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Review

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Voltammetric methods for determination of Allura Red AC in foods and beverages

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Abstract

Allura Red AC, is one of the azo group dyes, finds extensive application in foods and beverages such as fruit juices, baked goods, meat products, and confections. The substantial consumption of Allura Red AC has been associated with potential adverse effects on human health, like food intolerance, allergies, cancer, attention deficit hyperactivity disorder, multiple sclerosis, brain damage, cardiac diseases, nausea, and asthma, largely attributed to the reaction involving aromatic azo compounds. Therefore, controlling amount of the Allura Red AC in food and beverage is very crucial. The voltammetric analysis of Allura Red AC offers numerous benefits, including high sensitivity, selectivity, and rapidity. Furthermore, these methods are characterized by their robustness, reproducibility, and user-friendliness. This review is gathered to the comparison of voltammetric methods being in the literature for determination of Allura Red AC in foods and beverages.

Keywords: Allura Red AC, cyclic voltammetry, differential pulse voltammetry, square wave voltammetry, stripping voltammetry

1. Introduction

The rapid development of the food industry leads a rising number of specific products with certain shape, taste, smell, color, etc. For this reason, to improve the organoleptic properties of foods, various food additives are used such as coloring agents, sweeteners, thickeners and preservatives. Dyes have a special role in this regard, as the taste and quality of food are often associated with its color [1].

The persistent application of such colorants raises significant safety concerns, impacting both human well-being and environmental health. Recognizing the potential for enduring harm stemming from these food colorants, both the European Food Safety Authority and the US Food and Drug Administration have established stringent regulations governing their usage [2]. These regulatory bodies aim to ensure that consumers can enjoy food products being free from harm, defining precise threshold values that indicate safe levels of exposure to these additives [3].

Dyes utilized in food can be classified into two broad categories: natural dyes and synthetic dyes. Natural dyes are derived from plant, animal, or mineral sources. They have been used for centuries to impart color to various foods and beverages [1].

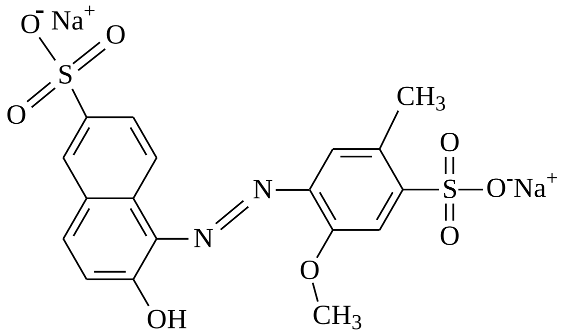

Figure 1. Molecular structure of Allura Red AC

Plant-based dyes are obtained from different parts of plants such as roots, berries, leaves, or flowers and they include turmeric (yellow), beetroot (red), spirulina (blue-green), and annatto (yellow-orange). Animal-based dyes; certain dyes are obtained from animal sources, particularly insects. One well-known example is carmine or cochineal, achieved from the bodies of female cochineal insects and provides a red color. Mineral-based dyes: these dyes are obtained from minerals and are often used in the form of pigments. For instance, titanium dioxide is used as a white pigment, and iron oxide provides shades of red, brown, and yellow. Natural dyes are generally considered safe, but they may

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have limitations in terms of stability, availability, and color range. Additionally, some individuals may have allergies or sensitivities to specific natural dyes [4].

Synthetic food dyes are chemically synthesized compounds that are specifically designed for use as food colorants. They offer a wide range of colors and are typically more stable compared to natural dyes. These dyes are classified into azo dyes, indigotine dyes, triphenylmethane dyes, xanthene dyes and quinoline dyes. Azo dyes have azo group in the molecular structure as the chromophore, which is largest group of color accounting more than half of global dyes production. They are commonly used in various industries, including the food industry, to enhance the appearance of products. Allura Red AC is one of the mostly used synthetic dyes in food industry that could be found in a variety of food and beverage products, including candies, beverages, and bakery products [5].

Allura Red AC designated as E-129 is a water-soluble monoazo dye, disodium 6-hydroxy-5-(6-methoxy-4-sulfo-m-tolylazo)naphthalene-2-sulfonate (Fig. 1). This synthetic dye has exhibited signs of both behavioral and physiological toxicity in laboratory rats when administered at extremely high doses, exceeding 10% of their diet [6,7]. In the past, numerous studies have investigated the toxicity and carcinogenic effects of Allura Red AC [8,9]. Allura Red AC has the potential to induce behavioral effects in both humans and animals, notably causing increased hyperactivity in children. Furthermore, certain studies have demonstrated the existence of aromatic amide or amine functional groups in the chemical compositions of Allura Red AC's degradation products. Allura Red AC is absorbed through the gastrointestinal tract and then entered the bloodstream, where it binds to proteins during its transportation and metabolism. It is essential to regulate the excessive use of Allura Red AC in food and beverage products. In numerous nations, the utilization of various food dyes, including Allura Red AC, has controlled, or prohibited due to its toxic properties. In the process of safety assessment, Joint FAO/WHO Expert Committee on Food Additives and the European Scientific Committee for Food, are the international scientific expert committees, established an acceptable daily intake for Allura Red AC ranging from 0 to 7 mg/kg of body weight per day. To address human health concerns, numerous analytical methods have been developed for the determination of Allura Red AC [5]. Allura Red AC has been determined by high-performance liquid chromatography [10], spectrophotometry [8], electrophoresis [11] and voltammetry [12].

