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Article 107682

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Review

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Evaluation of the ambiguity in second-order analytical calibration based on multivariate curve resolution. A tutorial

Alejandro C. Olivieri
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Microextraction techniques for sampling and determination of polychlorinated biphenyls: A comprehensive review

Vahid Jalili, Masoud Ghanbari Kakavandi, Alireza Ghiasvand, Abdullah Barkhordari
Article 107442

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State-of-the-art of convenient and low-cost electrochemical sensor for food contamination detection: Technical and analytical overview

Trias Mahmudiono, Dmitry Olegovich Bokov, Saade Abdalkareem Jasim, Walid Kamal Abdelbasset, Dinora M. Khashirbaeva
Article 107460

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Recent progress in the application of pH-responsive polymers in separation science

Herbert Musarurwa, Nikita Tawanda Tavengwa
Article 107503

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Remarks on use of the term “deep eutectic solvent” in analytical chemistry

FEEDBACK

Vasil Andruch, Patrycja Makoś-Chełstowska, Justyna Płotka-Wasylka
Article 107498

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Critical strategies to pinpoint carryover problems in liquid chromatography-mass spectrometry: A systematic direction for their origin identification and mitigation

Ashish Jogpethe, Tarang Jadav, Niraj Rajput, Amit Kumar Sahu, ... Pinaki Sengupta
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Smart materials for mercury and arsenic determination in food and beverages

Kamal K. Jinadasa, Elena Peña-Vázquez, Pilar Bermejo-Barrera, Antonio Moreda-Piñeiro
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Recent advances in turn off-on fluorescence sensing strategies for sensitive biochemical analysis - A mechanistic approach

Bui The Huy, Daniel T. Thangadurai, Mirkomil Sharipov, Nguyen Ngoc Nghia, ... Yong-Ill Lee
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HPLC in the discovery of plant phenolics as antifungal molecules against *Candida* infection related biofilms

Neha Jaiswal, Awanish Kumar
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A magnetic molecularly imprinted polymer hierarchical composite adsorbent embedded with a zinc oxide carbon foam nanocomposite for the extraction of sulfonamides

Ananya Kliangsuwan, Apichai Phonchai, Opas Bunkoed
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Exo-III enzyme based colorimetric small extracellular vesicles (sEVs) detection via G-quadruplex-based signal quenching strategy

JunLiang Zhang, MengHan Chang, Xin Shi, Xing Zhou
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Tuning of Co_3X_4 (X = O, S, Se) by anion substitution for highly electrochemical sensing of glucose

Xiao Bai, Hang Yin, Chongchao Zhang, Ziyin Yang
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An ultrasensitive electrochemical immunosensor based on in-situ growth of $CuWO_4$ nanoparticles on MoS_2 and chitosan-gold nanoparticles for cortisol detection

Cuijie Nong, Bo Yang, Xiaokun Li, Suxiang Feng, Hongxin Cui
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A shipboard method for catalytic kinetic spectrophotometric determination of trace Cu(II) concentrations in seawater using reverse flow injection analysis coupled with a long path length liquid waveguide capillary cell

Ting Wang, Yongming Huang, Yong Zhu, Jin Xu, ... Dongxing Yuan
Article 107441

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Magnetic solid phase extraction of lead ion from water samples with humic acid modified magnetic nanoparticles prior to its fame atomic absorption spectrometric detection

Xiaoyue Shan, Lei Zhang, Haipeng Ye, Ji Shao, ... Chengjian Cao
Article 107417

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Microwave-assisted one-pot synthesis of carbon dots for highly sensitive and selective detection of selenite

Guiping Liao, Jin Luo, Tengfei Cui, Jie Zou, ... Fujian Xu
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Development of a solvent-free micellar HPLC method for determination of five antidiabetic drugs using response surface methodology

Eman A. Bahgat, Hani M. Hafez, Heba M. El-Sayed, Naglaa Abdel Sattar Kabil
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Detection of $BRAF^{V600E}$ mutation of thyroid cancer in circulating tumor DNA by an electrochemical-enrichment assisted ARMS-qPCR assay

Jia Wei, Yaoqi Wang, Jiaxue Gao, Zhenshengnan Li, ... Xianying Meng
Article 107452

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Sequential decision fusion pipeline for the high-throughput species recognition of medicinal caterpillar fungus by using ATR-FTIR

Yun Li, Qirui Bi, Wenlong Wei, Changliang Yao, ... Dean Guo
Article 107437

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A smartphone based-paper test strip chemosensor coupled with gold nanoparticles for the Pb^{2+} detection in highly contaminated meat samples

Rossukon Srisukjaroen, Kanokorn Wechakorn, Siriwan Teepoo
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A method based on the conversion and determination of folpet and phthalimide residues in tea by GC-MS/MS and GC-TOF-HRMS

Chao Han, Chaoqun Huang, Qinting Jiang, Li Chen, ... Yan Shen
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Preparation of polyfluorinated azobenzene/niobate composite as electrochemical sensor for detection of ascorbic acid and dopamine

Tongtong Cao, Yicheng Zhou, Haoran Wang, Rongrong Qiao, ... Zhiwei Tong

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Switched electrochemical sensor for hydroquinone based on rGO@Au, monoclinic $BiVO_4$ and temperature-sensitive polymer composite material

