a wide concentration ROD of 10–1000 nM [325]. BPA concentrations can be detected as a function of the change in gray color intensity because of the catalytic activity of the 3,3',5,5'-tetramethylbenzidine and ZnFe$_2$O$_4$ peroxidase mimetics. Lee et al. [326] reported the conjugation of aptamer/AuNPs and fluorescing single-strand DNA (ssDNA) aptamer as a fluorescent probe for BPA detection with a LOD of 9 pg/mL. Moreover, the conjugation of DNA-QD$_{655}$ with magnetic beads-QD$_{565}$ and the aptamer is used to design a selective fluorescence sensor for BPA with a LOD of 1.0 ng/mL in the presence of bisphenol B and C [327]. A fluorescent sensor based on a field-effect transistor (FET) through aptamer-modified multichannel carbon nanofibers (MCNFs) is used for selective BPA detection with a LOD of 1 fM [328]. The integration of GO and anti-BPA aptamer (FAM-ssDNA) is applied to design a selective and sensitive BPA fluorescent sensor with a LOD of 0.05 ng/L in the ROD of 0.1–10 ng/mL [329]. Li et al. [330] developed a fluorescent sensor by using a specific BPA
aptamer, AuNPs, and CdTe QDs for the detection of BPA with a LOD of 1.86 ng/mL and a linear ROD of 10–80 ng/mL.

Most of the reported optical sensing methods have shown evidence supporting the sensitive and selective detection of BPA, but they suffer from some limitations, such as multiple step sensor design, time consuming, and short lifetime. The electrochemical sensing approach of BPA is widely available compared with optical detection strategies. Electrochemical BPA sensors have intense features, including the cost-effectiveness, short sensing time, on-site detection, high stability and reproducibility, and good sensitivity and selectivity [331-336]. The modified electrode of B-doped diamond and nanocarbon can serve as a potential sensor for BPA detection with good selectivity and sensitivity and LOD of 13 nM [331]. A carbon-based electrode of GO and MWCNTs functionalized by β-cyclodextrin is used for the selective monitoring of BPA in the ROD of 0.05–30 μM and a low LOD of 6 nM [332]. Moreover, a functionalization/immobilization of CNT materials with biomolecules (i.e., tyrosinase), carbon black, aptamers, and molecularly imprinting polymers offers simple and smart detection methods of BPA with good sensitivity and selectivity [333-336].

GCE modification with a Cu–Zn/GO nanocomposite shows high sensitivity and selectivity for BPA detection at pH 7.1 by using square wave voltammetry (SWV) with a LOD of 0.88 nmol/L and a linear ROD of 0.003–20 μM [337]. The modification of a single crystal TiO₂ electrode via inorganic framework molecular recognition is applied to monitor BPA with high accuracy, good stability, low interfering response, and rapid response time. This electrode sensor displays a ROD of 0.01–20 μM and a LOD of 3 nM [335]. AuNP-decorated TiO₂ NT nanosensors are designed for photoenhancing the electrochemical detection of BPA with a LOD of 6.2 nM and electrode sensitivity of 2.8 μAμM⁻¹ cm⁻² [338]. The coupling of Cu-MOFs with ball mill-exfoliated graphene is used to design an electrochemical sensor electrode for BPA detection [339]. Electropolymerized poly(pyrrole-nitrilotriacetic) acid and DNA aptamer with a pentahistidine peptide are functionalized and used for
BPA detection with high signaling stability, high signal affinity, wide linearity of 0.01–1000 nM, and high sensitivity of 372 Ω/unit [334].

Nanoscale materials are used in the development of various analytical approaches to meet the selective and sensitive sensor/biosensor designs of BPA. Progress in the field screening of BPA-based nanomaterial-sensors provides signaling enhancement, simple recognition, long-run durability and stability, fast response, and low cost. Significantly, the fabrication of portable sensor devices can save cost and time spent on analyses and minimize the risk of BPA contamination in food.

**Table 4**

8. Nanosensor-based devices for pesticide detection

One of the hot issues and global concerns in drinking and food safety are not only the treatment of chemicals, including pesticides, but also the detection of pesticide residues. Pesticides have a serious impact on the surface of water and food, thereby providing a driving force for major research efforts on effective ways to recover and manage these chemicals (Scheme 13A) [340-343]. Several pesticides are applied to agricultural fields each year to protect crops from damage and diseases, promote plant growth, and increase yield.

Lower than 0.1% of pesticides can prevent target pests from infesting crops. The remaining amount unnecessarily penetrates the environment, specifically contaminates the environment and enters the food system. Pesticides can be found in food or animal feed, and they may be indirectly or directly consumed by people. Eating pesticide-based nourishment is a principal method for pesticide pollution. As highly toxic species, some pesticide residues cause serious problems in aquatic life and food systems [340-343]. Because of the increased public awareness of the health and environmental risks of pesticides, detection, and monitoring of pesticide residue levels in the drinking and food chain has attracted great concern worldwide. Several techniques, including SERS, SPR, EISA, and electrochemical approaches, are used to determine the level of
pesticide residues [344-346]. Tracking and sensing of pesticide residues are essentially associated with food categories and pesticide species. Pesticide species can be classified as organic or inorganic substances and based on their physiological effects.

Given its ultrasensitive sensing, simple protocol, and cost-effectiveness, surface-enhanced Raman scattering is an effective strategy to detect pesticide residues in food chains. Nanomaterials based on noble metals, such as Ag and Au, have been used as a prevailing platform to detect pesticide residues with fast response and high sensitivity and selectivity [347, 348]. In addition, semiconductor–metal hybrid platforms have been utilized to improve the pesticide detection performance of SERS with good reproducibility, acceptable stability, and reusability. SERS may provide a promising approach for the direct pesticide detection in liquid samples, surface, and solid samples, but using it in complex solid food matrices is difficult.

Scheme 13

A great effort has been devoted to the engineering of flexible SERS frameworks based on the integration of active materials, such as carbon, and noble metals with flexible materials, including cellulose paper and polymer membranes that can be applied as a batch combined with food surfaces for SERS analysis [349-353]. Ma et al. [354] fabricated an integrated hybrid paper-based SERS for pesticide residue detection in vegetables and fruits by using Ag NPs and GO on cellulose paper through a screen-printing technique (Scheme 13B). The designed SERS paper offers a high sensing performance toward different pesticides in complex surfaces and LODs lower than permissible levels based on pesticide types, such as about 0.26 ng cm$^{-2}$ for thiram.

A SERS sensor is modified with Ag nanowires with M13 bacteriophages through a mixing and filtration process involving glass-fiber filter paper [355]. In this process, the glass-fiber paper is stacked with Ag nanowires, and the surface is decorated with a tryptophan–histidine–tryptophan
peptide sequence (BPWHW) as a bioreceptor to produce a selective sensor for paraquat pesticide (Scheme 13C). Cellulose filter paper-based sensors are modified with an alkyl ketene dimer to improve the hydrophilic nature of the filter with increasing density and dispersion of AgNPs in the filter paper for pesticide-based SERS [356]. The modified SERS sensor has LOD of 0.46 and 0.49 nM for thiram and ferbam, respectively.

Yao et al. [357] designed a magnetic molecular imprinting sensor based on polydopamine and Fe₃O₄ NP nanocomposites to improve the sensitivity and specificity of SPR to pesticide detection with a LOD of around 0.76 nM in the ROD of 0.001–10 μM. Multiple classes of pesticides are detected through the immobilization of glutathione-S-transferase amperometry biosensors [358]. Pesticides such as carbamate and OPs working as inhibitors for acetylcholinesterase (AChE) cause neurological diseases; AChE has the responsibility to terminate nerve impulse transmission at the synapse [359, 360]. da Silva et al. [361] constructed glassy carbon/rGO/AChE as a biosensor electrode for carbaryl monitoring in tomato. AChE is immobilized into Pd–Cu nanowires to construct a modified electrode for the quantitative monitoring of organophosphate pesticides (OP) in fruits and vegetables [362]. The proposed biosensor electrode displays a high sensitivity toward malathion in the ROD of 0.005–1 and 500–3000 ppb with a LOD of 1.5 ppt. Moreover, malathion is detected using an electrochemical sensor electrode based on AChE-modified transition metal carbide (Ti₃C₂Tx-based) nanosheets and chitosan [363]. The designed AChE/CS-Ti₃C₂Tx/GCE electrode displays a high electrochemical performance for malathion monitoring with a LOD of 0.003 pM. Lu et al. [364] immobilized AChE into 1D Pd@Au core–shell nanorods to prepare a highly sensitive biosensor electrode for detecting OP pesticides, such as paraoxon, with a LOD of 3.6 pM in a wide ROD of 3.6 pM–100 nM.

The photoelectrochemical (PEC) detection of targeted species provides many advantages, including high sensitivity with a completed separation between the photocurrent signal of the
detector and the excitation light source, low background noise, simple device, convenience, and low cost. Cheng et al. [365] designed a PEC biosensor to detect highly toxic OP pesticides widely used in agriculture. This electrode is constructed by immobilizing an AChE “bioreceptor” with glutaraldehyde as a cross-linking agent into a TiO$_2$NP “photoactive material” and a nitrogen-doped carbon QD “photosensitizer”-modified ITO electrode to produce a biosensing platform. Nitrogen-doped carbon QDs with a brilliant optical performance and a high electrical conductivity can improve the visible light of the TiO$_2$NP-ITO platform by 42 times. The designed PEC biosensor is used to detect chlorpyrifos in the ROD of 0.001–1.5 $\mu$g/mL and a LOD of 0.07 ng/mL. In this context, graphene QDs, AChE, and choline oxidase (CHOx) are designed as an active GQD/AChE/CHOx platform for the direct PEC detection of OPs (Scheme 14A) [366]. The mechanism of the sensor is based on the generation of H$_2$O$_2$ from the AChE–CHOx reaction. The reaction of H$_2$O$_2$ with GODs generates turn-off photoluminescence properties at 467 nm, and turn-on photoluminescence can be recovered in the presence of OP pesticides. The LOD of the designed electrode toward dichlorvos is 0.172 ppm. A Au nanostructure is commonly used as a sensitive substrate to detect amperometric dimethoate pesticides with LODs of about 3.9 nM and 16.4 $\mu$m [367-369]. Chen et al. [369] reported a fast colorimetric probe to detect pesticides, including dimethoate and terbuthylazine, with LODs of 20 nM and 0.3 $\mu$m, respectively, and necked eye visualization based on AuNPs (Scheme 14B). Various novel nanoscale materials are fabricated to develop the potential detection protocols of pesticides and to optimize and screen food safety and environmental quality.