2. Electroanalytical Methods

Electroanalytical methods typically involve in voltammetry, polarography, potentiometry, amperometry, and coulometry. Electroanalytical methods offer several advantages over other instrumental methods in analytical chemistry such as sensitivity, selectivity, speed, cost-effectiveness, and versatility, making them valuable tools for chemical analysis in numerous fields. Among these, voltammetry and polarography stand out as the most frequently employed electroanalytical methods. These techniques enable the identification and evaluating of organic compounds, with a detection limit as low as 10^{-7} M [13].

The voltammetric method effectiveness is influenced via the material of the working electrode, as the reply obtained relies on the electrochemical reactions taking place at the interface between the electrode and the solution. Moreover, it's crucial to take into account both the composition and the structure of the working electrode material, because they can impact on the voltammetric method performance. The physical form of the working electrode may also influence the diffusion and electron transfer processes associated with analyte detection [14–18]. The voltammetric techniques contain cyclic voltammetry (CV), linear sweep voltammetry (LSV), differential pulse voltammetry (DPV), square wave voltammetry (SWV), and stripping voltammetry [19].

CV serves as an initial and frequently employed voltammetric method due to its ability to offer valuable insights into both the thermodynamic and kinetic aspects of various chemical systems. This method excels at swiftly revealing the redox behavior across a broad spectrum of potentials. It stands out as the primary electroanalytical technique for gaining qualitative insights into the electrochemical reactions of electroactive samples [20–21].

LSV represents the most straight forward approach involving the systematic application of a varying potential to the working electrode to monitor changes in current over both potential and time. Also, the applied potential boundaries depend on factors such as the working electrode material, and the supporting electrolyte composition. This method entails a linear progression of the working electrode's potential over time, starting from an initial potential and concluding at a final potential. The direction of the scan rate can be denoted to demonstrate the potential scan direction, with positive for an anodic sweep and negative for a cathodic sweep. LSV is an exceptionally valuable electroanalytical technique, particularly when employing solid electrodes, as it offers swift analysis with a detection limit of approximately 10^{-6} M [22–25].

Pulse methods involve the application of varying potential pulses, with the corresponding current response measured at specific time intervals defined by the pulse duration. The DPV method proves highly effective in the detection of minute quantities of electroactive compounds. The waveform utilized in differential pulse voltammetry typically resembles a staircase. This method involves measuring of current at two key points: initially before the pulse is applied, and then at the end of the pulse. Thus, the differential pulse voltammogram is generated based on as a function of the potential, derived from the disparity between these two current measurements and then the dual current measurement approach enables the detection of analytes in solutions even at low concentrations. Because differential pulse voltammograms exhibit peak shapes, they are particularly well-suited for analytical purposes [26,27].

SWV employs a substantial amplitude differential approach characterized by symmetrical square waveforms. Within this technique, current measurements are taken twice during each cycle of the square wave, specifically at the end of both the forward and reverse pulses. The difference between these current values is graphed against the staircase potential. Peak currents yielded through SWV are nearly quadruple those observed in comparison to differential pulse voltammetry. The priority advantage of square wave voltammetry lies in its rapidity, as it can be completed within a matter of seconds, in contrast to the 2-3 minutes required for a full voltammogram using DPV. This rapidity and sensitivity make SWV particularly valuable for quantitative analyzes [20,28].

Stripping voltammetric techniques have gained significant popularity owing to their low quantification limit (typically around 10^{-8} or 10^{-9} M), as well as their precision, accuracy, and cost-effectiveness when compared to alternative analytical methods. These stripping methods are typically divided into two separate stages. First, there is the electrochemical deposition or accumulation step, during which the electroactive compound accumulates on the working electrode. Following the accumulation step, the subsequent phase is the stripping measurement, which entails the dissolution of the deposited analyte. Stripping voltammetry methods encompass various versions depending on how the preconcentration and stripping steps are applied. When the potential is initially maintained at a negative level before initiating a positive-direction scan, the technique is referred to as anodic stripping voltammetry. Following a predetermined deposition period, the potential is then systematically scanned in an anodic direction using a voltammetric approach. Cathodic stripping voltammetry involves the anodic deposition of the analyte followed by its stripping during a negative potential scan. In terms of application, cathodic stripping voltammetry can be seen as the inverse of anodic stripping voltammetry [29].

