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Recent advantages in electrochemical monitoring for the analysis of amaranth and carminic acid as food color

Marzieh Alizadeh [a,b], Ersin Demir [c,*], Nida Aydogdu [c], Najmeh Zare [d], Fatemeh Karimi [d,**], S. Masoud Kandomal [d], Hassan Rokni [d], Younes Ghasemi [a]

[a] Pharmaceutical Sciences Research Center, Shiraz University of Medical Sciences, Shiraz, Iran
[b] Laboratory of Basic Sciences, Mohammad Rasul Allah Research Tower, Shiraz University of Medical Sciences, Shiraz, 71348-14336, Iran
[c] Afyonkarahisar Health Sciences University, Faculty of Pharmacy, Department of Analytical Chemistry, 03030, Afyonkarahisar, Turkey
[d] Department of Chemical Engineering, Quchan University of Technology, Quchan, 9477177870, Iran

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ABSTRACT

This study provides a comprehensive review of the latest developments in the electrochemical impressions of the important dyestuffs including amaranth and carminic acid. Food colors are organic substances that have important effects on human health and food safety. While these substances do not pose a problem when used in the daily intake (ADI) amounts, they harm human health when consumed excessively. Amaranth and carminic acid are synthetic and natural food colors ingredients, respectively. Analysis of these substances in food, pharmaceutical, cosmetic and textile samples is extremely important because of their genotoxicity, cytostatic and cytotoxic effects. Electroanalytical methods, which have great advantages over traditional analytical methods, shed light on the scientific world. Electrochemical monitoring modules, which are fast, simple, accurate, reliable, and highly selective, are promising for the determination of both substances. Until now, amaranth and carminic acid food determinations have been carried out successfully with electrochemical monitoring techniques in many numbers in the literature. Voltammetric techniques are the most widely used among these electroanalytical methods. In particular, square wave and differential pulse voltammetric techniques, which have extraordinary properties, have been heavily preferred. Limits of detection (LOD) comparable to the standard analytical method have been achieved using these methods, which have very quick analysis durations, high precision and accuracy, do not require long preprocessing, and have great selectivity. In addition, more sensitive and selective analyses of amaranth and carminic acid in natural samples were carried out with numerous indicator electrodes. The merits of powerful electrochemical monitoring studies for the determination of both food colors during the last decade are presented in this study. Moreover, parameters such as analytical applications, detection limits, electrochemical methods, selectivity, working electrodes, and working ranges are summarized in detail.

1. Introduction

Dyestuffs are organic-based chemicals used to color materials such as paper, leather, fabric, fiber, plastic, and textile (Bijad et al., 2018; Cheng et al., 2020; Ghalkhani et al., 2022; Han et al., 2014; Tajik et al., 2022; Wang et al., 2015b). Dyestuffs become irreversible by interacting chemically or physically with the surface of the applied object. These items can be classified in terms of the way they can be produced naturally or synthetically (Ghanbari et al., 2019). Natural dyestuffs were first obtained in foods such as onions, eucalyptus, turmeric, and henna to be used in the textile industry. Classification of dyestuffs can be done with different methods depending on the chemical structure, solubility and dyeing method. However, it is preferable to categorize them based on their chemical structures. According to this classification, it is possible to classify them as nitro and nitroso, triphenylmethane, phthalene, and xanthene, azo, anthraquinone, indigo, and phthalocyanine (Fig. 1).

These dyestuffs are not only used in the industrial industry but also frequently used in foods, medicines, beverages, and cosmetic products that directly affect human health (Alghamdi et al., 2009; Jian and Li-Ping, 2013; Yilmaz et al., 2014). Especially the azo group dyes are the

* Corresponding author.
** Corresponding author.
E-mail addresses: dr.ersindemir@yahoo.com (E. Demir), fkm024@gmail.com (F. Karimi).

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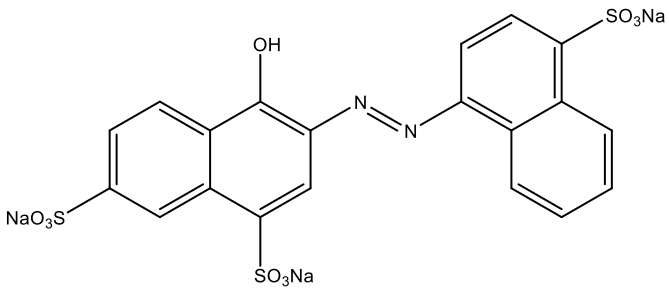
most preferred dyestuffs in the food, pharmaceutical, beverage, and cosmetic industries due to their high solubility in water, high stability, low microbiological contamination, and very stable against temperature, light, and pH (Brillas and Martínez-Huitle, 2015; Fajardo et al., 2016). Furthermore, dyes as food additives have been widely used in food processing in the food industry to improve the desired taste, appearance, and color of foodstuffs in recent years and to make them attractive to consumers. The utilization of these substances must be controlled by laws and regulations. However, excessive consumption of these substances can cause food anaphylaxis and allergy in individuals (El-Wahab and Moram, 2013; Shabani et al., 2009). Therefore, the analysis of these substances in samples that directly affect human health is of great importance.

Amaranth (E 123), whose IUPAC name is trisodium; 3-hydroxy-4-[(4-sulfonatonaphthalen-1-yl)diazenyl]naphthalene-2,7-disulfonate, widely used in soft drinks, syrups, ice creams, supplement mixes, alcoholic beverages such as wine, and salad dressings (Shabani et al., 2009; Wang et al., 2015b). It is a synthetic azo dye (N=N) commonly used to make chewing gums and chocolates more attractive and to give them a red color (Rovina et al., 2017). The term amaranth stems from the red-colored amaranth plant, which is high in edible protein. Amaranth shows an anionic dye property and contains azo (N=N) functional groups, and aromatic ring structures (Scheme 1) (Fajardo et al., 2016). Azo group dyes are not carcinogenic, but amines formed by the reduction of azo bonds can cause cancer (Brown and De Vito, 1993; Chung and Cerniglia, 1992; Ghanbari et al., 2019; Lopez-de-Alba et al., 2001). Azo group dyestuffs are also known as pollutants due to their effects on human health and the environment (Jian and Li-Ping, 2013).

