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Review

Recent advances in developing modified electrode interface for sensing tartrazine in real samples: a brief review

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Abstract

Many businesses, including food, textile, pharmaceuticals, and cosmetics sectors, employ tartrazine or other synthetic dyes. These dyes cause severe health effects if used or consumed frequently. Up to now, researchers have introduced many electrochemical methods for the determination of tartrazine. Among them, voltammetric methods were developed, mostly using effective and selective electrodes with modifiers to accurately sense tartrazine in baking products such as soft drinks, candy, jelly, and other beverages. In this review, we have covered several works on carbon-based, graphite-based and gold electrodes that have been offered for tartrazine detection with good recovery rates and detection limits. These works may aid future research and development toward more accurate tartrazine detection.

Keywords

Azo dyes; electrochemical sensors; modified electrodes; voltammetry techniques; foodstuff samples

Introduction

Soft drinks and other foodstuffs are prepared with flavorings and certain food colorings or dyes to make their appearance more appealing. The companies give importance to the visual aspect of the food throughout the production process of soft drinks, juices, jelly, candy, alcoholic and non-alcoholic beverages, desserts, and other sweeteners. These foods may contain synthetic or natural hues. Natural dyes come from fruits, vegetables, or other biological sources, and they are safe and healthy [1,2]. A broad category of dyes known as "synthetics" are produced chemically to impart persistent and vivid hues; they are toxic and carcinogenic. Synthetic dyes are present in different forms, like, azo, anthraquinone, and phthalocyanine dyes [3,4]. The printing and textile industries,

as well as the food, pharmaceutical, and cosmetics industries, use these dyes. They are less expensive than natural dyes and have excellent stability against light, oxygen, and pH variations [5].

Most artificial food dyes are called azo dyes. In the food sector, they are referred to as yellow and red dyes. The examples of yellow dyes are tartrazine and sunset yellow. Azo dyes have azo groups, also known as n-groups, as well as diazo and triazo groups, in their structures [6]. The baking, pharma, and textile industries all employ TZ. Synthetic azo dyes are frequently preferred by food industries over natural colorants because they provide intense color in less time without any unpleasant flavors, have a wide color range, are more affordable than other dyes, have versatile application areas, and can be used with different dyeing and printing techniques. However, they can also leave aromatic amines due to azo bond reduction and cleavage, which can negatively impact animal life and the biosphere. Azole dyes, which are carcinogenic for both humans and rodents, are made using aromatic amines like benzidine and 2-naphthylamine [7,8].

The azo dye called tartrazine (TZ) is frequently found in foods like cakes, candies, juices, fizzy drinks, soft drinks, jelly, and more. Sunset yellow, amaranth, allura red, ponceau, brilliant blue, carmoisine, acid violet 7, patent blue V, and other dyes are among those that react with TZ [6]. Numerous techniques, including spectrometry, capillary electrophoresis, chromatography, and electrochemical methods, can be used to detect these dyes in addition to TZ [9]. Because of their higher sensitivity, ease of handling, less wait, quick analysis times, and affordability, electrochemical techniques have drawn a lot of interest, showing a promise for use in food safety analysis [10].

Electrochemical methods for the detection of TZ offer several advantages over traditional techniques like spectrometry and chromatography. These include higher sensitivity, ease of handling, shorter analysis times, and lower costs [11,12]. Electrochemical sensors can be modified with various nanoparticles and compounds to enhance their selectivity and efficiency, making them highly effective for detecting TZ in food products with minimal interference from other substances [13]. Voltammetric methods like cyclic voltammetry (CV) and differential pulse voltammetry (DPV) are usually used to determine the linear concentration range, limit of detection (LoD), recovery rates of dyes, and much more [14-16]. Several electrochemical approaches with different working electrodes have been employed to detect TZ in food samples. The electrodes can be carbon paste electrode (CPE), gold electrode, glassy carbon electrode (GCE), graphite paste electrode (GPE), screen printed electrode (SPE), pencil electrodes, etc. [17-20]. These electrodes are frequently modified to enhance the efficiency and accuracy of the results. Modifiers can be nanoparticles such as zinc, graphite, gold, palladium, nickel, cobalt, acetylene black, chitosan, calcium, copper, and iron, where these nanoparticles may be present alone or in the complex of other nanoparticles [21]. Carbon nanotubes that can be multiwalled (MWCNTs) or single-walled (SWCNTs) nanotubes, and also metal-organic frameworks (MOFs), are frequently used as modifiers [22]. These modifiers are typically prepared by polishing and cleaning the glassy carbon electrode (GCE), followed by coating it with MWCNTs dissolved in an organic solvent. Once the solvent evaporates, a binder is applied, and the electrode is then dried and stored for future use in electrochemical applications. [22]. Since various ions may interfere with dye sensing in practical samples, a highly selective electrochemical sensor is essential for accurate analysis.