**Scheme 14**

9. **Monitoring protocols of foodborne pathogens**

Pathogenic microorganisms are common forms of environmental pollution because of population growth, human consumption, and poor waste management. A safe drinking water
supply is a basic requirement for human health. In consideration of many types of microorganisms can contaminate drinking water and beverages, the total number of bacteria is the main feature of quality assessment. Waterborne microbial agents, such as bacteria, pose a major risk and cause many serious diseases; consequently, they have become a major burden on public health over time. Therefore, a rapid, sensitive, and selective approach of detecting pathogenic microorganisms should be developed. Several studies have focused on developing a method for the fast and good selective monitoring of bacterial pathogens [370-375]. Current detection methods require a significant amount of time. Samples should be concentrated by reducing the sample size for rapid contamination and by adopting pathogenic bacteria. Various types of nanomaterials have been manufactured for the rapid detection and elimination of pathogens [370, 372, 376, 377]. However, the selectivity of these protocols poses challenges because target bacterial pathogens are usually present in complex samples at very low concentrations.

**Scheme 15**

Various techniques used for bacterial detection include RT-PCR, SPR, bulk acoustic wave impedance biosensing, microscopic examination and ATP measurements, ELISA, fluorescence and bioluminescence assays, and fluorescent in situ hybridization [378-383]. Extensive reports have focused on the importance of the nanostructured features of metal oxides. Mesoporous hexagonal MgO nanosheets with a highly active surface are designed as a powerful antibacterial agent for *Staphylococcus aureus*, *Pseudomonas aeruginosa*, and *Escherichia coli* in water (Scheme 15A) [384]. The underlying mechanism is based on the interaction with the cell wall proteins that affect its permeability, and ROS released from the designed material reacts with cell components, causing death (Schemes 15A–15C) [384, 385].
Tokel et al. [386] developed a portable label-free pathogen platform based on the integration between microfluidic and SPR technologies to detect and quantify *E. coli* and *S. aureus* (Scheme 16). A microchannel chip design consisting of a single microchannel with an outlet is created as a cartridge. This microchannel is located at the center of a slice consisting of two methyl methacrylate layers (PMMA) joined by a layer of a double-sided adhesive (DSA). A microchannel shaped from a second DSA layer and Au-plated platform, and a PMMA-DSA-PMMA-DSA-Au chip are obtained. The designed sensor displays a reliable ROD of *E. coli* at $\sim 10^5 - 3.2 \times 10^7$ CFU/mL in PBS. A simple DC process is used to manufacture a flexible SERS platform through AuNP deposition on dragonfly wings, thereby creating a promising bioplatform to detect pesticide residues [387]. The designed platform is utilized to detect cypermethrin (pesticide residue) in tomato husks through a pressure and peeling method. The platform can rapidly and reliably detect cypermethrin with a LOD of $10^{-3}$ ng cm$^{-2}$.

**Scheme 16**

Sedki et al. [370] fabricated an RGO-hyperbranched chitosan nanocomposite as an active electrode for the sensitive and real-time detection of bacterial cell viability based on microbial electrochemical systems (Scheme 17A). *E. coli* and *P. aeruginosa* are used as targeted pathogens to evaluate electrode performance. The results show that the designated nanocomposite electrode exhibits the fast response/detection of cell viability and enhances the bioelectrochemical detection limit. Optical scanning microscopy is used as a digital, rapid, accurate, and label-free strategy to evaluate the microorganism’s viability through the development of bacteria on a glass-slide cell culture device (Scheme 17B) [371]. Using *E. coli* as a model bacterium confirms the fast response of the device with a decreasing experimental duration from 13 h to 5 h with the accurate quantitative determination of bacteria and the automatic accurate counts of microcolonies. In addition, the designed system allows the use of small pieces of agar plates
instead of whole agar, thereby shortening bacterial counting. Højris et al. [388] provided an optical on-line bacterial sensor through the algorithmic analysis of 3D image recognition by considering 59 quantitative parameters of the obtained image. The designed sensor allows the individual quantification, counting, and classification of outstanding targets, such as bacteria or abiotics, in single and mixture suspension solutions. This study is presented for the rapid detection of bacteria that may contaminate a type of water sources used for food productions. Yu et al. [389] reported an accurate strategy for the total quantification of E. coli ER2738 in contaminated water through flow cytometry and PicoGreen nucleic acid fluorescence staining (Scheme 17C). Detection is based on the scattering and fluorescence signals of the targeted bacterial cells. On the basis of this background, the presence of a bacterial cell can be confirmed as a function of equivalent detection of signals.

**Scheme 17**

Phillips et al. [390] developed a sensing assay by using hydrophobically functionalized AuNPs for discriminating three E. coli strains. The mechanism of the sensing protocol involves the replacement of negatively charged conjugated polymers with pathogenic cells in the sensing protocol; as a result, polymer fluorescence changes. In addition, ultralow concentrations of volatile organic compounds can be detected easily by the human’s nose. Duncan et al. [391] reported a selective bacterial cell sensor composed of hybrid and functionalized NPs. The platform is constructed on the basis of three components, namely, functionalized AuNPs, pro-fragrance molecules that offer a “turn-on” response and produce distinctive odiferous, and anti-pro-fragrance enzymes that create an olfactory output (Scheme 17D). The displacement of lipase in the lipase–NP complex enhances the detection performance as the signal response increases, achieving high sensitivity within 15 min. Three types of quaternized magnetic nanoparticle–urease are fabricated and designed for the sensitive detection of Gram-positive or Gram-negative
bacteria (Scheme 17E) [392]. Detection can be achieved through the displacement of urease to hydrolyzed urea. The designed sensor has a high sensitivity of $10^2$ CFU mL$^{-1}$ with an accuracy of 90.7% within 30 min. In addition, Wan et al. [393] designed a nanocomposite platform for the quantitative recognition of bacteria by using three quaternized magnetic nanoparticle–fluorescent polymers. The designed sensor displays a high sensitivity of $10^7$ CFU mL$^{-1}$ and an accuracy of 87.5% within 20 min.

On the other hand, reports have confirmed that distinguishing and evaluating food-related viruses, which pose a serious threat to the food industry, is a priority task for food safety. Therefore, food safety experts have provided appropriate techniques and measures to detect and control viruses at all stages of the food chain and their control in the food industry [394-400]. Viruses obligate intracellular parasites and sensitive host cells for reproduction and the infection of their host. Incidentally, the design of the gastric extracellular matrix is simple; it consists of the nucleic acid, either single-stranded (ss) or double-stranded (ds) DNA or RNA, surrounded by a protein envelope. Viruses are classified as enveloped or non-enveloped types based on the presence or absence of an envelope of lipid bilayer derived from host cell membranes and viral proteins. In general, many different viruses, such as rotaviruses, astroviruses, saboviruses, enteroviruses, adenoviruses, and hepatitis E, are associated with the human digestive system. Subsequently, viruses cause a variety of diseases, such as gastrointestinal (stomach and intestinal) inflammation or hepatitis, which are transmitted through food or drink. Furthermore, the transmission of these gastrointestinal and liver diseases through food is associated with human norovirus (HNoVs) and HAV, respectively.

In 2012, approximately 21 million cases of acute gastroenteritis were detected in the United States, and 800 deaths were reported; moreover, the economic cost of infection with HNoVs is $5.8 billion annually in the United States [394]. Viral HNoV particles can survive for a long time
in various environments, including food products, and they are transmitted from the host via the fecal oral route, contaminated surface contact, and water resources [395, 396]. Electron microscopy, enzyme-linked immunosorbent assay (ELISA), immunochromatography, and biosensors have since been developed to detect the HNoVs in their resources [396]. Moreover, the development of fast, sensitive, and portable sensors for HNoV detection is in great demand [395]. The reverse transcription-polymerase chain reaction (RT-PCR) provides a good analytical technique for quantifying HNoVs. Despite the importance of the RT-PCR strategy, it has many drawbacks, including high cost, high time consumption, complicated sample preparation, and the need for experienced technicians. Well-designed biosensors, such as optical biosensors (i.e., Raman, IR, fluorescence, chemiluminescence, SERS, and SPR), can help to overcome most of the existing disadvantages related to sensitivity, selectivity, temporal response, and portability [397, 398]. Guo et al. [399] successfully designed an immunosensor by immobilizing the monoclonal antibody of HNoVs on electrode surface and performed photoelectrochemical assays for HNoV detection with LOD of $2 \times 10^{-10} \text{ g mL}^{-1}$ (4.9 pM) with a fast response time of 30 minutes. Another study [400] modified the microfluidic chip with monoclonal anti-HNoV. In the design, HNoV fluorescence intensity tracking was detected using a smartphone image after the background noise was filtered. A rapid, fast-response, low-cost, and easy-to-use technique was developed by using colorimetric HNoVs sandwich immunosensor and cotton swabs. The cotton swabs were functionalized by anti-HNoVs and then immersed on labeled anti-HNoVs with Au NPs for detecting HNoVs in a variety of food samples, including cucumber, lettuce, and chicken, with LOD of 10 and 53 pfu/mL [401].

A Au/Ag core/shell NP-based colorimetric biosensor was designed to detect HNoVs based on the improving of peroxidase-like performance of Au/Ag NPs. The HNoVs were captured by the antibodies immobilized on the 96-well microtiter plate surface; moreover, the oxidation reaction
between Au/Ag core/shell NPs and H₂O₂/3,3′,5,5′-tetramethylbenzidine (TMB) presented an intense blue color with LOD of 10.8 pg/mL, which could be attributed to the liberation of Ag from the Au/Ag NP surface [402].