Chenxi Wang, Pengcheng Zhao, Li Zhang, Yilin Wang, ... Junjie Fei
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F-doped silicon quantum dots as a novel fluorescence nanosensor for quantitative detection of new coccine and application in food samples

Yulu Liu, Minghui Zan, Lei Cao, Jiahui Peng, ... Wen-Fei Dong
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First application of microwave-assisted extraction in the analysis of polybrominated diphenyl ethers in human milk

Marija Dvoršćak, Karla Jagić, Lucija Besednik, Iva Šimić, Darija Klinčić
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Copper tape to improve analytical performance of disposable carbon electrodes in stripping analysis

Xiang-Yun Lin, Qing-Wen Zhou, Xiao-Lei Huo, Ning Bao
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Deep eutectic solvent-based extraction of metals from oil samples for elemental analysis by ICP-OES

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Urease-Functionalized Near-Infrared Light-Responsive Gold Nanoflowers for Rapid Detection of Urea by a Portable Pressure Meter

Yuan Yuan, Yuhan He, Dongni Pei, Liujuan Tong, ... Jianxiu Wang
Article 107450

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DES-based vortex-assisted liquid-liquid microextraction procedure developed for the determination of paraben preservatives in mouthwashes

Aslihan Dalmaz, Sezen Sivrikaya Özak
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Synthesis, characterization, and the investigation of the applicability of citric acid functionalized Fe_2O_3 nanoparticles for the extraction of carvedilol from human plasma using DFT calculations and clinical samples analysis

Mina Shaban, Hanieh Hayadokht, Jalal Hanaee, Jaber Jahanbeen Sardroudi, ... Somaieh Soltani
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Unexpected sensitivity enhancement in analysing alfatoxin M1 using LC-IDMS

Ádám Tölgyesi, Bence Tóth Kovács, Edgár Tóth, Andrea Simon, ... Virender K. Sharma
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Highly selective, rapid and simple colorimetric detection of Fe^{3+}in fortified foods by L-Cysteine modified AuNP

Nirangkush Borah, Amlan Jyoti Kalita, Ankur K. Guha, Manash R. Das, Chandan Tamuly
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Development and validation of a method for determining D-limonene and its oxidation products in vegetables and soil using GC–MS

Chenyu Huang, Chuanfei Bian, Long Wang, Wenwen Zhou, ... Baotong Li
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Highly sensitive and selective electrochemical detection of clothianidin using reduced graphene oxide-anionic pillar[6]arene composite film

Yanju Wu, Kainan Guo, Junqi Zhao, Qunpeng Duan, ... Kui Lu
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A novel salting-out extraction system for determination of ethylene glycol and diethylene glycol in liquid samples followed by GC-FID

Fu Wan, Shuping Liu, Li Wang, Shihui Si
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Self-assembled $Ti_3C_2T_X$ MXene/graphene composite for the electrochemical reduction and detection of *p*-nitrophenol

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Lucia Bartella, Marcos Bouza, Priscilla Rocío-Bautista, Leonardo Di Donna, ... Antonio Molina-Díaz
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Aneesh Koyappayil, Sang-ho Yeon, Sachin Ganpat Chavan, Ling Jin, ... Min-Ho Lee
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Ahmet Cetinkaya, S. Irem Kaya, M. Emin Çorman, Mustafa Karakaya, ... Sibel A. Ozkan
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Lelouda-Athanasia Koronaiou, Christina Nannou, Nikoletta Xanthopoulou, Georgia Seretoudi, ... Dimitra A. Lambropoulou
Article 107457

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Wenhao Peng, Ningning Song, Zilong Su, Jiajun Wang, ... Aijuan Xie
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Matheus Valentin Maia, Willian Toito Suarez, Vagner Bezerra dos Santos, João Paulo Barbosa de Almeida
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PCR-free electrochemical genosensor for *Mycobacterium tuberculosis* complex detection based on two-dimensional Ti_3C_2 Mxene-polypyrrole signal amplification

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Kobra Salimiyan Rizi, Behnaz Hatamluyi, Mahdieh Darroudi, Zahra Meshkat, ... Majid Rezayi
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Katja Schulz, Heiko Helms, Anne Janich
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Rapid and interference-free quantification of nine coumarins in *Cnidii fructus* using HPLC-DAD assisted with second-order calibration model

Bing-Bing Liu, Hai-Long Wu, Yue Chen, Tong Wang, Ru-Qin Yu
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Fatemeh Emambakhsh, Hamideh Asadollahzadeh, Nahid Rastakhiz, Sayed Zia Mohammadi
Article 107466

Chromatographic analysis of water and water-alcohol extracts of *Echinacea purpurea* L. obtained by various methods

Zaual Temerdashev, Elena Vinitskaya, Ekaterina Meshcheryakova, Oleg Shpigun
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Expanding the applicability of magnet integrated fabric phase sorptive extraction in food analysis: Extraction of triazine herbicides from herbal infusion samples

Natalia Manousi, Abuzar Kabir, Kenneth G. Furton, George A. Zachariadis, Erwin Rosenberg
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Determination of benzimidazoles in beef by molecularly imprinted boron nitride composite based dispersive solid phase microextraction and ultra performance liquid chromatography