Electrochemistry of tartrazine

Tartrazine is an organic dye with an azo group, which is yellow, and is an approved artificial food colorant frequently present in the food and pharmaceutical industries [6]. The chemical formula of TZ is $C_{16}H_9N_4Na_3O_9S_2$, while its chemical structure is drawn in Figure 1.

Figure 1. Chemical structure of tartrazine dye

Because TZ contains hydroxyl and azo groups, it possesses features related to both reduction and oxidation. Oxidation of TZ is an irreversible process where the OH oxidizes to the RCOR group with only one electron transferred [23,24]. As seen in Figure 2, this process takes place on the surface of the working/modified electrode and is controlled by both adsorption and diffusion [25].

Figure 2. Oxidation reaction of tartrazine dye

Kobun Rovina *et al.* [26] explored the electrochemistry of TZ by developing a modified glassy carbon electrode (GCE) using a nanocomposite film of chitosan (CHIT), graphene oxide (GO), multiwalled carbon nanotubes (MWCNTs), and gold nanoparticles (AuNPs). This CHIT/GO/MWCNTs//AuNPs/GCE composite showed improved electrocatalytic activity achieved through a uniform film distribution and increased surface area, which maximized the number of active sites on the electrode interface. The oxygen groups on GO promoted electrostatic adsorption of AuNPs, which, along with the high conductivity of MWCNTs and charge-carrying properties of AuNPs, significantly enhanced the oxidation peak signals of TZ compared to a bare electrode. Studies using CV technique confirmed these effects, noting that the modified GCE optimized TZ detection at pH of 7 and potential of 0.3 V vs. Ag/AgCl within 30 seconds. The sensor exhibited a linear TZ detection range from 10 to 100 mg/mL ($r^2 = 0.99037$) with a sensitivity of 0.018 μ A/ μ M and low limits of detection and limits of quantification at 1.45 mg/mL and 4.83 mg/mL, respectively.

Electrodes

Electrodes are commonly used to determine analytes and can be reactive and inert electrodes. Reactive electrodes are copper, zinc, silver, lead, etc., while inert electrodes are used as standard electrodes in experimentation and can be made of platinum, graphite, gold, carbon, etc. Usually, the inert electrodes have been modified by nanoparticles and their complexes, nanotubes, and microfibers of different compounds or metals. These modifiers enhance the sensor signals by giving more accurate results compared to bare or unmodified electrodes.

Carbon-based electrodes

Carbon-based electrodes are the most commonly used electrodes for the detection of dyes and other analytes as they are of low cost and have a high surface area. There are different forms of

carbon electrodes, such as carbon paste electrodes (CPE), glassy carbon electrodes (GCE), screenprinted carbon electrodes (SPCE), carbon ceramic electrodes, and others [17-22]. Each one of these electrodes has its unique advantage and features to determine TZ and other analytes.

Carbon paste electrode

Carbon paste electrode (CPE) is a low-cost electrode with a high surface area that can be further enhanced by modifying with surfactants, polymers, and nanoparticles like gold or zinc oxide. The modified electrodes could show detection limits in the nanomolar range [27-36]. Karim-Nezhad et al. [10] have proposed an electrochemical device to determine TZ in soft drinks using poly(p-aminobenzenesulfonic acid) (Pp-ABSA) and zinc oxide (ZnO) nanoparticles (ZnONPs) modified carbon paste electrode (CPE). First, ZnONPs were prepared using the thermal technique, and TEM and SEM analysis confirmed particle sizes. By using CV, ZnONPs/CPE was then characterized at potentials between -0.6 and +1.8 V vs. Ag/AgCl reference electrode. When compared to the bare electrode, the ZnONPs/CPE nanocomposite produced a notable improvement in the electrode surface structure. The anodic peak of TZ rises upon the introduction of the modified electrode, as shown in Figure 3, indicating that this electrode is a highly effective TZ sensor. The voltammetric responses of TZ showed a linear response in the concentration range of 0.349 to 5.441 μM, a detection sensitivity of 2.2034 μ A/ μ M, and a LoD of 80 nM. This electrode was utilized to measure TZ in actual samples, yielding satisfactory results and showing no interference from possibly interfering species. When the recovery value is greater than 98 %, it may be concluded that a synthetic sensor used to track TZ levels in actual samples is a reliable and simple method.