Aimed at the selective DNA detection of norovirus and influenza A, Lee et al. [403] designed an electrochemical sensor based on the Pt-interdigitated electrode modified by CNTs decorated by Au/ magnetic iron oxide nanoparticles (Au-MNP-CNT). The designed sensor was highly sensitive, with the selective assay presenting a wide DNA linear range of 1 pM to 10 nM and LOD of 8.8 pM for both viruses. Label-free electrochemical sensors have been designed for the fast-response, sensitive, selective, and onsite detection of HNoVs. Peptides of HNoVs were used for the selective binding of HNoV particles. The peptides were immobilized at the electrode surface of Au to design a sensitive selective HNoV electrochemical sensor. The LOD was 1.7 copies/mL and could be used in care bioassay to detect HNoVs in food samples [404]. Hong et al. [405] designed electrochemical biosensors based on the Au electrode modified by concanavalin A (ConA) for the sensitive and selective detection of HNoVs in food samples. The sensor offered a wide linear range of operation at 10^2–10^6 copies/mL and low LOD of 35. Hirano et al. [406] selected certain immobilization parameters, including immobilization time, concentration of aptamer, concentration of supporting electrolyte containing 4 mM K₃[Fe(CN)₆], and investigated their effects on an electrochemical aptasensor designed for the selective detection of HNoVs by using the SWV technique. Giamberardino et al. [407] performed an onsite monitoring of HNoVs with LOD of 180 virus particles and developed an HNoV aptasensor after immobilizing ssDNA at the Au electrode via the SELEX process. To date, the effective monitoring protocols of foodborne pathogens and the novel progress of materials that can enhance sensing analysis are still required to improve the risk assessment of bacteria in food contaminations.
10. Food adulteration screening

Food adulteration, which is mainly related to the failure of a food product to meet legal standards, is the result of the change in nature and quality of food with the addition or substitution of a food ingredient with another unspecified ingredient; this scenario poses a serious threat to healthcare, especially in developing countries [408]. An example of food adulteration is the contamination of Chinese infant’s milk with the addition of melamine in 2008, and global attention has since been drawn to the continuous analysis of all basic food products, such as milk, dairy products, and meat [409-414]. Milk and dairy products, which can easily adulterate, are important for adults and infants as they include key complementary ingredients, such as fats, proteins, carbohydrates, vitamins, and minerals. Therefore, different techniques have been sought to verify food adulteration according to the nature of the foreign components or sources of food. Milk-like products are widely replaced by various plant protein sources, such as soya, rice, and almond proteins, and sold as natural milk products [411]. However, some proteins (e.g., such as soya, wheat, almond pea, rice, lupin, and maize) are labeled or/and clinically recognized as allergens by the Food Allergen Labelling and Consumer Protection (FALCPA) [412, 413]. In addition, the production cost of plant proteins, such as soya milk, is cheaper by 70% than natural milk protein, suggesting the prevalence of milk adulteration [414].

Consequently, many approaches and techniques, such as RT-PCR, high-performance liquid chromatography (HPLC), near-infrared (NIR), ELIZA, spectroscopy, and electrochemical methods, have been designed to investigate food adulteration [410]. ELISA can detect various proteins other than milk protein by labeling the monoclonal/polyclonal antibodies in these products [415]. The purity and originality of pasteurized milk powder protein was studied to determine the foreign mix of proteins (e.g., soya, pea, and wheat) by using the polyclonal
antibodies of these proteins [415]. This method can help to match the polyclonal data with the theoretical value by using relative standard deviations (RSDs) in the range of 5% to 22%. Moreover, skimmed milk adulteration by other proteins was evaluated via the UHPLC technique with high efficiency [416]. In addition, the turbidity measurements of selective tetraborate-EDTA buffer approach was used to detect insoluble plant proteins mixed with milk proteins by means of their separation and identification via mass spectroscopy [417].

Milk source and animal species difference quantification is challenging because of their genetic and nongenetic dependency. Milk source originality (genetic variant) was detected using the capillary electrophoretic (CE) technique [418, 419]. Bovine, ovine, and caprine milk proteins were used as the basis for identifying their respective genetic variant sources [418]. Currently, the ELISA and PCR techniques are widely used to detect the genetic adulteration of other milk sources (e.g., goat, sheep, and buffalo milk) in addition to cow milk [420]. The PCR test was successfully used to detect cow milk adulteration with respect to caprine milk. Cow milk in goat cheese, ewe milk cheese, and buffalo mozzarella cheese was also detected using the PCR test [421-424]. Moreover, the geographical source of milk can be identified by analyzing the isotopes and metal contents of the milk via inductively coupled plasma emission spectroscopy (ICP-OES), HPLC, and other elemental analytic techniques [425]. Elemental analysis provides information about milk geographical originality and contamination by hazardous inorganic materials [426, 427]. Milk additive ingredients, such as melamine, urea, and other products, were detected using various techniques, such as spectroscopy NIR, GC-MS, HPLC, and SERS [428-433]. All of the abovementioned techniques can provide a sensitive and selective assay for detecting melamine and urea in the milk ingredients. In summary, many approaches have been developed to ensure milk quality assurance.
Meat adulteration, a major consideration in distinguishing meat sources, is a relevant topic because of cost and religious reasons. Meat adulteration attributable to other sources, such as mixing beef with donkey, pig, and horse meat, and labeling these meats as beef have been discovered globally. In 2013, horse meat was labeled as beef meat in European markets [434]. Various methods, such as ELISA, spectroscopy, and PCR, have since been used for food quality and content authentication [435]. Ha et al. [436] developed a PCR method to detect pork adulteration in meat products. The 294-bp mitochondrial DNA of pork was used for pig target-DNA sequencing to identify pig adulteration in various kinds of meats (i.e., sheep, cattle, and chicken meat). The developed technique could detect 1% pork in beef, chicken, and beef–chicken mixtures. Successive developments of the PCR test, including the multiplexed PCR, to detect different specific primers in a single run from various sources have since been reported [437]. Five meat specific-species primers (mitochondrial ND5, ATPase 6, and cytochrome b genes) were detected using the multiplexed PCR. The genes from pig, cat, rat, dog, and monkey meats were amplified as 141, 172, 108, 163, and 129 bp DNA fragments, respectively. The method provided a highly sensitive assay for DNA detection in the range of 0.01–0.02 ng with a low suspected meat rate of 1%. The method, which was intended for commercial use, helped to observe Halal food quality of Muslim food. The multiplexed PCR was also used to identify various meat species (i.e., buffalo, chicken, ostrich, lamb, horse, rat, and fox species) with high efficiency, low time consumption, and high selectivity [438-441]. In general, PCR tests can selectively detect meat adulteration, but they suffer from various drawbacks, such as sample contamination, high time consumption, expensive technique, and low quantity [442]. Various techniques (i.e., ELISA, HPLC, lateral flow immunoassay, electrochemical immunoassay, etc.) are used to detect proteins and identify meat adulteration. The different types of ELISA (direct, sandwich, and indirect competitive techniques) are widely used to detect food
proteins and distinguish the food sources [443-445]. Sensitive and selective ELISA assays have been developed to identify pork adulteration in meat products. The IgG was immobilized in a microplate and competed with the extracted IgG to decrease the testing time; the assay showed a fast response time of 45 minutes with low susceptibility (0.1%) of pork content [445]. The sandwich ELISA assay was developed to detect pork adulteration in cooked meats of lamb, beef, horse, chicken, and goat; this assay provided a highly sensitive technique of 0.00014% w/v and 0.0004% w/v for cooked and autoclaved pork, respectively [444]. Electrochemical sensors have various advantages, including rapid, sensitive, simple, and portable assays [446-448]. Label-free electrochemical immunosensors have been developed for the selective detection of porcine serum albumin (PSA). A study [449] functionalized the carbon nanofiber-modified SPE by 4-carboxyphenyl, and the PSA antibodies were immobilized at the modified electrode. The specific binding between the amino group of the antibodies and the carboxylic group at the 4-carboxyphenyl was determined. The LOD was 0.5 pg/mL, with a wide linear range of 0.5–500 pg/mL of PSA. Mansouri et al. [450] designed an electrochemical biosensor for meat adulteration detection and subsequent cooked sausage evaluation. The electrochemical biosensor was designed on the basis of a specific-species DNA probe. The electrochemical DNA probe could selectively detect donkey meat adulteration in sausage food. This technique provided a sensitive and selective assay with a fast response time, low LOQ of 148 pM, and RSD of 0.16%. Many other approaches of food quality and adulteration detection have since been developed, including the design of a portable chipset biosensor for onsite monitoring. These portable biosensors can detect multiple targets in a single run and obtain a fast response, hence their high economical value.

11. Portable sensor devices for daily food safety analyses
In principle, food assessment based on portable devices can be performed by self- and in-home or in-market and other places where a food sample is served. To date, many portable devices for food control are available, but the design of commercially available portable sensors should be further improved. Wireless electronic devices and portable sensor technologies for food and beverage products should be enhanced to improve the reliability of screen displays for visual monitoring and online acquisition, to address significant food safety and health management concerns.

Mixing alcoholic beverages with foods during eating is a passive way to suppress the direct adsorption of alcohol to blood stream. Alcohol is frequently used as a food ingredient. In addition, a large amount of alcohol intake is a health risk that leads to various diseases and about 2.5 million deaths annually worldwide. For example, alcoholism may cause significant health risks that may lead to deaths from possible traffic accidents. Therefore, an alcohol level detector is essential for human life and safety. Reliable microdetectors that can continuously detect the consumption and misuse of alcohol in liquids have attracted considerable attention [451, 452].

Blood alcohol concentration is an accurate measure that can be used for alcohol detection in the blood, but it is difficult to use because pain associated with blood sampling restrains alcohol level monitoring in the blood. Therefore, alcohol concentration is usually used to detect alcohol, but it needs a special case and requires frequent analysis because it can be affected by environmental changes. In addition, alcohol detection has been investigated through transdermal-based sensors in humans [453, 454]. Electrochemical-based alcohol sensors are used as a good non-invasive analysis tool because of the association between the interstitial fluid (ISF) and alcohol concentration in the blood to detect alcohol in sweat and estimate the corneal layer through vacuum pressure [455].

Scheme 18
In this context, a microneedle-based sensor has been reported as an attractive skin sensor for the continuous evaluation of alcohol in ISF based on electrochemical analysis (Scheme 18) [456-459]. This sensor is constructed through alcohol oxidase (AOx) immobilization on the integrated structure of a pyramidal microneedle design containing microcavity opening wires of Pt and Ag that have been modified with \( o \)-phenylene diamine through electropolymerization. Immobilization has been implemented in a medium chitosan layer with Nafion as an outer layer. Using AOx in a functionalizing working electrode enhances selectivity, stability, and continuous alcohol recognition from artificial ISF. Experimental results in an ex vivo skin mouse model show that the designed sensor is effective in subcutaneous alcohol detection. In addition, this design has potential for application in real-time noninvasive alcohol detection. A wearable sensor based on gold and zinc oxide as sensor electrodes can detect alcohol consumption by determining the amount of ethanol metabolite ethyl glucuronide (EtG) and by designing a LED-based device to detect EtG in human sweat (Scheme 19). This strategy is built on the basis of electrochemical EtG detection through the immobilization of monoclonal antibodies to EtG on the electrode in the ROD of 0.001–100 μg/L and a LOD of 0.001 μg/L [460].