Su Han Wang, Xiao Huan Zang, Shuai Hua Zhang, Jian Ping Wang
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Qiang Bai, Hongyang Luo, Xuetao Yi, Shugao Shi, ... Ning Sui
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High-Throughput Single-Step plasma sample extraction optimization strategies with experimental design for LC-MS and GC–MS integrated metabolomics and lipidomics analysis

Cemil Can Eylem, Emirhan Nemutlu, Aysegul Dogan, Vedat Acik, ... Nursabah Elif Basci Akduman
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Synthesis of β-cyclodextrin grafted rhombohedral-CuO antioxidant nanozyme for detection of dopamine and hexavalent chromium through off–on strategy of peroxidase mimicking activity

Salim Ali, Suranjan Sikdar, Shatarupa Basak, Debasmita Das, ... Mahendra Nath Roy
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Ultra-preconcentration technique for the determination of thallium (I) in water samples by a combination of thallium (I)-imprinted polymer and vortex-assisted liquid–liquid microextraction

Abolfazle Darroudi
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Kaewta Danchana, Hiroshi Iwasaki, Kenta Ochiai, Haruka Namba, Takashi Kaneta
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UiO-66-NH_2: An easily attainable and label-free turn-on probe for facile fluorescence sensing of alkaline phosphatase

Fujian Xu, Jinyi Zhang, Xiaoliang Zeng, Zhou Long, Xiandeng Hou
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Lanthanide metal doped organic gel as ratiometric fluorescence probe for selective monitoring of ciprofloxacin

Yiyang Sun, Pierre Dramou, Zhaorui Song, Limin Zheng, ... Hua He
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Article 107540

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Page 1 of 2

Previous vol/issue Next vol/issue

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Review Article

State-of-the-art of convenient and low-cost electrochemical sensor for food contamination detection: Technical and analytical overview

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ABSTRACT

Food safety issue is becoming as an international concern for human health around the world. Common substances in food can pose a significant threat to human health containing but not restricted to antibiotics resides, veterinary drugs, pesticides, mycotoxins, heavy metals, food borne-pathogens, illegal additives and allergens. A series of analytical methods were engaged for monitoring of food safety but imparts either practical limitations or analytical defects. Therefore, emerging of alternative methods like portable nanoprobes is of great importance. Over the last few years, sensor platforms have since been shifted towards wearable devices and being portable. Theses probes are movable, accessible, and tiny diagnostic devices which offer in-site analysis of biological contaminants in foods to guarantee their quality and safety. Among different types of portable sensors, low-cost, and flexible sensors as a novel development of portable consumer electronics are a great revolution in living sensors that can be exploited extensively in our daily lives. In this review, recent progress of lab-on-a-glove, leaf, and disc as groundbreaking electrochemical probes for food safety sensing will be discussed, importantly in the aspect of diverse contaminants encountered in food samples.

1. Introduction

The WHO (World Health Organization) strongly emphasized food safety as a global issue for providing innocuous and nontoxic food [1,2]. Food processing, preparation and storage as three significant and determinative steps of food safety standards, now broadly adopted in the worldwide food industry, such as HACCP, FSSC 22000, BRC and IFS [3]. A vast majority of food contaminations including heavy metals, pathogens, mycotoxins, pesticides, veterinary drugs, herbicides, and illegal additives can be analyzed by exploiting sensing platforms. Common conventional detection methods in food science such as HPLC (high-performance liquid chromatography), GC–MS (gas chromatography-mass spectrometer), MS (mass spectrometry) and ELISA (enzyme-linked immunosorbent assay) can provide high sensitivity and good selectivity sensing approaches for detection of various types of food contaminations [4,5]. On the other hand, traditional sensing platforms require skilled operators to implement samples and high-priced instruments, which is time-consuming and costly [6]. In addition, well-developed sensing technologies are commonly implemented in the lab, which cannot provide the essential requirement of on-site determination that requires portable sensors to gain the result immediately [7,8]. Hence, simple, fast, economic and portable analytical methods have attracted considered attention for the determination of various food contamination [9].

1.1. Electrochemical sensor

Sensors as an intelligent gadget that exploits specific biochemical

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reactions mediated to determine an analyte by measuring electrical, thermal or optical signals [10]. Different sensor technologies have been carried out based on sensing elements connected to a transducer to convert an observed response into a measurable signal. The level of signal is related to the concentration of an analyte target under investigation to which sensing element binds [11,12]. Among various types of sensors, invaluable advantages of electrochemical sensors including continuous real-time analysis, simplicity, high specificity and sensitivity, affordability, rapid response, relatively compact size and user-friendly operation have caused received numerous attention as an ideal sensing approach for food contaminations [13–15]. These types of analytical approaches may be categorized into label-free and label assays [16]. In addition, electrochemical sensors are broadly classified on the basis of the type of electrochemical electrode (including, GCE (glassy carbon electrode), Au (gold) electrode and SPCE (screen-printed carbon electrode)) used for detection in the field of food safety [17]. However, conventional sensing zones cannot carry out POC (point of care) testing in terms of connecting with heavy instruments [18,19]. Therefore, there is a necessity for portable electrochemical sensors able to POC detect food contaminations in the food samples and environmental resources without sample preparation [20]. Over the last few years, researchers all over the world have attempted to create an affordable and flexible sensing zone for the detection of various types of food contamination. According to the flexibility and stretchability of surface, glove and leaf have remarkable benefits for on-site real-time analysis of food contaminations. Secondly, LoD (lab-on-disc), LoG (lab-on-glove) and LoL (lab-on-leaf) have been exploited as newfangled sensing platforms in terms of affordability. Herein, in this overview, the recent advances of portable electrochemical sensors based on glove, disc and leaf for the detection of food contaminations have been discussed. Meantime, the benefits and restrictions of different kinds of novel and emergent recognition strategies to consider the most flexible, low-cost and sensitive platform for food contaminations were evaluated.