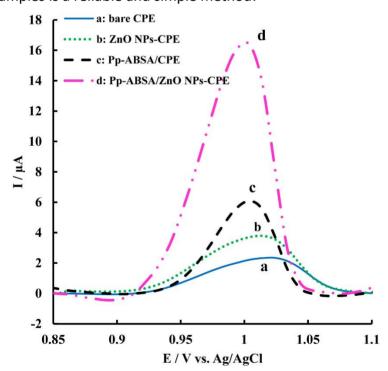


Figure 3. DPV curves for 1.49 μ M Tz in 0.1 M PBS, pH 3 at different types of CPE. Reprinted with the permission of Ref. [10]

Sensing TZ in drinks using a CPE that has been altered with gold nanoparticles (nAu-CPE) was proposed by Ghoreishi *et al.* [37]. Initially, absorptive UV-Vis spectroscopy created and characterized gold nanoparticles (nAu), with a maximum absorption peak measured at 520 nm. Afterward, graphite powder, colloidal Au, and paraffin oil were used to make the modified CPE [37]. The voltammetric method (DPV) measured the TZ oxidation peak at 1.02 V vs. Ag/AgCl. Modified

electrodes were found useful for detecting TZ in genuine samples. Soft drink samples may also be tested with this modified electrode. The results indicated that concentrations of interfering species have no discernible impact on the recovery rate of 104 %, with a detection limit of 2 nmol/l. These findings suggest that this method for TZ determination is both sensitive and effective [37].

To detect TZ in soft beverages, Zhang et al. [38] developed a novel sensor that uses modified alumina microfibers on carbon paste electrodes. Al(NO₃)₃ was used in a hydrothermal reaction with water and urea to create the alumina microfibers, which were then filtered, autoclaved for a day, and calcined for two hours at 500 °C [38]. The produced alumina microfibers were examined using SEM and TEM techniques, revealing porous surface features that were later applied to carbon paste electrode. Oxidation peak currents of TZ were measured using the voltammetric method at 1.01 V against a saturated calomel reference electrode (SCE), with a potential scan rate of 40 mV/s. Alumina microfibers were very porous and thus had a wide surface area, resulting in increased oxidation signals, LoD of 2 nM, and high accumulation efficiency to TZ. The electrochemical results obtained by alumina microfibers modified electrodes were in good agreement with the values acquired by the HPLC method [38].

Ghoreishi *et al.* [39] investigated the electrochemical determination of TZ using a chemically modified CPE enhanced with MWCNTs. The modified electrode, prepared with 78 % graphite powder and MWCNTs, provided a high surface area and improved adsorption capacity at the electrode interface. For TZ detection, differential pulse voltammetry (DPV) was employed, showing a significant oxidation peak at 1.06 V vs. Ag/AgCl sat. KCl reference electrode. In addition to DPV, CV and square wave voltammetry (SWV) techniques were used, providing complementary data on the electrochemical behavior of TZ. The optimized sensing conditions, particularly at pH 2.0, resulted in high sensitivity and low detection limits in the nanomolar range, making the MWCNT-CPE highly suitable for TZ analysis in real samples such as cool drinks. The excellent performance of this electrode, including its ease of preparation, regeneration, and long-term stability, emphasizes its potential for reliable, routine monitoring of synthetic dyes like TZ in food products.

By creating a metal-organic framework (MOF) sensor, Darabi *et al.* [40] prepared (Cu-BTC MOF), where BTC denotes 1,3,5-benzene tri carboxylate. Its mixture with 1-ethyl-3-methylimidazolium chloride as an ionic liquid (IL) altered CPE and detected TZ with other pigments in foodstuffs. First, Cu-BTC MOF was created by dissolving copper nitrite and H3BTC in ethanol, autoclaving the mixture, and centrifuging the blue powder result [40]. The structure of the product was then examined using SEM, EDX, XRD, and FTIR surface techniques. The findings showed that Cu-BTC MOF contained 20 to 37 nm porous nanoparticles and a crystal structure. After that, IL (1-ethyl-3-methylimidazolium chloride) was added, and the obtained was applied to the CPE surface. The modified electrode was used to measure TZ and other dyes, where oxidation peak current was increased at concentrations ranging from 0.08 to 900 μ M and LoD of 0.07 μ M. Several voltammetric methods assessed the modified electrode at pH 7. Based on these findings, it was concluded that the modified electrode shows good sensitivity and selectivity, with recovery rates ranging from 82 to 108 % for condiments, 94 to 110 % for candies, and 85 to 98 % for soft drinks. This confirms the effectiveness of the IL/Cu-BTC MOF/CPE sensor in detecting food dyes like TZ [40]. A detailed comparison of the limits of detection for tartrazine, as determined by different CPEs, is presented in Table 1.

