Voltammetric Determination of Various Food Azo Dyes Using Different Modified Carbon Paste Electrodes

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Abstract: The food industries are developing very fast by improving the taste, odor, appearance of various food products. To do so, they are adding a variety of preservatives, sweeteners, and coloring agents and attracting a huge community of people. Dyes or coloring agents play an important role in enriching food’s appearance, quality, and taste. Most used food coloring agents are synthetic azo dyes. They are cheaper than natural dyes, therefore, they are used in huge amounts very frequently. Most of the azo dyes like Amaranth, Tartrazine, Allura Red AC, and Metanil Yellow can cause serious health issues for all the living system, and therefore the use of them in food should be controlled, or the optimum level of these dyes in food and beverages should be measured. Many methods like thin layer chromatography, mass spectroscopy, spectrophotometry, capillary electrophoresis, cyclic voltammetry, high-performance liquid chromatography are used to detect azo dyes in the food. But cyclic voltammetry method is one of the simple, robust, highly selective, accurate, high speed, economical, and highly sensitive methods to determine the presence of azo dyes in foods. The main subject of this review article is the detection of several azo dyes utilizing modified carbon paste electrodes and cyclic voltammetry.

Keywords: azo dyes; modified carbon paste electrode; cyclic voltammetry; sensors; food colors.

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1. Introduction

Food colorants play an important role in meal selection since they improve the food’s stylish appearance, making it more desirable to buyers. However, many colorants become hazardous with prolonged usage, causing health problems. A few examinations have exhibited the relationship of some azo colors with unfavorably susceptible reactions, including asthma and contact hypersensitivity [1-8]. Also, some colorants prompt food bigotry, extreme touchiness, and conduct hyperactivity in youngsters. Cyclic voltammetry and linear sweep voltammetry [9-17] are commonly used along with various nanoparticles modified carbon paste electrodes for detailed studies [18-21]. Colors are grouped into common and engineered. Regular colors are removed from plant and creature sources by actual techniques. Manufactured colors are natural mixes that are combined falsely [2]. Colors are ordered into characteristics and engineered. Common colors are extricated from plant and creature sources.
by actual strategies. Manufactured colors are natural mixes that are integrated misleadingly. Engineered food colors have a few critical points of interest in correlation with normal ones. Azo colors structure the biggest gathering of counterfeit food colors [22-26]. They represent 70% of all-natural colors delivered on the planet. Tartrazine [25], Sunset Yellow, Azorubine, Allura Red, and Amaranth are the most recognized azo colors in the food sector [2]. However, they possess an electrochemically oxidizable -OH group on a sweet-smelling ring, allowing for anodic voltammetry verification. Much of the time, they are utilized for shading of a dessert shop, jams, confections, jams, frozen yogurt, soda pops, mixed refreshments, and so forth notwithstanding their broad use, a few negative impacts on the human wellbeing were recognized as the aftereffect of various examinations [2]. It is realized that azo colors rot items (for example, fragrant amines) are poisonous and cancer-causing. Furthermore, late distributions support the association of azo colors with human serum egg whites and hemoglobin. As per a few records, made food colorants can cause outrageous energy and hyperactivity in youngsters, particularly when utilized in huge amounts. Additionally, they have the potential to induce infections in people who are particularly susceptible to them, such as asthmatics. The European Food Safety Authority (EFSA) set up the most extreme passable degree of azo tones in drinks and staples. One of the most prevalent and widely used methods for determining diverse analytes is the cyclic voltammetric method [27-30].

Numerous strategies have been utilized for the assurance of different food azo colors. The Electrochemical techniques customarily have discovered wide applications in example examination because of the short investigation time, low force utilization, and economical hardware [3]. Cyclic voltammetry is the most broadly utilized electroanalytical strategy for the subjective data about electrochemical responses [31-34]. Carbon glue cathodes are generally utilized in electroanalysis because of their wide possible window, compound inactivity, ease, and appropriateness for different detecting location applications. Carbon paste electrode (CPE) was broadly utilized on the grounds that because of the minimal effort, getting another reproducible surface. Over the many previous years, CPEs containing different modifiers were generally utilized and applied in the goal of various analytes. CPEs can give a reasonable anode substrate to the planning of a changed cathode.

