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Review—Emerging Trends in the Development of Electrochemical Devices for the On-Site Detection of Food Contaminants

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Abstract

Traditional laboratory-based sensing strategies for food contaminant detection are often limited because they are time consuming and expensive and require trained personnel, which makes them unsuitable for routine sensing. Therefore, the scientific and industrial community is showing enormous interest in the design and development of portable sensing devices for the on-site and point-of-care detection of food contaminants. Portability is one of the chief characteristic features of designing contemporary analytical devices. Portable devices have received tremendous attention, as these novel devices have advanced the field of sensing. Various sensing strategies have been utilized for on-site detection of food contaminants. Among these, portable electrochemical devices have emerged vigorously in the past few years. Scientists and industrialists have worked effortlessly to develop portable electrochemical devices for a minute amount of food contaminant detection in water bodies and food products. The current work aims to demonstrate recent research progress related to the design, development, and improvement of portable electrochemical devices for detection of food contaminants.

1. Introduction

Over the past few years, there has been a substantial need for continuous and efficient monitoring of food and water quality. The World Health Organization (WHO) has stressed food safety as an international concern for providing nontoxic, innocuous, and clean food. A wide range of food contaminants, such as pesticides, phenolic compounds, illegal food additives, veterinary drugs, pathogens, herbicides, mycotoxins, and heavy metals, are widely affecting food products. The contamination of food results in severe and drastic health issues. Moreover, food contamination is a huge financial and medical burden globally. According to WHO assessment, many diseases ranging from common diarrhea to incurable cancer are caused by food contamination.[1-5] Hence, it is imperative to efficiently monitor and detect food contaminants to safeguard food safety and public health. Common and conventional food contaminant detection methods are liquid chromatography, gas chromatography, capillary electrophoresis, micellar electrokinetic capillary chromatography, gas chromatography-mass spectrometry, inductively coupled plasma mass spectrometry, thin-layer chromatography, fluorescence microscopy, quantitative real-time polymerase chain reaction, high-performance liquid chromatography, inductively coupled plasma atomic emission spectrometry, enzyme-linked immunosorbent assays, inductively coupled plasma optical emission spectrometry, atomic absorption spectroscopy, and liquid chromatography-mass spectrometry. These are highly sensitive and selective approaches. Characteristically, these are very costly large instruments with complex operational procedures, and they require time-taking sample detection procedures and professional manpower; they also restrict sample preconcentration procedures.[5-11] Hitherto, despite their outreach, these conventional strategies are being constrained within sophisticated laboratories. These limitations make them suitable for real-time, point-of-care (POC), in situ, and on-site detection. Additionally, the large size of these

strategies restricts them for field-based detection. In this regard, several analytical strategies have been designed for the detection of food contaminants. These include phosphorescence, colorimetric, electrochemical, fluorescence, chemiluminescence, surface plasmon resonance, surface-enhanced Raman scattering, and microfluidic strategies. These techniques are somewhat more advantageous than traditional strategies. Even though these strategies are good and efficient, they cannot be used in the respective fields for on-site detection, owing to the large size of the device. Hence, portable devices need to be designed for on-site detection of food contaminants. In this regard, scientists and technologists have designed and developed portable analytical strategies for the on-site detection of food contaminants. These devices are fast, economic, simple, and portable analytical techniques; these characteristics have helped them gain considerable attention for the on-site detection of various food contaminants.[1-10, 12]