Modifier Methods Operating parameters LoD, µmol/L Recovery, % Ref. Gold NPs CV, DPV pH 4.0; t_{acc} = 2 min; 0.6 to 0.8 V 0.017 94.1 [41] TZ-cetryltrimethyl pH 4.2 to 8.1; T = 20 to 55 °C Potentiometry 0.055 95.0 [42] ammonium bromide Response 5 to 8 min Cobalt complex[bis(3,5--dimethyl-1-pyrazolyl) EIS, CV, SWV pH 3.1; $t_{acc} = 30 \text{ s}$ 0.30 and 0.08 [43] >90.0 methane-NN] cobalt (II) 0.283 Poly (glycine) CV, DPV pH 7.0 94.5 to 98.2 [44] pH 2.0; $t_{acc} = 10 \text{ s}$, La₂O₃ and TiO₂ CV, SWV Pulse amplitude 0.05 V; 0.03 [45] microcomposite Frequency 10 Hz; Scan rate 0.1 V/s pH 3.0; $t_{acc} = 30 \text{ s}$; Near 100 Neodymium (III) oxide EIS, CV, SWV 0.02 [46] $E_{ACC} = 0.10 \text{ V}$ (94.5)

Table 1. Literature data on operating parameters and analytical properties of some electrochemical sensors for TZ, based on modified CPE

Glassy carbon electrode

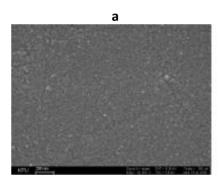
To measure TZ and sunset yellow dyes, Yu *et al.* [47] created a voltammetric sensor based on GCE modified by poly(diallyl dimethylammonium chloride) (PDDA)—dispersed graphene (Gr)—supported Pd nanoparticles. Graphene oxide (GO) was generated by graphite ultrasonication, and 5 mg of PdCl₂ was added with HCl to form H₂PdCl₄. After mixing and centrifuging a dark precipitate of PDDA-Gr-Pd composite was formed [47]. By using XRD and TEM surface techniques, the authors analyzed the composite, and observed development of Gr, PDDA-Gr-Pd composite, and uniformly dispersed PdNPs on the Gr surface, respectively. An infrared lamp was used to dry the produced dispersion after its deposition over a bare GCE surface [47]. Electrochemical impedance spectroscopy (EIS) technique was then utilized to analyze the resistance characteristics of PDDA-Gr-Pd/GCE, demonstrating a notable increase in electrical conductivity compared to bare GCE. The peak current was linear with TZ concentration in the 0.05-8.0 μ mol/L range, and LoD was 5.0 nmol/L. Detection of interfering foreign species was done using DPV, which revealed no significant interference and a recovery value of 95 %.

To modify GCE, Moarefdoust *et al.* [48] created three-dimensional raspberry-like In^{3+}/NiO hierarchical nanostructures doped with indium (In^{3+}/NiO RLHNSs). First, the hexahydrates of nickel and indium chloride ($NiCl_2 \cdot GH_2O$) were divided into ethylene glycol. Next, hydrazine monohydrate ($N_2H_4 \cdot H_2O$) was added to this green solution with NaOH, refluxed at 800 °C, cooled to room temperature, and dried at 600 °C for eight hours [48]. The surface of the In^{3+}/NiO RLHNSs was described by SEM, the arrangement was investigated by XRD, and the EDX method was used to determine its chemical composition. Subsequently, the resulting solid product was dissolved in distilled water using ultrasonication to create a suspension, which was then applied to GCE to obtain the modified electrode. The TZ dye was detected in real samples like edible drinks, jelly, and food colorants using voltammetric methods (DPV, LSV, and CV) in a pH 5.0 solution at the potential scan rate of 50 mV/s. The TZ oxidation peak potential was 1070 mV against Ag/AgCl, sat. KCl reference electrode with a peak current of 2.94 μ A and detection limit of 3 nM [48].

