Review

Electrochemical Sensors for Bisphenol A Analysis in Foods and Beverages – a New Approach in Food Quality Control

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Abstract

Modern nanofabrication technologies combined with electrochemical techniques offer benefits in terms of extremely high sensitivity, low limit of detection, minimum power requirement, simplicity, and low cost of the electrochemical sensors making them powerful for food quality evaluation. The limited number of electrochemical non-enzymatic sensing platforms for bisphenol A (BPA) successfully applied for safety assessment of foods and beverages, testifies that the food matrices present significant challenges and there is still a need to improve the analytical performances of these devices. This review systematically explores the contributions of diverse functional nanomaterials, metal–organic frameworks, ionic liquids, and molecular imprinting in improving the sensor performance. A critical discussion on the latest interesting innovations and most promising electrochemical tools for BPA analysis is presented. By addressing the electrochemical aspects and challenges in the design of BPA sensors and exploring innovative solutions, the article offers insights into future prospects and avenues, paving the way for advancements in this field.

Keywords: Bisphenol A, food analysis, food safety, electrochemical sensors, endocrine disrupting compound

1. Application and Toxicity of BPA

Bisphenol A (BPA) [4,4'-dihydroxy-2,2-diphenylpropane, CAS 80-05-7] was first reported by Aleksandr Dianin in 1891. In the mid-twentieth century it was observed that BPA could be polymerized to make polycarbonate plastic – low cost product that is lightweight, transparent, colorable, resistant to heat and chemicals, and easy to mold and thermoform.[1] Since then, BPA has been abundantly used as an integral raw material in the manufacture of polymers (polycarbonate, polysulfone, polyacrylate), and epoxy resins. Some of the applications of polycarbonate include reusable plastic bottles, baby feeding bottles, plates, cups, microwave ovenware, food storage containers. Epoxy resins are used for internal protective layer of metal cans and internal coating on metal lids for food in glass jars.[2] The coating on the inside of the can is used to protect the metal from corrosion and protect the food from metal contamination during sterilization and storage.[3,4] Various polymers used in dental sealants and tooth coatings also contain BPA.[5] There is evidence for leakage of BPA from some resin-based dental materials. Although there is a lack of studies analyzing the association between BPA exposure from dental materials and its adverse effects on human health, the potential effects of BPA release from dental material could not be negligible due to its very long-term exposure.

Human exposure does occur when BPA migrates from plastic bottles, plastic-lined food and beverage cans into the contents even under normal conditions of use due to incomplete polymerization.[6] Long-term storage or thermal treatment could fast the migration of BPA monomer residues from the above materials into foods.[7] Also, BPA leaching occurs when plastic and epoxy resin-containing bottles and cans are repeatedly washed. Harsh detergents cause BPA to leach into the food and beverages due to the changes in pH (alkaline conditions), which can hydrolyze the ester bond linking BPA monomers.[8] Therefore, the presence of BPA in foodstuffs could be attributed to the migration of residual BPA monomers after manufacturing (physical migration) or hydrolysis of the polycarbonate (chemical migration). The effect of migration of BPA from polycarbonate (PC) drinking bottles was illustrated in an intervention study where volunteers were requested to consume all cold beverages from PC drinking bottles during one week. An increase of 69% in urinary BPA concentrations was observed after one week compared with urinary levels obtained after a wash-out period of one week, where no use of PC bottles was allowed.[9]

BPA is considered an endocrine disrupting compound (EDC), capable of interacting with various biological receptors and affecting normal hormone signal pathway transduction.[10–15] Numerous studies have investigated the reproductive toxicity of BPA, and extensive reviews were conducted to address the strength of the evidence regarding BPA toxicity.[16–18] The phenolic structure of BPA has close relationship with endocrine hormones, such as estradiol and diethylstilbestrol, and causes linking with estrogen receptors.[19] Long-term BPA exposure has been associated with reproductive disorders including the incidence of infertility, sexual dysfunction, prostate and breast cancer, increased birth defects, and genital tract abnormalities.[8,17,20–21]

Exposure of women to BPA interferes with the reproductive system. BPA binds to estrogen receptors and causes irreversible alteration to the hypothalamic-pituitary-ovarian axis. BPA will provoke estrogen receptors and thus increase the chances of *polycystic ovary syndrome* (PCOS), endometriosis, and infertility.[23]

Epidemiological studies have provided results indicating that BPA also alters male reproductive function. Data revealed that men occupationally exposed to BPA had high blood/urinary BPA levels, abnormal semen parameters, reduced libido and erectile-ejaculatory difficulties.[24,25] BPA causes atrophy in the testis, apoptosis in Leydig cells and germ cells, and reduction in testosterone biosynthesis, which will either cause sperm quality and quantity alterations, retardation of testicular development, reduction in sperm motility, and infertility.[23] A literature review, published in 2023, summarises the existing information on the effects of BPA on human male infertility using the most recently published literature.[26] Moreover, the overall BPA effects on male reproduction appear to be more harmful if exposure occurs *in utero*. Clinical evidence suggests that developing fetus and neonates are most vulnerable to endocrine disruption. BPA is transported across the human placenta,[27] thus the fetus absorbs the chemical from maternal blood plasma (after maternal exposure). BPA has been detected in the urine and serum of pregnant women, as well as the plasma, serum, and placenta of newborn infants.[28]

BPA interferes with the function of thyroid systems and affects both immune and central nervous systems. The number of publications in the field of BPA and thyroid hormones has increased tremendously since 2000.[29] The impact of BPA on thyroid hormones, especially pregnant women and children, was the latest research frontiers. The potential negative effect of BPA on the developing thyroid gland of children may affect proper neurodevelopment, suggesting the need to focus future research on designing

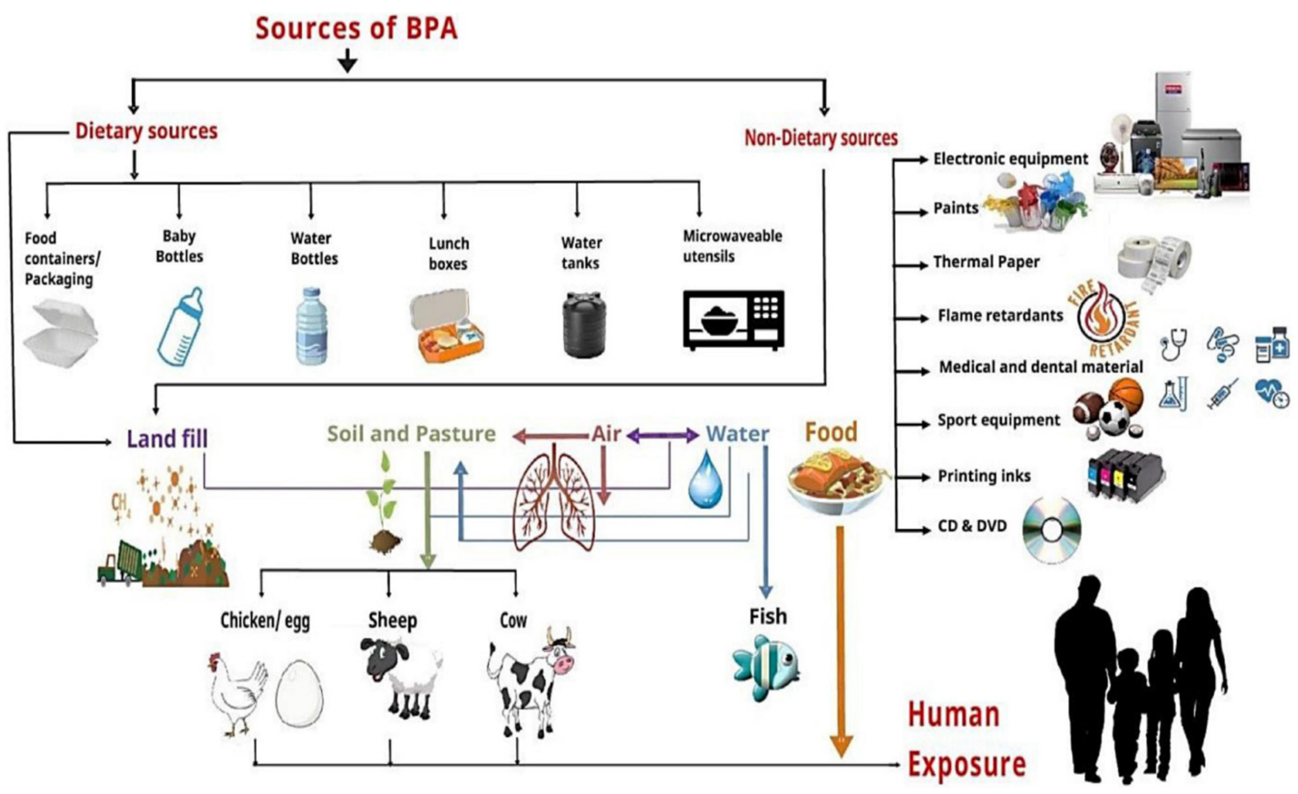

Figure 1. Human exposure to BPA *via* different sources and exposure routes. Dietary and non-dietary sources contaminate landfill, soil, air, water, and food that directly or indirectly affect the human through different exposure routes. Reproduced from reference[23]; permissions under Creative Commons Attribution License (CC BY). Copyright 2022 Manzoor, Tariq, Fatima, Sahar, Tariq, Munir, Khan, Nawaz Ranjha, Sameen, Zeng, and Ibrahim.

studies that elucidate the underlying mechanisms and the effects of BPA in thyroid function in early life.[30]

The first systematic review that investigated the relationship between BPA exposure, both prenatally and in childhood, and behavior in children up to 12 years of age, was published in 2017.[31] Descriptive analyses indicated that exposure to BPA was associated to higher levels of anxiety, depression, aggression, and hyperactivity in children.