Adsorptive stripping voltammetry offers notable advantages, including heightened sensitivity and superior selectivity when compared to alternative voltammetric techniques. When the concentration step involves adsorption, the method is categorized as adsorptive stripping voltammetry. In the realm of electrochemistry, the process of ions or molecules adhering to the electrode surface is referred to as adsorption. The quantity of adsorbate present on the electrode surface holds significant importance, as it directly influences the voltammetric response. Adsorptive stripping voltammetry stands out for its excellent selectivity and sensitivity, making it particularly well-suited for the precise determination of trace and ultra-trace concentrations of analyte in samples [30,31].

This paper includes studies of the voltammetric analysis of Allura Red AC in foods and beverages, and we have reviewed general voltammetric methods for selective detection of Allura Red AC to ensure the safety of food and beverage products. Voltammetric sensors were developed to detect the presence of Allura Red AC in a wide range of food and beverage products to provide a new perspective. For this purpose, the voltammetric determination studies of Allura Red AC in the literature are assembled, also these studies are chronologically listed in Table 1.

3. Voltammetric determination studies for Allura Red AC

A carbon paste electrode (CPE) modified by silica gel impregnated with cetylpyridinium chloride (SG/CPCl) was developed by Pliuta *et al.* This innovative electrode has proven highly effective for the precise determination of Allura Red AC in Britton-Robinson buffer (BRB) solution at pH 2.0. CV was used to examine the redox characteristics of Allura Red AC on the developed sensor, and it showed an oxidation peak at +0.95V. The electro-oxidation of Allura Red AC was defined irreversible and adsorption-controlled process. After the optimal conditions for SWV were determined, the calibration curve exhibited excellent linearity within two concentration ranges: 0.04 – 0.2 µM and 0.2 – 1.0 µM with a low limit of detection of 0.005 µM and a limit of quantification of 0.015 µM. The newly developed sensor has showed satisfactory results in analyzing both model solutions and jelly candies [12].

Table 1. The voltammetric studies for determination of Allura Red AC

Electrode Type	**Method**	**Medium**	**Linear Range (µM)**	**LOD (µM)**	**Recovery (%)**	**Ref.**
PAMI/GCE	SWV	0.5 M HCl	10.0 – 300.0	1.4	—	[49]
MWCNT/GCE	DPV	PBS, pH 7.0	0.1 – 1.2 (r = 0.991)	0.05	98.1 – 100.6 (soft drink)	[48]
Gr/TiO_2/CPE	SWV	0.1 M H_2SO_4	0.00067 – 0.21 (r = 0.997)	0.00034	100.8 – 101.6 (soft drink) 97.12 – 102.1 (sausage)	[40]
IL/EGPE	SWSV	BRB, pH 6.0	0.1 – 10.0 (r = 0.999)	0.00179	96.0 – 99.2 (soft drink)	[39]
SbF/SPCE	DPV	AcB, pH 4.0	1.0 – 5.0	0.3	—	[50]
MWCNT/IL/GO/GCE	SWSV	BRB, pH 7.0	0.0008 – 0.5	0.0005	100.0 – 106.8 (alcoholic beverages)	[51]
PC/GCE	DPV	PBS, pH 7.0	0.005 – 0.8	0.0034	-	[52]
PDDA/Gr/Ni/GCE	DPV	PBS, pH 3.0	0.05 – 10.0 (r = 0.9993)	0.008	95.0 – 97.6 (strawberry juice)	[38]
IRGO/Au/GCE	SWV	BRB, pH 5.0	0.0006 – 0.2 (r = 0.998)	0.00043	—	[53]
CPB/GCE	SWAdSV	PBS, pH 3.3	0.06 – 11.5 (r = 0.999)	0.032	95.5 (cherry gelatin) 104.2 (chili sauce) 97.85 (strawberry juice)	[37]
MWCNT/GCE	DPV	PBS, pH 6.0	0.1 – 9.0 (r = 0.996)	0.014	95.0 – 118.0 (red isotonic drink) 94.0 – 130.0 (lilac isotonic drink)	[35]
Co_3O_4/CPE	SWV	PBS, pH 3.1	0.1 – 1.0 (r = 0.995)	0.05	91.5 (chili sauce) 89.7 (isotonic drink) 96.3 (soft drink)	[36]
m-AgSAE	DPAdSV	AcB, pH 3.6	0.01 – 0.4 (r = 0.996)	0.0012	—	[54]
	DCAdSV		0.008 – 0.6 (r = 0.9926)	0.0034		
p-AgSAE	DCAdSV		0.08 – 0.6 (r = 0.9943)	0.03		
IL/CB/CTS/ECH/GCE	SWAdASV	PBS, pH 4.0	0.0398 – 0.909 (r = 0.9998)	0.00091	87.5 – 108.3 (soft drink powder)	[33]
TiO_2/ErGO/GCE	DPV	PBS, pH 7.0	0.3 – 5.0 (r = 0.991) 0.01 5.0 – 800 (r = 0.992)	0.05	98.42 – 106.0 (milk drink)	[34]
Er-BTC/CPE	DPV	PBS, pH 6.0	0.001 – 0.1 (r = 0.99)	0.0003	—	[47]
SG/CPCI/CPE	SWV	BRB, pH 2.0	0.04 – 0.2 (r = 0.992) 0.2 – 1.0 (r = 0.998)	0.005	—	[12]
Ptre/PGE	DPV	PBS, pH 7.0	0.25 – 100.0 (r = 0.992)	0.075	98.4 – 99.6 (energy drink) 99.4 – 100.8 (cherry juice)	[32]
MoO_3/CPE	SWV	PBS, pH 6.5	0.36 – 6.0	0.38	96.0 (gelatin) 95.0 (syrup)	[41]
In^{3+}/NiO RLHNSs/GCE	DPV	PBS, pH 4.0	– 700.0 (r = 0.9999)	0.0041	99.0 – 101.2 (soft drinks)	[45]
F-nanodiamond@SiO_2 @TiO_2/SPE	DPV	BRB, pH 3.0	0.001 – 0.12 (r = 0.985) 0.12 – 8.65 (r = 0.991)	0.00122	96.3 – 101.0 (soft drink) 95.0 – 106.0 (powder) 95.0 – 102.8 (orange juice) 99.4 – 102.7 (candy)	[43]
Mn_3O_4@C/SPM	DPV	—	0.1 – 168.4 (r = 0.92441) 168.4 – 1748.4 (r = 0.97599)	0.033	98.6 – 100.68 (sports drink)	[44]
SHU/PCFE	DPV	PBS, pH 4.0	0.001 – 0.1 (r = 0.9988) 0.1 – 2.0 (r = 0.9490)	0.00036	98.0 – 101.0 (beverages) 100.0 – 105.0 (pharmaceuticals) 100.0 (lollipop)	[42]
ErGO/GCE	DPAdASV	BRB, pH 2.5	0.1 – 0.8 (r = 0.997)	0.028	96.2 – 102.3 (soft drink)	[46]