To prepare a graphene layer-wrapped phosphotungstic acid (PTA) hybrid as a modifier on the surface of a GCE, where graphene is utilized as an electron transfer mediator, Gan *et al*. [49] used a one-step electrochemical technique. Using a modified version of Hummer's process, graphene oxide was made from natural graphite flakes. Graphene (GN) was then produced by reducing graphene oxide with NaBH₄. After cleaning the GCE, it was swept in a PTA and GN solution and dried to form GN-PTA/GCE [49]. SEM analysis of the GN-PTA film revealed that the continuous GN layer may firmly

anchor PTA crystals onto the electrode surface, enhancing GN-PTA/GCE repeatability. PTA expands the electrode surface area, while the GN layer serves as a conducting medium for the transport of electrons. CV was used to examine the electrochemical activity of the modified electrode, and the results showed that the electrode could accurately sense the electrochemical reaction. The TZ was detected with good selectivity and high sensitivity using DPV, showing good oxidation peaks with a peak potential separation of about 260 mV. The detection limit was 30.0 μ g/L, and possible interferents did not affect TZ detection in real samples. The recovery value ranged from 98 to 104 % in various soft drink samples, demonstrating the accuracy and viability of the method.

To detect TZ, Gimadutdinova et~al. [50] introduced an electrochemical sensor built on a GCE altered by the presence of MnO₂ nanorods. Utilizing ultrasonication, MnO₂ nanorod suspension was produced from a cetylpyridinium bromide solution. Then, the suspension was applied to the clean GCE surface and allowed to dry. SEM analysis (Figure 4) was used to examine the electrode surface morphology. The porous construction with channels increased the electrode surface area and accelerated the electron transfer rate. Voltammetric behavior of TZ on an electrode modified with MnO₂ nanorods, where the oxidation current is higher than on the bare GCE and the voltammogram shape is improved, was also noted. Two linear ranges of 0.10 to 2.5 and 2.5 to 15 μ M are obtained for TZ through DPV, with a LoD of 43 nM. Recovery values range from 99 to 100 % in various samples at pH 7.0, and a high selectivity response in the presence of interference agents is achieved, demonstrating an accurate and workable sensor for TZ detection [50]. A detailed comparison of the limits of detection for tartrazine, as determined by different GCEs, is presented in Table 2.



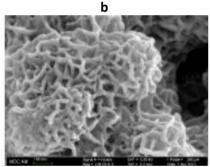


Figure 4. SEM images of (a) bare GCE and (b) MnO_2 nanorods/GCE. Reprinted from Ref. [50] with the permission of MDPI

Table 2. Literature data on operating parameters and analytical properties of some electrochemical sensors of TZ, based on the modified glassy carbon electrode

Modifier	Methods	Operating parameters	LoD	Recovery, %	Ref.
ZnO/cysteic acid	CV, DPV	pH 3 to 9; Accumulation time 120 s	10 ± 0.05 nM	More than 95	[51]
MWNT film	CV, DPV	pH 5.5 to 9; Accumulation time 4 min	0.1 μg / mL	91.2 to 96.5	[52]
Acetylene-black nanoparticle [AB-film]	CV, DPV	pH 5.6 to 7.0; Accumulation time 2 min	0.1 mg kg ⁻¹	92.4 to 104.8	[53]
MWCNTs using 1, 3-dioxolane as a dispersant agent	CV, DPV	pH 2.0 to 10, optimal 7.0; Accumulation time 2 min	0.22 μΜ	101 to 102	[37]
CHIT/graphene oxide/MWCNTs/AuNPs	CV, DPV	pH 6 to 8; Accumulation time 30 s	1.45 mg / mL	94.52 to 109.0	[38]
CuS-crystal morphologies [nanoflowers]	DPV	pH 1.5 to 10.3, optimal 2.4	0.012 μΜ	97.0 to 104.5	[54]
Ionic liquid (AMIM-CI) functionalized reduced graphene oxide supported Au nanoparticle [ILRGO-Au]	CV, SWV	pH 3.0 to 7.0; Accumulation time 300 s	0.83 nM	95.3	[55]