The daily intake (ADI) of this substance has been reported as 0–1.5 mg/kg according to the recommendation of the FAO and WHO Food Additives Expert Committee (Mpountoukas et al., 2010). They have the potential to be poisonous and pathogenic, especially if taken in large quantities. In addition, amaranth has been reported to have adverse effects on human health, such as high genotoxicity, cytostaticity, and cytotoxicity (Han et al., 2014; Mpountoukas et al., 2010; Sarıkaya et al., 2012). Moreover, it has been banned as a suspected carcinogen by the Food and Drug Administration (FDA) in the United States since 1976. Unfortunately, excessive amounts of amaranth are still known to be used in some food products. Analysis of this dye, which is extremely important for human health and food safety, must be carried out by analytical method correctly and routinely (Bijad et al., 2017).



Scheme 1. The molecular structure of amaranth.

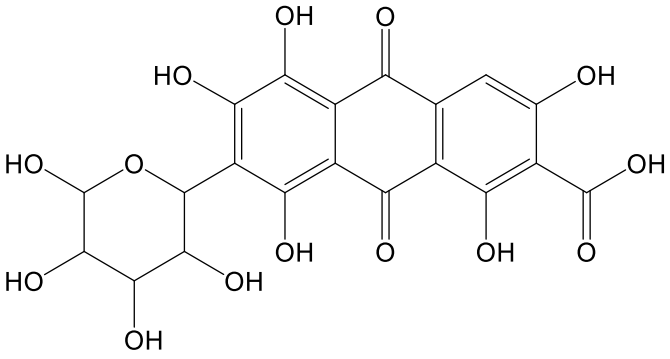
Carminic acid (CA) which is a natural dye and whose IUPAC name is "7-a-D-glycopyranosyl-9,10-dihydro-3,5,6,8-tetrahydroxy-1-methyl-9,10-dioxo-2 anthracenecarboxylic acid") is one of the most used substances as a red coloring agent (E120) in foods, beverages, pharmaceuticals, cosmetics, and textiles due to its high chemical and biological stability (Chung et al., 2001; Yilmaz et al., 2014). CA is generally obtained by extraction from the cochineal, which consists of the dried stems of the female *Dactylopius coccus Costa* beetle (Alghamdi et al.,

Fig. 1. Classification of dyestuffs according to their chemical structures.

2009; Reyes-Salas et al., 2011). The molecular structure of CA, it consists of an anthraquinone chromophore, a sugar residue, and a carboxyl group (Scheme 2). Carmine is the aluminum and calcium salt of carminic acid and has been used as a paint and lacquer pigment since ancient times and is one of the oldest natural dyestuffs (Alghamdi et al., 2009). Moreover, the use of carmine in foodstuffs is not recommended can still be found in supermarket foods such as Indian curries, although it has been licensed by European food safety authorities and predominantly for coloring in alcoholic beverages (Alghamdi et al., 2009). It is also employed as a food coloring in innumerable different products such as fruit juice, ice cream, soft drinks, yogurt, and candies (Alghamdi et al., 2009; Jiang et al., 2017).

Carminic acid is also known as a substance belonging to the types of antitumor and antibiotic anthracycline derivatives. It has been reported to have an increased hyperactivity effect in a few cases. Also, ingestion of carmine has been shown to cause food anaphylaxis and food allergy (Quirce et al., 1994; Wüthrich et al., 1997). Carmines in foods and beverages, and possibly CA, are known to cause allergic effects in some people. CA has been associated with urticaria, anaphylaxis, and angioedema via IgE (Sadowska et al., 2020). The fast, reliable, easy, and inexpensive new analytical methods are needed to better understand the unique red color provided by carminic acid, its chemical behavior, and to measure its amount in foods for quality control purposes.

In the literature search carried out so far, analytical methods such as high-performance liquid chromatography (HPLC), and spectrophotometry, as well as capillary electrophoresis, and thin layer chromatography (TCL) have been reported for monitoring of carminic acid amaranth. Moreover, for the carminic acid determination, chemiluminescence (Mokhtari et al., 2015), HPLC (Carvalho and Collins, 1997; Koizumi, 1996; Lim et al., 2014), HPLC/photodiode array detectors (PDA) (Nishizaki et al., 2018), microemulsion electrokinetic chromatography (MEEKC) (Huang et al., 2005), micellar electrokinetic chromatography (MEKC) (Maguregui et al., 2007), spectrofluorometry (VILCHEZ et al., 1994), spectrophotometry (Aznarez et al., 1985; Manzoori et al., 2000; Samari et al., 2010), inductively coupled plasma mass spectrometry (ICP-MS) (Floquet et al., 2017; Kuru et al., 2019), fluorimetry (Aznarez et al., 1985), whereas for the detection of amaranth, fluorescence spectrometry (Zhu et al., 2018), spectrophotometry (Pourreza and Elhami, 2009; Sha and Zhu, 2015), high performance capillary electrophoresis (Sun et al., 2011), TLC (Tonica et al., 2018), HPLC-UV (Chen et al., 2015; Floriano et al., 2018), HPLC-MS (Xian et al., 2013), capillary electrophoresis (Mahale et al., 2021) have been utilized. Although these aforesaid traditional analytical methods have been fully validated, widely used, and preferred in routine analysis, they have still shortcomings such as being expensive, long analysis times, requiring expertise, and crowded pre-processing requirements. Therefore, the academic community has focused on the development of a new, reliable, easy, portable, and sensitive analytical method for drugs, dyestuffs, pesticides, phenolic compounds, foodstuffs, metals, and electro-active compounds, etc. (DEMIR, 2019; Demir et al., 2021; Demir and İnam, 2014; Demir and Silah, 2020; Eren et al., 2015; Inam et al., 2020; Jamali et al., 2014; Karimi-Maleh et al., 2022). Electrochemical systems shine a light on the world of science as an analyzer for numerous samples testing, because they require inexpensive equipment and easy handling, can be miniaturized, and are accessible with simple dipstick sampling (Bijad et al., 2021; Cahuantzi-Muñoz et al., 2019; Hojjati-Najafabadi et al., 2020, 2022; Hussain et al., 2021a; Raeder et al., 2008). Moreover, they are powerful tools in the fields of biosensor and chemical sensor research (Karimi-Maleh et al., 2021a, 2021b; Mohanraj et al., 2020; Sohrabi et al., 2022). Electrochemical methods are the best candidates for the determination of coloring agents as dyestuffs since they are swift, cost-effective, available for on-site analysis, reliable, and sensitive (Nuñez-Dallos et al., 2018; Wang et al., 2015a).



