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Mini Review

Electroanalytical Sensors for Synthetic Food Colorants Determination in Beverage. Period 2010-2018

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Synthetic colourants are widely used in the food industry, especially in beverages. The excessive intake of synthetic colourants can be harmful to human health. Therefore, it is necessary to establish a simple and rapid means of their analysis and detection for food safety management. Electrochemical analysis is a fast, safe and low-cost method. In this review, we summarized the literature on electrochemical detection of synthetic colourants from 2010 to 2018 and found that sensor design is primarily based on carbon materials. In this review, we also compare the properties of different materials and discuss the future directions of this field.

Keywords: Beverage; Electrochemistry; Food chemistry; Sensor; Analytical chemistry

1. INTRODUCTION

Colourants are a kind of food additive, which can make food coloured or change the colour of food. There are two kinds of colourants currently used in the food processing industry. One is natural colourants, and the other is synthetic colourants. Natural colourants mainly come from animals and plants. Compared with synthetic colourants, natural colourants have the characteristics of nutrient conversion, high safety, non-toxicity and no side effects [1-6]. Generally, natural colourants are relatively safe. However, the chemical structures of most natural colourants contain unsaturated double bonds and other oxidizable groups, which can be oxidized under the action of oxygen and easily fade [7-13]. In addition, natural colourants are also very sensitive to metals, light, heat, pH and other factors, resulting in poor stability [14-19]. The extraction cost of natural colourants is relatively high, which limits their applications. After extraction and purification, their properties may be different from the original form, so natural colourants are not necessarily harmless. Artificial synthetic colourants are essentially tar colourants. Tar colourants are made from coal tar components such as benzene, toluene and naphthalene, so they are unsafe or harmful to the human body. Compared with natural colourants, tar colourants exhibit good stability, high resolution, easy colouring, bright colours, and can be arbitrarily toned. Most importantly, they are cheap, easy to use and easy to preserve [20-26]. Like other food additives, synthetic edible colourants require strict toxicological evaluation for safe use, including assessment of their chemical structure, physical and chemical properties, purity, existing form in food, degradation process, degradation products, retention distribution, metabolic transformation and excretion in tissues and organs. At present, many countries have strict control over the amounts, types and scope of application of edible artificial synthetic colourants. Therefore, it is necessary to effectively detect and monitor the contents of synthetic colourants in food.

The detection of synthetic colourants can be achieved using different methods. High-performance liquid chromatography (HPLC) is one of the most commonly used detection methods, offering the advantages of rapid analysis and high separation efficiency. It is often used for the separation and analysis of compounds with high boiling points, macromolecules, strong polarity and poor thermal stability. It is widely used in the detection of synthetic colourants. Ma et al. [27] demonstrated the detection of water-soluble synthetic colourants, such as sunset yellow, lemon yellow and amaranth, by HPLC. Yoshioka et al. [28] established a method for the determination of 40 synthetic colourants in beverages and candies by HPLC. Wu et al. [29] established a method for the determination of six synthetic colourants in beverages by HPLC. Long et al. [30] analysed six synthetic colourants in food by HPLC. The disadvantages of HPLC are its high cost, complex operation and long analysis time, which are not suitable for rapid on-site detection.

Spectrophotometry has many advantages, such as simple instrumentation, convenient operation and rapid determination. It offers great developments and applications in the field of colourant detection. Dinc et al. [31] reported the detection of azo colourants in beverages, such as sunset yellow, lemon yellow and temptation red, by spectrophotometry. Vidotti et al. [32] established a spectrophotometric method for simultaneous determination of several food colourants. Unsal et al. [33] established a sensitive method for the determination of sunset yellow by spectrophotometry. Turabik et al. [34] used spectrophotometry to determine two synthetic colourants at the same time. Spectrophotometry is widely used in the detection of colourants. The shortcoming is that the sensitivity and selectivity of the method are not high, and it is not suitable for rapid detection with high efficiency and sensitivity.