Screen-printed carbon electrode

Screen-printed carbon electrodes (SPCEs) were modified by Wu et al. [5] employing MOF of NiBTC and rGO, which resulted in the development of an electrochemical sensor rGO/NiBTC/SPCE. The first step involved heating a mixture of trimesic acid, ethanol, and nickel (II)-nitrate solution to 180 °C to produce green NiBTC powder. The second step involved heating graphene oxide and ethanol to 180 °C to produce reduced graphene oxide [5]. After ultrasonication, the suspension of rGO and NiBTC was applied to the surface of SPCE. XRD, TEM, SAED, SEM, and EDS techniques were used to characterize NiBTC. The results showed uniform mixing, rGO planar sheets, and stability, and they also proved that the surface of the rGO/NiBTC/SPCE electrode was hydrophilic. The TZ concentration of real samples was sensed utilizing the DPV method, which examined the SPCE within a pH range of 8.0. The accumulation time was 360 s, the linear range was 0.075 to 5.0 μM and LoD was 0.05 μM. The recovery ranged from 94.41 to 104.73 %, all without any interference from external sources. A new doublestranded copper(I) helicate (H) was synthesized and used in a screen-printed electrode modified by single-walled carbon nanotubes (SWCNT/SPCE) by Nuñez-Dallos et al. [56]. H was created by a selfassembly process between Cu(I) iodide and 1,3-bis(1H-benzotriazol-1-ylcarbonyl) benzene (L). Elemental and thermogravimetric analysis and single crystal and powder XRD were used to characterize the new H. Copper (I) helicate suspension was drop-cast onto the electrode to create the modified H-SWCNT//SPCE. To achieve a clean and consistent surface, CV was applied to the newly modified electrode at a potential of -0.3 to +0.8 V against Ag/AgCl reference electrode. A step potential of 10 mV, an amplitude potential of 50 mV, and a frequency of 15 Hz was applied to the square wave voltammetry measurements, and the accumulation time was 60 s [56]. The electrochemical responses of the modified electrode were investigated using CV and EIS techniques at a pH of 2.3. The results showed an increased oxidation current of 60 %, a detection limit of 60.0 nmol/L, and recovery values above 100 % in several real samples. The fact that the determination revealed no interference from outside sources suggests that the method is accurate and efficient.

Carbon ceramic electrode

To modify the carbon-ceramic electrode, Majidi et al. [57] created a MWCNT-ionic liquid (MWCNTs-IL) nanocomposite. The electrode was modified in two steps. Firstly, 300 mg of graphite powder and ormosil were combined, giving a homogeneous gel solution. The composite was then allowed to dry at room temperature before Cu wire was joined to provide electrical contact [57]. Alcohol and purified MWCNTs were used to create the MWCNTs-IL composite. The bare and modified carbon ceramic electrode surfaces were examined using the SEM technique. The surface of the modified electrode had several tubes evenly spaced throughout it, whereas the surface of the raw carbon ceramic electrode was porous and featured separated carbon layers. The TZ dye was detected using CV, showing a strong peak with an increased current response. At the other side, the peak potential on the bare carbon ceramic electrode was at 1.01 V against SCE reference electrode, with a low peak current height. The modified electrode was used to acquire TZ by DPV, with a linear range of 3 to 70 μ M and a detection limit of 1.1 μ M. pH 7.0 was taken into consideration. Compared with other modified electrodes, the suggested modified electrode shows high conductivity, quick electron transfer, antifouling qualities, ease of preparation, and reduced preparation time. The recovery rate for TZ was 93.2 to 102 %, showing that the MWCNTs-IL nanocomposite-modified carbon ceramic electrode demonstrates good selectivity of analytes in various soft drink samples. There was no interference from outside sources throughout the detection process [57].

The carbon ceramic electrode was modified by Majidi et al. [58] using an ionic liquid containing 1-allyl-3-methyl imidazolium tetrafluoroborate. The electrode was built in two stages. The first step involved drying the mixture of ormosil and graphite at room temperature, polishing the electrode surface, and connecting the copper wire. The 1-allyl-3-methylimidazolium tetrafluoroborate ionic liquid was dissolved in ethanol to create a modifier solution, which was then applied to the polished electrode surface in the second step. Finally, a suspension of graphene nanoplatelets was deposited onto the carbon ceramic electrode surface [58]. This procedure allowed the development of a modified electrode made of graphene nanoplatelets and 1-allyl-3-methyl imidazolium tetrafluoroborate nanocomposite. Using SEM and EDX techniques to characterize the bare and modified electrode surfaces confirmed that ionic liquid was successfully deposited on the carbon-ceramic electrode surface. CV was used to determine the surface area of the modified electrode and record the electrochemical behavior of TZ and sunset yellow at pH 3 of sample solutions with a scan rate of 50 mV s⁻¹. Due to the high ionic conductivity of the modified electrode, the electron transfer between TZ and the electrode is significantly improved, resulting in a higher peak current at a reduced peak potential (0.889 V vs. SCE) compared to the bare electrode (0.957 V vs. SCE). Using DPV, TZ was detected with a linear range of 0.1 to 20 μ M and a detection limit of 0.081 μ M. The peak-to-peak potential separation for simultaneous TZ and sunset yellow dye detection was 230 mV, ensuring no interference. These findings demonstrate the high conductivity, selectivity, and feasibility of the modified electrode for sensitive TZ analysis.