In their study focused on the electrochemical analysis of Allura Red AC, Uruc *et al.* prepared an anodically pretreated pencil graphite electrode (Ptre/PGE) which has oxygen-containing groups. The anodic pretreatment procedure involved the application of a constant potential of +1.8 V for a duration of 250 seconds in pH 9.0 phosphate buffer solution (PBS) accomplished by chronoamperometry method. When the studies were carried out with CV and DPV methods on Allura Red AC in pH 7.0 PBS, a significant and notable increase in the anodic peak current was observed for Ptre/PGE compared to the unmodified PGE. Under optimized conditions for DPV, the calibration curve, reflecting the response of Ptre/PGE to varying concentrations of Allura Red AC, showed a linear relationship within the concentration range from 0.25 to 100 µM, with a limit of detection and limit of quantification values of 0.075 µM and 0.251 µM, respectively. The developed sensor was effectively applied for assessing the presence of Allura Red AC in cherry juice and energy drink samples. It exhibited impressive performance, delivering relative standard deviation values within the range of 1.14% to 2.21% and recovery rates ranging from 98.4 to 100.8% [32].

In another study, a novel sensor for sensitive voltammetric determination of Allura Red AC and Brilliant Blue FCF was developed by incorporating carbon black (CB) nanoparticles and the ionic liquid (IL) (1-butyl-3-methylimidazolium tetrafluoroborate) within a crosslinked film made from chitosan (CTS) and epichlorohydrin (ECH). This film was applied over the surface of a glassy carbon electrode (GCE), allowing for efficient detection of Allura Red AC in soft drink powders. The electrochemical behavior of the Allura Red AC colorant was examined using CV in PBS at pH 4.0. The analysis revealed an irreversible oxidation peak occurring at approximately +0.9 V. Moreover, the redox process of Allura Red AC on the IL/CB/CTS/ECH/GCE was found to be controlled by a combination of diffusion and adsorption processes. The optimization of experimental parameters effecting the sensor response was carried out systematically using square-wave adsorptive anodic stripping voltammetry (SWAdASV). Under the ideal experimental conditions, the SWAdASV method demonstrated a linear analytical curve within the concentration range of 0.0398 to 0.909 μM, accompanied by a notably low limit of detection of 0.00091μM. The precision of the suggested sensor was assessed through intra and inter-day repeatability experiments and relative standard deviations was calculated as 3.5–6.7% and 7.6–11.1%, respectively. Furthermore, interference and recovery experiments were conducted, demonstrating the proposed sensor has good accuracy and selectivity, with recovery values ranging from 87.5 to 108.3% [33].

Li and co-workers prepared an electrochemical sensor by modifying a GCE with titania/electro-reduced graphene oxide nanohybrids (TiO2/ErGO) for the detection of Allura Red AC. Notably, when compared to pure ErGO and TiO2 nanoparticles, the TiO2/ErGO nanohybrids exhibited a significant enhancement in electrocatalytic activity and voltammetric response for Allura Red AC. In 0.1 M PBS at pH 7.0, which serves as the optimal supporting electrolyte, Allura Red AC showed an oxidation peak at +0.62 V in DPV. Additionally, cyclic voltammetric studies revealed that the electrochemical oxidation process of Allura Red AC was diffusion-limited quasi-reversible. Using the DPV method, it was observed that within the concentration range of 0.3–5.0 μM, the anodic peak currents of Allura Red AC exhibited a linear correlation with its concentrations. However, this linear relationship shifted to a semi-logarithmic relationship in a higher concentration region (5.0–800 μM). The detection limit was calculated as 0.05 μM. The suggested TiO2/ErGO/GCE exhibited good reproducibility and stability in both determination and storage. It reliably and accurately detected the concentration of Allura Red AC in milk drinks, with recovery results between 98.42–106.0% and relative standard deviation values within the range of 0.85 to 4.86 % [34].