Hafezi and Abdel-Rahman, in 2019 highlighted the findings linking BPA to cancer with a focus on the molecular mechanisms of this action and the emerging role of BPA in response to cytotoxic therapy.[32] They concluded that BPA induces resistance to various chemotherapeutics such as doxorubicin, cisplatin, and vinblastine *in vitro*. The development of chemoresistance to available therapeutics is an emerging significant aspect of BPA toxicity because it worsens the prognosis of tumors, including ovary, breast, prostate, and colon cancer. A review article, published in the same year, provides comprehensive data of BPA toxicity on human health and related mechanisms.[33]

Figure 1 illustrates human's exposure to BPA *via* different sources and exposure routes. Dietary sources (food containers/packaging, baby bottles, water bottles, lunch boxes, water tanks, and microwave utensils) and non-dietary sources (electronic equipment, paints, thermal paper, flame retardants, medical and dental materials, sports equipment, printing inks, and DVDs) directly or indirectly affect the human through different exposure routes.[23]

2. Legislations

BPA is authorised for use as a monomer in plastic food contact materials, in accordance with Commission Regulation (EU) No 10/2011/EU on plastic materials and articles intended to come into contact with foodstuffs.[34] The former specific migration limit (SML) of 3 mg/kg was reduced to 0.6 mg/kg in 2002 and then further reduced to 0.05 mg/kg following European Food Safety Authority opinion of 2015.[35] Commission Implementing Regulations (EU) No 321/2011[36] and (EU) No 2018/213[37] place restrictions on the use of BPA, respectively, in the manufacture of PC infant feeding bottles and in varnishes and coatings intended to come into contact with food and amending Regulation (EU) No 10/2011[34] as regards the use of that substance in plastic food contact materials. Additionally, usage of BPA in baby bottles has been banned in many countries like U.S.A,. Canada and China. In 2010, the Canadian Government forbade the commercialization of polycarbonate baby bottles containing BPA.[38] Two years later, the Food and Drug Administration (FDA) banned the use of BPA in baby bottles and also in children's drinking cups.[39] On May 23, 2011, the Ministry of Health of The People's Republic of China announced the ban of BPA in infant feeding bottles.[40]

A tolerable daily intake (TDI) should ensure that life-time exposure up to the TDI does not lead to appreciable adverse health effects in the general population (TDI = tolerable amount of an active ingredient that is considered safe for human health in case of lifelong daily intake). In 2015, as a respond to a refined risk assessment of BPA and its unwanted health effects, EFSA decreased the TDI value from 50 μg/kg body weight (BW) per day to 4 μg/kg BW/day.[41] In 2023, EFSA has re-evaluated the risk assessment for BPA and published a draft of a new scientific opinion on 19.04.2023, which sets the TDI value of 0.2 ng/kg BW/day.[42]

3. Conventional Analytical Methods for BPA Determination in Foodstuffs

The most predominant target for BPA exposure in human is via gastrointestinal tract, followed by respiratory and dermal tract.[33] For general population, the amount of BPA from dietary route accounts for more than 90% of total BPA.[43] Hence, canned foods and bottled drinking water are the main sources of BPA exposure. Various factors such as contact time, temperature, solar irradiation, manufacturing process or specific characteristics of the food (pH, fat content) have a direct impact on the migration of BPA into food.[4,23] For this reason, in order to protect human health, it is extremely important to analyze the concentrations of BPA in foodstuffs and water.

Currently, the chromatography methods such as liquid chromatography and gas chromatography, coupled with another detection technique, are the most employed for the detection of BPA in water and foodstuff samples. Martín-Pozo et al. have provided a comprehensive coverage of the principal methods for the determination of BPA in breast milk, food, and beverage samples by gas chromatography–mass spectrometry (GC-MS), and liquid chromatography coupled to tandem mass spectrometry (LC-MS/MS).[44] Authors summarize the particular methodologies developed for the analysis of these complex matrices, with an emphasis on the sample treatment. The different strategies used for the collection, elimination of interferences, extraction, and cleanup of the samples were discussed in detail. Generally, such techniques provide an excellent performance with high sensitivity and reproducibility for the determination of BPA. Nevertheless, these detection strategies are time-consuming and require complicated and expensive instruments, complex sample pretreatment steps, trained personnel, and high operational requirements, which may hinder real-time and rapid analysis of BPA. For these reasons, fast and convenient analytical methods that can realize direct and reliable quantitative detection of BPA in complex samples are urgently needed for controlling food quality and ensuring the consumers' health safety. The newly developed sensing methods should be easy to integrate with portable, hand-held devices, thus enabling on-site sample analysis.

4. An Overview on Emerging Materials and Technologies for Electrochemical BPA Analysis in Foods, Beverages, and Bottled Water

Recently, electrochemical sensors used for the quantitative determination of food ingredients, additives, chemical contaminants, and foodborne pathogens proved to be a great alternative to classical methods.[45,46] Generally, the electroanalytical methods have demonstrated advantages over conventional analytical methods: operational simplicity, cost effectiveness, high sensitivity and selectivity, rapid sample preparation procedures without complicated treatment, short analysis time, remarkable accuracy, low power requirements, high level of automation, and feasibility for an eco- and user-friendly assay.[47] In most cases the procedure requires only appropriate sample dilution followed by immediate analysis.[48–51] Moreover, the electrochemical signal is not affected by ambient illumination conditions and the color or turbidity of the sample. This is an extra advantage in the food control field, where food/beverage extracts often remain colored or exhibit a certain level of turbidity, which can interfere with optical measurements.[52] The coupling of exclusive principles of electroanalysis with the enormous possibilities of nanosized materials and natural materials (zeolites, clay minerals, chitosan, paper, biowastes, and non-toxic solvents including ILs) has led to powerful and environmentally friendly sensing systems.[53]

Due to the ability to easily integrate electrodes in microchips, electrochemical sensor is actually easy to be miniaturized into a hand-held device without compromising the analytical behavior. Thus, the electrochemical sensors have the potential to be miniaturized and compact portable devices and fabrication becomes easy.

Most of the advanced electrochemical sensors are equipped with the technology allowing them to be used as a part of the wireless network. Recently, there are significant developments in the integration of smartphone technology with electrochemical sensing platforms for real-time monitoring. Miniaturized electrochemical analyzers can be connected to smartphones for powering, processing, data analysis, and visualization (Figure 2). Recent advances in nanotechnology, surface modification, microfabrication, and signal processing have paved the way for the design of reliable microsensor devices.[54]

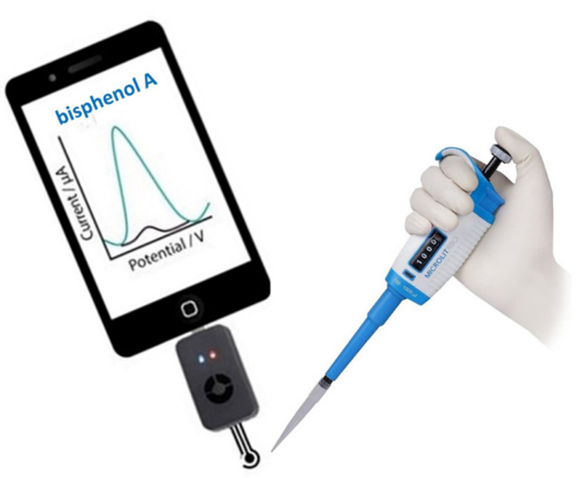

Figure 2. Portable sensing platform using all-in-one screen-printed electrode, hand-held commercial potentiostat, and smartphone.

Scientific community has continually investigated new sensing materials for BPA at lab level which show high sensitivity, fast response, and good reproducibility. Figure 3 shows the trend of publications in the recent years targeting electrochemical sensors for BPA. The data for this study was retrieved in March 2025 from the Scopus database using the keywords “bisphenol A” and “electrochemical sensor” and discovered a total of 445 papers dedicated to the study of electroanalysis of BPA from 2000 to 2025. The dataset suggests a rising interest in the development of novel electrochemical sensing platforms for BPA.

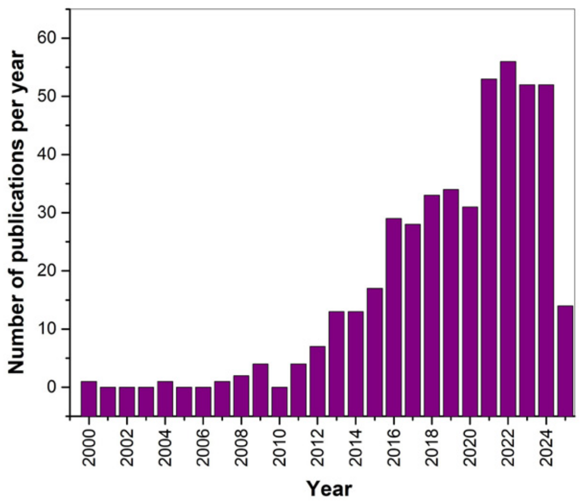

Figure 3. The number of papers published on BPA electrochemical sensors in the past 25 years. Data were retrieved from Scopus using the search terms “Electrochemical sensors” and “Bisphenol A” as search topics. Search date: 13 March 2025.

This paper purposefully delves into the fundamental electrochemical aspects, sensor designs, operational parameters, advantages, and drawbacks of these technologies, forecasting their potential applicability on a practical scale. With an emphasis on efficiency and selectivity, the manuscript contributes valuable insights to the design of electrocatalytical materials and sensing platforms, offering a glimpse into the promising future of sustainable electroanalysis. Thus, the review critically assesses the options for implementing these technologies in food control.

4. 1. Electrooxidation of BPA

Electrochemical detection of BPA is based on the well-known electroactivity of the phenolic groups present in the molecule.[55,56] The electrooxidation of BPA involves two electrons and two protons (Figure 4).

transfer rate, and improved peak separation ability between BPA and other interfering molecules in food matrices. Moreover, using SPEs allows for the miniaturizing of electrochemical sensors, making them highly portable.

The next parts of the paper provide an overview on how the advances in modern electroanalytical technology have contributed towards developing precise and easy-to-handle sensor devices for quantitative detection of BPA in food samples.

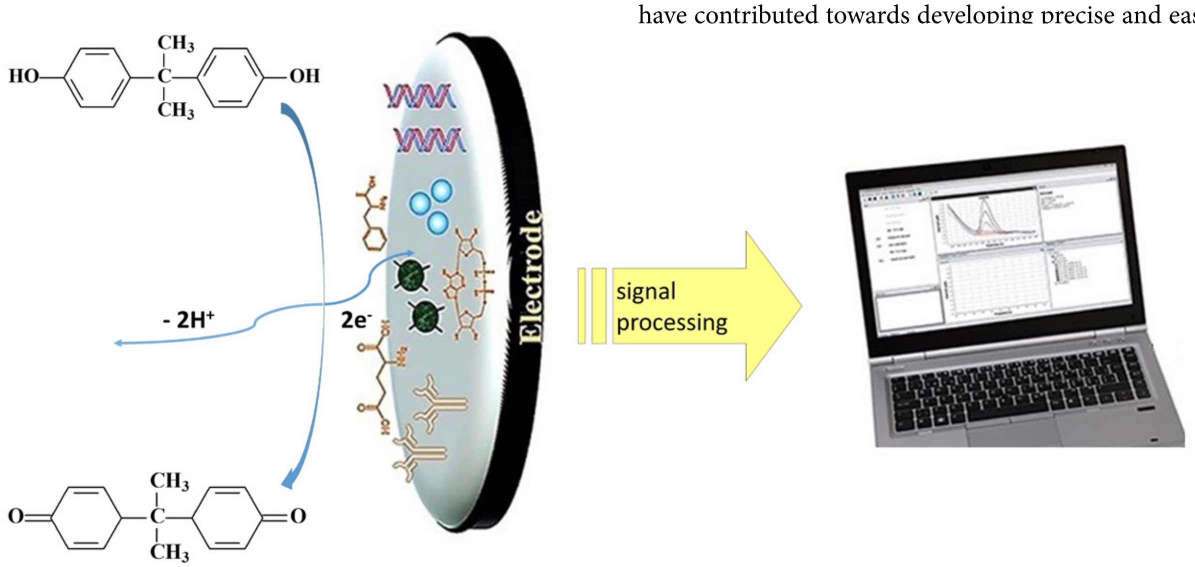

Figure 4. Schematic representation of the electrooxidation reaction of BPA via 2e⁻ and 2H⁺ pathway.