Among the various sensing strategies, colorimetric, fluorescence, and electrochemical assays are widely used candidates for the detection of contaminants.[3-7,9] These assays exhibited great promise for food quality and food safety, which are highly suitable for the execution of onsite sensing of food contaminants, especially in the resource-limited settings. Actually, up to now, onsite sensing strategies are still designated as significant desirable assays for simply attaining accurate information about the contaminants in foods, both rapidly and at the point of care. Real-time detection of food contaminants requires novel and affordable materials that can be easily prepared and deliver rapid and efficient outcomes without compromising sensitivity or accuracy. To attain this target, extensive scientific research is being conducted for the sensitive and selective detection of food contaminants. Recent advanced scientific research interests are focused on the electrochemical sensing strategies. Currently, these sensing strategies are being widely used to detect hazardous contaminants and to identify and quantify

specific target analytes, owing to their exceptional selectivity, sensitivity, reliability, fast response, low maintenance costs, and wide linear range.[13-18] Chief electrochemical sensing methodologies are voltammetry, potentiometry, impedimetric, conductometry, and amperometry. These include cyclic voltammetry, impedance, linear sweep voltammetry, differential pulse voltammetry (DPV), differential pulse anodic stripping voltammetry (DPASV), anodic stripping voltammetry, and square wave anodic stripping voltammetry.[1-5]

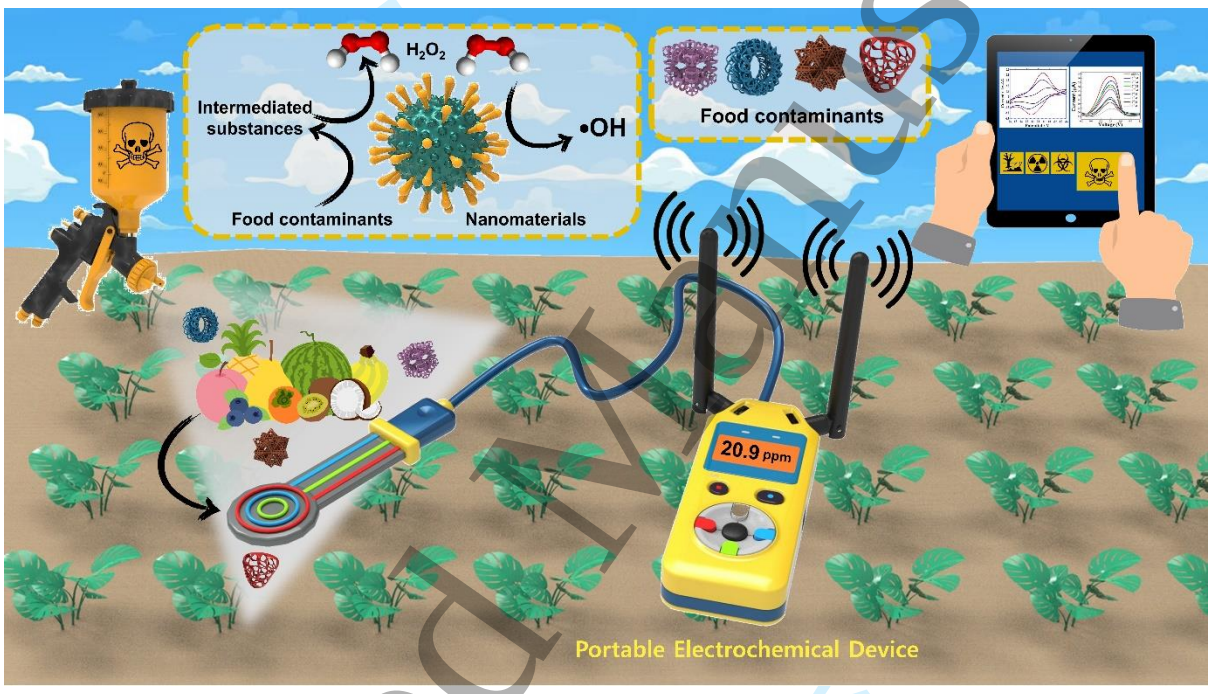

Figure 1. Schematic representation of the work.