Sierra-Rosales and colleagues have introduced a rapid and simple electrochemical sensor for the determination of Allura Red AC in isotonic sports drinks. This sensor was prepared from GCE modified with multi-walled carbon nanotubes (MWCNT). The electrochemical behavior of the colorant was investigated by CV on the MWCNT/GCE, revealing an anodic peak at +0.72 V, along with a small cathodic peak at +0.72 V on the reverse scan. This process was found to be mixed adsorptive-diffusion controlled. In differential pulse voltammograms, the sensor demonstrated a linear response to Allura Red AC within the concentration range of 0.1–9.0 μM, with a detection limit of 0.014 μM in isotonic sports drinks. The method also showed good recovery results in both red and lilac isotonic sport drinks, with relative standard values below 3 % and 5 %, respectively [35].

A novel CPE composed of cobalt (II, III) oxide was prepared by Penagos-Llanos et al. for the detection of Allura Red AC. The presence of Co3O4 resulted in a remarkable enhancement of the cathodic peak current for Allura Red AC, with an increase of over 50% in CV and a 65% boost in SWV, as compared to an unmodified CPE. In PBS at pH of 3.1 serving as the supporting electrolyte, Allura Red AC exhibited a linear response in the concentration range of 0.1 to 1.0 μM, with a remarkable detection limit of 0.05 μM. This technique was successfully applied to chili sauce, soft drink and isotonic drink beverages containing Allura Red AC, displaying the good accuracy of the developed method. Furthermore, studies have proven that the sensor is sensitive, stable, and selective [36].

Nagles and García-Beltrán have offered a new and simple approach for the detection of Allura Red AC in cherry gelatin, chili sauce, and strawberry juice. This method is based on utilizing square-wave adsorptive stripping voltammetry (SWAdSV) on GCE, with the presence of cetylpyridinium bromide (CPB). In PBS at pH 3.3, the oxidation peak of Allura Red AC occurred at 0.81 V. However, with the addition of small quantities of CPB, an aggregate of Allura Red AC and CPB formed, causing a notable shift in oxidation to nearly 0.14 V and a concurrent increase in current by approximately 50%. This method was effectively applied for the analysis of Allura Red AC, demonstrating linearity within the concentration range of 0.06 to 11.5 μM, and achieving a low limit of detection as low as 0.032 μM. Furthermore, this method has proven effective when applied to real samples, yielding satisfactory recovery results within the range of 95.5 to 104.2%. The authors also have highlighted that when utilizing CPB/GCE, it is possible to perform at least 60 measurements with little loss of reliability, and each measurement can be completed in just 60 seconds [37].

In another study, Yu et al. have introduced a novel and highly sensitive electrochemical sensor that based on a composite material consisting of poly(diallyldimethylammonium chloride) (PDDA) functionalized graphene (Gr) and nickel nanoparticles, which is utilized to modify a GCE for the detection of Allura Red AC. The sensor demonstrated a significantly enhanced electrochemical activity towards Allura Red AC. The observed improvement could be attributed to the synergistic effect arising from the enlarged active surface area and enhanced electron transfer efficiency achieved through the combined utilization of Gr and Ni nanoparticles. The electro-oxidation process of Allura Red AC on PDDA/Gr/Ni/GCE was detected irreversible and adsorption-controlled. The differential pulse stripping voltammetry (DPSV) responses to various concentrations of Allura Red AC in PBS at pH 3.0

exhibited a linear correlation within the range of 0.05–10.0 µM, with a detection limit as low as 0.008 µM. The method was successfully applied to strawberry juice samples and proved its accuracy with 96–97.6% recovery values and relative standard deviation lower than 5% [38].

Zhang et al. devised a practical and highly responsive technique for quantifying Allura Red AC and Ponceau-4R simultaneously. This approach involved the utilization of an expanded graphite paste electrode (EGPE) that was modified with an IL (1-butyl-3-methylimidazolium hexafluorophosphate), in combination with square-wave stripping voltammetry (SWSV). The high conductivity of IL resulted in a significant improvement in the electrocatalytic oxidation signal of Allura Red AC at +0.72 V when comparing it to EGPE. In BRB solution at pH of 6.0, the linear range for the concentration of Allura Red AC spanned from 1.0 to 10.0 µM, with an impressive detection limit of 0.0179 µM. To assess its practical applicability, the developed method was applied for the quantification of Allura Red AC in both orange and grapefruit juices under optimized experimental parameters. The recoveries achieved using this method for Allura Red ranged from 96.0 to 99.2%, which strongly indicate the good accuracy of the proposed method. The electrode design also demonstrated excellent characteristics in terms of reproducibility, stability, and reusability [39].