The sensitivity and accuracy of non-enzymatic electrochemical detection are closely related to the electrode materials. In the field of electroanalysis, the great potential of carbon-based electrodes as sensing platforms is exciting due to their unique properties, such as high electrical conductivity, chemical stability, and robust mechanical strength. However, the direct electrochemical oxidation of BPA on carbon electrodes suffers from slow kinetic and high potential requirement. Additionally, the oxidation of BPA molecule is an irreversible process and the products of its oxidation cause the surface fouling of the electrodes.[55] This is a major problem occurring during the electrooxidation of phenols due to the electropolymerization of phenolic compounds. The formation of polymeric product after BPA oxidation blocks the active surface area of the electrodes and delays important electrode processes.

In order to improve the electrode selectivity and sensitivity, as well as to inhibit the electrode surface fouling phenomena, surface modification of the working electrode has been extensively investigated. Literature survey reveals that common electrodes, such as glassy carbon electrodes (GCEs), carbon paste electrodes (CPEs), and screen-printed electrodes (SPEs), after appropriate modification by different electroactive materials have the ability to effectively detect BPA at low potentials. The modification of carbon-based electrodes offers sensing platforms with low limit of detection (LOD), wide linear range and remarkable selectivity, mostly attributed to the enhanced electron

4. 2. Nanomaterials-based Electrochemical Sensors

Nowadays, the combination of nanotechnology with modern electrochemical techniques offers new horizons for the application of nanomaterials in electrochemical analysis. The synthesis and application of various nanosized materials have contributed significantly to the design of reliable electrochemical sensor devices. Nanomaterials can serve as electrode modifiers or signal tags, greatly amplifying the signal. A huge number of nanostructured sensors have been realized exploiting different types of nanomaterials, including graphene (GR), graphene oxide (GO), carbon nanotubes (CNTs), metal/metal oxide nanoparticles (NPs), nanosized composites, etc. Additionally, the combination of two or more different electrocatalytic nanomaterials improves the sensitivity and selectivity of electrochemical sensors, making the assay results more satisfactory.

Research teams have reported nanoparticles-based powerful electrical devices for efficient determination of diverse target analytes with advantages such as increased specific surface area, high electrical conductivity, effective catalysis, and higher stability.[56–62] Metallic and metallic oxide nanostructured materials have attracted a considerable attention due to their enhanced surface-to-volume ratio, excellent electrocatalytic activity, electrical and thermal conductivity properties, which are highly relevant to applications in catalysis and electronic devices.

Lei et al. reported a portable and fully integrated electrochemical sensing device (lab-on-injector) that reduces the sampling pretreatment and detection to a simple on-step operation, making *in situ* detection of BPA in real samples much easier and more feasible.[63] Extra-small **Au nanodots** (1.6–2.0 nm) confined in **carbon nanofibers** (AuNDs@CNFs) were used to modify screen printed electrodes' surface in order to enhance detection performance of the sensor. Using this AuNDs@CNFs based lab-on-injector, BPA in commercial beverages (apple juice, coke, orange juice, and tomato juice) was detected directly with recoveries from 93.14% to 94.25%. The work provides a new platform for *in situ* detection of BPA in foodstuffs and holds a great promise for the development of smart devices in electroanalysis.

Recently, researchers have attempted to use cost-effective **non-precious metals** as an alternative to noble metals to develop reliable electrochemical sensors. Malakootian et al. designed a $FeNi_3/CuS/BiOCl$ nanocomposite that was used to modify CPE for BPA determination.[64] The sensor demonstrated high selectivity and sensitivity, which enable it to be used for the quantification of BPA in canned food samples like tuna fish, tomato paste, apple, and corn.

Electrochemical platform based on GCE, modified with **graphitic carbon nitride** (g-C_3N_4), combined with differential pulse voltammetry (DPV) technique, exhibited a linear current response in the concentration range 0.01 to 75.0 μM and LOD of 9.0 nM.[65] The good analytical performance of the sensor was confirmed by determining BPA in different real samples (drinking water, milk, and orange juice) with satisfactory recovery values (95.6–104%). In many respects, this work demonstrates that simplicity can be a good thing in electrode design for electroanalytical sensing devices.

Liu et al. designed an electrochemical BPA sensor based on GCE decorated with **$CoFe_2O_4$ nanoparticles** which were synthesized by a sol-gel combustion method.[66] A linear calibration curve range from 0.05 to 10 μM and a LOD of 3.6 nM were obtained. The sensor was successfully utilized to determine BPA in milk samples. Co_3O_4-CeO_2 composites prepared by the same method were also used as electrode modifier to construct sensitive and selective electrode for the analysis of BPA.[67]

Magnetite (Fe_3O_4) NPs have the apparent ability to increase electrode conductivity and facilitate electron transfer. Hou et al. developed a simple, relatively cheap platform with superior sensitivity (LOD = 0.031 nM) based on carbon black (CB) supporting magnetite nanoparticles-modified GCE.[68] It has been demonstrated that the resulting Fe_3O_4NPs-CB nanocomposite exhibits a high catalytic activity towards the electrooxidation of BPA and contributes to a substantial improvement in the analytical properties of the sensors, including stability and repeatability. After each measurement, the Fe_3O_4NPs-CB/GCE underwent 10 successive CV sweeps from 0.0 to 1.0 V in the blank supporting electrolyte 0.1 M PBS (pH 6.0) to refresh the electrode surface for successive measurements. The application in BPA determination in bottled water samples was satisfactory with recovery from 93.2–104.1%. The leaching study indicated that with increasing temperatures and incubation time, more BPA is released from the PC bottles. Detection results of the presented method are generally in good agreement with those of HPLC (recovery 96.8–99%), suggesting the potential application of this method for BPA detection in real samples detection. A few years later, screen printed electrode modified with functionalized Fe_3O_4NPs (ThTAA-Fe_3O_4NPs/SPE) was developed for quantification of BPA in bottled water and food samples.[69]

Zhan et al. detected BPA using GCE modified with ultrathin **exfoliated Ni_2Al layered double hydroxide** (ELDH) nanosheets achieving LOD of 6.8 nM.[70] Upon modification of GCE surface, the electrochemical peak current of BPA enhanced owing to: i) lower mass transfer resistance, ii) the increase in active sites, and iii) larger electrochemical effective surface area of the modified electrode. The applicability of ELDH/GCE to determine BPA concentration in real food sample (milk) was examined and results showed that recovery (95% to 116%) and RSD (≤ 5.1%) values were acceptable. The stability of the modified electrode was analyzed as well. After keeping it in refrigerator at 4 °C for two weeks, there was a low fluctuation in peak current (< 5%), confirming the appropriate stability of the sensing platform.

Several electrochemical sensors using **graphene (GR)** and/or **graphene oxide (GO)** as key components have been created. Graphene oxide is a monolayer with a high oxygen content, typically characterised by a C/O ratio of less than 3:1 and typically closer to 2:1.[71] GO contains a series of active oxygen functional groups, such as hydroxyl, carbonyl, and epoxide. Despite the hydrophilic nature of GO, its low conductivity restricts its use in the electrochemical sensor design. Reduced graphene oxide (RGO) is more often the material of choice for electrochemical sensors and associated applications. RGO is usually fabricated from the oxidation/exfoliation of graphite to GO and then its reduction via chemical, thermal, or electrochemical methods to reduce its oxygen content. The obtained RGO is a two-dimensional nanomaterial with excellent physicochemical properties, such as large surface area, fast electron transport, high electrical conductivity, good thermal conductivity, chemical stability, and flexibility. However, graphene-based electrodes tend to aggregate and are suffering from restacking of their sheets due to π-π interactions and van der Waals forces. Their combination with other electrocatalytic materials offers remarkable results regarding electrochemical sensing applications. Thus, the combination of RGO with metal/metal oxide NPs results in a synergistic effect, improving effectively the sensing performance. For example, Wang et al. reported the fabrication of a disposable electrochemical sensor (RGO/CNT/AuNPs-SPE) for highly sensitive detection of BPA with LOD of 800 pM.[72] The electrode showed good selectivity and could be used to fabricate a portable device for rapid, reliable detection of BPA content in food and environmental samples.

An electrochemical sensor based on **Cu_2O nanospheres** wrapped by RGO was prepared using electrochemical reduction method, which was carried out by one pot synthesis of Cu_2O nanocomposites and RGO by cyclic voltammetry.[73] The sensor was successfully evaluated to detect BPA in bottled water sample. The response currents were still up to 90% of the original currents after 14 days of storage at 4 °C in refrigerator, indicating good stability of the prepared electrode. In a later publication, the researchers fabricated a sensor based on Rh_2O_3–RGO composite nanomaterial-modified GCE for detecting BPA in beverages directly after simple filtration.[74] However, the stability of electrode Rh_2O_3–RGO/GCE was not satisfactory – after two weeks of storage at 4 °C, the peak current retained 77.2% of the initial response.

Iodine-doped graphene (I-RGO) was prepared by simultaneous iodine-doping and reduction of GO with hydrogen iodide solution at mild reaction conditions.[75] The modified electrode I-RGO/GCE showed a large electrochemical active surface area and excellent conductivity, making it promising for construction of an electrochemical sensor. The determination of BPA in real milk samples was achieved with satisfactory recovery results. The authors noted that the peak currents only reduced 1.66% of the initial value after 30 days of storage, demonstrating the excellent reliability of I-RGO/GCE for BPA sensing.

Carbon dots (CDs) are a kind of zero-dimensional (0-D) carbon nanomaterial. Recently, CDs have gained attention for sensing applications owing to their advanced electronic properties arising from the quantum size effect. Rajesh et al. have analysed the electrochemical behavior and sensing ability of CDs-V_2O_5 nanoporous modified GCE towards BPA.[76] The catalytically active phase CDs-V_2O_5 efficiently facilitated BPA oxidation at a relatively low operating potential (+0.55 V vs. SCE). The electrode CDs-V_2O_5/GCE revealed an excellent dynamic range for BPA concentration (5 nM – 9.2 mM). The detection limit was as low as 0.8 nM, which is superior to most of the previously reported BPA sensors. Stability of the modified electrode was investigated for 1000 min (measurements were recorded every 60 min) and the resultant current response of the sensor decreased by 5%. Authors have not presented data on long-term stability (days, weeks) of the electrode material.