Scheme 2. The molecular structure of carminic acid.

In the previous decade, electrochemical methods have been utilized extensively for the analysis of carminic acid amaranth as food colors, which have piqued the scientific community's curiosity (Nuñez-Dallos et al., 2018; Wang et al., 2015a). With the developing advanced nanomaterials (Deeksha et al., 2021; Hosseinzadeh et al., 2021; Joseph et al., 2020; Peiravi and Alinejad, 2021; Yassin et al., 2020; Yorseng et al., 2020), they enable a more sensitive analysis of electroactive materials and these substances (Babu et al., 2021; Ensafi et al., 2012; Karimi-Maleh et al., 2014; Nangare et al., 2021; Taherkhani et al., 2014). The spread of nanomaterials in various branches of science (Arefi-Oskoui et al., 2022; Keyikoglu et al., 2022; Taherian et al., 2021b; Xia et al., 2020), especially electrochemistry(Orooji et al., 2022; Taherian et al., 2021a), has led to the development of new approaches in new technologies(Arzaghi et al., 2021; Bai et al., 2020), especially electrochemical sensors(Ensafi and Maleh, 2010; Karimi-Maleh et al., 2014). Also, they provide not only analysis but also extraordinary selectivity with electrochemical monitoring methods at a level comparable to traditional methods(Akhgar et al., 2012; Baghayeri et al., 2013; Hussain et al., 2021b; John et al., 2021; Khand et al., 2021; Mirmomtaz et al., 2008; Raoof et al., 2011).

2. Overview of amaranth in electrochemical monitoring

Since amaranth a food colorant is not only an oxidizing substance but also a reducing agent (Fig. 2 and Fig. 3) (Ni and Bai, 1997; Sheikhshoaie et al., 2017; Sun et al., 2005), it results in a well-defined and sharp peak in both anodic, and cathodic voltammetric studies. The oxidation peak of amaranth can be observed at around 0.2 V depending on the pH of the supporting electrolyte, and type of the working electrode, while the reduction peak occurs at −0.5 V. In addition, mercury electrode has been commonly used in cathodic studies over a period of time, carbon paste electrode (CPE), glassy carbon electrode (GCE), and screen-printed electrode (SPE) and new generation composite sensors from these electrodes have been preferred in anodic studies for the analysis of food color amaranth. The large surface area, and the electrocatalytic properties of the developed modern sensors have enabled more precise determination of amaranth in food samples by electrochemical monitoring.

Among the various electrochemical methods, voltammetric techniques have been the most preferred analytical method for the determination of amaranth. These electrochemical monitoring techniques can be classified as cyclic voltammetry (CV), differential pulse voltammetry (DPV), linear sweep voltammetry (LSV), and square wave voltammetry (SWV) (Akbari, 2020; Al-Ghamdi, 2009; He et al., 2018; Jing et al., 2017; Ni and Bai, 1997; Pogacean et al., 2018; Sun et al., 2005, 2021; Tajik et al., 2021; Tvorynska et al., 2018; Wang et al., 2018, 2021; Wu et al., 2021; Zhang et al., 2013). Furthermore, there are a few studies by using stripping modules of square wave and differential pulse voltammetric techniques (SWSV and DPSV). Rarely used direct current adsorptive stripping voltammetry (DC-AdSV) (Sun et al., 2005), ratio derivative voltammetry (RDV) (Bijad et al., 2021), and microchip electrophoresis as electrochemical monitoring were also implemented for the determination of amaranth in natural samples. However, although it

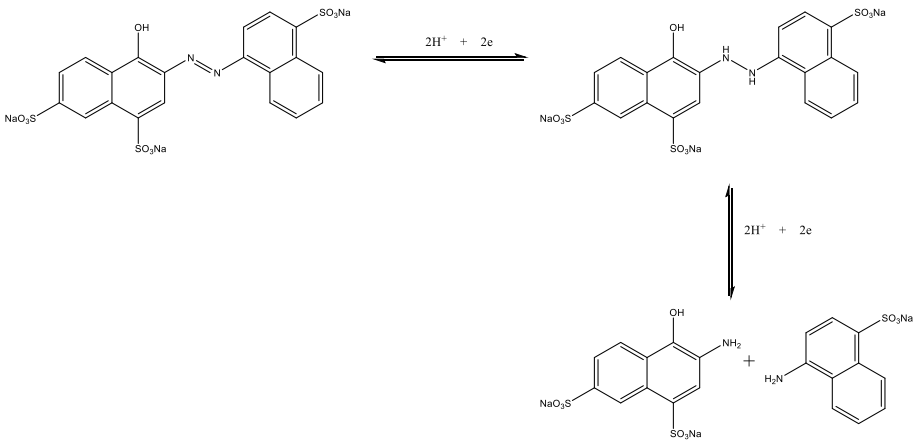


Fig. 2. Electrochemical reduction mechanism of amaranth (Al-Ghamdi, 2009).

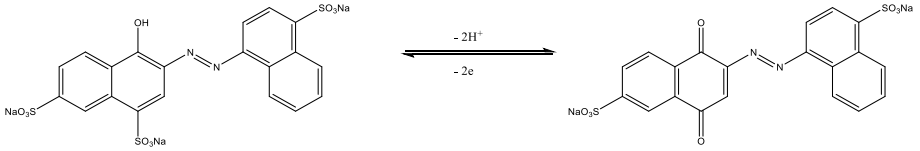


Fig. 3. Electrochemical oxidation mechanism of amaranth (Wang et al., 2018).

provides extremely important information about the electroactive material, the CV technique cannot be preferred for quantitative analysis. The main reason for this is that the sensitivity of this method is very passive compared to other electrochemical methods. Nevertheless, successful determination for the amaranth food color has been achieved at μM levels in natural samples using SPE and CPE based nanosensors (Char et al., 2008; Mahale et al., 2021) (Table 1).