Recent progress in portable electrochemical sensing assays has intensely enhanced selectivity, accuracy, sensitivity, and ease of handling, and they take less time for testing, require less sample size, consume fewer chemicals, and present simplified on-site detection. Moreover, electrochemical assays can be used for the in situ analysis. Further, modification of electrodes with specific nanomaterials can significantly improve overall electrochemical

behavior and the limit of detection (LOD). Major areas of interest for noncentralized and centralized investigations are the reduction of sample volume and the fabrication of portable devices. This can be achieved by combining electrochemical sensing assays with screen-printed electrodes (SPEs). The design and development of SPEs depend on thick-film technologies. SPEs have revolutionized decentralized electroanalysis owing to their capability to bridge the gap between lab-to-hand implementation.[19-24] Fig. 1. Schematically represents the outline of current work. The advantages of SPEs are as follows: economical, easy operation, on-site analysis, fast, environmentally benign, and sample treatment is not required. SPEs are widely used in various fields. Recently, SPEs have been widely used in portable electrochemical devices for on-site sensig of food contaminants. SPEs can generate a wide range of electronic sensing platforms that are cost-effective, real-time, rapid, and practical to utilize in environmental monitoring, agricultural monitoring, healthcare, and biotechnology. Screen printing technology will become a crucial component of future research. Fig. 2. represents schematically difference between the laboratory based electrochemical device and portable device. With the advancement and popularity of electrochemical sensing strategies and digital imaging features, potable electrochemical devices are being widely integrated with smartphones. This makes them an exceptional analytical platform to design POC testing devices. Electrochemical strategy appears to be promising and powerful with a substantial potential to meet the global market demand. Manufacturing of the portable devices is progressively increasing with a momentous impact in the scientific and technological community. Internationally, portable devices business has significant potential. Portable devices market is expected to grow rapidly in the coming years.

This perspective aims to provide a strategic idea for portable electrochemical devices

for on-site sensing of food contaminants. Effective portable sensing devices aim to achieve precise on-site monitoring of toxic food contaminants, which is immensely important for safeguarding food safety and human health.[3-9]

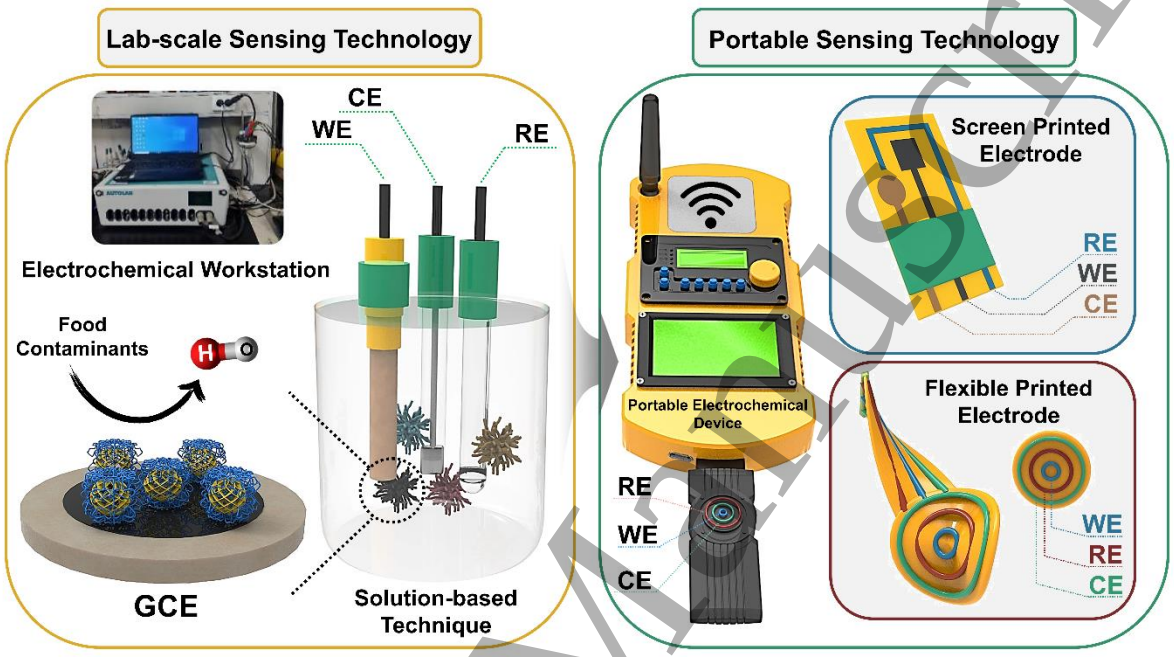

Figure 2. Comparison between laboratory based electrochemical device and portable device.[4]