Bimetallic alloy nanomaterials are being extensively used for electrode modification owing to their outstanding properties and unique electrocatalytic characteristics providing multifunctional active sites. Bimetallic NPs are considered more effective than monometallic counterparts because of their synergistic characteristics. Due to the exclusive structure and specific surface area, they act as high-energy surface sites enhancing the electron transfer kinetics between electrode and analyte. The adsorption energies of analyte and intermediates on the surface of bimetallic catalyst are changed, thus leading to better catalytic activity and selectivity of bimetallic catalysts. Additionally, the integration of transition elements to the noble metallic NPs improve the utilization efficiency of the noble metal and make them economic and user-friendly. A decisive survey of recent advancements in the field of bimetallic nanostructures was presented by Rajeev et al.[77]

A sensitive and selective sensor based on nanoporous (NP-PtFe) alloy and graphene was successfully developed for direct detection of BPA in water samples.[78] The next year, Mo et al. reported the development of bimetallic AuPd nanoparticles incorporated in carboxylic multi-walled carbon nanotubes (MWCNTs) with synergetic amplified current signal applicable for electrochemical determination of BPA.[79] MWCNTs were used to improve electron transport. To overcome the intrinsic van der Waals' force between the pristine MWCNTs and further increase the loading ability of metal NPs, a linear positively charged polyelectrolyte of poly(diallyldimethylammoniumchloride) (PDDA) was used as a dispersant for MWCNTs. The prepared electrode MWCNTs-PDDA-AuPd/GCE showed an enhanced electrocatalytic performance toward BPA electrooxidation, compared to those of homologous monometallic counterparts and MWCNTs-PDDA. The synergetic effect of catalysis, the accumulation effect of MWCNTs-PDDA-AuPd, and the increased surface of the nanomaterial, the modified electrode shows improved signal and better sensitivity to BPA detection. The good analytical performance of the sensor was confirmed by determining BPA in milk and water samples with good recoveries (96.0% to 101.1%) and an acceptable relative standard deviation (RSD of 3.5%, n = 3). New sensors based on three-dimensional nanoporous PtSi (NP-PtSi) alloy and graphene,[80] and bimetallic Pd-Cu aerogel[81] supply convenient platforms for determining BPA with satisfying results.

In the field of materials science "green" synthesis, using mild reaction conditions and natural resources as plant extracts, has received a lot of attention as a convenient, sustainable, cheap, and environmentally friendly approach for synthesis of a wide range of nanomaterials. Recently, extensive studies have been performed to explore the electrochemical behavior of biosynthesized metal NPs and their potential in sensor design. There is convincing evidence that green synthesis of metal and metal oxide nanoparticles has a potential to provide a new direction in the fabrication of cheap and highly effective electrocatalysts applicable in food, clinical, and environmental analysis. A few review articles emphasize the significance of **biosynthesized metal nanoparticles** in the field of electrochemical sensing.[82–84]

An electrochemical sensor for BPA determination based on carbon black (CB) and gold nanoparticles (AuNPs) nanocomposite-modified sonogel-carbon electrode (SNGCE) was fabricated.[85] The AuNPs were synthesized by a novel green approach employing olive leaves extract and assisted by high energy ultrasound. The CB/AuNPs/SNGCE sensor showed good sensitivity at concentration range of 0.5–15 μM (LOD = 60 nM), and it was applied for the determination of BPA in mineral water samples. Here it should be pointed out that the sensing performance is highly dependent on the electrode surface morphology and may differ from electrode to electrode. Generally, the experimental data show that the surfaces modified with biosynthesized nanoparticles still remain challenging as they are not often as reproducible and stable as one would hope. According to the data reported in the commented paper, the deviation of current intensity of three electrodes prepared independently was 1.61% (RSD, n = 3). The repeatability, examined by registering the response of BPA employing the same modified electrode, gave a RSD of 5.70% (n = 3). The authors do not report the long-term stability of the material.

In conclusion, overall review indicates that NPs modified carbon-based electrodes are presenting some shortcomings. The main drawbacks are difficulties in maintaining the same size and shape of metallic/metallic oxide NPs during different batches of synthesis, as well as difficulties in achieving the same dispersion of NPs onto the electrode surface. A further complication is that many of the research teams used the drop-casting method to prepare the modified electrodes. The evaporation of droplet containing suspended nanoparticles forms a ring-like pattern. This unwelcome phenomenon labelled as "coffee ring effect", as well as uncontrolled aggregation of the coating materials, directly influence the morphology, which affects negatively the sensing performance, reproducibility, and stability of the electrodes modified by drop casting layers.[86] Thus, the problem of uniformity in colloidal film fabrication is a serious issue of functionality in the industrial context. Therefore, developing reproducible carbonaceous electrodes modified with NPs is a challenging task that remains to be investigated and addressed.

4. 3. Electrochemical Sensors Based on Metal–Organic Frameworks

Over the past decade conductive metal-organic frameworks (MOFs) are becoming increasingly important and attractive as electrocatalytic modifying materials in electrochemical applications because of their diversity in construction and composition. MOFs are a class of highly porous materials, constructed from the self-assembly of metal ions/clusters and organic ligands through coordination bonding resulting in 1-D, 2-D, or 3-D network structure.[87] MOFs are innovative and emerging sensing platforms as they provide unique structural benefits, including large specific surface area, tunable porosity, chemical stability, non-toxic nature, low density, and adjustable chemical functionalities. The high porosity and large surface area of MOFs are helpful for high-efficiency concentration and mass transfer of the analytes, which can effectively increase the signal response and detection sensitivity. On the other hand, MOFs have a tuneable pore size based on the choice of metal clusters and organic linkers. The practical shape and size of the available channels and vacant sites increase the selectivity towards specific analyte through size exclusion effects. For the MOF with open channels, the size and shape of the MOF channels must match that of guest molecule (BPA) to ensure the successful encapsulation. The common host-guest interactions usually involve hydrogen bonding, π–π stacking, electrostatic interactions, van der Waals, hydrophobic/hydrophilic interactions, and coordination interactions between open metal centers and the guest molecules.[88] The host-guest interactions in the proper channel (cavity) determine the ease with which the guests enter the host MOFs and can affect the position and orientation of guests in the host frameworks. Thus, controlled host-guest interactions are of crucial importance to the construction, properties, and functions of host-guest MOF sensing systems. These features provide MOFs with appropriate catalytic properties, making them helpful as effective coating materials for electrocatalytic electrodes that are used in sensing devices.[87] Further modifications improve the properties of MOFs materials so that they can be better applied to electroanalysis. One of the effective methods is to combine MOFs with other conductive materials for improving the performance of the electrodes, and another way is to derive various complexes from MOFs as templates.[89] Motivated by these fascinating properties, various pristine MOFs with excellent electrocatalytic activity toward BPA have been used as electrocatalysts to detect BPA in foods and beverages.[90–95]

Modification of MOF surfaces with GO further enhances their electrochemical conductivity that improves the overall activity of the modified electrode. A novel metal-organic framework (UiO-66-NDC/GO) has been synthesized, characterized, and used as a sensing material for the development of electrochemical sensor for the detection of BPA.[91] UiO-66-NDC is formed by substituting the organic linker in UiO-66 (zirconium-based MOF, chemically stable in aqueous solutions) with 1,4-naphthalene dicarboxylic acid. The experimental findings suggest the acceptable applicability of the developed electrode UiO-66-NDC/GO/CPE for the detection of BPA in different real samples (drinking water and milk).

A sensitive electrochemical BPA sensor was fabricated based on Fe_3O_4 NPs and ZIF-4 (zeolitic imidazolate framework-4) synergistic effect.[92] The modified screen printed graphite electrode (Fe_3O_4/ZIF-4/SPGE) demonstrated satisfactory results toward BPA detection in water and canned foods.

Zhang et al. prepared hierarchical Ce-MOF modified with a cationic surfactant (cetyltrimethylammonium bromide, CTAB), for the construction of an ultrasensitive electrochemical BPA sensor.[93] In electroanalytical chemistry, the modification with surfactant was used to enhance analyte adsorption and the electron transfer rate, which lowered the detection limit of the proposed sensor system. CTAB/Ce-MOF/GCE showed an improved electrochemical response because of the high adsorption capacity and electrocatalytic activity of Ce-MOF, and enhanced preconcentration of BPA by the long alkane chain in CTAB via hydrophobic interaction. The proposed sensor showed a low detection limit of 2.0 nM. It was employed to analyze BPA in twelve real samples (four types of fresh liquid milk, four types of PC drinking package and four types of PET drinking bottle) to evaluate the potential application. The recoveries determined for BPA ranged between 96.2% and 104.6% demonstrating its practicability and reliability. The DPV peak current of BPA oxidation at CTAB/Ce-MOF/GCE showed 98.6% and 97.6% of its original signal after one and four weeks, respectively, confirming the good stability of the modified electrode.

Finally, the development of MOFs-based sensors from the structural design to the fabrication of the sensing device is a multidisciplinary task and the primary concern for the researchers is insufficient stability and low electrical conductivity of the conventional MOFs. One straightforward technique to increase the charge transfer in MOFs is creating the structure with intrinsic electrical conductivity. 2-D MOFs (MOF nanosheets) are also promising candidates for sensor applications due to their good conductivity (fully in-plane π-delocalization and out-of-plane π-conjugation), large surface area, easy diffusion, and more available active sites for the analyte. In order to obtain better fundamental knowledge regarding the role of MOFs in the particular electroanalytical process *in situ* spectroscopic studies should be applied to investigate the host-guest interactions. Additionally, theoretical calculations as an emerging powerful research tool should also be applied to explore the detailed electrocatalytic mechanism.