Among the electrochemical monitoring models, the most used method for the determination of amaranth food color is DPV and its stripping technique, so-called DPSV (Table 2). This voltammetric technique, which is sensitive, reliable, and reproducible, has been analyzed with very low detection limits of amaranth in fruit juices, soft drinks, and wine and natural water samples [63–75]. Furthermore, in these studies, numerous bare and modified composite sensors have been developed, including GCE (Ghanbari et al., 2019), CPE (Chandran et al., 2014), SPE (Wang et al., 2010), screen-printed carbon electrode (SPCE) (Ni and Bai, 1997), and modified new generation sensors (Al-Ghamdi, 2009; Capoferri et al., 2017; Char et al., 2008; Fereiduni et al., 2019; Hu et al., 2018; Huang et al., 2017; Steinfeld et al., 2015; Wang et al., 2018). Many materials with outstanding properties have been used in the production of composite electrodes for the analysis of amaranth food color. This hybrid electrode; reduced graphene oxide (rGO) (Ni and Bai, 1997), multi-walled carbon nanotube (MWCNT) (Fereiduni et al., 2019; Steinfeld et al., 2015), Fe_3O_4 nanoparticles (NPs) (Han et al., 2014), single-walled carbon nanotube (SWCNT) (Capoferri et al., 2017) Co_3O_4–CeO_2 nanoparticles (NPs) (Hu et al., 2018), graphene quantum dots–ionic liquid (GQD-IL) (Huang et al., 2017), Al_2O_3-microfibes [73], Ni–Mo metal–organic framework (MOF) [74], and mercury meniscus-silver solid amalgam electrode (m-AgSAE) (He et al., 2015; Sun et al., 2005) have taken place in the literature on the field of electrochemical monitoring. When these studies are examined, the detection limits are generally at the micromolar level (μM). Electrochemical studies developed for the determination of amaranth at nanomolar levels (nM) have also been found in the literature. Especially thanks to the well-defined anodic peak in the reduction direction of amaranth, the DPV, and DPSV as electrochemical monitoring using mercury or amalgam electrodes have equivalent determination limits according to the chromatographic, spectrophotometric, and even sensitive analytical methods. Furthermore, thanks to the catalytic feature of these developed sensors, they allow a more selective, easier, and specific analysis of amaranth substances.

The linear sweep voltammetry (LSV) is an electrochemical method that is frequently used not only for qualitative analysis but also for quantitative analysis of natural samples of electro-active species. LSV is one of the electrochemical models applied for the determination of amaranth food coloring matter (Akbari, 2020; Tajik et al., 2021; Tvorynska et al., 2018; Wang et al., 2021; Zhang et al., 2013). While bare electrode operation is rarely observed in this unique technique, there are many studies with composite sensors (Akbari, 2020; Tajik et al., 2021; Tvorynska et al., 2018; Wang et al., 2021; Zhang et al., 2013). A new generation of sensors has been developed using hybrid materials such as Co_3O_4 nanoparticles (NPs) (Akbari, 2020), molecular imprinted polymer (MIP) (Zhang et al., 2013), and multi-walled carbon nanotube (MWCNT) (Zhang et al., 2013) treated with bare electrodes. As a result, the low detection limits of amaranth with electrochemical monitoring were a substantial benefit. Moreover, LSV has promoted sensitive, reliable, and rapid analysis for the determination of amaranth food color amaranth in soda varieties, juice samples, and soft drinks (Akbari, 2020; Tajik et al., 2021; Tvorynska et al., 2018; Wang et al., 2021; Zhang et al., 2013) (Table 3).

The square wave (SWV) and its stripping (SWV) voltammetry techniques are among the most effective electrochemical monitoring

Table 1
Determination of amaranth by CV technique.

Analyte	Method	Electrode	Linear range (μM)	LOD (nM)	LOQ (nM)	Samples	Optimum pH	Interference	Peak potential (V)	Ref
AM	CV	SPE	$0.15 \times 10^6 - 1.20 \times 10^6$	–	–	–	pH 3.2 (PBS)	–	+0.6	Mahale et al. (2021)
AM	CV	SDS/CPE	–	–	–	–	H_2SO_4	–	–	Char et al. (2008)

Table 2
Determination of amaranth by DPV and DP-AdSV techniques.