2. Determination of Amaranth Dye using Cyclic Voltammetry

Amaranth (E123. AM) is one of the important azo dyes easily soluble in water and mainly used in beverages, cakes, frozen yogurts, oats, confections, espresso, and in a variety of pharmaceuticals and beauty care products [4, 35-40]. The first impression of a food’s flavor is given through shading. Amaranth is commonly used to give beverages, syrups, and desserts a reddish or caramel tone.

2.1. Electrochemical behavior of amaranth on screen-printed carbon electrode as a function of pH.

In general, azo dyes can be electrochemically reduced in two processes, causing the signals of distinct dyes to overlap. The use of cyclic voltammetry and Screen-Printed Carbon Electrode (SPCE) can resolve them only at anodic peak currents in the range of 3.2 to 6.8 in the presence of nitric acid and phosphate buffer solutions. The electrochemical oxidation of AM has been calculated to comprise an electron and one proton.
Figure 1. (A) Cyclic voltammograms of Amaranth solution in the presence of nitric acid and phosphoric buffer solution. (B) Effect of pH on anodic peak potentials of amaranth using screen-printed carbon electrode. Adopted from [4].

The cyclic voltammograms (CV) for amaranth (17.7 mol L\(^{-1}\)) at pH 3.2 are shown in Figure 1A. (bend a). This picture depicts two all-around defined oxidation tops, one at 0.66 V (ipa 0.37 A) and the other at 0.78 V (ipa 0.65 A), connected to Amaranth oxidation. The contrary output has no decline top, illustrating the irreversible oxidation of these colors. When the pH was increased, the apex potential shifted in the wrong direction (bend b), showing that protons are involved in AM’s electrochemical process. The only evidence of AM at pH 6.8 was a minor articulation (0.54 V, ipa 0.01 µA). The flows decreased after increasing the pH from 3.2 to 6.8. To achieve a delicate and secure method, pH 3.2 was used. Figure 1B shows plots that show the relationship between amaranth’s oxidation top possibilities and pH. For amaranth, the inclination obtained for the Epa versus pH plot is 18mV.

2.2. Effect of supporting electrolyte (SE).

The SE lowers the cell's resistance and, as a result, lowers the Ohmic drop effect; meanwhile, it also inhibits the movement of electro-active species towards the electrodes through electrostatic attractions to achieve diffusion-controlled currents [4]. The cyclic voltammograms of AM were observed using SPCE in different SE in the presence of 300 µL of nitric acid (curve a), acetic buffer (curve b), or phosphoric buffer (curve c), all of them at a concentration of 0.01 mol L\(^{-1}\) (Figure 2). This resulted in strong 2 oxidation peaks. For amaranth, oxidation signals were recorded at 0.66 V (0.36 µA) with nitric corrosive, 0.60 V (0.69 µA) with acetic buffer, and 0.56 V (1.34 µA) with phosphoric buffer. According to these findings, the most remarkable pinnacle flows for amaranth (1.34 µA) and the best variation of ∆Epa occur when the phosphoric buffer is used. The phosphate buffer of 3.2 pH was used to transport all the resultant allot since it provided sufficient affectability and selectivity for determining amaranth.
Figure 2. Cyclic voltammograms of amaranth (17.7 µmol L⁻¹) at pH 3.2; adjusted with nitric acid (curve a), acetate buffer (curve b), and phosphate buffer (curve c). Scan rate 50 V s⁻¹. Adopted from [4].

2.3. The effect of scan rate (υ).

Using SPCE, recognition of the mass transport process was performed to study the effect of sweep rate (υ) on anodic peak current for various azo dyes. The relation between Ipa vs. υ is as depicted in figure 3. For amaranth, the oxidation peak current increases linearly with υ over the range of 10–100 mVs⁻¹. This suggests that the electrode reactions are adsorption controlled.