4. 4. Molecularly Imprinted Electrochemical Sensors

Molecularly imprinted polymers (MIPs) and their incorporation with various transducer platforms are among the most promising approaches for detection of several analytes.[96] The increasing number of publications on molecularly imprinted electrochemical sensors (eMIPs) for BPA detection reveals the potential in this field. Briefly, molecular imprinting involves the creation of specific recognition sites within a polymer matrix that can selectively bind the target molecule. This is achieved via self-assembly of functional monomers around a BPA template molecule, followed by the polymerization process that solidifies the structure. The template is then removed, leaving behind specific cavities that are complementary to BPA in terms of shape, size, and chemical functionality.[97] Extremely high affinity and selectivity enable MIPs to be used in electrochemical sensing strategies.[98] Electrochemical detection measures changes in electrical signals upon BPA's rebinding to imprinted cavities. This allows eMIPs to offer high selectivity towards BPA even in complex sample matrices. Using simple instrumentation BPA can be reliably detected within minutes.[97] Owing to the improved separation efficiency and negligible cross-reactivity, the eMIPs are enabled to provide the lowest reported hitherto detection limits of BPA in real food samples. Picomolar (pM/10^{-12} M) concentration LOD was achieved using eMIPs. Additionally, eMIPs have been integrated into compact portable sensing devices for on-site analysis. Therefore, unique features including high specificity, robustness, and low cost production makes them attractive in food safety evaluation. The different production methods of MIPs and the varied types of electrochemical and optical sensors that employed MIPs to detect BPA have been reviewed extensively by Hamed and Li in 2022.[99] The next year Pan et al. have provided a comprehensive overview on recent advances in eMIPs for BPA detection discussing the operating principles, fabrication strategies, materials, and methods used in eMIPs.[97]

MIP was prepared using BPA as a template, 2-hydroxyethyl methacrylate (HEMA) as the functional monomer and ethylene glycol dimethacrylate (EGDMA) as a crosslinker with a ratio (1:2:30) and was incorporated in carbon paste electrode modified with MWCNTs.[100] Linearity within the working dynamic range 1×10^{-10} to 1×10^{-4} M (0.023–23 × 10^3 ng/mL) was observed with a LOD of 8×10^{-11} M (0.02 ng/mL) and LOQ of 2.4×10^{-10} M (0.05 ng/mL). BPA was successfully quantified in water stored in a baby bottle and soft drink samples with recovery ranges 97.60–102.0% with RSD values 0.34–2.45% indicating the possible applicability of the electrode in real samples.

MIP film coated carbon nanomaterials and metal nanoparticles were used as transducer platforms with good electrical conductivity and increased surface area. A novel electrochemical sensor was prepared for the detection of BPA, based on a MIP/CNTs-AuNPs/boron-doped ordered mesoporous carbon composite.[101] CNTs functionalized by poly-diallyldimethylammonium chloride (PDDA) were employed as support materials. The introduction of functionalized CNTs not only increased the specific surface area, but also improved the electrostatic adsorption of AuNPs. On the other hand, boron doping is a viable strategy for improving the electrochemical activity of ordered mesoporous carbon (OMC), and boron doped ordered mesoporous carbon (BOMC) becomes a promising electrochemical sensing material. According to the data presented, the modified electrode MIP/CNTs-AuNPs/BOMC/GCE exhibited good reproducibility, reasonable stability, and selectivity towards some structural analogs of

BPA. The resulting sensor showed a linear range of 0.01–10 μM and a LOD of 5 nM determined by using differential pulse voltammetry. It was applied to detect BPA in milk samples.

Electrochemical sensor based on an acetylene black paste electrode modified with molecularly imprinted chitosan–graphene composite film for sensitive and selective detection of bisphenol A (BPA) has been developed by Deng et al.[102] The fabricated sensor (MIP-CHIT-GR/ABPE) showed an excellent specific recognition of the template molecule among the structural similarities and coexistence substances. It was successfully employed to detect BPA in plastic bottled drinking water and seven types of canned beverages (four soft drinks, Red Bull energy drink, tea, and beer). In a more recent study Tan et al. reported sensor based on molecularly imprinted chitosan film doped with acetylene black (AB) modified GCE as a recognition element.[103]

Highly advanced and modern MIP electrochemical sensors supported on graphene materials were developed.[104,105] Graphene oxide (GO) is an oxygenated graphene sheet with high percentages of –OH, –COOH, –CO, and epoxy groups obtained by graphite exfoliation or oxidation. Interesting work utilising GO has reported a new composite of amino-functionalized GO and MIP (GO/APTES–MIP) which has been applied to the in-situ determination of BPA in milk and mineral water without any pretreatment and matrix interfering effects.[106]

Combining both RGO and MIP materials can significantly improve the sensing performance. For instance, PPy-based imprinted polymer film was synthesized on electrochemically reduced graphene oxide (ERGO) by electrochemical polymerization.[107] The results prove that the developed electrode MIP/ERGO/GCE is selective and sensitive for the detection of BPA in water and milk with good recovery and reproducibility. The authors report that the almost identical signal was obtained for 25 cycles of analysis. After 30 cycles, a slight decrease in signal was noted. This response of the MIP/ERGO/GCE sensor indicates its appreciable stability.

Compared with MIPs in other shapes, such as monolithic polymer rods and amorphous polymeric particles, molecularly imprinted polymeric microspheres (MIPMSs) are of better uniformity and often higher surface area, which are beneficial to enhance fabrication control and sensing response.[108] A highly sensitive electrochemical sensor based on CPE modified with MIPMSs was developed. MIPMSs/CPE exhibited a linear range for BPA $1 \times 10^{-11} - 1 \times 10^{-7}$ M with detection limit of 2.8×10^{-12} M under the optimal experimental conditions. This sensor showed accurate results comparable to HPLC, providing a reliable method to monitor BPA in real milk and water samples.

Gold-doped MIP,[109] MIPs/MWCNTs-AuNPs,[110] CPE modified with BPA-IP sol-gel and MWCNTs,[111] and flexible MIP electrochemical sensor based on the carbon felt[112] were also used to detect BPA in food samples, obtaining satisfactory recoveries: (96.7–107.6%),[109] (92.7–97.8%),[110] (92–105%),[111] and (91.3–112%),[112] respectively.

Since their introduction by the George Whiteside's group, microfluidic paper-based analytical devices (μPADs) provide alternative tools for point-of-care testing.[113] A novel μPAD based on molecularly imprinted curcumin nanoparticles platform for dual-modal electrochemical and fluorescence sensing of BPA was developed by Mars et al.[114] To enhance the electrochemical activity of μPAD, the working electrode surface was modified with Carbon black/Prussian blue nanocomposite. Fluorescence, as well as electrochemical measurements, revealed a sensitive response toward the presence of BPA. The feasibility of the MIP-μPAD was demonstrated for the sensing of BPA in seawater, foods, and polycarbonate plastic packaged water with recovery values of 97.2 and 101.8%.

Zhang et al. developed MIPs that consisted of carboxylated CdTe quantum dots (CdTe QDs) grafted with aminated MWCNTs as a carrier, BPA as a template, and 3-aminopropyltriethoxysilane (APTES) as a monomer.[115] The successful combination of QDs and MWCNTs enables nanotubes to exhibit higher sensitivity on the basis of excellent electrical conductivity. The proposed sensor showed extremely low LOD of 15 pM. Its applicability has been demonstrated by analyzing water samples. The electrode has stable characteristics within 20 days.

Beduk et al. presented a simple, low-cost imprinted sensor based on laser scribed graphene (LSG) technology combined to MIPs for highly sensitive and selective detection of BPA in water and plastic samples.[116] However, the proposed sensor has problematic reusability due to the loss of accuracy after measuring four times. The authors note that the electrochemical response decays by 23% of its initial current at the fifth cycle. Two years later, the research team presented a homemade portable potentiostat device integrated to LSG sensor for BPA detection as a practical food monitoring tool.[117] LSG platform is combined with MIP matrix, gold nanostructures and PEDOT polymer to create a specific MIP biomimetic receptor for ultrasensitive BPA detection. The sensing device has a Bluetooth connection, wirelessly connected to a smartphone (Figure 5). The reliability of the sensor has been validated by measuring BPA in commercial bottled water samples, commercial milk, and baby formula samples, as well as the commercial plastic samples with good recoveries. This customized sensor allows users to monitor BPA existence in the commented food samples on-site. In order to determine the reusability of the same sensor, authors investigated the possibility of reperforming the removal procedure for the same sample (three removal-rebinding cycles). The data show that the oxidation current response of sensor is slightly reduced after every cycle. Finally, the 10% of the initial sensor response was lost after three cycles. Authors explained this current loss by the slight damage coming from using an acetic acid/methanol (3:7) mixture as solution to remove BPA from the previously created cavities.

To overcome these drawbacks (time-consuming regeneration of electrode surface and loss of activity), magnetic molecularly imprinted polymers (MMIPs) provide an effective method for immobilization and MIP renewal from the electrode support.[118] MMIPs combine the synergic advantages of MIPs and magnetic materials, including low cost of production and stability. Compared to classical MIPs, regeneration of MMIPs is more convenient via magnetic actuation. These materials are synthesized by a core-shell procedure, in which the molecular imprinting surface (shell) is deposited over a paramagnetic core that provides magnetic actuation.[119] The resulting MMIPs show a high adsorption capacity, recognition with high efficiency and specificity, reusability, and outstanding magnetic properties. Therefore, MMIPs as a separable material by an external magnet can be easily incorporated on magneto-electrode, greatly simplifying the experimental procedures and enhancing the analytical performance of the sensor device.

Among the different types of magnetic materials, Fe_3O_4 is the most widely employed material in sensing devices, due to its strong superparamagnetism, good catalytic activity, and simple preparation procedure. A novel electrochemical technique for the determination of BPA was performed by synchronous extraction and preconcentration of BPA onto MMIP, with subsequent readout on a magneto-actuated glassy carbon electrode (MGCE) by differential pulse voltammetry.[118] This sensor enabled BPA determination procedures, including isolation, clean-up, enrichment, and qualification, to be completed in one step. BMMIPs exhibited good adsorption capacity in acidic and neutral conditions, which encompass most food samples. Based on the recoveries in real samples (tea drink, milk, and cabbage), the BMMIPs@MGCE sensor demonstrated

