

A Review of Extraction and Analytical Methods for the Determination of Tartrazine (E 102) in Foodstuffs

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ABSTRACT

Tartrazine is an azo food dye, which is orange-colored and water soluble. It is usually used in foods, pharmaceuticals, cosmetics, and textiles. Tartrazine has the potential to cause an adverse health effect on humans, such as hyperactivity in children, allergy, and asthma. Joint FAO/WHO Expert Committee on Food Additive and EU Scientific Committee for Food have standardized the acceptable daily intake for tartrazine that is 7.5 mg kg⁻¹ body weight. Many researchers have detected the presence of tartrazine for monitoring the quality and safety of food products. In this review paper, we highlighted various tartrazine detection and extraction methods. Some of the analytical methods are available such as high-performance liquid chromatography, electrochemical sensor, thin-layer chromatography, spectrophotometry, capillary electrophoresis, and liquid chromatography-tandem mass spectrometry. Also, we discuss following extraction steps: liquid-liquid extraction, solid-phase extraction, membrane filtration, cloud point extraction, and other extraction method. In addition, a brief overview is presented explaining the synthesis process and metabolism of tartrazine and the maximum permitted level in different countries. This review paper will give an insight into different extraction and analytical methods for the determination of tartrazine in healthy foods, which will attract the attention of public toward food safety and quality, and also the interest of food industry and government bodies.

KEYWORDS

Chromatography;
electrochemical sensor;
extraction; synthetic color;
tartrazine

Introduction

Food colorants are added to foods and beverages to restore the color of food lost during processing and to enhance the color of the food or to uniform the color of the final product. Food colorants have been classified as natural or synthetic colorants. Most natural colorants are originated from plant sources such as anthocyanins, betanin, β -carotene, and chlorophyll, whereas some colorants such as carmine or carminic acid are extracted from the insects. However, natural colorants are unstable toward pH, heat, and light. Synthetic colorants are obtained from a chemical process and mostly classified as azo dyes, quinolone, xanthene, triarylmethanes, and indigoid. Synthetic colorants are widely used due to their stability toward pH, heat and light, water solubility, and lower production cost (Titus, 2013).

Tartrazine (E 102) is an azo dye, which is orange-colored, water-soluble powder. It is commonly used in food products, drugs, cosmetics, and pharmaceuticals (Tanaka, 2006). In food, tartrazine is used in soft drinks, juices, jellies, candies, cakes, cereal, soups, and other products (Demirkol and Ercal, 2012). It is known as Food Drug & Cosmetic Yellow No. 5, C.I. No. 19140 and Food Yellow No. 4 with European Community (EC) number E 102. When combined with sodium benzoate, a high concentration of tartrazine present in food may cause health complication such as hyperactivities in children. The

Acceptable Daily Intake (ADI) of tartrazine is 7.5 mg/kg bw/day by Joint FAO/WHO Expert Committee on Food Additive (JECFA) in 1966 and EU Scientific Committee for Food (SCF) in 1975 and 1984 (EFSA Panel on Food Additives and Nutrient Sources 2009). The maximum level of tartrazine in non-alcoholic beverages should not be more than 0.01 g/mL (De Andrade et al., 2014).

Numerous analytical techniques have been developed in determining tartrazine including spectrophotometry (Antakli et al., 2015, Sahraei et al., 2013, Olgun et al., 2012), thin-layer chromatography (TLC) (Tang et al., 2014, Kartsova et al., 2009), capillary electrophoresis (CE) (Česla et al., 2007, Prado et al., 2006), and high-performance liquid chromatography (HPLC) (Bento et al., 2015, Tavakoli et al., 2013, Bonan et al., 2013, Hajimahmoodi et al., 2013). Yamjala et al. (2015) used the analytical methods in determining azo dyes in food products, and Kaur and Gupta (2012) have determined synthetic food dyes and lakes in food products using spectrophotometry determination methods. Rebane et al. (2010) have analyzed the Sudan I-IV in various food matrices using LC-UV-Vis and liquid chromatography-tandem mass spectrometry (LC-MS) and determined synthetic dyes by chromatographic methods (Kucharska and Grabka, 2010). Ahlström et al. (2005) have developed the analytical methods for the determination of

banned azo dyes in consumer products. In our knowledge, there are no reviews previously which summarize the extraction and advanced analytical techniques for the determination of tartrazine in food stuffs. This review paper is focused on the available methods of extraction and analytical for determination of tartrazine; it will be worthwhile for food analysts and regulatory authorities to monitor and control the use of tartrazine in food and beverage products.