2. Novel portable electrochemical sensors

In today's world which has suffered from myriads of efficient and movable methods for assessment of food quality and contamination. Most portable electrochemical sensors are ideal for the analysis of liquid samples such as milk, water and fruit juices. Besides, detection of food contamination in solid samples require pretreatment steps, which are time-consuming and very complicated. However, LoG as a novel developed POC electrochemical detection platform can able to the determination of food contamination on the solid samples [21,22]. Integration of electrochemical detection with the disc as another economic substrate and novel microfluidic system has provided several advantages including the possibility to miniaturize the transducer (electrode) and alongside that automation and POC detection of food contamination without losing the performance [23,24].

2.1. Lab-on-a-disc

Over the last few years, LoD as a type of centrifugal microfluidic device has received considerable attention in the field of food safety in terms of both low-cost required instruments and high ability to combination with electrochemical techniques. In many cases, the transport and manipulation of liquids have been provided by centrifugal forces of rotating the device. Thus, these types of sensing approaches suggest an affordable technology instead of traditional pressure-driven microfluidic systems due to requiring minimal instrumentation for liquid transport and handling [25,26]. Secondly, integration of electrochemical system with LoD have developed of sample-to-answer POC detection method for analysis of various food contaminations. In addition, LoD based on electrochemical method offers benefits including the capability to miniaturize both instrumentation and the transducer (electrode) as well as automation and multiplexing of analysis of food safety [27]. For instance, Sanger et al. [28], fabricated an LoD device with eight electrodes printed on a plastic substrate for electrochemical detection of p-Coumaric acid which is a biomolecule produced by genetically modified strains of E-Coli bacteria. In this concept, microporous filter membranes (0.2 μm) were utilized in each section for simple sample filtration of eight sensing zone (5 min, 12 Hz). The filtration liquid is metered and then shift into the electrochemical detection units (detection chamber). Simultaneous electrical connections to eight sensing zones conduct of three-electrode arrays were achieved using a printed circuit board to the slip ring (Fig. 1A). Under the static condition, the developed electrochemical platform could be able to quantify p-Coumaric acid with acceptable linear range (125 μM to 3 mM) in Au pseudo-RE in 10 mM Ferri/Ferrocyanide using PBS (pH 7.4) as supporting electrolyte. In 2020, Rajendran and co-workers, [29] attempted to solve another important problems of LoD based on the installation of the electrode on the disc by reporting a novel and user-friendliness electrochemical sensor for the detection of ascorbic acid (Fig. 1B). The Challenge of connection between potentiostat and electrodes is another important matter in the creation of LoD based on the electrochemical system. Although slip rings were used for overcoming the minor difficulty of this type of portable sensor, the inherent noise is inescapable towards the movement of contact points during the rotation [30,31]. In the order to solve the noise of rotation, wirelessly powered potentiostat-on-a-disc was exploited in the structure of developed lab-on-a-chip (Fig. 1B). In the fabricated electrochemical sensor, screen-printed electrodes were successfully connected to a custom-made designed potentiostat for measuring the concentration of ascorbic acid with a detection limit of 32 μM. A combination of potentiostat-on-a-disc and LoD can offer an appropriate portable electrochemical device for the detection of food contamination. On the other hand, there is still a necessity for additionally simplification due to the complexity and cost of this hybrid sensing platform. It should be emphasized, a comparison of the result of LoD based on the electrochemical method which is summarized in Table 1 demonstrated that only a few approaches have been developed based on the integration of LoD and electrochemical techniques for detection of food contaminations.

Chief amongst the reasons to support the integration of the electrochemical system with LoD is that of its myriads advantages. This type of portable sensing platform has carried out several tests without losing performance. The appropriate stability of the disc can provide a reusable substrate for conducting food contamination detection. Thirdly, a common feature of LoD approaches is their capability to contain multiple, identical testing sites. On the other side of the argument, direct detection of food contaminations in real samples, which contain a complex sample matrix, may be challenging in electrochemical sensors based on the disc. The second disadvantage is that such potential interfering compounds would impact the performance of this type of sensing device. Furthermore, the possibility for surface fouling of the transducer is taken into consideration. In addition, LoD sensing approaches have suffered from low flexibility in many cases. Overall, there is a massive shortage in the application of this novel and portable electrochemical based on disc in the field of food safety. Detection protocols should be developed and optimized based on nanomaterials with antibodies and aptamer for enhancing the performance of LoD.