<u>Carbon black-polyethylene composite electrode</u>

Lipskikh et al. [59] have synthesized a novel carbon black-polyethylene composite electrode (CBPCE) by immobilizing a carbon ink (CI) containing microcrystalline graphite, polystyrene, and 1.2-dichloroethane, on the surface of the CBPCE. CV of the redox pair $[Fe(CN)_6]^{3-}/[Fe(CN)_6]^{4-}$ on the bare CBPCE showed poorly defined peaks, whereas the modified CI/CBPCE showed sharper, well-defined redox peaks. Electrochemical properties of TZ and brilliant blue FCF dye were studied with CV and linear sweep voltammetry (LSV) in a pH 2.0 solution at a scan rate of 100 mV/s. The accumulation potential for TZ was 0.1 V. Accumulation time is an important factor in electrochemical analyses since it can increase sensitivity and specificity of the measurement. The increase in sensitivity and specificity results because time is given for the accumulation of the analyte on the surface of the electrode, raising its concentration locally and thus enhancing the electrochemical signal. Long accumulation times could also lead to saturation, but mainly due to interference from other species present in the solution. Real samples such as green candies and soft drinks exhibited excellent performance of the developed electrode with TZ recovery percentages of 103.2% and 97.2%, respectively. The detection limit is 0.019 μmol L-1 with a linear range between 0.037 and 1.38 μmol/L, thus showing better sensitivity, stability, and reproducibility compared to the unmodified electrode. This showed that TZ can be detected accurately by the CI/CBPCE modified electrode, with no interference effects.

Carbon paper electrode

An electrochemical sensor based on graphite powder (GrP) modified carbon paper (CP), used to detect synthetic food colorants, including TZ, was introduced by Stozhko $et\ al.$ [60] in a study from 2022. The GrP/CP sensor showed strong electron transfer, which, together with low resistance, points towards high sensitivity and selectivity for TZ. The electrochemical behaviour of TZ on bare and modified CP electrodes was recorded using CV (Figure 5). The sensor was capable of attaining a detection limit of 8.2 nM for TZ, with a linear range from 0.02 to 7.5 μ M and excellent reproducibility,

with RSD \leq 0.072, obtained by DPV. Modifying graphite powder significantly increased the electroactive area and the efficiency of charge transfer in comparison to carbon-based materials, such as carbon nanotubes and graphene. The GrP/CP sensor is an economical, fast, and efficient tool that can be used to detect TZ in both alcoholic and non-alcoholic beverages.

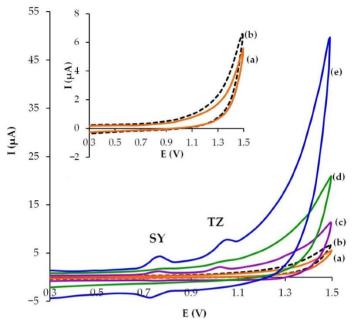


Figure 5. CVs (50 mV/s) of 10 μ M SY and TZ in the phosphate buffer solution, pH 5 at: (a) GR/CP, (b) bare CP, (c) MWCNTs/CP, (d) CB/CP, (e) GrP/CP. Reprinted with the permission of Ref. [60]

Graphite-based electrodes

Graphite powder-based electrodes exhibit electrochemical properties comparable to quasi-noble metal electrodes, with distinct advantages such as easy modification for improved selectivity and a renewable surface that eliminates the need for harsh chemicals for cleaning. Unlike commercial electrodes like screen-printed or sputtered-chip variants, graphite powder-based electrodes can be fabricated in a laboratory with a customizable form and characteristics [61,62]. This adaptability, combined with ease of modification using advanced materials and reliable reproducibility, makes graphite electrodes versatile for a wide range of analytical applications.

Graphite paste electrode

Zhang *et al.* [63] developed a new electrochemical sensor based on an ionic liquid (IL) modified expanded graphite paste electrode (EGPE) for the simultaneous determination of Ponceau 4R and TZ in soft drinks. The combined use of ionic liquid with the expanded graphite resulted in an increase in the electrocatalytic oxidation signals generated by the IL-EGPE due to the extraordinary conductivity and functional properties of the ionic liquid. The sensor showed wide linear response ranges of Ponceau 4R from 0.01 to 5.0 μ mol/L, and of TZ from 0.01 to 2.0 μ mol/with detection limits of 1.4 nmol/L or 0.85 ng/mL and 3.0 nmol/L or 1.6 ng/mL, respectively, at a signal-to-noise ratio of 3. The sensor showed high reproducibility, stability, and reusability and is practical for applications.