**Scheme 19**

The real-time detection of ethanol levels in the blood is developed through a prototype device as a noninvasive strategy for ethanol detection in the blood through the transdermal alcohol concentration strategy (Scheme 20A) [455]. Ethanol concentration is detected through the amperometry measurement of ethanol in sweat. The sensitivity of this device is higher than that of breathalyzers, and the device can work in a wide ROD of 0.0005–0.6 g/L. Droplet microdroplet screening is utilized to determine ethanol levels in cell cultures (Scheme 20 B) [461]. The prototype is based on an electrochemical alcohol oxidase/HRP sensor, and it provides a useful device for detecting the alcohol consumption of drivers. Kim et al. [462] used a wearable
tattoo-based sensor as a noninvasive strategy to detect alcohol in sweat (Scheme 20C). A flexible wireless electronic device based on an integrated biosensor iontophoretic tattoo can transgenically deliver a pilocarpine drug that prompts perspiration to detect ethanol through amperometry by utilizing Prussian Blue and Aox. The proposed design as a wireless skin strategy shows potential for the real-time monitoring of alcohol consumption and can be easily expanded for further detection analysis.

**Scheme 20**

With the increasing attention of the self-monitoring of food quality and safety, portable sensor devices contribute effectively in visual reading out and on-time and in-place analyses of bacterial contamination levels in food products with minimal restrictions. Portable electronic devices for the indoor and outdoor screening of pathogenic targets in food are important because of their distinct characteristics, including cost effective, providing fast and quantitative results on site. Based on visual detection through the fluorescence imaging of chemicals and biomolecules, an integrated system of a UV LED and a smartphone camera is used as a detector. In this context, the fluorescence imaging platform of a smartphone camera is utilized to detect *E. coli* concentrations at which excitation antibodies immobilized on quantum dots through UV LEDs are applied to detect bacterial concentrations based on fluorescence signals [463]. The results reveal a LOD of around 5–10 CFU mL\(^{-1}\). In addition, Ludwig et al. [464] monitored milk antibodies, such as antirecombinant bovine somatotropin, by using an integrated system between a white LED and a cellphone-based diagnostic platform (Scheme 21A).

Peanut allergen in food is tested and detected through colorimetric assays based on a cellphone-based system (Scheme 21B) [465]. This strategy is based on the development of a two-tube mobile phone facility for test and control detection processes. Light intensity is detected using a smartphone application developed to measure the concentration of allergens relative to reference
concentrations. Color tests are conducted using an ELISA test kit for peanuts. This method provides a digital tool to detect and measure peanut levels in food samples with a LOD of approximately 1 ppm. Clenbuterol (CLB) can be used to increase the growth rate in livestock, but this procedure is illegal. Therefore, a CLB detection system is developed on the basis of a mobile electromechanical device with a LOD of 0.076 ng mL$^{-1}$ in 6 min [55]. CLB is prohibited as a feed additive because its aggregation in animal tissues has several health risks. Giordano et al. [466] reported an integrated data processing on a smartphone system for the successful reorganization of Brazilian honey (Scheme 21C). The designed sensor substrate is equipped with a USB connector and an integrated Bluetooth module in potentiostat devices to share data via various net applications.

**Scheme 21**

Portable sensor devices for the detection of pesticides have posed a great concern because of health risks associated with these compounds. For instance, OP nerve agents, such as VX and sarin, are highly toxic substances that affect the nervous system. Therefore, the presence of OP pesticides should be controlled to prevent illnesses caused by exposure to these compounds. In this context, a wearable glove sensor is developed for the electrochemical sensing of collected residues when the thumb touches the printed enzyme-based OP sensor (Scheme 22A). Xu et al. [467] monitored OP pesticides by using a wearable glove-based sensor of an integrated “lab-on-a-glove” system containing two fluorescent centers with an aerogel flexible host material (Scheme 22 B). The designed sensor provides a fast real-time response (30 s) with a LOD of 89 Nm and allows detection with the naked eye. Chen et al. [351] proposed AuNPs as an easy sampling of complex surfaces of a proof-of-concept SERS platform for the direct and fast sensing and extraction of 4-mercaptopyridine and pesticides (Scheme 22C). In this report, numerous pesticide residues, including chlorpyrifos, thiram, and parathion–methyl, are qualitatively
measured in vegetable, cucumber, orange, and apple. Wang et al. [468] reported a gecko-inspired nano tentacle SERS as a fast-sampling technique that can be used for pesticide detection based on the “press and peeled-off” strategy (Scheme 22D). A flexible and free-standing platform is constructed on the basis of 3D poly(dimethylsiloxane) nano tentacles and AgNPs for the recognition of three types of pesticides. In addition, Au nano islands on dragonfly wings are used as a SERS platform through a DC magnetron sputtering system for detecting pesticide residues from tomato peels with a LOD of $10^{-3}$ ng cm$^{-2}$ (Scheme 22E) [387].

**Scheme 22**

**Scheme 23**

The increased accumulation of UA and low secretion in human bodies are closely related to the onset of gout and excessive UA. They are strongly influenced by the intake of large quantities of food rich in purine. UA is found in higher plants and microorganisms (Scheme 23A). A real-time sensor for UA detection is designed on the basis of the integration system of a low-energy Bluetooth communication system-on-chip and a novel salivary metabolite mouthguard biosensor (Scheme 23B) [469]. Kassal et al. [470] designed a wireless smart bandage biosensor for UA detection (Schemes 23C and 23D) and constructed it based on the immobilization of urease with Prussian blue as a printed catalytic transducer to enable the detection of UA at low concentrations. The sensor bandage is integrated with a wearable potentiostat that provides reading out of the UA status on a smartphone, a tablet, or a computer.

New technology-based food analysis techniques, device-integrated smartphones, and on-time data acquisition tools are needed for the precise and rapid assessment of food quality. The development of portable food assessment devices represents a promising area with a significant scientific and commercial impact. The emerging effects of active nanomaterials in a portable sensor provide promising prospects for direct sensing, on-demand scanning, and self-analysis systems for home
testing. So far, wireless smart devices should respond to direct food assessment for applicable, accurate, and precise point-of-care analysis. Therefore, the incorporation of smart nanomaterials into sensor array devices may provide sensor designs with precise and customized properties. When integrated with smartphones, these portable sensor devices may be used for on-site testing. With effective advances in nano- and micro-metric devices, real assessment protocols should be developed for the enhancement of the quality of food and agricultural products with high sensitivity and accuracy.

12. Commercial/industrial market deployments of portable sensor devices for food assessment

To ensure the quality of food, the design of materials-based sensor devices (Table 5) in terms of readability (easy-to-read) and practicability needs special standards. In this regard, high-sensitivity wearable and implantable biosensors are needed to help improve real-time monitoring and food-sensitive detection. To date, the primary drawbacks of the traditional traceability approach in food quality assessment are as follows: (i) it requires multi-step inspections; (ii) it involves a time-consuming process; (iii) the method is labor-intensive, and well-trained personnel are needed; (iv) the method is unsuitable for onsite product analysis; (v) the results have low sensitivity; and (vi) risk of microbial contamination is common. Although non-invasive nanoscale/microscale designs with high efficiency and reliability and cyclic analyses are areas of interest in the field of disposable sensing platforms, the fully printable and wearable biosensing system is still in the research and development stage. Portable biosensors offer promising prospects for direct biosensing systems, on-demand screening, and home analysis for self-testing. However, a number of disadvantages need to be resolved, including the following: (i) high cost of sensor manufacturing devices, (ii) losing of its accuracy, sensitivity and selectivity in the presence of the interference species, (iii) short home and personal self-testing and on-time analysis, and (iv) special precautions for sterilization. Therefore, the recent developments in food sensor designs are required to improve the provision of (i) personal
monitoring use with compact device size, (ii) direct inspection, and (iii) accurate on-target screening as a means of preventing adulteration and improving food quality.

Personalized monitoring and checking of the quality of agricultural products, foodstuffs, and food beverage industry and their processing have received much attention to develop approaches for personal consumption and market determination. Therefore, the development and provision of analysis methods and electronic devices for testing the quality of food products are particularly interesting. Self-tester and portable readout sensor devices are on-demand, inexpensive, and convenient to attract consumers, researchers, and food manufacturers. To date, effective monitoring systems remain vital concerns to conduct regular checks for hazards and food quality, ensure food ingredient safety, and maintain public health. According to the increase in the global market growth rate and the requirements of the food control market, a number of companies introduced various devices and analytical methods that contribute effectively to food quality control (Table 5). Challenges remain for the commercial development of electronic sensor devices with miniature reading out, digital easy-to-read without supplying power, and wireless signal transmission into smartphones. Furthermore, the scalable manufacturing of electronic food sensor devices with actual and cheap cost in commercial/industrial market deployments has substantial issues.

Table 5

Conclusion and future outlook

With the need for today’s food safety and management, assessment should be centered on the whole food cycle to determine and confirm food and microbiological quality. Providing safe food and water is an essential prerequisite for human wellbeing. A wide range of pesticides, microbial bacteria, hazardous contaminants, and essential food additives may exist in drinking and food chains. Thus, overall quality assessment is a key milestone of human health from the point of view of care. Given the prevalence of food safety and its impact on health, economic,
and social sectors, advancements in an ultrasensing, early detection, and real- and on-time monitoring strategy of vital reactive species, biomolecules, chemicals, and even hazardous agents for the food quality sector are urgently needed.

This review provides real evidence on the key impact of the recent building block design of the engineering of nanodynamic early sensory detectors in the improvement of standard food quality through the full control of (i) an extraordinary food safety test in terms of developed sensing protocols and tools; (ii) numerous and extensive scale auto-examining evaluations of reactive food species, including \( \text{H}_2\text{O}_2 \), UA, AA, sulfites, and nitrite anions; (iii) extensive monitoring and assessment of targeted hazardous species, including bacteria and pesticides; and (iv) conceivable, portable, and compact nanosensors for real-time analysis via a small sample size.