2.2. Lab-on-a-glove

Doubtless, the efficiency of the hand (as one of the most important parts of the body) is undeniable in our lives. This important part of the human body can turn to an intelligent sensing approach through its ability to execute complex operations by exploiting engineered gloves. Several benefits of electrochemical sensors including cost-efficiently, simplicity with high accuracy and alongside that demonstrate high sensitivity have caused introduced a functional and attractive analytical performance method. Besides, the integration of smartphones with gloves-based electrochemical sensors have acted as an intelligent gadget

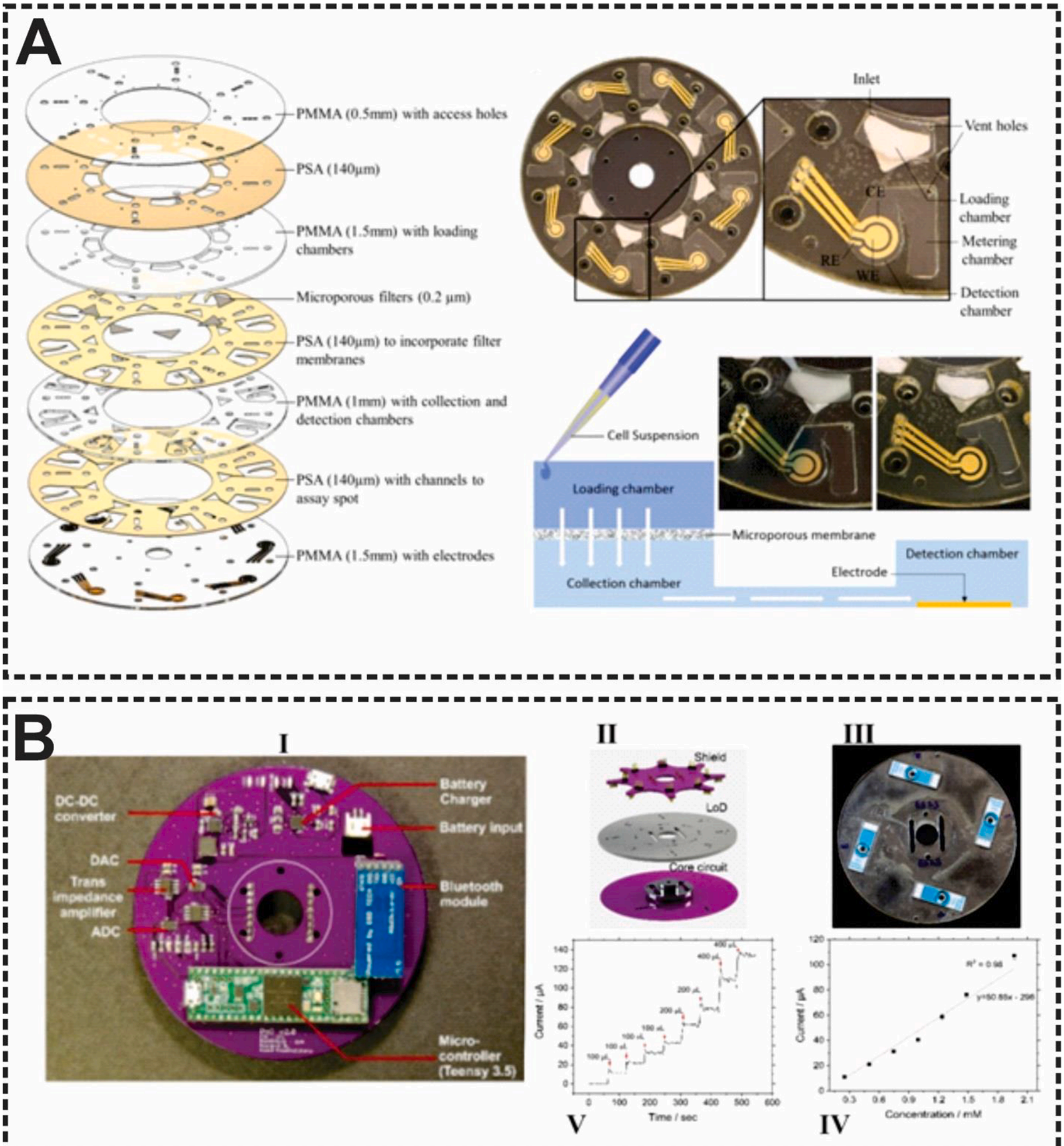

Fig. 1. **(A)** Photograph of the microfluidic disc based on three-electrode system on the disc [28]. **(B) I-III)** Schematic of potentiostat-on-a-disc for determination of ascorbic acid **V-IV)** Electrochemical response of various concentration of ascorbic acid and related calibration curve [29].