Analyte	Method	Electrode	Linear range (μM)	LOD (nM)	LOQ (nM)	Samples	Optimum pH	Interference	Peak potential (V)	Ref
AM	DP-AdSV	m-AgSAE	0.004–0.089	2.2	–	Soft drink	pH 3.6 (ABS)	–	−0.1	Steinfeld et al. (2015)
AM	DPV	rGO-methionine/SPCE	10–100	57		Sprite, SPY classic red	pH 4.0 (ABS)	NaCl, KCl, glucose, ascorbic acid, citric acid, sucrose, glycine	+0.8	Fereiduni et al. (2019)
AM	DPV	PSS-GR-Pd/GCE	0.1–9	7	–	Fanta drink	pH 2.0 (PBS)	Citric acid, tartaric acid, ascorbic acid, caffeine, theophylline, histidine, sudan I tartrazine, carvacrol, vanillin	+0.8	Capoferri et al. (2017)
AM	DPV	PLA-ERGO/GCE	0.75–75	250	–	Soft drink	pH 2.5 (PBS)	I^-, Cl^-, NO_3^{2-}, $C_2O_4^{2-}$, SO_4^{2-}, Na^+, K^+, Ba^{2+}, Mg^{2+}, Zn^{2+}, Ca^{2+}, Al^{3+}, Cr^{3+}, Cu^{2+}, Fe^{2+}, oxalic acid, starch, folic acid, L-cysteine, dopamine, L-serine, L-histidine	–	Hu et al. (2018)
AM	DPV	GS/GCE	0.0025–0.125	0.75	–	Soft drink	pH 4.6 (ABS)	Fe^{3+}, Zn^{2+}, Mg^{2+}, glucose, sucrose, caffeine, benzoic acid, glycine, quinoline yellow, citric acid, vitamin C, allura red, sudan red, sunset yellow	+0.7	Huang et al. (2017)
AM	DPV	MWCNT/GE	1–10	68	–	Soft drink	pH 5.0 (ABS)	NaCl, Na_2SO_4, $NaNO_3$, glucose, tartrazine, citric acid, ascorbic acid	+0.7	Chandran et al. (2014)
AM	DPV	Fe3O4@rGO/GCE	0.05–50	50	–	Grapes flavor, watermelon flavor, peach flavor	$[Fe(CN)_6]^{3-/4-}$	K^+, Na^+, glucose, sucrose, citric acid, erythrosine, new coccine, allure red	+0.1	Han et al. (2014)
AM	DPV	MWCNT/GCE	0.04–0.8	35	–	Soft Drinks	pH 5.0 (ABS)	Fe^{3+}, Fe^{2+}, Ca^{2+}, glucose, sucrose and glycine, citric acid, vitamin C, tartrazine	+0.7	Wang et al. (2010)
AM	DPV	SWCNT-TiN/GCE	0.1–100	40	–	Beverage	pH 5.0 (ABS)	Glucose, sucrose, citric acid, glycine, nitrite, acetaminophen, cysteine, glutathione, erythrosine, new coccine, allure red	+0.7	He et al. (2015)
AM	DPV	Co_3O_4–CeO_2/Gr nanocomposite	2–96	159.1		Soft drinks	–	–	–	Wang et al. (2021)
AM	DPV	GQD-IL/CPE	0.1–400	30	–	Tap water, well water, river water, apple juice, orange juice	pH 7.0 (PBS)	Na^+, Cl^-, L-threonine, alanine, glycine, glucose, guanine, ascorbic acid, tryptophan, tyrosine, Sudan I, vitamin B_6, vitamin B_9	+0.7	Akbari (2020)
AM	DPV	Al_2O_3-microfibes/CPE	1–150	0.75	–	Drink samples	pH 6.0 (PBS)	–	+0.7	Zhang et al. (2013)
AM	DPV	PDDA-Gr-(Pd–Pt) @MIPPDA/GCE	0.006–10	2	–	Soft drink	pH 3.0 (PBS)	Sunset yellow, tartrazine	+0.8	Shabani et al. (2009)
AM	DPV	Ni–Mo-MOF/SPE	0.15–500	50	–	Fruit juices, water resources	pH 7.0 (PBS)	Na^+, Cl^-, K^+, Mg^{2+}, NO_3^-, sudan I, glucose, sucrose, L-threonine, alanine, glycine, guanine, tryptophan	+0.6	Tajik et al. (2021)
AM	DPV	OMC/GCE	0.1–3	32	–	Wine	pH 7.0 (PBS)	Sunset yellow, kermes red	+0.7	Jian and Li-Ping (2013)
AM	DPV	GCE	1–283.7	500	–	Red grapes, orange	pH 4.0 (PBS)	Tartrazine, quinoline yellow, sunset yellow	+0.7	Ghanbari et al. (2019)
AM	DPV	m–AgSAE	0.004–0.089	2.2	–	Soft drink	pH 3.6 (ABS)	–	−0.1	Tvorynska et al. (2018)

techniques. It is of great interest in the scientific world since they are both highly sensitive, and the measurement process can be conducted in the order of seconds. SWV and SWSV are the most employed electroanalytical method in the establishing of amaranth (He et al., 2018; Jing et al., 2017; Pogacean et al., 2018; Sun et al., 2021; Wu et al., 2021). With this superb method, it is possible to detect the food colorant at a very low detection limit as nanomolar (nM) (He et al., 2018; Hojjati--Najafabadi et al., 2020, 2022; Jing et al., 2017; Pogacean et al., 2018; Raeder et al., 2008; Sun et al., 2021; Wang et al., 2015b; Wu et al., 2021). The determination of this substance in these concentrations in a few seconds is unbelievable. Moreover, with this electroanalytical method, validation parameters such as accuracy, precision, and reproducibility have been obtained with satisfactory low relative error for the determination of amaranth food color amaranth in soda varieties, juice samples, soft drinks, and other foodstuffs (see Table 4).

In the SWS and SWSV electroanalytical methods, bare SPE (Pogacean et al., 2018), expanded graphite paste electrode (EGPE) (Jing et al., 2017), and modified electrochemical indicator electrodes (He et al., 2018; Hojjati-Najafabadi et al., 2020, 2022; Nuñez-Dallos et al., 2018; Sun et al., 2021; Wang et al., 2015b; Wu et al., 2021) have been utilized

Table 3
Determination of amaranth by LSV technique.

Analyte	Method	Electrode	Linear range (μM)	LOD (nM)	LOQ (nM)	Samples	Optimum pH	Interference	Peak Potential (V)	Ref
AM	LSV	GTA/AuE	0.3–100 1–10	100	–	–	pH 4.4 (ABS)	–	+0.7	Pogacean et al. (2018)
AM	LSV	PSS-GN/ Co_3O_4/GCE	0.01–6.0	4		Soft drink	pH 2.5 (PBS)	Mg^{2+}, Ca^{2+}, Cl^-, SO_4^{2-}, brilliant blue, sodium benzoate, glucose, glycine sucrose, citric acid, tartrazine	+0.8	Jing et al. (2017)
AM	LSV	MIPs/ MWCNTs/ GCE	0.007–1.0 0.4–17.0	0.4	–	Watermelon juice, grape juice, orange juice	pH 6.0 (PBS)	K^+, Na^+, Ca^{2+}, Mg^{2+}, glucose	+0.3	Wu et al. (2021)
AM	LSV	Ti/ nanoSnO_2-α-Fe_2O_3	–	–	–	–	0.1 M Na_2SO_4	–	–	Sun et al. (2021)
AM	LSV	MnO_2NRs-ErGO/GCE	0.02–400	1	–	Grape soda, watermelon soda, candy, lemon soda	–	Lemon yellow, sunset yellow, rhodamine B, ponceau 4R	+0.8	He et al. (2018)

Table 4
Determination of amaranth by SWV and SW-AdSV techniques.