2. Portable electrochemical devices for on-site detection of food contaminants

Portable electrochemical devices have shown significant potentiality in the point of care testing technology. Detection of hazardous contaminants using portable electrochemical devices has become more strategic, highly efficient, faster to operate, executable, and cost-effective. Further, these characteristics features offer massive potential for the precise and early assessment and detection of food contaminants. Portable electrochemical devices are emerging the sensing platforms, which can satisfy the expectations of the user. With the aid of miniatured

electrochemical tools, analysis can be done outside the lab by a non-specialized person. Recently, portable electrochemical tools have been designed for on-site detection of pesticide residues in foods. For instance, Jin *et al.*[25] developed a dual-mode (colorimetric and electrochemical) readout for the on-site sensing of pesticides. In this system, they used all-in-one enzyme-inorganic hybrid nanoflowers to design a high-performance artificial enzyme cascade. The fabricated affordable and sensitive lab-on-paper biosensor was incorporated with disposable screen-printed carbon electrode (SPCE). Paraoxon was detected with a sensing platform with an LOD of 6 fg mL^{-1}. Pereira *et al.*[26] demonstrated the selective and sensitive detection of carbendazim, diuron, paraquat, and fenitrothion pesticides using glove-embedded sensors printed on the fingers of rubber gloves. Fabricated sensors are flexible, wearable, glove-embedded, and nonenzymatic. LODs (DPV) for the diuron and carbendazim were found to be 9.2×10^{-7} and 4.7×10^{-8} mol L^{-1}. The LODs (SWV) for the fenitrothion and paraquat were 6.4×10^{-7} and 2.4×10^{-8} mol L^{-1}. Appreciable electrochemical performances have been attained in real samples of cabbage and apples by touching with the glove and with the immersion of the fingers in the orange juice. Mishra *et al.*[27] reported the fabrication of wearable, stretchable, disposable, and flexible electrochemical glove biosensors for the on-site screening of organophosphorus nerve agents on fruits and vegetables. Electrochemical profiles were obtained using a portable wireless potentiostat. The “lab-on-a-glove” sensor was integrated with a wireless data transmitter and smartphone device and immobilized with the enzyme. The sensor has excellent resiliency and is mechanically robust. Deformations can be accomplished by coupling ink materials with serpentine-printable patterns. A wearable point-of-use device can be used in food security, forensics, and defense applications. Hildebrand *et al.*[28] deliberated on the electrochemical behaviors of portable biosensors for the screening of carbamate and organophosphorus insecticides in food and water. The biosensor was fabricated by inhibiting