Tartrazine (E 102)

Tartrazine (E 102) is an azo group food dye that is characterized by N linkage ($-N=N-$) (Figure 1), orange-colored, and hydrophilic powder, with a molecular weight of 534.36 g/mol. This colorant was first discovered in 1884. Tartrazine is essentially the 3-carboxy-5-hydroxy-1-(4'-sulfonatophenyl)-4-(4'-sulfonatophenylazo)-*H*-pyrazol-3-carboxylate (EFSA Panel on Food Additives and Nutrient Sources, 2009) and the molecular formula is $C_{16}H_9N_4Na_3O_9S_2$ with density 0.70 g/mL. The solubility in water is 20.0 g/100 mL at 25°C, in glycerol is 18 g/100 mL at 25°C, in propylene glycol is 7.0 g/100 mL at 25°C (Marmion, 2001), ethanol is 0.8 mg/mL, and in ethylene glycol monomethyl ether is 20 mg/mL (Green, 1990). Melting point of tartrazine is greater than 300°C, and it can be detected at the wavelength of 425 nm in water (National Center for Biotechnology Information (NCBI) 2016). Large concentration of tartrazine in food may be harmful, and in a small portion of population, even a small dose when combined with sodium benzoate can give rise to health complication such as hyperactivities in children.

Synthesis of tartrazine

Tartrazine in foods is widely found in soft drinks, candies, jellies, jams, flavored chips, cakes, ice cream, soups, sauces, and cereals (Demirkol and Ercal, 2012) and non-food products found in cosmetics, medicine capsules, plastics, paint additives, and textiles (Stolz, 2001). Tartrazine is synthesized by condensing phenylhydrazine-*p*-sulfonic acid with oxalacetic diethyl ester that is paired with diazotized sulfanilic acid. The product ester is hydrolyzed with sodium hydroxide. The process is shown in

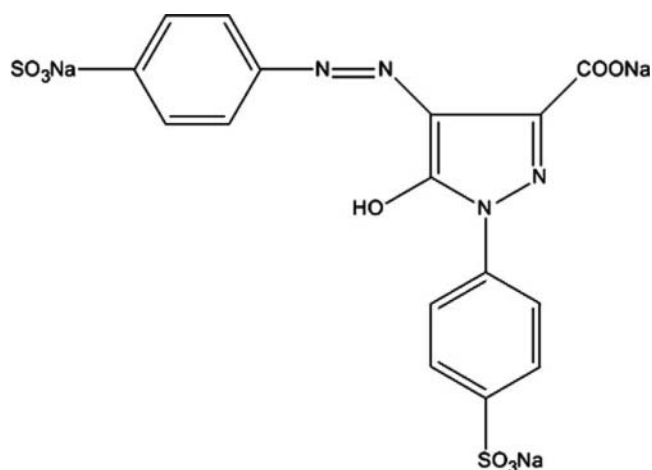


Figure 1. Chemical structure of tartrazine.

Figure 2. Besides the above-mentioned steps, Tartrazine can also be synthesized by condensing two moles of phenylhydrazine-*p*-sulfonic acid with one mole of dihydroxytartaric acid to form 3-carboxy-5-hydroxy-1-(4'-sulfonatophenyl)-4-(4'-sulfonatophenylazo)-*H*-pyrazol-3-carboxylate (Deshpande, 2002). Additionally, tartrazine can be converted to the similar aluminum lake by reacting aluminum oxide with the coloring matter under aqueous conditions. Undried aluminum oxide is prepared by reacting aluminum sulfate or aluminum chloride with sodium carbonate or sodium bicarbonate, or aqueous ammonia. Later, the product is filtered, washed with water, and dried (National Center for Biotechnology Information (NCBI) 2004).