Analyte	Method	Electrode	Linear range (μM)	LOD (nM)	LOQ (nM)	Samples	Optimum pH	Interference	Peak Potential (V)	Ref
AM	SW-AdSV	SPE	0.15–1.20	26	–	Orange jelly, soft drink	pH 3.2 (PBS)	Sunset yellow, allura red, sudan I, sudan II	+0.6	Arancibia Moya et al. (2017)
AM	SW-AdSV	copper(I) helicate-SWCNT/SPE	–	30	–	orange juice, soft drink	pH 3.2 (PBS)	PonceauR-4, allure red, sunset yellow, sudan I, sudan II	+0.6	Nuñez-Dallos et al. (2018)
AM	SWSV	EGPE	0.008–4	36	109.1	Grape juice	pH 7.0 (BRB)	SO_4^{2-}, Cl^-, Ca^{2+}, Mg^{2+}, sucrose, glucose, sodium benzoate, vitamin C, glycine, tartrazine, citric acid, Triton X-100	+0.6	Zhang et al. (2014)
AM	SWSV	CNT–ppy/ GCE	0.005–0.5	0.5	–	Fruit drinks	pH 7.0 (BRB)	Fe^{2+}, Ca^{2+}, Cu^{2+}, Mg^{2+}, SO_4^{2-}, NO_3^-, Cl^-, citric acid, sodium oxalate, oxalic acid, tartrazine, quinoline yellow, saccharin, glucose, glycine, vitamin C, sodium citrate	+0.7	Wang et al. (2015a)
AM	SWV	RuO2/NR/ DPIBr/CPE	0.008–550	3	–	Soft drink, orange juice, orangeade juice, Apple juice	pH 7.0 (PBS)	Na^+, Cl^-, L-threonine, glucose, alanine, glycine, guanine, tyrosine, ascorbic acid, tryptophan, Sudan I, vitamin B_6, vitamin B_9	+0.7	Sheikhshoaie et al. (2017)
AM	SWV	1-M-3-BIBR/CuO/ SWCNTs/ CPE	0.004–750	1	–	Orange juice, apple juice, Sausage	pH 7.0 (PBS)	Mg^{2+}, F^-, NO_3^-, K^+, Na^+, Cl^-, glucose, vitamin B_9, ascorbic acid, sucrose, alanine, methionine, isolucin, sudan I, bisphenol A, vitamin B_6, starch	+0.8	Bijad et al. (2017)
AM	SWV	CNT/GO-IL/ GCE	0.0005–4.0	0.1	–	Bacardi breezer, reinnbow rum cocktail drinks	pH 7.0 (BRB)	Zn^{2+}, Ca^{2+}, Cu^{2+}, Mg^{2+}, Na^+, SO_4^{2-}, NO_3^-, Cl^-, quinoline yellow, tartrazine, glucose, sucrose, saccharin, vitamin C, sodium citrate, oxalic acid, sodium oxalate	+0.7	Wang et al. (2015b)
AM	SWV	GNM/GCE	0.005–1.0	0.7	–	Chocolate beans, fruit drink	pH 7.0 (PBS)	Fe^{3+}, Zn^{2+}, Mg^{2+}, Ca^{2+}, Cl^-, glucose, sucrose, citric acid, nitrite, glycine, cysteine, glutathione, allure red, quinolone yellow	+0.7	Wang et al. (2018)
AM	SWV	EGPE	0.01–4.0	5	–	Changyu cabernet, Great Wall dry white wine, and Dynasty white wine	pH 7.0 (BRB)	Ca^{2+}, Mg^{2+}, SO_4^{2-}, Cl^-, triton X-100, sucrose, sodium benzoate, glycine, vitamin C, citric acid, tartrazine, sunset yellow, glucose	+0.7	Wang et al. (2013)
AM	SWV	*f*GO/CS/IL/ NPG/GCE	0.008–1.2	23	–	Watermelon-flavor, grape flavor, rose flavor	pH 6.0 (BRB)	Glucose, sucrose, potassium sorbate, sodium hexametaphosphate, sodium citrate, tartrazine	−0.1	Zhang et al. (2022)

as working electrodes for the analysis of amaranth. However, numerous hybrid sensors have been produced for the assessment of amaranth in natural samples by using SWS and SWSV in electrochemical monitoring. Carbon nanotubes (CNT), and its derivatives (SWCNT or MWCNTP), metal oxide nanoparticles (MO NPs), nanorod (NR) graphene oxide (GO) and its derivatives like porous graphene nanomesh (GNM), poly pyrrole (ppy), and ionic liquids (ILs) have been preferred in the construction of composited sensors with indispensable characteristics (Table 4). Thus, the amaranth substance have not only allowed for sensitive determination but also selectively determined in the natural sample at low relative standard deviation (see Table 4).

In electrochemical monitoring, direct current adsorptive stripping voltammetry (DC-AdSV), direct current voltammetry, microchip electrophoresis, and ratio derivative voltammetry (RDV), and polarography techniques are rarely used for the determination of amaranth food color (Tables 5 and 6). Mercury-based indicator electrodes are commonly used in these methods. There is one study that utilized a glassy carbon electrode (Zhang et al., 2014). The lowest limit of detection (LOD) has been obtained at a very sensitive level of 0.41 nM using the hanging mercury drop electrode (HMDE) (Sun et al., 2005). Analytical application of amaranth food color in soft drinks and fruit juices has been realized successfully with these less applied methods (Arancibia Moya et al., 2017; Bijad et al., 2021; He et al., 2015; Sun et al., 2005; Wang et al., 2010; Zhang et al., 2014).

3. Overview of carminic acid in electrochemical monitoring

Although there are not many studies like amaranth dyestuff, electrochemical impression techniques using polarography and square wave voltammetry and its stripping technique (SWV and SWSV) have been found in the literature for the determination of carminic acid food color (Tables 7–9). Although rarely seen in the determination of carminic acid, electrochemical methods including potentiometric and conductometric titrations have been encountered in the literature (Reyes-Salas et al., 2011). When these studies are examined in detail, it can be observed that carminic acid exhibits both anodic and cathodic peaks (Fereiduni et al., 2019; Jiang et al., 2017; Sun et al., 2006; Yilmaz et al., 2014). Therefore, it is possible to analyze it using the reduction and oxidation peaks of carminic acid food colorant (Fig. 4 and Fig. 5). While mercury indicator-based polarography studies were intensively carried out in cathodic studies (Table 7), only one study was found in the anodic direction using the modified graphite electrode (GE) as a working sensor (Jiang et al., 2017). When we look at the sensor type, we come across two different working electrodes. The first of these is the mercury electrode, which also forms the basis of polarography (Alghamdi et al., 2009; Reyes-Salas et al., 2011; Yilmaz et al., 2014). Mercury sensors based on two different mechanical working principles, the drop mercury electrode (DME) and the hanging mercury drop electrode (HMDE) have been used for the analysis of carminic acid in foodstuffs (Alghamdi et al., 2009; Reyes-Salas et al., 2011; Yilmaz et al., 2014). The other is the solid electrode coated with Pd–Au bimetallic nanoparticles and polyproline (Poly(Pr)) of the graphite electrode, which is an important type of carbonaceous material (Jiang et al., 2017). Comparing the detection limits of these electrodes for carminic acid, it is seen that HMDE is the most sensitive one at the 1.43 nM level (Alghamdi et al., 2009). Interestingly, it can be said that the sensitivity of the constructed Pd-AuNPs/Poly(Pr)/GE modified sensor is almost as good as HMDE (Jiang et al., 2017). The recommended electrochemical modules of carminic acid in milk, candy, and soft drinks have been successfully determined to demonstrate its accuracy and precision. Moreover, the carminic acid has been selectively detected by using proposed nanosensors in the presence of interfering species such as some anions and cations, and various organic substances. As a result, accurate, fast, reliable, and on-site analyzes of an important natural food color as carminic acid has been carried out to detect with both anodic and cathodic studies at levels comparable to other validated analytical methods.