With significant advances in the diverse and competitive food and beverage market, consumer concerns about food safety, ingredients, contaminations, and processed foods have emerged. Food contamination has caused prevalent diseases and impacts on social, financial, and health sectors. Thus, sensory gadgets for accurate quantity screening, early explicit monitoring, assessment, and real-time detection should be developed. This review highlights new development monitoring technologies for a comprehensive treatment strategy, outstanding screen monitoring pattern, and restorative conditions for potential food safety. For the proximity control of food safety and management, special attention and efforts are still needed to provide appropriate sensing protocols and strategies that can be configured in sensitive and smart gadgets linked to basic, quick, convenient, and continuous target recognition devices.

In this context, we focus on engineering a powerful nanoscale sensor design and protocol that investigates the explicit restricting partiality to a wide scope of the food safety, periodical transduction into readable output signals, high-precision assessment of food contamination, dead-end-use examination, and fast response analysis. AA, \( \text{H}_2\text{O}_2 \), UA, nitrite, and sulfite anions, pathogenic
bacteria and viruses (i.e., *E. coli*, H1N1, HuNoN, and SARS-CoV-2), food adulteration, and pesticides have been covered as targeted species for food safety assessment. Thus, we provide details on advanced sensor development in monitoring, analysis, and evaluation sectors for food safety applications. Moreover, this review highlights the key principle for the future steady progress required in food safety and quality, biotechnological, analytical, and sensing applications.

Reducing the spread of COVID-19 is the first priority worldwide. Although more than a year has passed since the COVID-19 pandemic began, information and explanations about the nature of the infection remain scarce and unclear. Currently available information indicates that COVID-19 does not directly infect humans through food, but the hypothesis indicates the emergence of the virus in bats and its transmission to this organism used in food. With the emergence of new evidence, measures should be taken to follow up on even frozen foodstuffs and their preparation stages. In addition, as of writing this review, the design of any sensitive device for detecting COVID-19 in foods has not been disclosed, but efforts should be devoted to providing a sensor to assess food with high selectivity and sensitivity and prevent the spread of COVID-19 through the food production process and its manufacturing stages.

Nanodynamic portable sensors can be used to monitor contamination and assess environmental food with short-time consumption and simple operation. The precision control of present propels in the tangible transformation of nanomanufactured sensor innovation into a food test home-use facility can be assessed via the following capacities:

(i) Advanced detecting appraisal and precise assessment of natural examination parameters (e.g., sensitivity, real-time monitoring, calibration line, stability, quality, and reproducibility)

(ii) Timely and real-time investigation protocol, early checking and precaution treatment, and board analysis tests that can be connected to shorten treatment duration (e.g., quick reaction, early visualization and finding, brief span treatment, and precise detecting examination)
(iii) Simple design of a compact nanobiosensor in daily use (e.g., chipset sensor, well-planned reconciliation, wearable, adaptable, and brilliant material-based gadget stages) with proficient examination precision, security, and viability of self-customized observation of targeted levels in the food chain

(iv) Well-controlled, scalable configuration, and low-cost manufacturability of sensory gadgets

This review gives proof-of-evidence that the engineering and design of a powerful nanosensor are ongoing acquisitions, offering considerable future avenues for wide-range opportunities to control healthcare initiatives. With the headway of human advancement and progress in nanoscience and nano-engineering, the manufacture of effective and operative detecting devices for the early recognition and inhibition of food contaminations will be vital in food analysis and environmental monitoring. Up to date, one of the urgent nanodevelopment of the future is the building blocks of nanoscale sensor devices in sensitive portable, touchscreen, and wearable end-user products to enable the direct assessment of food quality. Continuous acquisitions and significant future orientations provide broad opportunities for food assessment and control. Next-generation nanoscale wearable sensors are urgently needed for rapid, simple, and on-time biosignature monitoring assays to ensure food safety. However, the controlled construction of the sensing, surface functionality, and on-site monitoring of target molecules are a challenge for the driven noninvasive sensor promotion of organic and nonorganic agents, targeted biomolecules, bacteria, pesticides, and pollutants that are naturally associated with food safety. This review gives evidence that the powerful engineering of nanosensor in handy IoT devices is an ongoing acquisition, offering considerable future avenues for wide-range contributions to perspective healthcare initiatives in Society 5.0.
List of Schemes:

Scheme 1: (A) Essential applications of nanotechnology in the food sector, and the key elements for food assessments. (B) Schematic representation of sensor workability and design in terms of fabrication and sensing assay through various transducers that converted this reaction or binding into digital signal.

Scheme 2: Nanomaterials-based sensor platforms with a wide range of domains of hierarchical control of the virtual and anisotropic orientations including the dendritic nanoarchitecture nanostrands inorganic metal oxide, nanoscale sheet orientation building blocks, metal organic frameworks (MOFs) materials, nanocomposite and hybrid nanocomposite materials, and uniform, multi-directional mesoporous accessibility of the nanoarchitecture was doped microporous carbon microspheres. The sensory design, clear orientation and formation such as cavities, geodes, tubes, spherical bodies, cubes, and multi-surface structures profoundly affect the sensitivity and selectivity of target species among competitive species.

Scheme 3. (A) Colorimetric sensor design based on AgNPs for quantitative AA detection in commercial and pharmaceutical samples, reproduced with permission [99]. (B) A “turn-on” Eu (III) luminescent complex sensory probe for AA visual imaging in the living cell using time-gated luminescence microscopy (TGLM) system, reproduced with permission [100]. Fabrication of a fluorescent switchable N, S-CDs sensor for AA detection in fruits based on the mechanism of N-,S-CDs/Fe$^{3+}$ system (C,D), reproduced with permission [110].

Scheme 4. Electrochemical electrode design with a sulfur-doped microporous carbon microsphere (S-MCMS) for AA detection in its real samples. (A-C) Analysis of the morphological surface structure of using FE-SEM images, (D) the electrooxidation design of AA on the S-MCMS electrode
surface, (E, F) representing the real amperometric detection of AA in Lemon juice and pharmaceutical products (Vitamin C tablets), Reproduced with permission [11].

Scheme 5. (A) Synthesis of NPC/Co₃O₄ nanocomposites through direct thermal treatment of ZIF-67 precursor as a sensitive electrode material for voltammetric detection of AA in vitamin C tablets, Reproduced with permission [121]. (B) Electrochemical detection of AA and DA with high sensitivity using a fabricated microelectrodes of carbon fiber/ZnO core–shell hybrids material, Reproduced with permission [122]. (C) Modification of pencil graphite electrode (PGE) by raphene-iron oxide-polyvinyl alcohol (PIG) for AA detection (C) [132].

Scheme 6: (A) Design of fiber optic sensor through the modification of plastic clad silica fiber core (FC, 0.6 nm in diameter) with Ag layer and nanocomposite of AgNPs-PVA. (B) Fixing of the designed fiber optic probe, FC/Ag film/Ag NPs-PVA nanocomposite, into a flow cell for H₂O₂ detection, Reproduced with permission [140]. (C, D) Schematic design for the detection of H₂O₂ using a fiber-optic probe based on the PB/PW response during the oxidation and reduction of sample, Reproduced with permission [142]. (D) The designed illustrated the detection mechanism of H₂O₂ using a fiber-optic PB/PW probe, the generated white light traveling through arm1 arriving at the sensor film of PB/PW, then the collected light directed back through the arm2 to measure the intensity of the reflected spectrum to confirm the PB/PW response during the oxidation and reduction process.

Scheme 7: (A) Selective detection of H₂O₂ species based on the turn-on fluorescence aggregation/deaggregation behavior of tetraphenylethylene based on the aggregation induced-emission during the reaction with H₂O₂ species, Reproduced with permission [145]. (B) Two-photon fluorescent probes mechanism for detect H₂O₂ detection in living cells and its successful use in imaging, Reproduced with permission [154]. (C) A turn-on fluorescence for selective H₂O₂ in living cells based on oxidizing the mitochondria-targetable ratiometric fluorescent probe, HBTPB (as a NIR-
emitting fluorescent probe), Reproduced with permission [155]. (D) The design of mitochondria-targetable modified with a HKB ratiometric fluorescent chelating agent for detecting H$_2$O$_2$, with the fluorescence analysis using 0–500 μM of H$_2$O$_2$, Reproduced with permission [156]. Schematic designed of the proposed fluorescence sensing process of H$_2$O$_2$ molecules via the mitochondria-targeted modification using coumarin moiety (E), Reproduced with permission [157]. Design of the real in vivo H$_2$O$_2$ detection using young angelfish (age 20 days) as a real example of living organisms (F), Reproduced with permission [159].

**Scheme 8** (A) Schematic design of the enzymatic sensor design by controlling the immobilization of targeted enzymes into the electrode surface platform such as nickel foam, and (B) the full steps of Heme proteins modified Ni foam electrode for enzymatic electrochemical detection of H$_2$O$_2$, Reproduced with permission [172-174].

**Scheme 9.** Schematic sensor design through hem-proteins immobilized of on the NiO nanostructures surface. The proposed mechanism of non-enzymatic H$_2$O$_2$ sensor based on the modification of Ni foam electrode through the immobilization of Cyt.C, Reproduced with permission [175].

**Scheme 10.** Design of a non-enzymatic H$_2$O$_2$ selective electrochemical sensor by the electrodeposition of PtNPs into modified RGO/GCE electrode for real and in-vitro cell application (A), Reproduced with permission [31]. The electrochemical real monitoring of H$_2$O$_2$ released from live cells (A549) using a GCE modified by MoS$_2$ NS/N-doped carbon NWs nanocomposite (B), Reproduced with permission [186]. Engineering of an encapsulated Au@ZIF-67 electrode sensor for electrochemical detection H$_2$O$_2$ released from stimulated living organisms to screen cancer cells (C), Reproduced with permission [187] [163]. The fabrication and proposed mechanism of Ni(OH)$_2$/RGO/Cu$_2$O@Cu electrode for glucose and H$_2$O$_2$ detection, (D), Reproduced with permission [189].
Scheme 11. (A) Schematic diagram of the main steps of the an encapsulated CdS-QDs-Uricase/HRP fluorescence biosensor engineering for UA detection, in addition to the mechanism of detection based on the quenching properties of H₂O₂ produced from the biosensor system, Reproduced with permission [202]. The turn-off design of Co²⁺/H₂O₂–AUR fluorescence sensor for UA detection, and effect of different UA concentrations on the Co²⁺/H₂O₂–AUR sensor (B), Reproduced with permission [206].