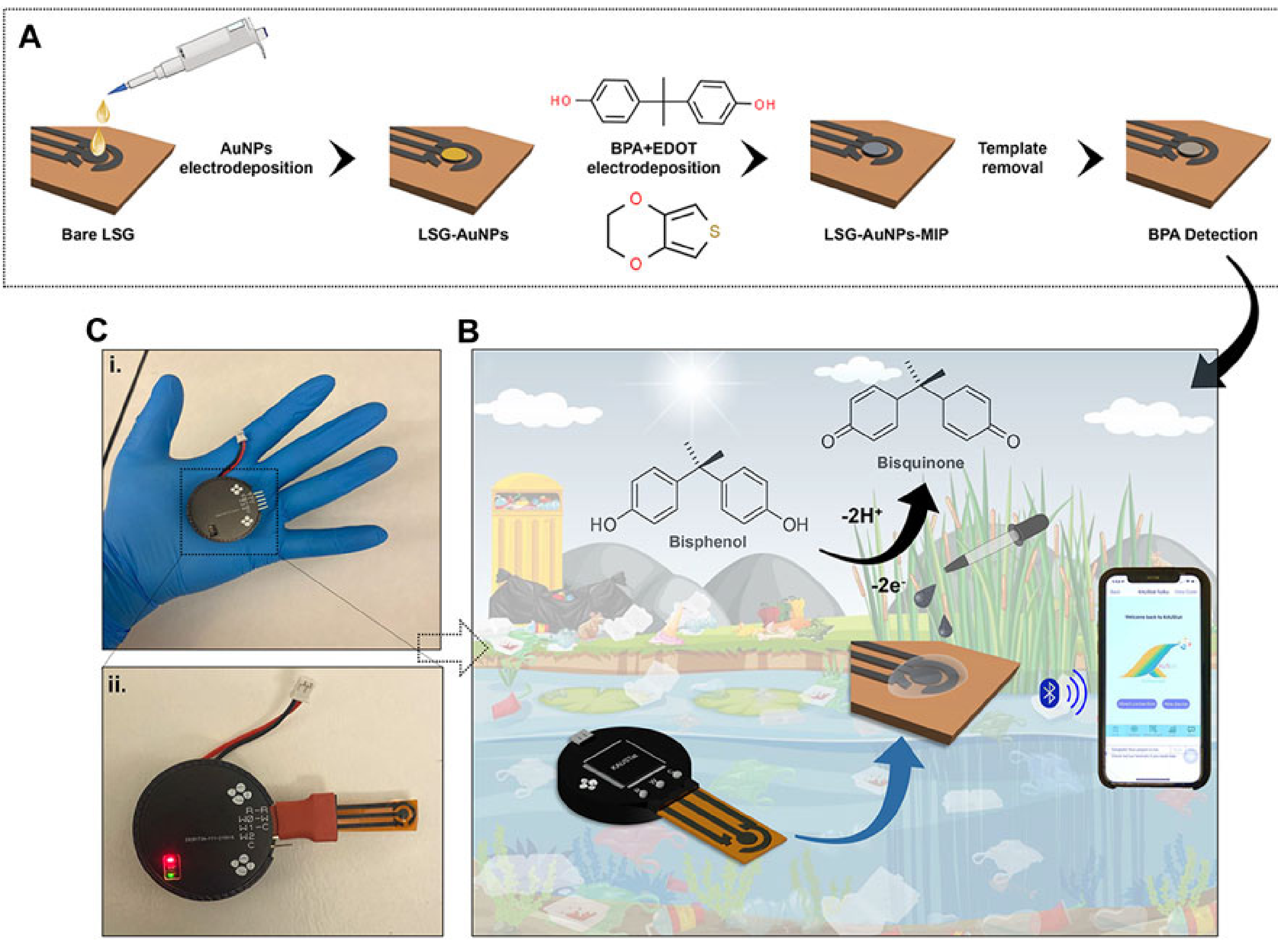

Figure 5. (A) Preparation of the molecularly imprinted laser-scribed graphene (LSG) sensor including electropolymerization of AuNPs, EDOT monomer in the presence of BPA, and template removal to create BPA cavities. (B) Schematic illustration of the portable potentiostat attached to the AuNPs/LSG sensor and connected to a smartphone via Bluetooth for detecting BPA in environmental samples. (C) The photos of (i) the potentiostat, (ii) the device combined with the LSG-MIP sensor. Red light indicates an active Bluetooth connection. Reproduced from reference[117]; permissions under Creative Commons Attribution License (CC BY). Copyright 2022 Beduk, Gomes, De Oliveira Filho, Shetty, Khushaim, Garcia-Ramirez, Durmus, and Ait Lahcen abd Salama.

potential for simple, quick, and low-cost determination of BPA in food applications. Moreover, the BMMIPs exhibited excellent temperature stability, as evidenced by the minimal influence on the adsorption capacity of BMMIPs in the temperature range of 5–45 °C. Reusability (or regeneration) and associated cost benefit is another key factor in the actual application of solid-phase extraction materials. Here, BMMIPs were used for the adsorption of BPA in five consecutive adsorption/desorption cycles. Based on the experimental data presented, there is no significant tendency towards a decrease in the binding capacity of the BMMIPs after five adsorption/desorption cycles, indicating that BMMIPs could be applied for even more cycles.

Zhu et al. introduced another MMIPs sensor for BPA designed by integrating magnetite nanoparticles and surface imprinting with electrode magnetization techniques.[120] Cationic surfactant CTAB was applied as an antifouling agent to avoid the direct contact of electrochemical oxidation product of BPA on the electrode. The prepared sensor displayed good stability, sensitivity, rapid electrochemical response, and strong antifouling ability.

Ultrasound-assisted magnetic molecularly imprinted polymer (US-MMIP) combined to an electrochemical sensor modified with a nanocomposite of carbon black nanoparticles (CBNPs) and AuNPs was developed for highly selective and sensitive detection of BPA.[121] The analytical approach was applied successfully for determination of BPA in mineral water samples.

Deveci et al. fabricated e-MIPs based on boron-functionalized graphitic carbon nitride (B-g-C_3N_4) and graphene quantum dots (GQDs) for selective determination of BPA with an extremely low LOD of 3.0×10^{-12} M.[2] Superior sensitivity and LOD may be due to the following: (i) the doping treatment of GQDs and the boron element into g-C_3N_4 resulted in the specific surface area, indicating more interactions with BPA molecules; (ii) the synergistic effect between GQDs and the boron element increased electron transfer rate on the electrode surface. The nanocomposite also showed repeatability and selectivity in food sample (orange juice).

In conclusion, MIPs can be categorized as one of the advanced synthetic methods for designing robust recognition materials that offer promising potential in the development of electrochemical sensors for BPA.[122,123] Although there is a genuine market need for such devices, MIP-based technology has remained mostly in the academic field. Therefore, particular attention must be paid at the later stage of laboratory research, and transfer to large-scale industrial production.

4. 5. Electrochemical Sensors Based on Ionic Liquids

The discovery of water and air-stable ionic liquids (ILs) by Wilkes in 1992 has opened a new field of interest within the advanced multifunctional materials community.[124] ILs exist in liquid state at around room temperature. They are organic salts often composed of big organic cations and small inorganic anions. During the last years, more focus has been paid to ILs because of their novel and adjustable physical and chemical features: facile design, high chemical stability, high ionic conductivity, and wide electrochemical window. Therefore, they have been demonstrated to be effective modifiers for improving the sensing performance of electrodes. Up to the present, the most common ionic liquids used in electrochemical BPA sensors were: 1-hexyl-3-methyl-imidazolium hexafluorophosphate,[126] 1,3-dipropylimidazolium bromide,[127] 1-methyl-3-octylimidazolium tetrafluoroborate,[128] 1-ethyl-3-methylimidazolium tetrafluoroborate,[129] and 1-butyl-2,3-dimethylimidazolium tetrafluoroborate.[130] For example, a novel composite of conductive metal organic framework and molecularly imprinted poly-(ionic liquid) (CMOF-MIPIL) was prepared for highly sensitive detection of BPA in water and milk samples.[125] Under optimal conditions, CMOF-MIPIL sensor exhibited a linear range of BPA detection from 0.005 to 5.0 μM (LOD = 4.0 nM). Authors concluded that the concept of conductive MOF as support in surface imprinting may provide a new way of preparing imprinted composites with excellent electrocatalytic activity and good selectivity. Next year, Nasehi et al. developed CPE amplified with 1-hexyl-3-methyl imidazolium hexafluorophosphate (HMH) and TiO_2-SWCNTs.[126] Wider linear dynamic range of 3.0 nM – 450 μM and a lower limit of detection of 1.0 nM was achieved. This platform was utilized for voltammetric sensing of BPA in water and soft drinks.

1,3-dipropylimidazolium bromide modified ZnO/CNTs carbon paste electrode was also used to investigate the electrochemical behaviour of BPA and Sudan I.[127] Sudan I (1-phenylazo-2-naphthol) is a synthetic compound used as a food additive, namely in chili powders and beverages due to its profound red-orange color. The International Agency for Research on Cancer (IARC) has categorised Sudan I as a category 3 carcinogen, underscoring the potential risks this substance posess to human health. The ZnO/CNT/IL/CPE successfully resolves the overlapped voltammetric peaks of BPA and azo dye Sudan I by 240 mV, so that the modified electrode displays high selectivity in the SWV measurement of BPA and Sudan I in their mixture solutions. The electrode was successfully used for the determination of BPA and Sudan I in a variety of food samples (tomato paste, corn, stew, tuna fish, chili sauce, and bottled water). Authors reported that the electrode can be immersed in an aqueous media for 2.5 h with stable response.

1-methyl-3-octylimidazolium tetrafluoroborate (MOITFB) was also used as an ionic liquid to fabricate modified electrode CdO/NPs-MOITFB-CPE applied in the determination of BPA in canned foods with acceptable recovery data.[128] An electrochemical sensor based on ionic liquid/chitosan/graphene nanosheets (IL/CS/GNs) has been successfully applied for the recovery tests of BPA in milk samples.[129] Another sensing platform based on a composite of graphene nanoplatelets (GNPs) and ionic liquid as a modifier for glassy carbon paste electrodes (GCPEs) exhibited a linear relationship between signal and BPA concentrations ranging from 0.02–5.0 μM, with LOD and LOQ of 6.4 nM and 0.02 μM, respectively.[130] The sensor was successfully applied to the determination of BPA in bottled water samples and the results were in agreement with the reference values from a standard HPLC method.

4. 6. Other Types of Electrochemical Sensors Used for The Determination of BPA

In the recent years, more attention has been gained in using polymers for functional modification of the working electrodes because of their adhesion, specific selectivity and the ability to provide more active sites. Conducting polymer nanocomposites are functional advanced materials due to synergistic effect of two individual components with high conductivity and fine structure at nanoscale. A facile electrochemical sensor based on RGO-Ag/poly-L-lysine nanocomposites (RGO-Ag/PLL) modified GCE was proposed for the quantitative determination of BPA in drinking water with satisfactory results.[131] Highly sensitive sensor (LOD = 0.4 nM) based on polyaniline (PANI) and AuNPs composite as an electrode material was utilized to detect BPA in bottled water and canned beverages.[132]

Stable biopolymer dispersions combined with nanomaterials are a viable alternative for modification of electrode surface. Vieira Jodar et al. fabricated a new material for BPA detection composed of casein (CAS) and carbon black (CB).[133] The proposed film showed good conductivity and adsorption capability resulting in high electrocatalytic activity.