4. Conclusions

Amaranth and carminic acid food colors are important substances in terms of both human health and food safety. Although these substances do not have a direct carcinogenic effect, they have negative effects on human health. Therefore, scientists need new analytical methods for accurate, sensitive, fast, and reliable analysis of these substances in samples from foods, medicines, cosmetics, and other uses. Electrochemical monitoring has aroused a substantial interest in the scientific world in the last 20 years due to its simple, portable, sensitive, and reproducible results. Electroanalytical procedures, which are mostly suggested voltammetric techniques for amaranth and carminic tests, have yielded excellent outcomes. The analyzes in natural samples of these two food color substances, which give both anodic and cathodic peaks, were performed satisfactorily. In these studies, the most preferable electrode for the cathodic studies is the mercury electrode, while the carbon indicator electrodes mostly preferred to appear in anodic studies. In addition, new composite sensors with countless features have been produced by coating the surfaces of bare electrodes with polymers, metal oxide nanoparticles, ionic liquid, carbon nanotubes (CNTs, SWCNT and MWCNT), graphene oxide (GO and rGO), molecularly imprinted polymers, and metal-organic framework (MOF). Moreover, with the developing technology and new generation sensors in recent years, electrochemical monitoring has limits of detection (LOD) comparable to conventional analytical levels for both food colors. In the order of seconds, the determinations of these two important food color substances have been carried out at nM levels by proposed methods on the nanosensor. Furthermore, hybrid electrodes with catalytic capabilities have been used to achieve more selective and sensitive electrochemical tests. The amaranth and carminic acid have been also selectively detected on the nanosensors in the presence of interfering species such as anions, cations, and organic substances. In addition, in terms of green chemistry, electrochemical methods, which are very environmentally friendly, allow a sensitive, reliable, and on-site analysis

Table 5
Determination of amaranth by polarography technique.

Analyte	Method	Electrode	Linear range (μM)	LOD (nM)	LOQ (nM)	Samples	Optimum pH	Interference	Peak potential (V)	Ref
AM	Polarography	HMDE	0.001–0.3	0.69	–	–	pH 3.6 (ABS)	–	–	Steinfeld et al. (2015)
AM	Polarography (RDV)	HMDE	0.05–0.266	50	–	Orange juice, fruit juice, merida orangeade	pH 6.0 (McIlvane buffers)	–	−0.4	Ni and Bai (1997)
AM	Polarography	HMDE	0.01–0.11	1.7	–	Soft drink	pH 10 (CBS)	Sunset yellow, gelatin, natural and artificial sweeteners, tartrazine, preservatives, antioxidants	−0.6	Al-Ghamdi (2009)

Table 6
Determination of amaranth by different electrochemical technique.

Analyte	Method	Electrode	Linear range (μM)	LOD (nM)	LOQ (nM)	Samples	Optimum pH	Interference	Peak Potantial (V)	Ref
AM	DC-AdSV	p-AgSAE	0.089–0.7	30	–	–	pH 3.6 (ABS)	–	−0.2	Steinfeld et al. (2015)
AM	DC-AdSV	m-AgSAE	0.004–0.089	2.1	–	Soft drink	pH 3.6 (ABS)	–	−0.2	Steinfeld et al. (2015)
AM	DC-AdSV	HMDE	0.001–0.3	0.41	–	–	pH 3.6 (ABS)	–	−0.2	Steinfeld et al. (2015)
AM	DCV	m–AgSAE	0.008–0.6	3.3	–	Soft drink	pH 3.6 (ABS)	–	−0.1	Tvorynska et al. (2018)
AM	CA	Ni–Mo-MOF/SPE					pH 7.0 (PBS)	–	–	Tajik et al. (2021)
AM	MCE	GCE	–	15100	–	Soft drinks, candies	pH 11 (PBS)	–	–	Dossi et al. (2007)

Table 7
Determination of carminic acid by polarography technique.

Analyte	Method	Electrode	Linear range (μM)	LOD (nM)	LOQ (nM)	Samples	Optimum pH	Interference	Peak Potential (V)	Ref
CA	CP	DME	50–300	18000	–	–	Ethanol-water (1:1)	–	–	Reyes-Salas et al. (2011)
CA	DPP	DME	4–300	1100	–	Cochineal	Ethanol-water (1:1)	–	–	Reyes-Salas et al. (2011)
CA	DPP	HMDE	0.05–0.125	1.43	–	Ice cream and soft drinks	pH 3.0 (ABS)	Amaranth, tartrazine, sunset yellow, erythrosine B, allura red	−0.3	Alghamdi et al. (2009)
CA	DPP	DME	1–90	160	550	Milk, candy	pH 2.0 (BRB)	Cu^{2+}, Fe^{3+}, Pb^{2+}, Zn^{2+}, Cd^{2+}, NO_2^-, SO_3^{2-}, Se^{4+}, K^+, Na^+, NH_4^+, Ca^{2+}, Mg^{2+}, NO_3^-, SO_4^{2-}, Cl^-	−0.4	Yilmaz et al. (2014)

Table 8
Determination of carminic acid by SWV and DPV techniques.