Thin-layer chromatography (TLC) is one of the commonly used experimental methods for rapid separation and analysis. Because of its simple operation and fast analysis time, TLC is often used for the analysis and detection of food colourants. Soponar [35] used TLC to analyse and determine the colourants in several foods. Sherma et al. [36] established a TLC method for the determination of colourants in food and agricultural products. Ford et al. [37] established several methods for the determination of azo colourants by TLC. Rhee et al. [38] used TLC to analyse and detect several common colourants. Van Berkel et al. [39] used TLC to detect azo colourants commonly used in food. The disadvantages of TLC are its low sensitivity and accuracy. It needs to be used in cooperation with various detection methods to improve the sensitivity of detection.

Capillary electrophoresis (CE) has become one of the fastest-growing liquid phase analysis methods in recent years. It offers the advantages of high sensitivity, fast analysis speed and simple sample pretreatment. It can simultaneously separate and determine many components; thus, it has great potential in the detection of food colourants. Frazier et al. [40] used CE to analyse and detect azo colourants in beverages at the same time. Huang et al. [41] used CE to analyse and detect eight kinds of colourants in milk. Chiu et al. [42] obtained good results in the determination of colourants in bulk samples by CE. Del Giovine et al. [43] established a method for the determination of synthetic colourants in ice cream by CE. Jager et al. [44] used CE to separate and determine several food colourants. The disadvantage of capillary electrophoresis is its low reproducibility in sample separation.

The electrochemical method is an analytical method for studying the electrochemical properties of substances in solution. It has the advantages of high sensitivity and accuracy, simple instrumentation and equipment, and fast and convenient operation [45]. Chemically modified electrodes are a frontier field of electrochemistry. By affixing molecules, ions and polymers with excellent chemical properties on the surfaces of electrodes, the design and improvement of electrodes can be achieved and the selectivity and sensitivity of the methods can be improved [46]. Because synthetic colourants contain active groups such as N = N and C = O, they can be oxidized and reduced on the electrode under certain conditions [47]. Therefore, a variety of modified electrodes have been established to determine synthetic colourants in food by electrochemical methods. In this review, we reviewed the detection of the use of colourants in beverages according to different kinds of electrode modifiers based on the literature published from 2010 to 2018. At the same time, we discuss the future development trends of this field.

2. NOBLE METAL NANOPARTICLES

Noble metal nanomaterials are an important class of nanomaterials. Gold, platinum, silver, and palladium-based nanomaterials have attracted more attention due to their excellent corrosion resistance and catalytic properties. Their nanostructures also show potential research and application value in many fields due to their unique optical, electrical, physical and catalytic properties. Until now, noble metal nanomaterials have been successfully reported to have nanorod, nanoflake, nanowire, dendritic, star and many other morphologies. They have been widely used in biomedicine, electronics and optical devices. Noble metal nanomaterials have remarkable advantages in the construction of electrochemical biosensors, such as accelerating electron transfer capability, effectively improving response speed and increasing response current. As sensitive elements, they exhibit good electrocatalytic performance, good biocompatibility and effective immobilization of enzymes. Furthermore, they have a large specific surface area, which can significantly improve the sensing performance and sensitivity. The useful and fascinating characteristics have stimulated people’s interest in fabricating immunoassays by employing gold nanoparticles as the skeletons for carrying antibodies or antigen biomolecules. Generally, gold nanoparticles can bind substances to build up efficient electrochemical transducer surfaces containing ionic liquids [48], metal nanoparticles [49], mesoporous silica [50], amino acids [51], proteins [52], polymers [53], carbon nanoparticles [54], and electron mediators [55]. Table 1 summarizes recently developed noble metal nanoparticle-based electrochemical sensors for synthetic colourants determination. It can be seen that only several reports focused on the direct use of noble metal nanoparticles for synthetic colourants determination. This can be attributed to some disadvantages of the noble metal nanoparticles, such as propensity for aggregation and poor anti-fouling ability. Therefore, the use of the noble metal nanoparticles for electrochemical sensor design is commonly combined with a substrate, such as select carbon materials.