Figure 3. Effects of the scan rate on the anodic peak current of AM and TZ. Adopted from [4].

2.4. Effect of frequency, step potential, and amplitude.

Recurrence, step adequacy, and heartbeat plentifulness were the square wave boundaries analyzed. amaranth’s anodic pinnacle current grew as all the boundaries grew. In any case, when the recurrence was higher than 10 Hz, the pinnacle of amaranth is expansive, losing a goal. A stage abundance of 10 mV and heartbeat adequacy of 10 mV at a recurrence of 10 Hz were utilized for additional analyses.

2.5. Linear range, detection limit, and repeatability of the method.

Phosphate support pH 3.2 (0.01 mol L⁻¹), Eads 0.00 V, and smidgens 30 s were discovered to be ideal scientific conditions. The pinnacle current corresponded to Amaranth clustering over the 0.15–1.20 mol L⁻¹ zone under these conditions (Figure 4).
Figure 4. Adsorptive stripping voltammetry and calibration curves for increasing concentrations of AM with SPCE. Adopted from [4].

3. Cyclic Voltammetric Detection of Tartrazine Dye

Tartrazine is perhaps the most well-known colorant utilized in a wide scope of nourishments items [41-47]. Today, it is a run-of-the-mill added substance found in types of food, drinks, prescriptions, supplement upgrades, magnificence care items, toiletries, and other non-sustenance things. Colorants are typically allowed up to 200-500 sections per million, with a few exceptions for certain sustenance types with lower limitations [5]. Tartrazine usually causes hazardous side effects and is associated with leading problems as frequently as feasible.

3.1. Fabrication of carbon paste electrode.

The bare carbon paste electrode (BCPE) was made by mixing graphite powder (150 mesh) and silicone oil in a 70:30 percent weight-to-weight ratio [18-21]. The glue was then packed into the depression of a scratch-built cathode that had been streamlined on gauging paper.

3.2. Preparation of the poly glycine modified carbon paste electrode (PGMCPE).

The BCPE was electrochemically activated 10 times by cycling the potential sweeps in the range of 500 to 1800mV in PBS. The modified terminal was made in 1mM glycine under the same circumstances as the cathode actuation approach. The corrected terminal was flushed with purified water after electropolymerization.

3.3. Results and discussions.

According to Manjunatha [5], cyclic voltammograms were estimated for cathodes flooded in pH 7 PBS containing Tz (1×10^{-4} M). Tz demonstrated a fair electrochemical reaction, equivalent to BCPE. Tz’s oxidation potential and current were 883 mV and 11.32 mA at BCPE, and 918 mV and 30.46 mA at PGMCPE, respectively. These findings suggest that PGMCPE can accelerate the movement of Tz electrons. Differential pulse voltammetric tests
were also carried out by Manjunatha [5]. Tz’s electrochemistry on the BCPE revealed a little oxidation peak at around 870 mV. In any instance, a sharp peak at 911 mV was observed at the PGMCP, indicating that the PGMCP showed significant electrocatalytic action toward Tz oxidation.

4. Investigation of Metanil Yellow Dye using Cyclic Voltammetry

Synthetic colors are more common in food products nowadays, which affect human health. The banned food color Metanil yellow is a known mutagen and a class II toxin according to the World Health Organization [6]. The development of cancer and the degradation of stomach mucus have been connected to this color. Nanomaterial-based electrochemical sensors are a unique detecting technology that may be used in various monitoring and analytical applications.

4.1. Differential pulse voltammetry (DPV) of Metanil Yellow (MY) and Fast Green (FCF).

The DPV is one of the important electrochemical methods generally used to detect the analytes with high resolution. Following that, in the current study, DPV was used to record the voltammetric marks of MY and FCF at bare and modified electrodes at a rate of 5mV/s at a potential of 0 to 1.2 V and maintaining the amplitude and width respectively at 50 mV and 70 ms. The figure reported by Shah [6] depicts that Metanil yellow and FCF oxidize at 0.58 and 0.84V, individually.