Recently, researchers have started to focus on the development of MOFs as precursors to prepare NiNPs/carbon nanocomposite, and the nanocomposite has been demonstrated to have good electrochemical performance owing to the protection of the carbon supporting nanomaterials from the oxidation of NiNPs, and the strong interaction and synergistic effect between NiNPs and carbon nanomaterials. Three-dimensional hierarchical cylinder-like NiNPs/nitrogen-doped carbon nanosheet/chitosan nanocomposite (NiNPs/NCN/CS) is used for the modification of GCE.[94] This composite demonstrated to have good electrical conductivity, electrocatalytic activity, and high antifouling ability, which resulted in high sensitivity and improved reproducibility for BPA detection. The NiNPs/NCN/CS/GCE based sensor had strong anti-interference ability and good stability (94.1% of the original signal after 15 days), and can be employed to detect BPA in milk samples.

Naik et al. reported for the first time 2,6-bis (2-benzimidazoyl) pyridine (BZPY) as a catalyst for BPA detection.[134] The existence of three active electron-donating moieties in the structure of BZPY (two imidazolic nitrogens and one pyridine nitrogen) makes the molecule a suitable candidate for sensing application. The results of molecular dynamic simulation reveal that BPA can interact with BZPY through hydrogen bonding. The developed BZPY/CPE sensor was electrochemically characterized and employed for the detection of BPA in bottled water. The authors reported noteworthy stability of the sensor – the decrease in the peak intensity of BPA after one month storage was 5.8%.

Graphene nanoplatelets (GNP) were dispersed in water with the assistance of a tri-azo dye, Direct Blue 71 (DB) and a stable thin film of GNP/DB on GCE was formed.[135] The resulted electrode GNP/DB-GCE was used for selective and sensitive detection of BPA in commercial milk, cranberry juice, and lemonade.

Cyclodextrins are often used to create sensors for various purposes.[136] Alam and Deen developed an electrode that combines the high surface area of GO and carbon nanotubes, and the superior host-guest interaction capability of β-cyclodextrin (βCD).[19] The proposed electrode GO-MWCNT-βCD/SPE provides opportunities for developing a smart, low-cost, and easy-to-use system for water quality monitoring. The materials cost for a single GO-MWCNT-βCD/SPE electrode was as low as \$0.2 (authors have shown a breakdown of materials cost to fabricate a single electrode). Thus, the combined efforts and achievements of screen printing, nanotechnology, electrochemistry, analytical chemistry, and organic chemistry led to a new generation of promising sensors.

Dual-signal sensor by coupling nanoporous gold leaf with thiolated beta-cyclodextrin exhibited a promising performance in analyzing milk samples, and the accuracy of results was validated by the standard method HPLC.[137] The manufacture of sensor was facile and highly controllable, which benefited the cost efficiency and reproducibility.

In 2024, a novel stirrer-assisted liquid-phase microextraction method using switchable hydrophilic solvents was developed for the extraction and preconcentration of BPA from real samples prior to electrochemical analysis.[138] In this work, SSHS-LPME was successfully used for the extraction and preconcentration of BPA in water samples prior to electrochemical analysis. It was shown that changing the stirrer speed can be used to dissolve the switchable hydrophilic solvent (*N*,*N*-dipropylamine) during the extraction process. The proposed method was used for the determination of bisphenol A in milk, fruit juice, and water samples.

Table 1 summarizes the recent reports of electrochemical sensors for BPA determination in foods and beverages. It provides a comparative study of linear range, detection limit, recovery, and stability of electrodes.

Table 1. Electrochemical sensors used for BPA sensing in foods and beverages.

Sensing electrode	Technique	Linear range	LOD	Real sample	Recovery, %	Stability (period)
Nanomaterials-based electrochemical sensors						
$FeNi_3$/CuS/BiOCl/CPE[64]	DPV	0.1–300 μM	0.05 μM	tuna fish, tomato paste, apple, corn	96.8–103.9	92 % (30 days)
$CoFe_2O_4$/GCE[66]	DPV	0.05–10 μM	3.6 nM	milk	98.5–102	95 % (30 days)
Fe_3O_4NPs-CB/GCE[68]	DPV	0.1 nM–50 μM	0.031 nM	bottled water	93.2–104.1	–
ThTAA-Fe_3O_4NPs/SPE[69]	DPV	0.03–700 μM	10 nM	bottled water, tuna fish, tomato paste, corn, stew, chili sauce	98–103.2 (15 days)	97.6 %
Rh_2O_3-RGO/GCE[74]	CV	0.6–40 μM	0.12 μM	beverages	93–98	77.2 % (14 days)
I-RGO/GCE[75]	DPV	0.04–4.5 μM	0.02 μM	milk	95–110	98.3 % (30 days)
NP-PtSi/GR/GCE[80]	DPV	0.3–85 μM	0.11 μM	water, milk	99.2–103.5	94.7 % (14 days)
CB/AuNPs/SNGCE[85]	DPV	0.5–15 μM	60 nM	mineral water	95–102	–
Electrochemical sensors based on metal–organic frameworks						
Ce-Ni-MOF/MWCNTs/GCE[90]	DPV	0.1–100 μM	7.8 nM	bottled water	97.4–102.4	–
UiO-66-NDC/GO/CPE[91]	DPV	10–70 μM	25 nM	milk	94.8–99.3	–
Fe_3O_4/ZIF-4/SPGE[92]	DPV	0.06–300 μM	15 nM	tomato paste, stew, tuna fish	97.3–103.1	–
CTAB/Ce-MOF/GCE[93]	DPV	0.002–50 μM	2 nM	milk	99.2–103.4	97.6 % (30 days)
Molecularly imprinted electrochemical sensors						
MIP/GQDs/B-g-C_3N_4/GCE[2]	DPV	0.01–1 nM	3 pM	orange juice	99.7–100.3	7 weeks
MIPs/CNTs-AuNPs/BOMC/GCE[101]	DPV	0.01–10 μM	5 nM	milk	98–107	93.3 % (15 days)
MIP-CHIT-GR/ABPE[102]	LSV	8 nM–20 μM	6 nM	bottled water, 7 types of canned beverages	92–106	91.7% (10 days)
MIPs-AB/GCE[103]	DPV	0.005–10 μM	20 nM	bottled water	94.1–105.7	92.8 % (20 days)
GO/APTES-MIP/GCE[106]	DPV	0.006–20 μM	3 nM	milk, mineral water	96.8–106.2	94.8 % (21 days)
MIP/ERGO/GCE[107]	DPV	0.5–750 nM	0.2 nM	bottled water, milk	91–99	–
MIPMSs/CPE[108]	CV	10 pM–0.1 μM	2.7 pM	bottled water, milk,	95–98.3	94% (15 days)
Au@MIP/Au/GCE[109]	DPV	0.5–100 μM	52 nM	milk, orange juice, bottled water	96.7–107.6	93.8 % (5 days)
MIPs/MWCNTs-AuNPs[110]	Amp.	0.1 μM–8.2 mM	3.6 nM	honey, grape juice	92.7–97.8	–
BPA-IP/MWCNTs/CPE[111]	DPV	4 nM–50 μM	3.1 nM	bottled water	92–103	94.6 % (30 days)
MIP@CF[112]	DPV	0.5–300 nM	0.36 nM	milk	91.3–112	12 days
MIP/CdTe QDs-MWCNTs/GCE[115]	DPV	0.05–50 nM	15 pM	water	87.1–102.6	91.6% (20 days)
MIP@PPy/LSG[116]	DPV	0.05–20 μM	8 nM	mineral water	88–110	90.7 % (20 days)
MIP/LSG-AuNP-PEDOT[117]	DPV	0.01–10 μM	3.97 nM	bottled water, milk, baby formula	97.3–112.2	–
BMMIPs@MGCE[118]	DPV	0.8–8 μM	0.133 μM	tea drink, milk, cabbage	81.3–119.8	–
MMIPs NPs /CTAB/CPE[120]	CV	0.6–100 μM	0.1 μM	bottled water	95–112	91.4 % (30 days)
MIP/AuNPs/CBNPs/SPCE[121]	DPV	0.07–10 μM	8.8 nM	mineral water	96.4–98.4	88 % (30 days)

Sensing electrode	Technique	Linear range	LOD	Real sample	Recovery, %	Stability (period)
Electrochemical sensors based on ionic liquids						
CMOF-MIPIL/GCE[125]	DPV	0.005–5 μM	4 nM	water, milk	95.3–104	97.2 % (14 days)
ZnO/CNTs/IL/CPE[127]	SWV	0.002–700 μM	9 nM	tomato paste, corn, stew, tuna fish, chili sauce, bottled water	–	(30 days)
CdO/NPs-MOITFB[128]	DPV	0.01–280 μM	1 nM	tuna fish, tomato paste, stew	98–103.4	92 % (30 days)
IL-GNP/GCPE[130]	DPV	0.02–5 μM	6.4 nM	bottled water	95.3–104.5	80 % (14 days)
Other electrochemical sensors						
RGO-Ag/PLL/GCE[131]	DPV	1–80 μM	0.54 μM	water	91–107.3	90.4 % (14 days)
PANI-AuNPs/GCE[132]	DPV	0.003–45.7 μM	0.4 nM	bottled water, canned beverages	98–104.6	95.7 % (28 days)
CAS-CB/GCE[133]	LSV	0.49–24 μM	0.25 μM	milk	96.9–107	–
BZPY/MCPE[134]	DPV	2–18 μM	29 nM	bottled water	94.7–101.2	94.2 % (30 days)
GNP/DB/GCE[135]	DPV	10 nM–25 μM	1.23 nM	milk, cranberry juice, lemonade	99.3–102.3	–
GO-MWCNT-βCD/SPE[19]	LSV	0.05–30 μM	6 nM	bottled water	97–104.7	–
SH-β-CD/NPGL/Au[137]	SWV	0.3–100 μM	60 nM	milk	99.7–105.3	89.8 % (14 days)
SSHS-LPME/GCE[138]	DPV	0.01–250 μM	3 nM	bottled water, milk, fruit juice	96.8–102.3	–
Cu/CuO-N-Cs[139]	DPV	1–11 μM	31 μM	milk	90–102	(22 days)
Tween 80/VXC-72R/GCE[140]	DPV	0.08–40.0 μM	0.087 μM	milk	101.8–102.3	(12 days)
CdTBrPP/AuNPs/SPCE[141]	SWV	10^{-11}–10^{-2} M	9.5 pM	water	104.6–106%	87 % (5 days)

5. Advantages and Limitations of Electrochemical Sensors for Determination of BPA in Food Samples.

Although all the sensor platforms discussed here were successfully applied in real food samples analysis, there is a considerable gap between the laboratory tests and the fabrication of commercial analytical devices for practical applications. The next section highlights the advantages, limitations, and approaches for improving the electrochemical determination of BPA in food samples. Even though there are still some drawbacks to using this type of equipment for food control, the benefits outweigh the shortcomings.