Table 1. Recently developed noble metal nanoparticle-based electrochemical sensors for the determination of synthetic colourants.

Materials	Method	Target	Reference
Palladium-ruthenium nanoparticles	DPV	Sunset yellow	[56]
Au NPs	DPV	Sunset yellow	[57]
Silver-poly (L-cysteine)	DPV	Sunset yellow	[58]
Au NPs	DPV	Methylene blue	[59]

3. CARBON BASED MATERIALS

Carbon, which is located in the IVA family of the second cycle of the periodic table, is one of the most widely distributed basic elements in nature. It has many hybridization states including sp, sp^1, sp^2, and sp^3. Because of its special structure and properties, carbon materials have always represented a research hotspot in the fields of sensors, environmental detection, energy storage and energy conversion. Carbon materials primarily include graphene, graphene oxide, fullerene, diamond, activated carbon and carbon nanotubes. Among them, carbon nanotubes and graphene, as the most potential representatives of the new carbon materials family, have a series of unique properties. Expanded graphite is a relatively new carbon material made by immersing natural flake graphite in a bath of chromic acid, then concentrated sulfuric acid, which forces the crystal lattice planes apart, thus expanding the graphite [60].

Unique properties of carbon nanotubes are frequently exploited in chemical sensors. They are characterized by high strength and, at the same time, by good deformational stability [61]. The main classification of nanotubes is carried out according to the number of constituent layers. SWCNTs - the simplest form of nanotubes - have a diameter of approximately 1 nm, while the length may be many thousands of times larger. MWCNTs are composed of several graphene layers in a concentric cylindrical arrangement. MWCNTs differ from SWCNTs by possessing a broader variety of shapes and configurations [62]. Both SWCNTs and MWCNTs are widely used for different types of electrode modifications.

Graphene is another promising carbon nanomaterial based on a two-dimensional structure consisting of carbon atoms arranged in hexagonal lattices [63]. Graphene oxide (GO) is a kind of modified graphene in which carbon atoms can be linked to hydroxyl or epoxy groups and the surface boundary can be modified with carboxyl and carbonyl groups. Due to such attractive properties as electrical conductivity, mechanical strength, and high specific surface area, GO finds its application in the fabrication of chemically modified electrodes [64].

For example, He and co-workers reported a TiO_2-reduced graphene oxide composite for the detection of tartrazine [65]. Tartrazine is a water-soluble synthetic colourant. It is mostly used for colouring foods, beverages, medicines, cosmetics, feeds, tobacco, toys and food packaging. As shown in Figure 1, the oxidation of tartrazine exhibited an irreversible process involving one electron and one proton. Table 2 summarizes recently developed carbon materials-based electrochemical sensors for synthetic colourants determination.

Table 2. Recently developed carbon materials-based electrochemical sensors for the determination of synthetic colourants.

Materials	Method	Target	Reference
Graphene wrapped-phosphotungstic acid hybrid	DPV	Sunset yellow and tartrazine	[66]
Multiwall carbon nanotube	DPV	Amaranth	[67]
CuO/single-wall carbon nanotube	DPV	Amaranth	[68]
Reduced graphene oxide	CV	Sunset yellow and tartrazine	[69]
Expanded graphite	DPV	Sudan I	[70]
IL-reduced graphene oxide–Au NPs	CV	Sudan I	[71]
Graphene-mesoporous TiO_2	CV	Sunset yellow and tartrazine	[72]
Pt/carbon nanotubes	CV	Sudan I	[73]
β-cyclodextrin-coated PDDA-functionalized graphene	LSV	Sunset yellow and tartrazine	[74]
Au NPs/graphene	DPV	Sunset yellow	[75]
Exfoliation of graphite	DPV	Tartrazine	[76]
CHIT/GO/MWCNTs/AuNPs	DPV	sunset yellow	[77]
Graphene oxide and multi-walled carbon nanotubes	LSV	Sunset yellow and tartrazine	[78]
Graphene-mesoporous TiO_2	DPV	Ponceau 4R and allura red	[79]
Reudced graphene oxide	DPV	Pyrosine	[80]
Fe_3O_4@SiO_2/MWCNTs	DPV	Sunset yellow and tartrazine	[81]
Single-walled carbon nanotube-TiN	DPV	Amaranth	[82]
Au-Pd-reduced graphene oxide	DPV	Sunset yellow	[83]
Multi-walled carbon nanotubes	DPV	Quinoline yellow	[84]
Multi-walled carbon nanotube/chitosan	LSV	Sudan I	[85]
Ionic liquid-expanded graphite	LSV	Brilliant blue and tartrazine	[86]