that can provide an efficient source of the signal process (voltage and potential) and monitoring of electrochemical curves [32,33]. In 2017, Mishra and colleagues [34], connected glove (as a green substrate) with smartphone device (as intelligent monitoring device) by the compact electronic interface for creation of a unique and portable electrochemical biosensor. To elaborate, the stress-enduring ink was utilized to conduct a three-electrode system on the surface of the index finger (detection finger) by stencil for in-site and fingertip determination of organophosphate compounds concentration on the surface of fruits and vegetables (Fig. 2A). The organophosphorus hydrolase enzyme was immobilized on the surface of the sensing zone by the CV technique for increasing the selectivity and sensitivity of the developed biodevice. Mishra's group proved that stretching and bending of the developed electrodes do not meaningfully compromise the performance of the reported glove-based biosensor. Furthermore, the innovative transduction mechanisms for applications in food samples was introduced. In another same application of glove in sensing platform, Farshchi and co-workers [35], fabricated a green platform based on Ag ink on the surface of the glove for quantification of trifluralin residue on the surface of various substrates (Fig. 2B). For this purpose, the synthesized metallic based ink (Ag nano-ink) was drawn on the index finger of the glove to prepare a three-electrode system (reference, counter, and working electrode). The developed LoG could be able to successfully detect concentration of trifluralin residue on the solid surface of tomato and mulberry leaves with linear range and LOD of 0.01 μM to 1 mM and 0.01 μM, respectively. The most pivotal argument in favor of Farshchi's reported sensor is that of inundated high sensitivity and selectivity they have in comparison to the same glove-based electrochemical sensors. Based on the fact that excellent stability and conductivity of Ag nanoparticles in the structure of portable electrochemical sensors can be able to significantly

Table 1
Figures-of-merit of the reported novel electrochemical portable sensors for efficiently quantification of food contaminations.

Strategy	Detection technique	Printed method	Sensing platform nanomaterials	Type of food contamination	LOD or LLOQ	Linear range	Samples	Ref
LoL	DPV, CV and SWV	Direct writing	Ag and GQDs	Trifluralin	0.005 mM	0.005 to 0.04 mM	Apple skin	[40]
LoL	DPV	Direct writing	MWCNTs, silica and chitosan	Organophosphorus pesticide residuals	0.02 mM	–	Sweet potato leaf, chinese cabbage and cucumber	[41]
LoG	SWV	Stencil	Carbon and Ag	Organophosphorus chemical threats	–	–	Apples, grapes, green peppers, tomatoes, wood, stainless steel, plastic, and glass	[34]
LoG	DPV and SWV	Direct writing	Ag nano particles	Trifluralin	0.01 μM	0.01 μM to 1 mM	Standard, mulberry leaves and tomato	[35]
LoG	ChA	Direct writing	Au@Ag coreshell/GQDs	Trifluralin	10 nM	10 nM to 1 mM	Standard, leaf and apple	[36]
LoG	DPV and SWV	Photo-emulsion printing screen	Carbon based nanomaterials	Diuron (phenylamide), carbendazim (carbamate), fenitrothion (organophosphate) and paraquat (bipyridinium)	9.2×10^{-7}, 4.7×10^{-8}, 6.4×10^{-7} and 2.4×10^{-8}		Standard, apple and cabbage	[37]
LoG	SWV		Carbon and Ag	Pyocyanin and pyoverdine	3.33 nM and 1.66 μM	0.01 to 0.1 μM and 5 to 50 μM	Furniture, medical scrapper and sink	[38]
LoD	SWV	Stencil or shadow mask	Gold	p-Coumaric acid	–	125 μM to 3 mM	Standard and clinical samples	[28]
LoD	Amperometric	–	–	Ascorbic acid	32 μM	–	–	[29]
LoD	CV	Stencil	Gold	p-Coumaric acid	–	–	–	[42]
LoD	SWV	Stencil-lithography and electron beam evaporation	Gold	p-Coumaric acid	3 μL	–	Standard and clinical samples	[43]

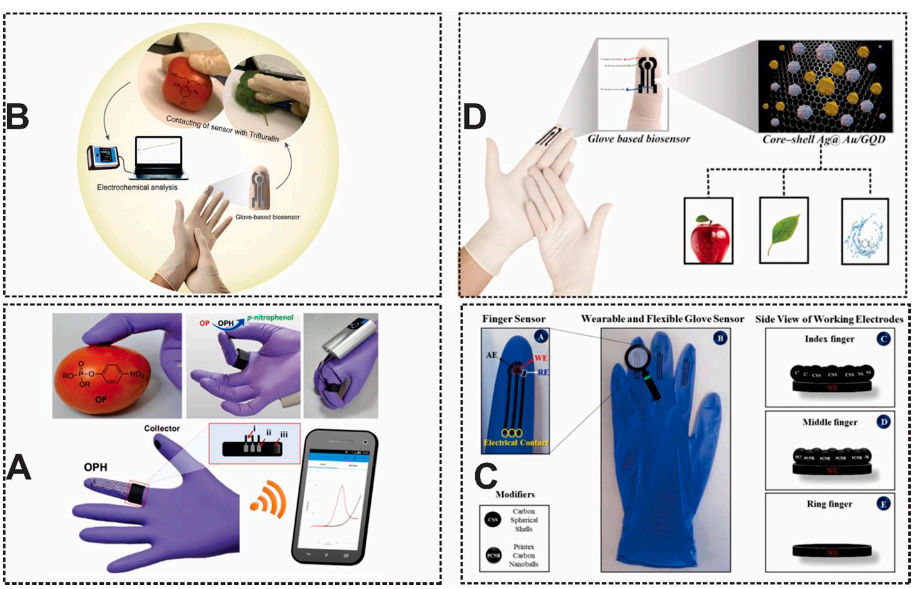

Fig. 2. **(A)** Components schematic of organophosphorus chemical threats detection by glove-based electrochemical sensor [34]. **(B)** Illustration of trifluralin residue determination on the surface of different substrates by using Ag nano-ink for construction of sensitive and selective LoG [35]. **(C)** Representation of glove-based electrochemical sensor based on Au@Ag coreshell/GQDs for measuring concentration of trifluralin residue [36]. **(D)** Schematic of electrochemical sensor based on glove for determination of four classes of pesticides including diuron (phenylamide), carbendazim (carbamate), fenitrothion (organophosphate) and paraquat (bipyridinium) [37].