the acetylcholinesterase enzyme using potentiostat and SPEs. Farshchi *et al.*[29] developed a wearable glove-based electrochemical sensor using silver nanoconductive ink, which was developed for the on-site screening of trifluralin pesticide residues. A three-electrode sensing system with optimum thicknesses was developed on the finger surface of the rubber glove. The designed electrochemical sensing assay detected trifluralin in the 0.01 μM to 1 mM range on the mulberry leaves and tomato surfaces using SWV and DPV techniques. The sensor is easy to use and portable and has a potential environmental capability that can be applied for screening chemical threats in water and soil. Nagabooshanam *et al.*[30] developed a low-cost, portable, economical, and user-friendly electrochemical biosensing strategy for the ultrasensitive monitoring of chlorpyrifos pesticide residues in tomato juice. The sensor module was prepared on paper to eliminate the contamination of plastics, as well as to reduce costs; this, in turn, provides environmentally friendly and economic solutions. The sensor attained 3 ng/L of LOD and 0.521 k Ω/ng L^{-1}/mm^2 of sensitivity. The sensor requires 100 μL of reagent and can be tested with a 10 ng/L to 1,000 ng/L linear concentration range with a 5 s response time. Significantly, 96% accuracy was noticed with the fabricated sensor. Rajaji *et al.*[31] developed a yttrium iron garnet and graphitic carbon nitride (YIG-GCN) electrocatalyst for the picomolar-level sensing of mesotrione oranges and grapes. The electrocatalyst has shown appreciable catalytic and surface properties. The modified electrode attained an LOD of 950 pM for mesotrione. The modified YIG-GCN electrode was integrated with a portable KAUST at potentiostat and connected with a smartphone. Mahmoudpour *et al.*[32] fabricated wearable and flexible glove-based sensors for point-of-use sensing in food security and defense applications. Conductive designs on the rubber glove fingers were drawn using gold–silver-modified graphene quantum dot nanoink with optimum thickness. The designed sensing assay was integrated with a portable electrochemical tool for on-site sensing of trifluralin in the fruits

at 10 nM of the limit of quantification in the 10 nM–1 mM range. The appreciable distinction and high efficiency of the pesticide at quantified concentrations in the real apple and leaf samples were accomplished by simply touching and immersing with the glove. Nagabooshanam *et al.* [33] designed a micro-electrochemical analytical tool for the screening of chlorpyrifos in foods. The gold microelectrode was modified with acetylcholinesterase, and a zinc metal–organic framework was utilized as the exceptional electroanalytical transducer. Cyclic voltammetry, differential pulse voltammetry (DPV), and electrochemical impedance spectroscopy (EIS) were performed with the fabricated portable micro-electrochemical device. Two µL of the analyte is enough for analysis and can be tested in the 10–100 ng/L linear range. The device detected the pesticide with a sensitivity and LOD of 0.598 µ A/ng L^{-1}/mm^2 and 6 ng/L, respectively. The designed sensor has appreciable stability with a 20-day shelf life. The portable electrochemical tool is also integrated with a low-cost handheld potentiostat. Zhao *et al.*[34] fabricated a wearable smart plant biosensor for in situ detection of pesticides. A Serpentine three-electrode was prepared using laser-induced graphene technology. The fabricated flexible and stretchable electrode can be used effectively on the irregular surfaces of fruits and leaves. Organophosphorus hydrolase modification and integration with semisolid biocompatible electrolyte, wearable smart plant biosensor, selectively detected the methyl parathion, and the results can be communicated wirelessly to a smartphone. This feature enables the in situ and real-time electrochemical detection of pesticides. Liu *et al* [20] constructed portable novel electrochemical biosensor by integrating laser-induced graphene electrode on the polyimide foil and MnO_2 nanosheets were loaded on paper for point-of-care testing of pesticide residues. Portable biosensor displayed satisfactory electrochemical behaviors for the pesticide assay with 3 to 4000 ng/mL linear range and 1.2 ng/mL of LOD. Arduini *et al.*[35] demonstrated a 3D origami paper device for the screening of pesticides using a combination of various enzyme-

inhibiting biosensors. The device was integrated with office paper and filter paper pads based SPEs to load the enzymatic substrates and enzymes. Multipurpose examinations of various pesticides were carried out by unfolding and folding the filter paper structures without incorporating the reagents, filtration, dilution, and pH adjustment. A paper-based sensing platform was used to screen paraoxon, atrazine, and 2,4-dichlorophenoxyacetic acid and to inhibit butyrylcholinesterase, tyrosinase, and alkaline phosphatase. The degree of inhibition of the quantity of pesticides was assessed chronoamperometrically using a portable potentiostat. To enhance the sensitivity, the paper electrodes were altered using carbon black for the detection of atrazine and 2,4-dichlorophenoxyacetic acid and modified with Prussian blue nanoparticles for paraoxon screening. The accuracy of the 3D origami paper electrochemical sensing assay was assessed in river water. The obtained recovery values were 90 ± 1% and 88 ± 2% for 10 and 20 ppb of paraoxon, respectively.