Polypyrrole (ppy) and single-walled carbon nanotube	CV	Sunset yellow	[87]
Graphene-ZnSe quantum dots	CV	Sudan I	[88]
Poly(Sodium 4-Styrenesulfonate)-graphene-Co$_3$O$_4$	DPV	Amaranth	[89]
PDDA-graphene-Pd	DPV	Sunset yellow and tartrazine	[90]
Expanded graphite paste	CV	Amaranth	[91]
Pd-doped graphene	DPV	Amaranth	[92]
Bimetallic nanoparticle functionalized graphene	DPV	Sunset yellow	[93]
Au-IL-reduced graphene oxide	DPV	Sunset yellow and tartrazine	[94]
MnO$_2$-reduced graphene	CV	Sunset yellow	[95]
Graphene	DPV	Sudan IV	[96]

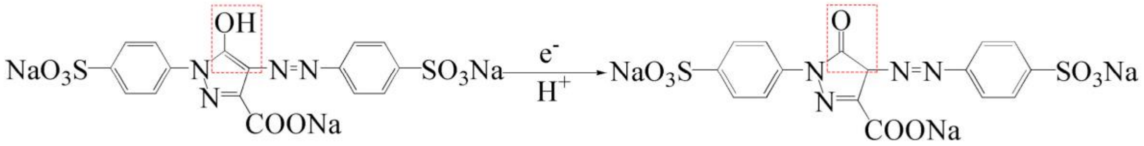

Figure 1. Oxidation mechanism of tartrazine.

4. MOLECULAR IMPRINTED AND POLYMER BASED ELECTROCHEMICAL SENSORS

Molecular imprinting technology (MIT) is a technique for synthesizing polymers containing imprinted sites complementary to template molecules in structure, size and functional groups. In the presence of template molecules, functional monomers copolymerize with cross-linking agents to form highly cross-linked polymers, and then remove template molecules, resulting in recognition sites complementary to template molecules. The polymer can select and recognize template molecules from molecular structure analogues. Compared with other recognition systems, MIPs have many unique properties, including low cost, simple preparation and excellent reproducibility. They can be used in harsh environments and attract the attention of researchers. Polymers formed by MIT can specifically adsorb template molecules. The principle can be explained in the following three steps: (1) Functional monomers interact with template molecules in appropriate solvents to form reversible monomer-template molecular complexes. (2) Functional monomers interact with crosslinkers to form a highly crosslinked polymer around template molecules. (3) The imprinted sites left by the template molecule after elution can selectively recombine with the template molecule.

Several works have been dedicated to the use of polymers for determination of synthetic colourants. For example, Sakthivel et al. [97] reported the synthesis of poly(3,4-

ethylenedioxythiophene)-terbium hexacyanoferrate for the determination of tartrazine. Li and co-workers demonstrated the use of poly(p-aminobenzene sulfonic acid) for sensitive Sudan I detection [98]. Acetylene black nanoparticle-modified electrode has been used for the determination of Ponceau 4R and tartrazine [99]. Table 3 summarizes recently developed molecular imprinted-based and polymer-based electrochemical sensors for synthetic colourants determination. For dye determinations by DPV and/or SWV, MIPs are used alone or in combination with other materials [100], e.g., molecularly imprinted polypyrrole alone [101], MIP polypyrrole/SWCNTs [102], MIP−rGO-IL/GCE [103], and MIP–MWCNTs-IL@PtNPs/GCE [104]. The LOD values of tartrazine and sunset yellow by means of such sensors vary from 3·10−9 to 8·10−9 mol/L. Nevertheless, the practical application of these promising sensors is still limited by their somewhat complicated and time-consuming preparation.