Analyte	Method	Electrode	Linear range (μM)	LOD (nM)	LOQ (nM)	Samples	Optimum pH	Interference	Peak Potential (V)	Ref
CA	SWV	Pd-AuNPs/Poly (Pr)/GE	0.01–1	5.9	19.7	Popping candy	pH 8.0 (PBS)	Glucose, lactose, saccharose	+0.7	Jiang et al. (2017)
CA	DPV	rGO-methionine/SPCE	1–20 20–60	36	–	Sprite, betagan, ice-cream	pH 3.0 (PBS)	NaCl, KCl, glucose, sucrose, ascorbic acid, citric acid, glycine	+0.5	Fereiduni et al. (2019)

Table 9
Determination of carminic acid by different electrochemical technique.

Analyte	Method	Electrode	Linear range (μM)	LOD (nM)	LOQ (nM)	Samples	Optimum pH	Interference	Peak Potential (V)	Ref
CA	Potentiometric titration	–	–		–	–	Ethanol-water (1:1)	–	–	Reyes-Salas et al. (2011)
CA	Conductimetric titration	–	–	–	–		Ethanol-water (1:1)	–	–	Reyes-Salas et al. (2011)
CA	MCE-PD	Membrane driving electrode	0.2–1.0	69	207	Model sample, soft candy, hard candy, radler, saffron, medicinal lollipops, lozenges	pH 6.0	–	–	Masár et al. (2020)

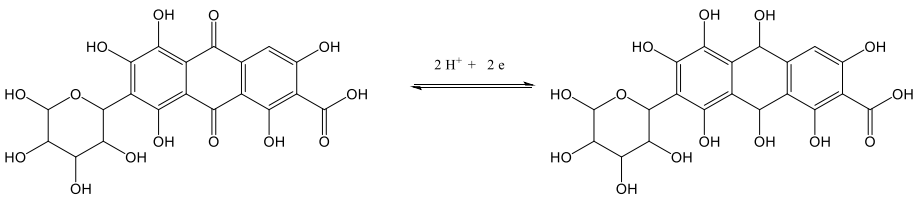


Fig. 4. Electrochemical reduction mechanism of carminic acid (Alghamdi et al., 2009).

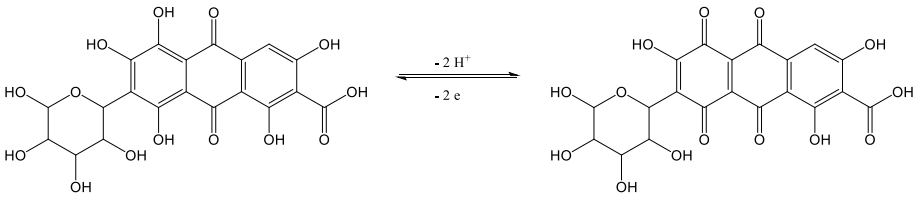


Fig. 5. Electrochemical oxidation mechanism of carminic acid (Jiang et al., 2017).

of these dyestuffs. Consequently, accurate, fast, reliable, and in-situ analysis of crucial agents such as amaranth, and carminic acid have been performed with both anodic, and cathodic studies at levels comparable to other validated analytical methods.

The biggest shortcoming of electrochemical methods with their outstanding capabilities is that they are not yet fully used in routine analysis. The reason for this is that although electrochemical methods have gained great momentum in the last three decades, expert analysts are not often encountered in the field. However, electrochemistry is the method that will provide the greatest on-site analysis of the future and offers great hope to the scientific world.

CRediT authorship contribution statement

Marzieh Alizadeh: Writing – original draft, preparation and revise of paper. **Ersin Demir:** Writing – original draft, preparation and revise of paper. **Nida Aydogdu:** Writing – original draft, preparation and revise of paper. **Najmeh Zare:** Writing – original draft, preparation and revise of paper. **Fatemeh Karimi:** Writing – original draft, preparation and revise of paper. **S. Masoud Kandomal:** Writing – original draft, preparation and revise of paper. **Hassan Rokni:** Writing – original draft, preparation and revise of paper. **Younes Ghasemi:** Writing – original draft, preparation and revise of paper.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Abbreviations

ABS Acetate buffer solution
CA Chronoamperometry
CBS Carbonate buffer solution
CNT Carbon nanotube
CNT–ppy/GCE Carbon nanotube and polypyrrole composite modified GCE
CP Classic Polarography
CPE Carbon paste electrode
CV Cyclic voltammetry
DC-AdSV Direct current absorptive voltammetry
DCV Direct current voltammetry
DME Mercury dropping electrode
DPP Differential pulse polarography
DPV Differential pulse voltammetry
EGPE Expanded graphite paste electrode
FDA Food and Drug Administration
GCE Glassy carbon electrode
GE Graphite electrode
GNM Graphene nanomesh
GO/CNT-IL: Graphene oxide, carbon nanotubes and ionic liquid
HMDE Hanging mercury dropping electrode
HPLC High-performance liquid chromatography
ICP-MS Inductively coupled plasma mass spectrometry
LOD Limit of detection
LOQ Limit of quantitation
LSV Linear sweep voltammetry
m-AgSAE Silver solid amalgam electrode modified by mercury meniscus
MCE-PD Microchip electrophoresis with photometric detection
MEEKC Microemulsion electrokinetic chromatography
MEKC Micellar electrokinetic chromatography
MIP Molecular imprinted polymer
MO NPs Metal oxide nanoparticles
MOF Metal–organic frameworks
MWCNT Multiwall carbon nanotube
nM Nanomolar
NR Nanoroad
OMC Mesoporous carbon
p-AgSAE Polished silver solid amalgam electrode modified by mercury
PBS Phosphate buffer solution
PDA Photodiode array detectors
Pd-AuNPs/Poly(Pr) Pd–Au bimetallic nanoparticles on a polypyroline
PSS-Gr-Pd Pd-doped polyelectrolyte functionalized graphene
RDV Ratio derivative voltammetry
rGO-methionine: Reduced graphene oxide and methionine
SDS Sodium dodecyl sulfate modified
SPCE Screen printed carbon electrode
SWCNT-TiN Single-walled carbon nanotube-Titanium nitride nanocomposites
SWSV Square wave stripping voltammetry
SWV Square wave voltammetry
TLC Thin Layer Chromatography

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