Portable electrochemical tools have been designed for on-site detection of illicit drugs. For instance, Parrilla *et al.*[36] reported a low-cost and sensitive electrochemical strategy for the detection of heroin, cocaine, 4-chloro-alpha-pyrrolidinovalerophenone, 3,4-methylenedioxymethamphetamine, and ketamine illicit drugs in oral fluid using surfactant solution. Specifically, the surfactant was adsorbed on the surface of the carbon electrode and produced the adsorption of illicit drugs; this allowed for an improved electrochemical signal. The developed analytical approach exhibited 1–30 μM of linear concentration range along with high sensitivity and sub-micromolar LODs. Finally, the assay was assessed in diluted oral fluid sample solutions spiked with heroin, cocaine, 4-chloro-alpha-pyrrolidinovalerophenone, 3,4-methylenedioxymethamphetamine, and illicit ketamine drugs. The proposed sensor is a rapid,

simple, sensitive, and portable system for screening illicit drugs. Echelpoel *et al*[13] demonstrated on the electrochemical strategy for on-site multidrug screening at the festivals. Parrilla *et al.*[37] developed a portable electrochemical tool for on-site and rapid recognition of illicit drugs in smuggled samples. The portable device uses affordable SPEs for profiling the electrochemical behaviors of various illicit drugs by SWV. An illicit drug was identified based on the oxidation potential of the analyte. The electrochemical profile library was built after the analysis of common cutting agents and illicit compounds. The library helps in designing a tailor-made script that allows for the identification of the drug through a mobile phone or laptop interface. Saisahas *et al.*[38] fabricated a nano coral polyaniline-modified graphene-based paper-based portable device for electrochemical detection of xylazine. The paper-based electrochemical device was integrated with a mobile phone. The fabrication procedure for the paper-based electrochemical device involves low-tack transfer tape, wax printing, screen printing, and cutting techniques. The substrate was coated with graphene ink and modified with nano coral polyaniline; this provides transfer of electrons with an effective and large surface area, which promotes the transfer of charges. Conductive graphene ink on the paper-based electrochemical device presented with 25.0 ± 0.9 μm of thickness for 0.374 cm^2 of effective surface area. A fabricated sensor was examined to detect xylazine using DPV. Linear responses were obtained from 0.2–5 μg mL^{-1} and from 5–100 μg mL^{-1}. The obtained LOD was 0.06 μg mL^{-1}. Recoveries were observed in the 84 ± 4 to 105 ± 2% range. The fabricated sensor confirmed excellent accuracy in xylazine detection in trace amounts.

Portable electrochemical devices have been established for on-site detection of pathogens. Roy *et al.*[39] explicitly demonstrated the emerging developments in POC pathogen diagnostic and detection methods and further discussed the incorporation and integration of

microfluidics, nanomaterials, SPEs, lateral flow tests, and smartphones into POC devices. Challenges and prospects were provided to develop sample-to-result POC tools for the detection of pathogens. Silva *et al.*[40] comprehensively elucidated electrochemical POC techniques for the detection of waterborne pathogens. Nordin *et al.*[41] developed a portable and simple electrochemical biosensor to monitor the Vibrio parahaemolyticus pathogen in real samples. The reader was based on the SPCE and modified with methylene blue and gold nanoparticles. Detection was performed using a microprocessor to examine the current under controlled potential. The sensor can precisely distinguish complementary, mismatched, and noncomplementary oligonucleotides. DNA was examined in the 2.0×10^{-8} – 2.0×10^{-13} M range with 2.16 pM of LOD. Furthermore, cross-reactivity measurements against different foodborne pathogens displayed a consistently sensitive detection of the pathogen.