Table 3. Recently developed molecular imprinted-based and polymer-based electrochemical sensors for the determination of synthetic colourants.

Materials	Method	Target	Reference
MIT-multiwalled carbon nanotubes - ionic liquid supported Pt NPs	DPV	Tartrazine	[105]
MIT-functionalized multi-walled carbon nanotubes	CV	Sunset yellow	[106]
MIT- Pd-Cu-graphene	DPV	Amaranth	[107]
MIT	CV	Tartrazine	[108]
Poly(L-phenylalanine)	CV	Sunset yellow and tartrazine	[109]
Poly(crystal violet)	LSV	Sunset yellow	[110]

5. OTHER ELECTROCHEMICAL SENSORS

In addition to the abovementioned materials, some other materials have also been applied for synthetic colourants determination. For example, Zhang and co-workers demonstrated the use of alumina microfibres as sensitive electrode for the determination of Ponceau 4R [111]. Several other reports used alumina as modifier determination of amaranth [112] and sunset yellow [113]. Medeiros et al. [114] reported the use of boron-doped diamond electrode for the determination of tartrazine, sunset yellow and brilliant blue. Wang and co-workers synthesized a montmorillonite calcium-modified electrode for the determination of Ponceau 4R [115]. Graphitic carbon nitride has also been used for the determination of tartrazine [116]. La^{3+}-doped Co_3O_4 has also been used for the detection of Sudan I [117].

One of the modern trends in electrochemistry is the search for new materials with useful physicochemical properties to be used as solvents, base electrolytes, modifiers or proper electrode materials. From this point of view, ionic liquids (ILs) that are liquid at room temperature are very promising [118]. In response to their unique properties, such as thermal stability, high conductivity, inflammability, low volatility, and ionic nature, these compounds are extensively used for fabrication of electrochemical sensors [119]. The most important advantage of ILs is the possibility of varying their physico-chemical properties by replacing the anion or cation of the salt. Typically, for improving conductivity and electron transfer promotion, ILs are used in combination with various compounds: graphene [120], expanded graphite [121], GO-MWCNTs [122], and metal nanoparticles [123]. The widespread practical use of ILs is still slightly hindered by their relatively high price.

Metal and metal oxides have been selected as stable materials for electrode surface modification. Dorraji and Jalali demonstrated a sensitive electrochemical sensor for detection of sunset yellow and tartrazine based on the ZnO/cysteic acid nanocomposite [124]. Ji and co-workers reported a Cu-BTC framework-based electrochemical sensor for the detection of sunset yellow and tartrazine [125]. Bismuth has been selected for electrode surface modification and subsequently used for the detection of sunset yellow, carmoisine and tartrazine [126,127]. Huang and co-workers reported MnO_2 microspheres/chitosan for Ponceau 4R detection [128].

6. CONCLUSIONS AND OUTLOOKS

In this review, we summarized the studies on the electrochemical detection of synthetic colourants published from 2010 to 2018. Three types of materials, including noble metal nanomaterials, carbon materials and polymer materials, are described in detail. In terms of the number of literature publications, carbon materials have been widely used in the electrochemical detection of synthetic colourants, especially for the design of carbon-based nanocomposites. Molecular imprinting technology can be used to detect synthetic colourants more specifically, but the entire preparation process is tedious. Current synthetic colourant sensors detect at most two pigments at the same time, but beverages often contain several colourants. Therefore, one of the future research directions is to build sensors that can detect multiple colourants at the same time. In addition, improving the stability, specificity and repeatability of electrochemical sensing is also the key to the research.

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