increase the electron transfer. In this work, the shortage of innovative transduction mechanisms for wireless transmission of data to the electrochemical device is tangible. Most recently, Mahmoudpour et al. [36], designed another glove-based electrochemical sensor by exploiting hybrid ink (Au@Ag coreshell/GQDs (gold@silver-modified graphene quantum dots)) for detection of trifluralin residue (Fig. 2C). The presence of a target on the surface of an agriculture product has caused the difference of electrochemical signal in three-electrode glove-based sensor on the surface of a rubber glove (detection finger) which was measured by ChA (chronoamperometry) technique. Carbon and metallic based nanomaterials nano-ink (as hybrid ink) have demonstrated phenomenal sensitivity in the structure of LoG. Another interesting example of glove-based electrochemical sensor application for on-site pesticide determination in food is reported by Raymundo-Pereira [37]. Contrary to previous reports, Raymundo-Pereira's groups utilized three glove-embedded sensors printed on three fingers of a rubber glove for providing a portable and non-enzymatic electrochemical sensing platform of various pesticides including carbamate (carbendazim), phenylamide (diuron), bipyridinium (paraquat) and organophosphate (fenitrothion). The index finger, middle finger and ring finger were functionalized with carbon spherical shells, Printex carbon nanoballs and 0.1 mol L − 1 sulfuric acid solution, respectively. The reported three glove-implanted sensors printed on three fingers introduced multiplex analysis of pesticides for the first time. In addition, it could be able to the measured concentration of different pesticides in solid and liquid samples by touching and immersing (apples, cabbage and orange juice) with recoveries varied between 90 and 110%, which revealed that fabricated LoG sensor is effective and selective to detect various pesticides in the real matrix. The illustration of developed LoG (design and working principle) was demonstrated in Fig. 2D. Thereby, electrochemical glove-based sensors can be able to analysis food contaminations on different surfaces. Interestingly, Ciui et al. [38], developed carbon-based and Ag/AgCl ink on the substrate of glove for pathogens quantification by pyocyanin and pyoverdine (as two known virulence factors of the bacteria Pseudomonas aeruginosa). The reported portable sensor could be able to indirect detect Pseudomonas aeruginosa in under four minutes by measuring the concentration of pyocyanin and pyoverdine with LOD of 3.33 nM and 1.66 μM, respectively.

Dissection of developed electrochemical glove-based sensors demonstrates that all of them have been carried out based on a three-electrode system and removing the use of the thumb finger to complete the electrochemical circuitry in most of them. The analytical performances and detailed outline of most of the portable systems based on the glove for food contaminations detection are summarized in Table 1. This type of novel electrode provided a handily, affordable and green platform for in-situ testing of different types of pesticide residues on the solid and liquid samples. On the other hand, there is a massive shortage in construction LoG for detection of other types of food contaminations on the surface of different types of foods such as vegetables and fruits. All in all, LoG can be the window of hope in wearable sensor applications in the field of food safety and alongside the development of chemical sensing robotic skin.

2.3. *Lab-on-leaf*

Electrochemical sensors based on the leaf as an environmentally friendly sensing platform have been designed on the surface of agricultural products by using conductive inks for the determination of food contaminations [39]. The high wicking capability of the leaf is a beneficial feature since it enhances the adhesion of the conductive ink and alongside that prepares solution flowing on the surface of the leaf without require for external pumping. For instance, Saadati et al.[40], developed a novel LoL based on the exploitation of specific nano-ink (Ag-citrate/GQDs (silver-citrate/graphene quantum dot)) on the flexible substrate for detection of trifluralin. For this purpose, nano-ink was drawn on the surface of the leaf by direct writing method for creating a three-electrode platform (Fig. 3A). In this context, all three electrodes were developed based on Ag-citrate/GQDs. The carbon-based nanomaterials (GQDs) and noble metal materials (Ag) in the structure of nano-ink could provide an appropriate sensing zone with excellent conductivity and high surface area for the efficient capture of analytes. Under optimized experimental conditions, DPV, CV and SWV techniques could successfully measure the concentration of trifluralin with linear range and LLOQ of 0.005–0.04 mM and 0.005 mM, respectively. In another similar application of ink on the leaf was exploited two-electrode electrochemical analysis system [41]. In this concept, MWCNTs (multi-walled carbon nanotubes) and chitosan as nanomaterial ink and as a stabilizer was used in the structure of a synthesized nano-ink, respectively. The designed conductive and carbon-based ink was drawn on the surface of the leaf (as a working electrode) by direct writing technique (writing by Chinese brush) for on-site direct detection of parathion residuals from vegetable products via painted electrode (Fig. 3B). Besides, the reference/counter electrode was made from silver nanowires ink. The reported sensing device could be able to detect a target with a detection limit of 20 ng mL^{-1}. Furthermore, the recovery of target analyte in real samples varied between 76.0% and 96.2%, validating the potential of this developed LoL for the determination of parathion in real agricultural products. Comparison of the developed platform with the same composition of the three-electrode platform revealed that the current response of the two-electrode electrochemical analysis system is almost 4 times lower.