Scheme 12 (A) Mechanistic signaling of fluorescent and colorimetric sensor based on CDs during the visual detection of nitrite anions, Reproduced with permission [220]. The glass fiber strip modified with the synthesized N-(1-naphthyl) ethylenediamine (1-Nap) preloaded PEG-hydrogel colorimetric detection of different concentrations of NaNO₂ (B), Reproduced with permission [222].

Scheme 13: (A) Schematic design showing the adsorption of different types of pesticides using the mesocage collector cavities (MCC) nanopocket materials with high efficiency (left side), and the complete process of carbofurane pesticides uptake into MCC nanopocket (right side), Reproduced with permission [341]. (B) Screen printing sensor design for detection of pesticide residues in vegetables and fruits by integrating cellulose paper with hybrid AgNPs and GO based on SERS-based approach, Reproduced with permission [354]. (C) A flexible glass-fiber paper patch of BPWHW/AgNWs for selective detection of paraquat pesticide in the apple, Reproduced with permission [355].

Scheme 14: (A) Design of active GQDs/AChE/CHOₓ platform for direct detection of OPs based on photoluminescence properties at 467 nm, Reproduced with permission [366]. (B) A Colorimetric detection of pesticide residue of terbutylazine (TBA) and dimethoate (DMT) in different real samples (apple juice, green tea and tap water) using AuNPs-based colorimetric sensor, Reproduced with permission [369].
**Scheme 15.** Schematic design of the bactericidal activity in the presence of hexagon MgO nanosheets (A), and the TEM images of *E. coli* growth in free Lysogeny broth (LB) environment (a) and after treatment with hexagon MgO nanosheets (b, c) (B), Reproduced with permission [370, 372, 376, 377]. The mechanistic reaction of hexagonal micro/macroporous MgO nanosheets and their potential with cellular components and the membrane of pathogenic bacteria (C), Reproduced with permission [370, 372, 376, 377].

**Scheme 16.** Design of a portable plasmonic label-free platform for pathogen detection using a modified microchannel chip SPR platform with layer of Au (50nm) along the disposable microchip surface, EDC (N-(3-Dimethylaminopropyl)–N’-ethylcarbodiimide Hydrochloride), MUA (11-Mercaptoundecenoic Acid), NHS (N-Hydroxysuccinimide) and with LPS (anti- Lipopolysaccharide) antibody for the for quantify detection of *E. coli*, Reproduced with permission [386].

**Scheme 17.** (A) A schematic diagram of the electrode construction for microbial electrochemical detection based on RGO-hyperbranched chitosan nanocomposite for the sensitive and real-time detection of bacterial cell viability, Reproduced with permission [370]. (B) Construction of a microbial culture device on a glass slide in a Petri dish with details of device integration, Reproduced with permission [371]. (C) The scattering and fluorescence of total bacteria detection in real tea beverages and drinking water using a HSFCM technique, Reproduced with permission [389]. (D) Design of bacterial sensor based on the AuNPs/anionic enzyme catalytic system of pro-fragrance molecules to produce “turn-on” response with odiferous fragrance, Reproduced with permission [391]. (E) Detection of the bacterial as a relation of pH changes due to the displacement of urease to hydrolyzed urea and increase the pH value using a litmus dye, Reproduced with permission [392].

**Scheme 18.** Schematic representation of different steps for the construction of integrated microneedle array for alcohol sensor based on the biocatalytic reaction (A, B). Photos of the microneedle array on the fingertip (C), and integrated design of Pt & Ag wires with the microneedle array (D). The design of alcohol sensing assay in artificial ISF through the mice skin by the microneedle (E), the images of the sensor, the mice skin and the penetrated sensor over the tested sample to detect alcohol (F), and the real-time alcohol detection through the mice skin, Reproduced with permission [451].

**Scheme 19.** Schematic representation of the integrated platform sensor for alcohol: general design of sweat evaluation test, and platform design (A), the immobilization strategy of the Dithiobis [Succinimidyl Propionate] (DSP) and EtG antibody into the electrode surface (B). Photos of the Au and ZnO sensors fixed on glass and polyimide substrates (C). Optical LED display for classification of measured EtG concentration and different alcohol consumption performance (D, E), Reproduced with permission [460].

**Scheme 20:** Wearable non-invasive sweat-based sensors for alcohol detection electrochemically using a bi-enzymatic monitoring cell (A), Reproduced with permission [455], Droplet microdroplet screening strategy, Reproduced with permission [461], the microfluidic chips for alcohol detection in sweat, and the noninvasive strategy for alcohol detection based on a wearable Tattoo-Based sensing system (C), Reproduced with permission [462].

**Scheme 21:** A schematic outline of the cellphone attachment for advanced fluorescence diagnosis (A), Reproduced with permission [464], iTube platform based on smartphone for colorimetric digital detection (B), Reproduced with permission [465], and Portable platform deployed for point-of-use analyses (C), Reproduced with permission [466].

**Scheme 22:** (A) Schematic overview of the printable, stretchable glove biosensor for direct and Bluetooth OP agent detection. (B) A non-invasive wearable glove-based sensor strategy for on-site
Op’s detection on the surfaces of agricultural crops including tomato, green pepper, pear and apple based on porous CMC aerogel and EuMOFs, Reproduced with permission [467]. (C) SERS-tape strategy for tracking the surface quality of fruit and agricultural crops, Reproduced with permission [351]. Schematic preparation of G-SERS substrate for SERS analysis (D), Reproduced with permission [468]. The DC magnetron sputtering method for Au sputtering on dragonfly wing as SERS platform (E), Reproduced with permission [387].

**Scheme 23:** A collage image showing some foods that contain uric acid (A). Design of an integrated amperometry Bluetooth wireless mouthguard sensor for real time detection of UA in saliva samples (B), Reproduced with permission [469]. Engineering of a wireless wearable bandage for UA detection according to: (i) Integrated smart bandage design for screen printing by fixing uricase to PB modified carbon electrodes which are placed on commercial bandage; (ii) a wireless wearable potentiostat determines connected with a computer or Smartphone for UA detection; and (iii) show the amperometry detection mechanism of UA detection using wireless wearable bandage. Image pf a wireless wearable smart bandage used in the UA detection (D). Reproduced with permission [470].

**References**


[37] H. Khalifa, S.A. El-Safty, A. Reda, M.A. Shenashen, A.I. Eid, Anisotropic alignments of hierarchical Li2SiO3/TiO2@ nano-C anode/LiMnPO4@ nano-C cathode architectures for full-cell lithium-ion battery, Nat. Sci. Rev. 7 (2020) 863-880.


[162] Y. Sheng, H. Yang, Y. Wang, L. Han, Y. Zhao, A. Fan, Silver nanoclusters-catalyzed luminol chemiluminescence for hydrogen peroxide and uric acid detection, Talanta, 166 (2017) 268-274.


Scheme 1
Scheme 3
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Table 5. A list of commercial mobile sensor devices used in the assessment of food quality.
Table 1: Various nanoscale sensory systems, and sensing protocols, and analytical techniques reported for sensitive detection of AA targets

<table>
<thead>
<tr>
<th>AA sensor protocols and designs based materials</th>
<th>Techniques</th>
<th>LOD (µM)</th>
<th>Linear Range (µM)</th>
<th>Ref.</th>
</tr>
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<tbody>
<tr>
<td>AgNPs</td>
<td>SERS</td>
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<td>AgNPs</td>
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<td>0.04</td>
<td>15.18 – 23.48</td>
<td>[93]</td>
</tr>
<tr>
<td>SiNPs/BSA/MnO</td>
<td>Fluorescence</td>
<td>0.102</td>
<td>1 – 400</td>
<td>[95]</td>
</tr>
<tr>
<td>Carbon nanocages</td>
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<td>0.0972</td>
<td>2 – 12</td>
<td>[96]</td>
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<tr>
<td>N, S-CDs</td>
<td></td>
<td>4.69</td>
<td>10 – 200</td>
<td>[110]</td>
</tr>
<tr>
<td>GQDs</td>
<td></td>
<td>0.094</td>
<td>0.3 – 10</td>
<td>[97]</td>
</tr>
<tr>
<td>polypyrrole/GQDs</td>
<td></td>
<td>0.005</td>
<td>0.02 – 5.66</td>
<td>[104]</td>
</tr>
<tr>
<td>CDs</td>
<td></td>
<td>0.136</td>
<td>0.136 – 0.23</td>
<td>[105]</td>
</tr>
<tr>
<td>GQDs</td>
<td></td>
<td>0.32</td>
<td>1.11 – 300</td>
<td>[106]</td>
</tr>
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<td>N-CDs</td>
<td></td>
<td>0.3</td>
<td>1 – 750</td>
<td>[107]</td>
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<tr>
<td>P-, N-, doped CQDs</td>
<td></td>
<td>1.35</td>
<td>5 – 200</td>
<td>[108]</td>
</tr>
<tr>
<td>N-, S-doped CDs</td>
<td></td>
<td>0.17</td>
<td>0.57 – 264</td>
<td>[109]</td>
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<td>polyallylamine-AuNCs/MnO2 nanosheets</td>
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<td>0.0032</td>
<td>0.01 – 200</td>
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<td>SiQDs</td>
<td>Chemiluminescence</td>
<td>0.57</td>
<td>2 – 200</td>
<td>[102]</td>
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<tr>
<td>o-phenylenediamine/CDs</td>
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<td>0.009</td>
<td>0.6 – 40</td>
<td>[103]</td>
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<td>AuNPs/Luminol</td>
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<td>20000</td>
<td>10000 – 1</td>
<td>[115]</td>
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<tr>
<td>Cu²⁺/NGQDs/H₂O₂</td>
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<td>0.5</td>
<td>1 – 100</td>
<td>[117]</td>
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<td>CdSe/CdS QDs</td>
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<td>0.0067</td>
<td>10 – 100</td>
<td>[118]</td>
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<tr>
<td>NiO/GR</td>
<td>Electrochemical</td>
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<td>0.05 – 1100</td>
<td>[120]</td>
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<td>NPC/CO₃O₄</td>
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<td>Carbon fibers/ZnO</td>
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<td>14.63</td>
<td>600 – 2000</td>
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<td>AgNPs</td>
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<td>0.01 – 10000</td>
<td>[125]</td>
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<td>PEDOT:PSS/OEXTS</td>
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<td>0.263</td>
<td>1 – 1000</td>
<td>[127]</td>
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<tr>
<td>MIPs (o-phenylenediamine and pyrrole)</td>
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<td>36.4</td>
<td>0.1 – 10000</td>
<td>[128]</td>
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<tr>
<td>poly(o-phenylenediamine-co-o-aminophenol) (PoPDoAP)</td>
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<td>1.19</td>
<td>25 – 725</td>
<td>[129]</td>
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<tr>
<td>TiO₂/rGO</td>
<td></td>
<td>1.4</td>
<td>0.05 – 0.75</td>
<td>[130]</td>
</tr>
<tr>
<td>pNR/rGO</td>
<td></td>
<td>0.234</td>
<td>40 – 4100</td>
<td>[132]</td>
</tr>
<tr>
<td>graphene-iron oxide-polyvinyl alcohol</td>
<td></td>
<td></td>
<td></td>
<td></td>
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Table 2. Various nanoscale sensory systems, and sensing protocols, and analytical techniques reported for sensitive detection of H$_2$O$_2$ molecules