A CPE was effectively modified using mesoporous TiO2 combined with graphene (Gr/TiO2). This modification resulted in a notable enhancement effect, significantly increasing the oxidation signal of Allura Red AC at +0.89 V when analyzed using SWV. The electro-oxidation of Allura Red AC was found irreversible and adsorption-controlled electrode process. 0.1 M H2SO4 solution was used as the supporting electrolyte for detecting Allura Red AC. Under the optimal conditions, it was observed that the oxidation current of Allura Red AC exhibited proportionality to its concentration within the range of 0.00067 to 0.21 µM, and the limit of detection was obtained as 0.00034 µM. To assess its practical utility, this developed method was put to the test in detecting Allura Red AC within soft drink and sausage samples. The authors also emphasized the excellent reproducibility in fabricating the method and its precision in detection [40].

In a study presented by Nagles et al., a novel application involving MoO3 combined with CPE was introduced to create a micro-composite electrode. This electrode was designed for the purpose of quantifying Allura Red AC and Paracetamol simultaneously, using SWV. The anodic peak current for Allura Red AC was 80% higher when compared to an unmodified CPE. The relationship between concentration of Allura Red AC and oxidation peak current was linear in the range of 0.36–6.0 µM with the detection limit of 0.38 µM in PBS at pH 6.5. The accuracy of the proposed method was evaluated by testing its ability to detect Allura Red AC in real samples of syrup and strawberry-flavored gelatin, indicating recovery values of 95% and 96%, respectively. Stability and potential interferences were thoroughly assessed, and the results obtained were very acceptable [41].

In another study, shungite (SHU), a natural mineral as a modifier on a planar carbon fiber electrode (PCFE), has proven to be advantageous for creating a favorable electrode interface for the electrochemical oxidation of Allura Red AC. This modification has led to several positive effects, including an expansion of the electroactive surface area, a reduction in electron transfer resistance, and the enhancement of electrocatalytic effects. In analytical applications employing DPV, PBS with a pH of 4.0 was used as supporting electrolyte. The developed sensor exhibited the capability to quantify Allura Red AC within the concentration ranges of 0.001 to 0.1 µM and 0.1 to 2.0 µM, with impressively low detection limit of 0.00036 µM. The practical utility of the SHU/PCFE was demonstrated by testing it with drink samples, lollipops, and pharmaceuticals. The results demonstrated its accuracy, as evidenced by excellent recovery values with relative standard deviations consistently lower than 5%. Additionally, this convenient and cost-effective sensor exhibited good repeatability, stability, and anti-interference capacity [42].

Mehmandoust et al. developed a sensitive and innovative electrochemical sensor designed for the detection of Allura Red AC in the presence of Tartrazine. This sensor was prepared by modifying a screen-printed electrode with functionalized nanodiamonds coated with silicon dioxide and titanium dioxide nanoparticles (F-nanodiamond@SiO2@TiO2/SPE). The electrochemical response of F-nanodiamond@SiO2@TiO2/SPE in the electro-oxidation of Allura Red AC was investigated by using CV and the results revealed that the oxidation reaction occurring on the electrode surface was a diffusion-controlled process. In BRB at pH 3.0 using DPV, the fabricated electrode exhibited two extensive dynamic ranges for Allura Red AC quantification: 0.01–0.12 µM and 0.12–8.65 µM and it achieved a detection limit as low as 0.00122 µM. The successful detection of Allura Red AC in various food samples such as soft drinks, powders, orange juice, and candy demonstrated the sensor's practical applicability for determining Allura Red AC with satisfactory recovery rates. Furthermore, the modified electrode indicates outstanding attributes in terms of repeatability, reproducibility, selectivity, and stability [43].

A core-shell architecture was ingeniously crafted, featuring $Mn_3O_4@C$ nanocubes, to construct an exceptionally sensitive screen-printed microchip (SPM) for the precise detection of Allura Red AC. This nanocomposite capitalizes on the strengths of $Mn_3O_4@C$, characterized by its high electrocatalytic activity and chemical stability. The electrochemical behavior of the prepared $Mn_3O_4@C$ nanocubes was compared with that of both the unmodified electrode and MnCO3 nanocubes by CV. Notably, the $Mn_3O_4@C$ nanocubes exhibited the highest redox currents, proving the robust redox capabilities of coexisting Mn^{2+} and Mn^{3+}. The newly Allura Red AC sensor exhibited excellent selectivity, featured with impressively large linear detection ranges from 0.1 to 168.4 μM and from 168.4 to 1748.4 μM, with a low detection limit of 0.033 μM. The accuracy of the prepared Allura Red AC sensor was assessed in real samples of various sports drinks using a standard addition method. The obtained recovery results ranged from 98.60% to 100.67%, indicating the sensor's excellent performance in accurately quantifying Allura Red AC in these samples [44].