Perhaps the most formidable argument in favor of portable electrochemical sensors based on the leaf is that it advantages. The charge-free of this surface is undeniable. A variety of plants in our environment can provide an inexhaustible source of substrate for developing this type of green electrochemical sensor. On contrary, if this type of sensing approach will be growing rapidly, the detriments that it imposes on the natural is incontrovertible. Second, in terms of importance is no complication of the printed process due to the exploitation of direct writing on this type of substrate. Annually, billions of dollars are invested in developing an appropriate printed technique including a microfluidic dispensing system, inkjet printing and screen-printing. Hence, there is a massive shortage in use this type of portable analytical device for determination of sever types of food contaminations. It is worth noting that although LoL has acceptable flexibility, the instability in the high temperature, bending and stretching is the most limitation of this type of sensing device.

3. Conclusion and future prospective

Several disadvantages and limitations of conventional electrochemical methods for in-site detection of food contaminations have caused the effect of the loss of analytes and test results. Although there are several high technology and experience mobile sensing platforms

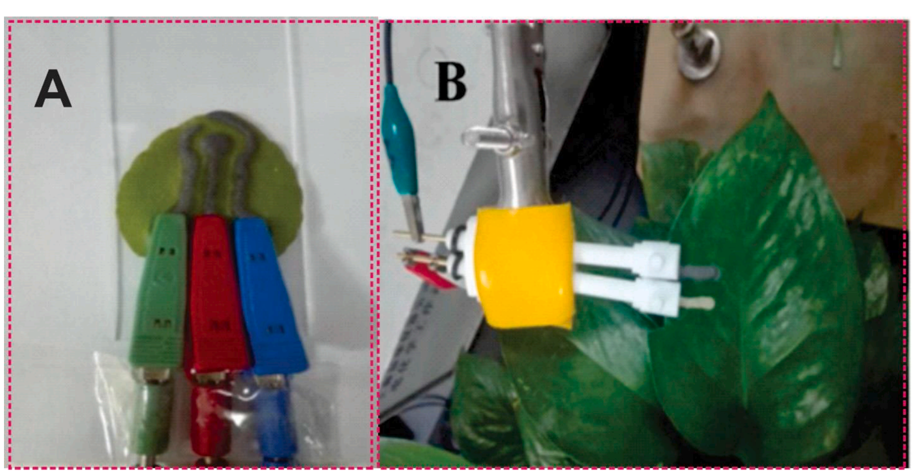

Fig. 3. **(A)** Representation of portable electrochemical sensor based on three-electrode platform on the leaf [40] **(B)** Image of two-electrode electrochemical analysis system for LoL [41].

based on different electrochemical electrode for food safety such as laser-fabricated graphitic carbon electrodes, SPE (screen-printed electrode) and ITO (Indium tin oxide) electrode, integration of electrochemical system with disc, glove and leaf (as an affordable and portable substrates) have created powerful laboratory approaches for low-cost and green lab-on-a-chip devices. In addition, electrochemical performance of the non-toxic, economic, green and flexible electrodes in safety and quality control of food requires further enhancement.

Over the last decade, most of the development of portable sensors including lab-on-chip, wearable sensors and traditional microfluidic systems so far focus on healthcare and in compound with a lack of diversity in the important food safety reported. Although a few electrochemical systems have been developed on the surface of leaf as sustainable and green POC detection of food contaminations, there is a great shortage on the application of several ink and bioreceptors for the detection of various types of food contamination by high performance. LoG as another novel type of portable electrochemical sensing platform has demonstrated great robustness for on-glove sampling/sensing operation due to the high resistance of glove based sensor from mechanical stress (bending and stretching). Although this type of mobile sensing approach is in the early development stages, it suggests noticeable opportunities for the determination of food contaminations in solid samples. It should be noted, the sensing zone of this mobile sensing approach was prone to physical damage, for which there is direct contact with the solid surface of the food. Despite the development of a few eco-friendly sensing platforms (LoG and LoL) for detection of food contamination, direct determination of target analytes in a complex sample matrix can be challenging due to interfering compounds and the possibility for substrate fouling of the transducer. Subsequently, many researchers have introduced LoD as another in-site detection platform for food safety. Although disc-based electrochemical approaches have several advantages such as low cost and reusability, the flexibility and toxicity of these types of novel electrochemical sensors should be taken into account.

We hope this review can burden our understanding of user-friendly and low-cost techniques based on electrochemical in the field of food safety. Last but not least, a revolutionary era in green, flexible and low-cost electrochemical sensor will go a long way to support the possibility of such portable sensing approaches and is anticipated to encourage expert researchers in the field of portable electrochemical sensors in this direction.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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