<table>
<thead>
<tr>
<th>H$_2$O$_2$ sensor protocols and designs based materials</th>
<th>Techniques</th>
<th>LOD (µM)</th>
<th>Linear Range (µM)</th>
<th>Ref.</th>
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<tbody>
<tr>
<td>MP-11/Tb@mesoMOFs/CHIT-AuNPs/3D-KSCs</td>
<td>Electrochemical</td>
<td>0.996</td>
<td>3.02 – 640</td>
<td>[113]</td>
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<tr>
<td>Pt/Pb/G</td>
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<td>0.05</td>
<td>0.05 – 30</td>
<td>[172]</td>
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<tr>
<td>MP-11/SnO$_2$</td>
<td></td>
<td>0.996</td>
<td>3.02 – 640</td>
<td>[173]</td>
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<tr>
<td>α-Fe$_2$O$_3$/rGO</td>
<td></td>
<td>0.002</td>
<td>0.002 – 2500</td>
<td>[133]</td>
</tr>
<tr>
<td>Fe$_2$O$_3$/ITO</td>
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<td>1</td>
<td>5-4495</td>
<td>[181]</td>
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<tr>
<td>Co$_3$O$_4$-MCNFs</td>
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<td>0.3</td>
<td>1-360</td>
<td>[182]</td>
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<tr>
<td>Co$_3$O$_4$/SPCE</td>
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<td>0.5</td>
<td>1-2580</td>
<td>[179]</td>
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<tr>
<td>Co$_3$O$_4$/MWCNTs</td>
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<td>0.145</td>
<td>0.1-50</td>
<td>[178]</td>
</tr>
<tr>
<td>Co$_3$O$_4$ nanowalls/GCE</td>
<td></td>
<td>2.46</td>
<td>20-430</td>
<td>[177]</td>
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<tr>
<td>3D GO–Co$_3$O$_4$ PHs/SPCE</td>
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<td>10</td>
<td>0-5350</td>
<td>[168]</td>
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<tr>
<td>MP-11/PCN-333 (Al)/ 3D-KSC</td>
<td></td>
<td>0.7664</td>
<td>250–4750</td>
<td>[166]</td>
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<tr>
<td>3D GO–Co$_3$O$_4$</td>
<td></td>
<td>0.015</td>
<td>0.05–400</td>
<td>[167]</td>
</tr>
<tr>
<td>Cytochrome C/Ni-Foam</td>
<td></td>
<td>0.127</td>
<td>0.387 – 1725</td>
<td>[174]</td>
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<tr>
<td>Co$_9$S$_8$ hollow spheres</td>
<td></td>
<td>0.02</td>
<td>0.1 - 11110</td>
<td>[183]</td>
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<tr>
<td>Au@C-Co3O4</td>
<td></td>
<td>0.2</td>
<td>50 – 128000</td>
<td>[175]</td>
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<tr>
<td>Ni(OH)$_2$/RGO/Cu$_2$O</td>
<td></td>
<td>0.143</td>
<td>1 – 30000</td>
<td>[189]</td>
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<tr>
<td>MoS$_2$/CN NWs $^9$</td>
<td></td>
<td>0.35</td>
<td>0.5 – 7500</td>
<td>[186]</td>
</tr>
<tr>
<td>Ag NPs</td>
<td>Colorimetry</td>
<td>0.0294</td>
<td>0.01–10000</td>
<td>[140]</td>
</tr>
<tr>
<td>Ag-NPs/optical fiber</td>
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<td>10</td>
<td>10 – 10000</td>
<td>[139]</td>
</tr>
<tr>
<td>Au-Ag/C NC</td>
<td></td>
<td>0.00864</td>
<td>1 – 120</td>
<td>[147]</td>
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<tr>
<td>GO-PtNi-3,3′,5,5′-tetramethylbenzidine</td>
<td></td>
<td>0.3</td>
<td>0.8–90</td>
<td>[148]</td>
</tr>
<tr>
<td>Chitosan</td>
<td></td>
<td>5</td>
<td>8 – 1500</td>
<td>[150]</td>
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<tr>
<td>HRP/BSA/glutaraldehyde</td>
<td>Fluorescence</td>
<td>1.55</td>
<td>10 – 10000</td>
<td>[146]</td>
</tr>
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<td>Coumarin</td>
<td></td>
<td>0.5</td>
<td>0.5 – 100</td>
<td>[143]</td>
</tr>
<tr>
<td>Iodophenol blue-luminol</td>
<td>Chemiluminescence</td>
<td>0.01</td>
<td>0 – 80</td>
<td>[157]</td>
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<tr>
<td>Ag (I)-Luminol-CoFe$_2$O$_4$</td>
<td></td>
<td>0.014</td>
<td>0.025–10</td>
<td>[152]</td>
</tr>
<tr>
<td>Bovine serum albumin (BSA) caped silver nanoclusters (Ag NCs)</td>
<td></td>
<td>0.15</td>
<td>0.5 – 100</td>
<td>[161]</td>
</tr>
<tr>
<td>Luminol/H$_2$O$_2$/GNPs CL</td>
<td></td>
<td>0.14</td>
<td>0.14 – 100</td>
<td>[162]</td>
</tr>
<tr>
<td>GO–CdS nanocrystal nanocomposites</td>
<td></td>
<td>1.7 µM</td>
<td>5 µM–1 mM</td>
<td>[164]</td>
</tr>
<tr>
<td>Hexythiazox (HXTZ)</td>
<td></td>
<td>0.011 µg mL$^{-1}$</td>
<td>0.017-0.42 µg mL$^{-1}$</td>
<td>[165]</td>
</tr>
</tbody>
</table>

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Table 3. Various nanoscale sensory systems, and sensing protocols, and analytical techniques reported for sensitive and selective detection of food ingredients and additives of nitrite and sulfite species

<table>
<thead>
<tr>
<th>Sensor protocols and designs based materials</th>
<th>Techniques</th>
<th>LOD (µM)</th>
<th>Linear Range (µM)</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Detection of nitrite species</td>
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<td></td>
<td></td>
<td></td>
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<tr>
<td>AgNPs</td>
<td>Colorimetry</td>
<td>0.1</td>
<td>0.1 – 50</td>
<td>[219]</td>
</tr>
<tr>
<td>Carbon dots (C-dots) and neutral red</td>
<td></td>
<td>0.196</td>
<td>0.145 – 4.34</td>
<td>[220]</td>
</tr>
<tr>
<td>N-(1-naphthyl)ethylenediamine/PEG</td>
<td></td>
<td>10</td>
<td>10 – 5000</td>
<td>[222]</td>
</tr>
<tr>
<td>CoOx/CNT/GCE</td>
<td>Electrochemical</td>
<td>0.3</td>
<td>0.5 – 249</td>
<td>[231]</td>
</tr>
<tr>
<td>CoNS/GO/PPy/GCE</td>
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<td>0.015</td>
<td>1 – 3200</td>
<td>[232]</td>
</tr>
<tr>
<td>Pd/rGO/GCE</td>
<td></td>
<td>23</td>
<td>1 – 1000</td>
<td>[233]</td>
</tr>
<tr>
<td>Ag-rGO/GCE</td>
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<td>0.012</td>
<td>0.1 – 120</td>
<td>[234]</td>
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<td>Ag/HNT/MoS₂/CPE</td>
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<td>0.7</td>
<td>2 – 425</td>
<td>[235]</td>
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<td>rGO-C60/AuNPs</td>
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<td>0.013</td>
<td>0.05 – 1175.32</td>
<td>[236]</td>
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<tr>
<td>Au-HNTs-GO</td>
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<td>0.03</td>
<td>0.1 – 6330</td>
<td>[237]</td>
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<tr>
<td>a-Fe₂O₃ NAs/CF</td>
<td></td>
<td>0.12</td>
<td>0.5 – 1000</td>
<td>[238]</td>
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<tr>
<td>Detection of sulfite species</td>
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<td></td>
<td></td>
<td></td>
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<tr>
<td>TSP ¹⁰</td>
<td>Fluorescence</td>
<td>0.2</td>
<td>0.5 – 150</td>
<td>[248]</td>
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<tr>
<td>Naphthofluorescein</td>
<td></td>
<td>1.74</td>
<td>0 – 300</td>
<td>[252]</td>
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<td>Carbon-dots</td>
<td></td>
<td>0.35</td>
<td>10 – 50</td>
<td>[255]</td>
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<tr>
<td>CPE/Au-Si4Pic+Cl</td>
<td>Electrochemical</td>
<td>0.88</td>
<td>2.54 – 48.6</td>
<td>[256]</td>
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<tr>
<td>n-CoNP/CPE</td>
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<td>40</td>
<td>1 – 59, 2000 – 8000</td>
<td>[257]</td>
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<tr>
<td>PrHCF</td>
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<td>2.15</td>
<td>600 – 8000</td>
<td>[258]</td>
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<td>CoHCF/ graphitic carbon</td>
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<td>1.7</td>
<td>4 – 128</td>
<td>[259]</td>
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<td>PTZ-IL/MWCNT/GCE</td>
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<td>9.3</td>
<td>30 – 1177</td>
<td>[260]</td>
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<td>CTAB-chitosan-CNTs</td>
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<td>9.6</td>
<td>30 – 800</td>
<td>[261]</td>
</tr>
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</table>

¹⁰ TSP: Temporal Switching Protein
Table 4. Various nanoscale sensory systems, and sensing protocols, and analytical techniques reported for sensitive detection of hazardous organic and inorganic species such as heavy metals and bisphenol A.