Pyrolytic graphite electrode

Song *et al.* [64] have demonstrated an elegantly simple electrochemical method for determining TZ in drinks using pyrolytic graphite electrodes (PGE) modified by MWCNTs. The regression plot demonstrated a clear relationship between the peak currents of reduction and concentrations of TZ, showing linear ranges of response at 2.0 to 70.0 mg/L and 70.0 to 230.0 mg/L, as well as a

detection limit of 0.5 mg/L. The MWCNT/PGE system ensured stable and reproducible voltammetric measurements and is, therefore, supposed to become a great tool for trace-level detection of TZ in food and drink products. It also demonstrates several advantages of electrode modification with nanomaterials, such as enhancing the performance of electrochemical sensors in complex matrices.

N-grap (nail polish - graphite) composite electrode

De Lima et al. [65] reported a sensitive, low-cost electrochemical sensor for detecting TZ with the aid of a composite material comprising graphite powder mixed with colorless nail polish called N-grap, coated onto an Ecoflex substrate designed to improve field analysis of beverage dyes. The mixture of 52:48 graphite to nail polish (ratio) was prepared by grinding. Compression of the sensor under high force resulted in a highly porous and rough surface, as confirmed by the SEM image, maximizing the area concerning electroactivity and electron transfer rate at 58.2 µm/s. Additional electrochemical techniques, such as CV and EIS, validated the high porosity of the sensor, which enhances trace TZ level detection capability. Testing with sports drinks and juice under optimized pH (4) and temperature (25° C) at the N-grap electrode presented a sensitive linear detection range from 2.0 to 50.0 µmol/L, having very low LoD of 21.0 nmol/L by employing square-wave voltammetry (SWV). The results from the N-grap device were consistent, indicating a relative standard deviation of 4.87 %, which is more reliable than UV-vis spectrophotometry, in which characteristic absorption at 430 nm of TZ was found. This study also shows that N-grap composite electrodes can become viable electrochemical sensors for the detection of TZ in complex matrices, awaiting to be effective, disposable, and portable, wherein their applications for food and drink monitoring may indeed find it alluring to cater to any field setting.

Pencil graphite electrode

To determine TZ, Tahtaisleyen *et al.* [25] created a sensor that included poly(L-phenylalanine) and graphene (Gr) pasted on a pencil graphite electrode surface (PGE). Two stages were involved in preparing the modified electrode. First, L-phenylalanine was electropolymerized to yield PLPA/PGE, and then the graphene electrode was modified to yield Gr/PLPA/PGE [25]. The modified electrode exhibited a faster electron transfer rate than the bare electrode, as demonstrated using CV. SEM results revealed that the altered electrode had graphene layers and a polymeric amino acid structure forming an interface. EIS results indicated low charge transfer resistance at Gr/PLPA/PGE compared to PLPA/PGE [25]. Then, utilizing a three-electrode system, a voltammetric measurement was carried out to identify TZ in the juice samples at pH 7.0. The obtained recovery values were from 98.71 to 104.444%, and the limit of detection was 1.54 μ M. Peak currents did not significantly alter during the TZ determination process as a result of interfering species, and they demonstrated great selectivity.

Gold electrode

To adapt gold electrodes for the sensitive analysis of TZ, Kobun *et al.* [66] employed chitosan (CHIT), MWCNTs, and calcium oxide nanoparticles (CaONPs). After dissolving CHIT, CaONPs were added, and the mixture was sonicated. To obtain the CHIT/CaONPs/MWCNTs nanocomposite suspension, MWCNTs (1 %) were added to the mixture [66]. With a LoD of 0.9 ppm, the DPV technique was able to sense in the 10-1 ppm range, demonstrating the excellent analytical performance of the prepared sensor. The recovery rate in the genuine samples (soft drinks, candies, and jelly) was calculated to assess the accuracy of this sensor. It was found to be 93.2 to 96.6 %, and

the RSD was less than 1 %, indicating good accuracy and an appropriate technique for detecting TZ in food and beverage goods.

Conclusions

This review article emphasizes the detection of synthetic dye tartrazine, usually used in food products, by various voltammetric (CV, DPV, SWV, LSV) methods. A wide variety of literature data on electrodes such as CPE, SPCE, carbon paper electrode, GPE, GCE, pencil graphite, gold electrode, and others, and their modifications by nanoparticles, MWCNTs, MOFs, and ionic liquids have been presented. Among them, CPE and GCE are the most used and efficient electrodes because of their availability, low cost, sensitivity, selectivity, and recovery rate. When modified, these electrodes provide active electron transfer compared to other electrodes and can detect very low concentration levels of TZ. Selecting suitable electrodes and modifiers with appropriate propositions could effectively detect dye in various foodstuffs with accurate results.

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