In their recent research, Moarefdoust and colleagues aimed to identify Allura Red AC through the creation of a novel electrochemical sensor by developing a raspberry-like In^{3+}/NiO hierarchical nanostructure (In^{3+}/NiO RLHNSs) modified GCE. The electrochemical behavior of Allura Red AC was investigated with three different electrodes (In^{3+}/NiO RLHNSs/GCE, NiO RLHNSs/GCE, and bare GCE) in 0.1 M PBS at pH 4.0 by CV. Notably, the anodic peak current obtained using the In^{3+}/NiO RLHNSs/GCE exhibited a significant increase, approximately 3.4 and 1.5 times higher than that of the bare GCE and NiO RLHNSs/GCE, respectively. These results indicated the successful grafting of In^{3+}/NiO RLHNSs onto the electrode surface, leading to a substantial enhancement in electrode sensitivity for Allura Red AC. Also, it was determined that Allura Red AC responses on the developed electrode surface continued with a diffusion-controlled process. Using DPV, a linear relationship was evident between the concentration of Allura Red AC and the corresponding peak current (0.01 – 700.0 μM). The calculated values for the limit of detection and the limit of quantification were 0.0041 and 0.0136 μM, respectively. The proposed sensor's applicability was evaluated using strawberry and fruit juice samples, and the mean recovery rate for Allura Red AC was found in the range of 99.0% to 101.2%, which confirms the good accuracy of the developed method [45].

The electrochemical characteristics of Allura Red AC on a GCE that had been modified with ErGO were studied using CV and differential pulse adsorptive anodic stripping voltammetry (DPAdASV). The electrochemical behavior of Allura Red AC on the ErGO/GCE indicated semi-reversible oxidation, and it was observed that Allura Red AC adsorbed onto the surface of the ErGO/GCE. In the supporting electrolyte consisting of a BRB solution with a pH of 2.5, the anodic peak current exhibited a linear increase with the concentration of Allura Red AC, ranging from 0.1 to 0.8 μM. The method's calculated limit of detection and limit of quantification were 0.028 and 0.093 μM, respectively. The ErGO/GCE was successfully employed for determining Allura Red AC in soft drinks, offering both high sensitivity and accuracy. This method is characterized by its simplicity and cost-effectiveness [46].

Cai et al. developed a modified CPE using erbium nitrate and 1,3,5-benzenetricarboxylic acid (Er-BTC) for the sensitive electroanalytical analysis of azo dyes and flavonoids in drink and tea samples. PBS with a pH of 6.0 served as the supporting electrolyte for the simultaneous detection of Allura Red AC and Rhodamine B using DPV. It was observed that as the concentration of Allura Red AC ranged from 0.001 to 0.1 μM, there was a linear increase in the oxidation peak currents. The detection limit was obtained as low as 0.0003 μM. When the practical applicability of the method was tested on various drink and tea samples, it was found that the relative standard deviation remained below 5%. This indicates the method's reliability and consistency in different sample types [47].

An electrochemical sensor was developed for the rapid and simple detection of Allura Red AC and Ponceau 4R. This sensor was created by modifying a GCE with a film of MWCNT. The MWCNT film sensor, owing to its substantial surface area and efficient accumulation capability, significantly enhances the oxidation signal of Allura Red AC. The electro-oxidation process of Allura Red AC on the MWCNT/GCE was determined to be irreversible and diffusion-controlled. The detection parameters, including pH value, the amount of MWCNT, accumulation potential and time were optimized for the best results. By DPV, it was observed that the obtained peak currents showed a linear relationship with concentrations of Allura Red AC in the range of 0.1 to 1.2 μM and the limit of detection for this analysis was determined to be 0.05 μM. To evaluate its performance in real sample analysis, the new method was applied to detect Allura Red AC in several soft drinks. The recovery values that obtained ranged between 98.1% and 100.6% suggest that the detection of Allura Red AC using MWCNT/GCE is accurate and feasible [48].

In their research, Silva et al. described the fabrication process of a polyallylamine (PAMI) modified GCE. They subsequently employed this modified electrode for the electro-reduction of Allura Red AC within complex food samples using SWV. Given the complex nature of the sample matrices to be analyzed, a multi-commutated flow system was devised. This system facilitated the seamless execution of the standard additions method, all while operating automatically and maintaining a continuous flow of samples. After optimizing the parameters of SWV, it was observed that the linear range for the quantification of Allura Red AC in a 0.5 M HCl solution extended from 10.0 to 300.0 μM, achieving a detection limit of 1.4 μM. The method was successfully applied to determine the concentration of Allura Red AC in gelatin powder and alcoholic beverages [49].

Rodríguez et al. demonstrated the effectiveness of combining sequential injection analysis with DPV detection, using Antimony film modified screen-printed carbon electrodes (SbF/SPCE), for the analysis of Allura Red AC in food samples. Acetate buffer (AcB) solution of pH 4.0 was selected as the supporting electrolyte for this analysis. The study revealed a linear relationship between the peak height and the concentration of the dye within the range of 1.0 to 5.0 μM, with a detection limit calculated as 0.3 μM. The method was put into practical use for the determination of Allura Red AC in real samples, including flavored cornflour, tomato bouillon cubes, candy, gelatin powder, and isotonic drinks [50].