<table>
<thead>
<tr>
<th>Sensor protocols and designs based materials</th>
<th>Target</th>
<th>Techniques</th>
<th>LOD (µM)</th>
<th>Linear Range (µM)</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>SWCNT- Thiophenol (PhSH)/Au electrode</td>
<td>Hg (II)</td>
<td>Electrochemical</td>
<td>0.003</td>
<td>0.005 – 0.09</td>
<td>[303]</td>
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<tr>
<td>GSH@Fe₃O₄ Glutathione functionalized  magnetic nanocomposite</td>
<td>Pb (II)</td>
<td>Electrochemical</td>
<td>0.00087</td>
<td>0.0024 – 0.5</td>
<td>[275]</td>
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<tr>
<td>GQDs/ Au electrode</td>
<td>Hg (II)</td>
<td>Electrochemical</td>
<td>0.00002</td>
<td>0.00002 – 0.0015</td>
<td>[274]</td>
</tr>
<tr>
<td>GQDs/ Au electrode</td>
<td>Cu (II)</td>
<td>Electrochemical</td>
<td>0.00005</td>
<td>0.000005 – 0.5</td>
<td></td>
</tr>
<tr>
<td>CDs@g-C₃N₄ Carbon dots@graphitic-carbon  nitride</td>
<td>Cr (VI)</td>
<td>Electrochemical</td>
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<td>[273]</td>
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<tr>
<td>CDs@g-C₃N₄ Carbon dots@graphitic-carbon  nitride</td>
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<td>Electrochemical</td>
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<td>Electrochemical</td>
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<td>CDs@g-C₃N₄ Carbon dots@graphitic-carbon  nitride</td>
<td>Cu (II)</td>
<td>Electrochemical</td>
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<td>[272]</td>
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<tr>
<td>PVDF-g-PAA-CDs</td>
<td>Hg (II)</td>
<td>Fluorescence</td>
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<td>PVDF-g-PAA-CDs</td>
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<td>Fluorescence</td>
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<td>PVDF-g-PAA-CDs</td>
<td>Fe (III)</td>
<td>Fluorescence</td>
<td>17</td>
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<td>[296]</td>
</tr>
<tr>
<td>Ratiometric fluorescent chemosensors (RFCs)/Zr-MOF</td>
<td>Zn (II)</td>
<td>Fluorescence</td>
<td>0.000447</td>
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<tr>
<td>S1/Zr-MOF ¹</td>
<td>Zn (II)</td>
<td>Fluorescence</td>
<td>0.001</td>
<td>0.008 – 31</td>
<td>[294]</td>
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<tr>
<td>PHBS-sensor ²</td>
<td>Fe (III)</td>
<td>Fluorescence</td>
<td>17</td>
<td>--</td>
<td>[293]</td>
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<tr>
<td>38-mer and 12-mer aptamers/AuNPs</td>
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<td>Colorimetry</td>
<td>0.0876</td>
<td>0.4 – 657</td>
<td>[321]</td>
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<td>38-mer and 12-mer aptamers/AuNPs</td>
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<td>Colorimetry</td>
<td>7.6 x10^6</td>
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<td>[322]</td>
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<tr>
<td>38-mer and 12-mer aptamers/AuNPs</td>
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<td>Colorimetry</td>
<td>14.41 x10^6</td>
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<td>[322]</td>
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<tr>
<td>24-bp aptamer/AuNPs</td>
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<td>Colorimetry</td>
<td>0.004</td>
<td>0.0000004 – 4</td>
<td>[323]</td>
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<tr>
<td>BSA-spefic aptamer/AuNPs</td>
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<td>Colorimetry</td>
<td>0.0015</td>
<td>--</td>
<td>[324]</td>
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<tr>
<td>ZnFe2O4 and cellulose fibers/MIP</td>
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<td>Colorimetry</td>
<td>0.0618</td>
<td>0.01 – 1</td>
<td>[325]</td>
</tr>
<tr>
<td>DNA–QD₆₅₅/ magnetic beads (MBs)</td>
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<td>Fluorescence</td>
<td>0.74 x10⁻⁹</td>
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<td>[327]</td>
</tr>
<tr>
<td>FAM-ssDNA/GO</td>
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<td>Fluorescence</td>
<td>0.02</td>
<td>0.4 – 40</td>
<td>[329]</td>
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<td>CdTe QDs/AuNPs</td>
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<td>Fluorescence</td>
<td>0.008</td>
<td>0.004 – 0.35</td>
<td>[330]</td>
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<tr>
<td>Boron-doped diamond</td>
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<td>Fluorescence</td>
<td>0.005</td>
<td>0.1 – 50</td>
<td>[331]</td>
</tr>
<tr>
<td>β-cyclodextrin/MWCNTs</td>
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<td>Fluorescence</td>
<td>0.006</td>
<td>0.05 – 5</td>
<td>[332]</td>
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<tr>
<td>Tyrosinase/MWCNTs</td>
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<td>Fluorescence</td>
<td>0.03</td>
<td>0.05 – 3</td>
<td>[333]</td>
</tr>
<tr>
<td>TiO₂</td>
<td></td>
<td>Fluorescence</td>
<td>0.003</td>
<td>0.01 – 20</td>
<td>[335]</td>
</tr>
<tr>
<td>Carboxylic acid functionalized carbon black (CB)/f-MWCNTs</td>
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<td>Fluorescence</td>
<td>0.08</td>
<td>0.1 – 130</td>
<td>[336]</td>
</tr>
<tr>
<td>Cu–Zn/GO/GCE</td>
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<td>Electrochemical</td>
<td>0.00088</td>
<td>0.003 – 0.1</td>
<td>[337]</td>
</tr>
<tr>
<td>TiO₂/Au NTAs</td>
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<td>Electrochemical</td>
<td>0.0062</td>
<td>0.1 – 28.9</td>
<td>[338]</td>
</tr>
</tbody>
</table>

¹ (2,2’-(1Z,12Z)-5,9-dithia-2,12-diazatriodeca-1,12-diene-1,13 diyl) diphenol), (S1)/Zr-MOF
² 4-(2-(3-Methyl-5-oxo-1-tosyl-1H-pyrazol-4(5H)-ylidene) hydrazinyl)-N-(pyrimidin-2-yl) benzenesulfonamide (PHBS-sensor)
³ 3', 6'- bis(diethylamino) -2- ((3,4,5 trimethyl benzylidene) amino) spiro [isoindoline-1,9'-xanthen] -3-one
Table 5. A list of commercial mobile sensor devices used in the assessment of food quality.

<table>
<thead>
<tr>
<th>Sensor devices</th>
<th>Target</th>
<th>Ref.</th>
<th>Device image</th>
</tr>
</thead>
<tbody>
<tr>
<td>Handheld ATP fluorescence detector</td>
<td>Rapid detection food microorganism bacteria detection instrument</td>
<td><a href="https://www.aliexpress.com/item/4001269652463.html">https://www.aliexpress.com/item/4001269652463.html</a></td>
<td>![Device2]</td>
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<tr>
<td>HD Pesticide residue detector</td>
<td>Detection of pesticides in vegetable, fruit, meat, tea, household food safety, heavy metal rapid test instrument</td>
<td><a href="https://www.aliexpress.com/item/1005001389512206.html?spm=a2g0o.detail.1000023.3.2d48141ded8XHS">https://www.aliexpress.com/item/1005001389512206.html?spm=a2g0o.detail.1000023.3.2d48141ded8XHS</a></td>
<td>![Device6]</td>
</tr>
<tr>
<td>Food / Hygiene Temperature Meter PCE-ST 1</td>
<td>Food / hygiene temperature</td>
<td><a href="https://www.pce-instruments.com/english/measuring-instruments/test-meters/food-hygiene-kat_42003.htm">https://www.pce-instruments.com/english/measuring-instruments/test-meters/food-hygiene-kat_42003.htm</a></td>
<td>![Device9]</td>
</tr>
<tr>
<td>Ascorbic Acid Portable Test Kits (Hach)</td>
<td>Ascorbic Acid</td>
<td><a href="https://www.weberscientific.com/ascorbic-acid-portable-test-kits-hach">https://www.weberscientific.com/ascorbic-acid-portable-test-kits-hach</a></td>
<td>![Device11]</td>
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<tr>
<td>Code</td>
<td>Description</td>
<td>Link</td>
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</tr>
<tr>
<td>---------</td>
<td>--------------------------------------</td>
<td>----------------------------------------------------------------------</td>
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<tr>
<td>HI 767</td>
<td>Marine checker Low concentration nitrite nitrogen</td>
<td><a href="https://hanna.co.jp/item-hi767.html">https://hanna.co.jp/item-hi767.html</a></td>
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<tr>
<td>HI 3854</td>
<td>Zinc test kit</td>
<td><a href="https://www.e-hanna.info/cart/shop/HI3854.html">https://www.e-hanna.info/cart/shop/HI3854.html</a></td>
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<tr>
<td>HI 3850</td>
<td>Ascorbic acid (vitamin C) test kit</td>
<td><a href="https://www.e-hanna.info/cart/shop/HI3850.html">https://www.e-hanna.info/cart/shop/HI3850.html</a></td>
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<tr>
<td>HI 3822</td>
<td>Sulfite test kit</td>
<td><a href="https://www.e-hanna.info/cart/shop/HI3822.html">https://www.e-hanna.info/cart/shop/HI3822.html</a></td>
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<tr>
<td>HI 3844</td>
<td>Hydrogen peroxide test kit</td>
<td><a href="https://www.e-hanna.info/cart/shop/HI3844.html">https://www.e-hanna.info/cart/shop/HI3844.html</a></td>
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<td>HI 3847</td>
<td>Copper test kit</td>
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<td>HI 3864</td>
<td>Phenol test kit</td>
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