Electrochemical sensors for BPA demonstrate irreplaceable advantages in food applications, including fast response time (few seconds) and quick data collection, high selectivity and sensitivity for trace levels determination, extremely low LODs and LOQs (picomolar range), high levels of repeatability and accuracy. The electrochemical analysis requires low sample volumes (microliters), minimum sample pretreatment, low-cost equipment, and ultra-low power consumption. Additionally, these systems are attractive candidates for miniaturized analytical devices, as they have a high compatibility with modern micro- and nanofabrication technologies. High benefit/cost ratio, multiplexing capabilities, and ability to be integrated as a detection module in a variety of analytical systems are other significant advantages. With the committed resources and pragmatic strategies, electrochemical sensors could enable routine BPA detection and contribute significantly to the food safety.

Limitations of electrochemical sensors for BPA should also be specified:

– **Biofouling and chemical fouling of electrodes**. This process is associated with gradual passivation of the transducer surface due to accumulation of fouling compounds (matrix components or electrochemical reaction products). The composition of food samples is very complex, as they contain a variety of substances in addition to the target analyte to be measured, such as proteins, lipids, carbohydrates, etc. The non-specific physisorption of biological macromolecules on the electrode surface negatively affects sensor's performance – it results in higher background signal, low accuracy, and low sensitivity. In the electrooxidation of BPA, reaction products also tend to accumulate at the electrode surface leading to the loss of catalytic activity.

- **Limited calibration standards.** Food matrices, rich in interfering substances, pose difficulties, demanding robust and selective portable electrochemical sensors for effective on-site, real-time monitoring. Accurately monitoring BPA concentrations in food samples presents a challenge due to the limited calibration standards.
- **Estimating for portability**. Laboratory-grade sensing devices often lack adequate exploration on their portability. Therefore, electrochemical sensors need transducers assembled within a carefully designed sensing interface that can be fabricated into a portable unit.
- **Fabrication challenges**. Most articles give a brief description of the fabrication process and raw materials used for electrode preparation, but they don't provide a detailed manufacturing workflow that is specific to industrial production. The analytical performance is highly dependent on the electrode surface morphology and may differ from device to device even though the catalytically active material originates from the same fabrication protocol. Additionally, most of the preparation procedures are not suitable for rapid large-scale production of cost-effective sensors. Commercialization and broad on-site applicability of BPA sensors will require improvements in stability, reproducibility, and, importantly, low-cost materials and easy fabrication methods.

6. Summary of Approaches for Improving the Electrochemical Determination of BPA and Implementation of Sensor Tools for Food Safety Applications

The effects of fouling can be sufficiently minimized by: (i) use of antifouling layers, nanoporous materials, permselective membranes, and hydrogels in order to provide a physical barrier between the electrode surface and fouling agents; (ii) development of novel electrode materials more resistant toward passivation; (iii) incorporating the electrochemical activation of electrode surface at regular intervals as a part of the analysis procedure. A single use of cathodic/anodic potential or a train of pulses may reduce adsorption of fouling agents or remove already attached substances.

Despite many technological advancements, most sensor platforms commented in this paper are still at the proof-of-concept stage. The limited validation of these new electroanalytical devices will continue to be a barrier to their use in real-world BPA detection. In this regard, additional efforts should be made by authors to evaluate the BPA assay beyond recovery experiments from spiked food/beverage samples. The analytical parameters (LOD, LOQ, and linear dynamic range) should be established in the specified food matrix.

Future studies must focus on the fabrication process to significantly improve intra- and inter-electrode reproducibility. With the use of advanced manufacturing techniques and automated procedures, variety of electrodes-catalysts may be consistently produced, lowering the overall production costs and facilitating manufacturability.

3D-printing is a hot topic with promising characteristics in electroanalysis as it provides an economical and robust alternative to current strategies. 3D-printing allows for a diverse range of materials (e.g., conductive polymers, nanoparticle- and metal-based formulations) where the cost of high-quality filaments can be higher than with traditional inks used in screen-printing.[142] Novel electrochemical devices for BPA should be manufactured by 3D-printing in order to exploit to the full the advantages of this technology (great flexibility in the sensor's design in terms of size and geometry, high batch-to-batch precision and uniformity). Moreover, 3D-printing reduces waste generation and it has enormous potential for large-scale fabrication in a mechanized/automatized process. Robust and large-scale produced 3D-printed electrochemical sensors combined with chemical modifiers can be envisaged for the development of improved BPA electroanalytical systems.

New approaches should be applied in order to manufacture integrated electrochemical devices for multi-component detection. A sensor that can simultaneously monitor the levels of BPA and other bisphenol analogues (i.e., B, C, E, F, G, M, P, S, Z, AF, AP, BP, PH, TMC, which are suspected to have endocrine-disrupting properties similar to BPA) in a single miniaturized and user-friendly device will offer the promise of practical applications. Multichannel electrochemical platform which supports both parallel signal stimulation and on-line electrochemical analysis functions will provide a more complete picture of exposure to these compounds.

Artificial intelligence (AI) algorithms should be introduced to power the electroanalytical methods. Convergence of machine learning and multimodal electrochemical readout can help achieve multiplexing with high accuracy using a single device/material. A database containing inputs and outputs may be used to build a regression model to resolve the overlapping peak problem in multiplexed quantification with a low technical threshold. However, to date AI-assisted electrochemical sensing is still in the initial stage.

Beyond researchers, there is also a significant role for editors and reviewers of high impact journals. In order to satisfy the novelty requirements to publish in these journals, research teams are pushed towards complicated designs, more complex methodologies, and exotic electrode materials. If editors/reviewers can evaluate on merit the value of real-world applicability of the sensor system, even if its design is not particularly innovative, this could considerably accelerate the progress in this field.

7. Conclusion Remarks

This study covers the current state of electrochemical sensing technology as well as its application as a reliable analytical platform for the detection of BPA in foodstuffs. Electrochemical methods are affordable, dependable, and appropriate for field use. Therefore, transforming an electrochemical sensor into a portable device from lab-scale research is the driving force for the commercialisation of these devices. Nevertheless, despite the substantial advancements made, a number of enhancements are still required to transfer electroanalytical technology for BPA determination from the lab to the real world.

Some shortcomings, such as unsatisfactory long-term stability, electrode fouling and problematic device-to-device reproducibility make the proposed sensor platforms difficult to apply commercially. Furthermore, conflicting requirements, such as the use of appropriate functional materials, cost effectiveness, simplicity, and speed in fabrication, need to be considered. Therefore, creating high-performing electrodes with simple procedures is crucial to develop electrochemical sensor devices that are commercially aviable. Recent achievements in microfabrication, multiplexing, electronics integration, and machine learning can help realize the potential of these sensor platforms. In the foreseeable future researchers should be utilizing the ability of artificial intelligence to process large datasets quickly and accurately in order to optimize the overall performance of the electrochemical sensors, addressing the issue of multi-component detection.

In conclusion, electrochemical methods are useful instruments that have the potential to revolutionize food quality control. Nowadays, smartphones, 5G communication and cloud computing allow the digitalization of food quality information obtained by integrated electrochemical sensors. The cross-disciplinary integration inspires innovative thinking and provides new solutions, thus promoting innovation and rapid development in the field of food electroanalysis.

Declaration of competing interest

The author declares that she has no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Abbreviations

AB, acetylene black; **ABPE**, acetylene black paste electrode; **APTES**, 3-aminopropyltriethoxysilane; **BOMC**, boron-doped ordered mesoporous carbon; **BPA**, bisphenol A; **BW**, body weight; **BZPY**, 2,6-bis(2-benzimidazoyl) pyridine; **CAS**, casein; **CB**, carbon black; βCD, β-cyclodextrin; **CdTBrPP**, [(5,10,15,20-tetrakis(*p*-bromophenyl) porphyrinato] cadmium(II); **CF**, carbon felt; **CHIT**, chitosan; **CNFs**, carbon nanofibers; **CNTs**, carbon nanotubes; **CPE**, carbon paste electrode; **EDC**, endocrine disrupting compound; **eMIPS**, molecularly imprinted electrochemical sensors; **ERGO**, electrochemically reduced graphene oxide; **GC**, gas chromatography; **GCE**, glassy carbon electrode; **GNPs**, graphene nanoplatelets; **GO**, graphene oxide; **GPE**, graphite paste electrode; **GR**, graphene; **IL**, ionic liquid; **LC**, liquid chromatography; **LOD**, limit of detection; **LOQ**, limit of quantification; **LSV**, linear sweep voltammetry; **MGCE**, magneto-actuated glassy carbon electrode; **MIPs**, molecularly imprinted polymers; **MMIPs**, magnetic MIPs; **MS**, mass spectrometry; **MWCNTs**, multi-walled carbon nanotubes; **NPs**, nanoparticles; **NPGL**, nanoporous gold leaf; **OVs**, oxygen vacancies; **PANI**, polyaniline; **PC**, polycarbonate; **PCOS**, *polycystic ovary syndrome*; **PPy**, polypyrrole; **RGO**, reduced graphene oxide; **RSD**, relative standard deviation; **SCE**, saturated calomel electrode; **SEM**, scanning electron microscope; **SH-*β*-CD**, thiolated beta-cyclodextrin; **SHS-LPME** switchable hydrophilic solvent-based liquid-phase microextraction method; **SML**, specific migration limit; **SNGCE**, sonogel-carbon electrode; **SPE**, screen printed electrode; **SPGE**, screen printed graphite electrode; **TDI**, tolerable daily intake; **ThTAA**, 2-(4-((3-(trimethoxysilyl) propylthio)methyl)1-*H*-1,2,3-triazol-1-yl) acetic acid.

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Povzetek

Sodobne tehnologije izdelave nanodelcev nudijo v kombinaciji z elektrokemijskimi tehnikami prednosti, kot so izjemna občutljivost, nizke meje zaznave, minimalne energetske zahteve, preprostost in nizka cena, kar naredi elektrokemijske senzorje pomembne pri presojanju kvalitete živil. Omejeno število neencimatskih elektrokemijskih senzorskih platform za bisfenol A (BPA), ki bi bile uspešno uporabljene za oceno varnosti hrane in pijače, priča, da živilski vzorci predstavljajo precejšen izziv in da je potrebno še izboljšati analizni odziv teh naprav. Ta pregledni članek sistematično raziskuje prispevek različnih funkcionalnih nanomaterialov, kovinsko-organskih ogrodij, ionskih tekočin in molekulskega vtisnjenja k izboljšanju odziva senzorjev. Prinašamo kritično razpravo o najnovejših zanimivih inovacijah ter najbolj obetavnih elektrokemijskih napravah za analizo BPA. Članek naslavlja elektrokemijske vidike in izzive pri izdelavi senzorjev za BPA ter raziskuje inovativne rešitve in s tem ponuja vpogled v prihodnje smeri razvoja, kar omogoča nadaljnji napredek na tem področju.