Both analytes were evaluated simultaneously after ascertaining the state of the oxidation signals of each MY and FCF. Shah [6] describes the voltammetric reaction of MY and FCF in conjunction. These findings are in good agreement with the EIS findings as reported by Shah in his publication [6]. The component of calixarene and silver nanoparticles can be thought of as a stepping stone that allows electrons to pass from the transducer to the analytes. Due to this, the modifier will attract electrons from the oxidizable entity because of the positive potential being applied via a potentiostat, and so the modifier acts as an extension between the electron-giving entities of the analytes and the acceptor anode. This collaboration between the modifier and the cooler improves the voltammetric response by upgrading the signs of the colors at the modified GCE by encouraging host-guest associations that lead to a larger grouping of the colors near the cathode area.

4.2. Cyclic voltammetry.

By varying the scan rate from 25 to 250 mV/s the voltammetric behavior of MY and FCF was recorded using calix8/Au NPs/GCE. The resulting voltammograms depicted the diffusion-controlled electrochemical processes. As per the published reports, if the slope of the log of peak current and a log of scan rate is equal to 1, the electron transfer mechanism is adsorption controlled. If the same is less than 1 then it is a diffusion-controlled mechanism [9, 18]. During the investigation of condition optimization (effect of supporting electrolyte, pH, and accumulation time), Shah [6] reported that the electrolyte can influence the peak current and shape. He also reported that the increased accumulation time could lead to the intensification of peaks. He has also found that 70 seconds of accumulation time results in the highest peak current and reaches saturation; therefore, no further increase in peak current occurs.
5. Electrochemical Determination of Allura Red AC Dye Using Cyclic Voltammetry

The crimson tone of Allura Red AC is widely used to shade dessert stores, alcoholic beverages, and non-mixed beverages [48]. Despite a lack of clear information on the hazards of azo colors [49, 50], recent studies reveal that the increased use of azo dyes can cause hyperactivity in youngsters and cancer and mutation.

5.1. Preparation of a carbon paste electrode modified with cetylpyridinium chloride.

The 0.01 M Allura Red AC solution was prepared using distilled water. The preparation of bare and cetylpyridinium chloride modified carbon paste electrodes were explained by Pliuta et al. [7] in their previous publication.

5.2. Determination of Allura Red AS in jelly sweets.

The laboratory mill was used to grind the jelly sweets containing the Allura red dye. Weighed exactly one gram of the ground jelly and dissolved in 100 ml of distilled water. An aliquot of the sample was taken in a 25 ml standard flask and adjusted to the mark with Britton-Robinson buffer of pH 2. A known quantity of the sample solution was added to the electrochemical cell, and a potential of 300 mV was maintained for 300 seconds, followed by the potential sweep with a scan rate of 250 mV/s from 0 mV to 1100 mV.

5.3. The electrochemical behavior of Allura Red AC using cetylpyridinium chloride modified carbon paste electrode.

Pliuta et al. [7] used cyclic voltammetry to further investigate the electrochemical behavior of the dye using bare and modified electrodes. They reported that the oxidation current increased when they used cetylpyridinium chloride modified carbon paste electrode; meanwhile, the oxidation peak potential shifted towards the cathode and attained 950 mV. The oxidation peak current in these modified electrodes is less than the bare electrode due to the blocking of silica gel. The voltammograms of such electrodes show only one peak indicating the irreversibility of the oxidation of the azo dye [7].

6. Conclusions

We have reviewed the cyclic redox behavior of various synthetic dyes and their kinetic studies. Nowadays, most food companies are using these synthetic azo dyes, which are unhealthy and cause many diseases. Therefore, determining these dyes in food is very important and can be carried out using cyclic voltammetry. It is one of the sophisticated methods used to sense the presence of azo dyes. Various researchers have reported the selectivity, sensitivity, robustness, and efficiency of azo dyes' and the efficiency of azo dyes' electrochemical sensing using cyclic voltammetry. The various modified carbon paste electrodes, especially nanomaterials and surfactant-based electrodes, are popular and widely used to detect the Amaranth, Allura Red AC, and Mentil Yellow dyes.

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