A modified GCE was fabricated by dispersing MWCNTs into an IL and graphene oxide (GO) mixture. The MWCNTs were effectively dispersed within the IL-GO mixture, leading to excellent dispersion properties and a significantly increased surface area for the composite. This enhanced the electrode's capability to absorb Allura Red AC, making it a promising platform for analytical applications. In BRB with a pH of 7.0, the oxidation peak current of Allura Red AC exhibited a linear increase as its concentration was raised (0.0008–0.5 μM), and the limit of detection was found 0.0005 μM. To assess its potential application, MWCNT/IL/GO/GCE was employed for the concurrent detection of Allura Red AC, Ponceau 4R, and Tartrazine in alcoholic beverages using SWSV technique, and the obtained recovery values were between 100.0–106.8% for Allura Red AC [51].

A series of porous carbon (PC) was synthesized by utilizing calcium carbonate nanoparticles as a hard template and starch as the carbon source. Through the surface modification of PC on a GCE, Cheng et al. successfully established a highly sensitive electrochemical sensing platform for the detection of Allura Red AC. As a result of the calibration study performed with DPV, the linearity range and limit of detection values for Allura Red AC in PBS at pH 7.0 were found to be 0.005–0.8 μM and 0.0034 μM, respectively. Reproducibility and interference studies were carried out with PC/GCE, demonstrating satisfactory results. To evaluate its practical applicability, the developed method was utilized in various drink samples [52].

In another study, to synthesize high dispersible nanocomposites, 1-allyl-3-methylimidazolium chloride functionalized reduced graphene oxide (IRGO) was used to load Au nanoparticles (Au). The prepared IRGO/Au nanocomposites were employed to modify a GCE. SWV was performed to investigate the electrochemical behavior of Allura Red AC on the surface of the IRGO/Au/GCE in BRB at pH 5.0, and the results indicated that the developed sensor could greatly facilitate the electro-oxidation of Allura Red AC. The SWV peak current for Allura Red AC exhibited an increase with rising concentrations within the range of 0.0006 to 0.2 μM, demonstrating a low detection limit of 0.00043 μM. To validate the practical applicability of the proposed sensor, the IRGO/Au/GCE was utilized for the quantification of Allura Red AC in chocolate, jelly, tea, and juice samples [53].

Lastly, a novel method was successfully developed to determine Allura Red AC in commercial beverages by Tvorynska et al. This method utilized two types of silver solid amalgam electrodes: the mercury meniscus modified amalgam electrode (m-AgSAE) and the liquid mercury-free polished amalgam electrode (p-AgSAE). The analysis was conducted using both differential pulse adsorptive stripping voltammetry (DPAdSV) and direct current adsorptive stripping voltammetry (DCAdSV) techniques. Due to the substantial increase in the reduction peak current of Allura Red AC observed on both m-AgSAE and p-AgSAE, which is a result of its adsorption at the electrode surface, the amalgam electrodes demonstrate wide linear ranges and high sensitivity for the quantification of Allura Red AC. In AcB with a pH of 3.6, the peak current of Allura Red AC exhibited a linear relationship with its concentration in the range of 0.01 to 0.4 μM when using DPAdSV and 0.008 to 0.6 μM when employing DCAdSV on the m-AgSAE electrode. The calculated detection limits for this analysis were determined to be 0.0012 and 0.0034, respectively. When utilizing the p-AgSAE, the linearity for the analysis of Allura Red AC was from 0.08 to 0.6 μM and detection limit was obtained 0.03 μM, by the DCAdSV technique [54].

4. Conclusion

Allura Red AC, which is a group of azo dyes in the food and beverage coloring, plays a key role in the food processing sector. Because the excessive consumption of this synthetic toxic dye can cause adverse effects on the human health, voltammetric methods were developed for detecting the presence of Allura Red AC in food and beverage samples. No study compiling the voltammetric determination of Allura Red AC in foods and beverages has been found in the literature. The voltammetric studies carried out for determination of Allura Red AC from food and beverage samples in the literature have been reviewed and evaluated on account of used method, obtained linear range and limit of detection values. Considering the methods used in these studies, DPV method was used the most, followed by stripping methods and SWV. When compared in terms of the linear range values have been obtained in these studies, it has been seen that the widest range was acquired with DPV method and the In^{3+}/NiO RLHNSs/GCE. Then limit of detection values have been evaluated for these studies, the lowest limit of detection value was obtained SWV and Gr/TiO_2/CPE. The concentration of Allura Red AC in foods and beverages can be determined at very low limits via voltammetric methods. Also, these methods provided good recovery, accurate and precise results. In conclusion, the determination of Allura Red AC can be carried out accurately and reliably by voltammetric methods in food and beverage samples.

Conflicts of Interest:

The authors declare no conflict of interest.

Author Contributions

EŞ: Writing-original draft.

FA: Supervision, writing-review & editing.

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