Portable electrochemical devices have been developed for the on-site detection of mycotoxins. Soares *et al.*[42] explicitly deliberated on the emerging advances, opportunities, and challenges for the point-of-need detection of mycotoxins in feeds and foods. Abnous *et al.*[43] demonstrated an amperometric aptasensor for the detection of ochratoxin A mycotoxin. The fabricated sensor was based on a modified gold electrode comprising methylene blue as a redox indicator, aptamer as a sensing ligand, aptamer complementary strands as assisting DNA, and single-walled carbon nanotubes as electrochemical peak amplifiers. In the absence of a mycotoxin, the duplex designed between the aptamer and the complementary strands remains intact. Hence, efficient electrochemical behavior will be noticed due to the presence of redox marker methylene blue in the duplex. In the presence of mycotoxin, the duplex will be disassembled, and methylene blue and single-walled carbon nanotubes will be moved from the gold electrode surface, resulting in the weakening of electrochemical signals. The proposed

strategy is highly specific for the detection of ochratoxin A and has an LOD of 52 pM. A fabricated aptasensor was effectively used to detect ochrotoxin A in the spiked grape juice and serum samples, with LODs of 58 and 134 pM, respectively. Uludag *et al.*[44] fabricated a lab-on-a-chip biosensor for the real-time sensing of aflatoxins. Goud *et al.*[45] designed a portable and disposable electrochemical aptasensor for the label-free sensing of aflatoxin B1 in alcoholic beverages. This sensing is based on the specific recognition of aptamers by covalent binding as a compact monolayer on the SPCE through a diazonium coupling reaction. Aflatoxin B1 quantification was obtained using EIS. The quantification range was observed from 0.125 ng mL^{-1} to 16 ng mL^{-1}. The obtained LODs were 0.25 ng mL^{-1} and 0.12 ng mL^{-1} for seqB and seqA, respectively. For the real-time detection of the fabricated aptasensors confirmed in wine and beer samples, the sensor showed appreciable recovery levels in the 92–102% range for the detection of aflatoxin B1. Recent advancements have enhanced the electrocatalytic behaviors of food sensors. Novel nanomaterials have shown potential in potentiometric, voltammetric, impedimetric, amperometric, and conductometric devices for sensing food contaminants. Advantages of portable devices and on-site sensing strategies are portability, high-throughput simplicity, affordability, ease of carrying, ease of handling, less laborious, low engineering cost, simple sensor designing, automatized detection, less weight, rapid analysis of results, and simple operation procedure.

3. Conclusions and future perspectives

Portability is the chief essential key for the design and development of advanced analytical devices. SPEs are economical, simple, highly selective, sensitive, and rapid response approaches that are highly suitable for on-site detection. Novel nanomaterials have shown potential for designing efficient electrochemical sensing platforms. Several research efforts

have been efficiently devoted to fabricating user-friendly and portable sensing tools with origami papers, SPEs, glove-based sensors, wearable sensors, and strip/paper/card-based electrochemical sensing strategies. Moreover, significant enhancements have been designed with electrocatalytic procedures for the on-site sensing or POC testing of food contaminants. This review highlights emerging trends in the development of electrochemical devices for the on-site detection of food contaminants. Even though electrochemical sensing strategies have prospered in monitoring various food contaminants, many challenges still need to be resolved. For example, multifunctional, miniaturized, wearable, intelligent, stable, implantable, adaptable, sensitive, integrated, selective, and inexpensive electrochemical devices should be fabricated for the detection of food contaminants. Fabricated portable tools should be upgraded with scientific automation and computation. More progressive, wearable, and reusable SPEs should be designed with well-functioned geometries. Efficient lab-on-a-chip, lab-on-a-glove, lab-on-a-disc, and lab-on-a-leaf models should be developed for the detection of food contaminants. Metabolites, biomolecules, microorganisms, specific antibodies, aptamers, and biomarkers should be coupled with SPEs for the therapeutic monitoring of various target analytes. We anticipate that with continuous advancements in advanced sensing strategies, the above-stated challenges will be efficiently solved.

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