Metabolism of tartrazine

The metabolisms of azo dyes in the liver and gastrointestinal tract are usually initiated by a reductive cleavage of the azo bonds by azo reductases that form aromatic amines (Demirkol and Ercal, 2012). These aromatic amines are oxidized to an *N*-hydroxy derivative by P450 enzymes (de Aragao Umbuzeiro et al., 2005). The carcinogenic activation mechanisms lead to the reduction and cleavage of the azo dyes, oxidation of azo dyes, and direct oxidation of the azo linkage to reactive electrophilic diazonium salt (Chen, 2006, Bardi and Marzona, 2010). Tartrazine is metabolized by gastrointestinal microflora to sulfanilic acid and aminopyrrolazalone. The compounds are cleaved to sulfanilic acid and α -amino- β -ketobutyric acid fragments by intermediary metabolism with the release of carbon dioxide (EFSA Panel on Food Additives and Nutrient Sources 2009). Excretion of 4-sulfophenylhydrazine metabolite labeled with sulfur-35 differs with the route of administration. After 48 hours of oral administration of this metabolite, 35% is excreted in urine and 49% in feces. When specified intraperitoneal, 90% and 5% of the metabolites is excreted in urine and feces, after 48 hours of administration. Through oral administration, 69% of the urinary radioactivity excreted is sulfanilic acid and 21% is 4-sulfophenylhydrazine. Through intraperitoneal administration, 9% of urinary radioactivity excreted is sulfanilic acid and 73% is 4-sulfophenylhydrazine. The outcome suggested the conversion of 4-sulfophenylhydrazine to sulfanilic acid occurs in the gut lumen.

Toxicology study of tartrazine

Numerous studies have shown the health effects of tartrazine, whose clinical significance is still unclear; however, the intolerance reaction toward tartrazine may happen. Adverse health effects of tartrazine are found as asthma, allergy, and hyperactivity in children. Stenius and Lemola (1976) have tested 140 asthmatics with acetylsalicylic acid (ASA) with tartrazine. They mentioned that one-quarter of the patients found a positive reaction to one of the two tested agents and that most of the patients are sensitive to ASA. Furthermore, a study conducted on 122 patients with a variety of allergic disorder found that most of the patients experienced general weakness, heatwaves, palpitations, blurred vision, rhinorrhoea, feeling of suffocation, pruritus, and urticarial when given 50 mg of tartrazine orally (Neuman et al., 1978). McCann et al. (2007) have reported that an increase in hyperactivity was observed in 3-year old and 8- to 9-year old children when given two mixtures of four

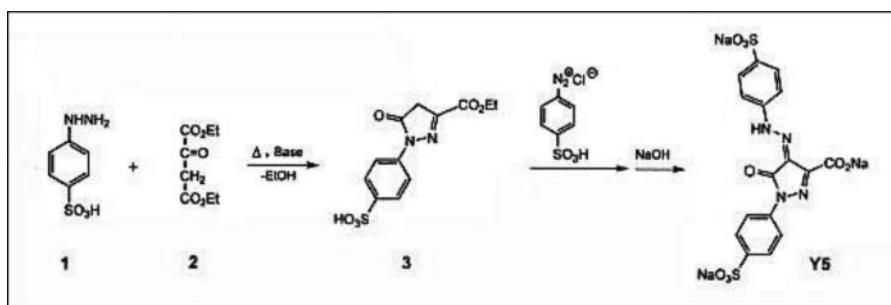


Figure 2. Condensation of (1) phenylhydrazine-p-sulfonic acid with (2) oxalacetic diethyl ester to form (Y5) tartrazine (Weisz et al., 2014).

synthetic colors with sodium benzoate. About 166 atopic women with medical history of asthma, rhinitis, urticaria, and with hypersensitivity to non-steroidal anti-inflammatory agents were tested to check the ability of tartrazine to cause hypersensitivity reactions. The results showed that tartrazine has the capability of provoking IgE and non-IgE dependent reactions in 6% of the volunteers from 99 volunteers who fulfilled all the requirements (Caldas et al., 2007).

A first case study was conducted by Trautlein and Mann (1978); they reported systemic anaphylaxis secondary to a standard enema preparation. After the patient was given an enema containing tartrazine and sunset yellow, the patient developed asthma, urticaria, and anaphylactic shock. The mechanisms are the immunoglobulin E mediated type hypersensitivity and the non-immunologic prostaglandin synthetase inhibition that are normally observed in allergic reactions to Tartrazine. Orchard and Varigos (1997) reported that 11 years old girl had a repeated fixed drug eruption to Tartrazine on the back of the left hand. The results of oral provocation test to two suspected foods, an artificially colored cheese crisp and tartrazine, have been found to be positive. Besides, Bhatia (2000) studied food allergy or intolerance to psychotropic drug. Tartrazine is often times inculpatated in causing the allergic reaction. The total patients were given tartrazine containing drug and were observed. The results showed that 83 out of 2210 patients have found that tartrazine-containing drug developed an allergy reaction and the symptoms lessen within 24 to 48 hours after stopping the drug. No patients found the allergic reaction toward non-Tartrazine-containing drug.

Maximum permitted level of tartrazine

According to European Parliament and Council Directive 94/36/EC, the maximum permitted level of tartrazine is in the range of 50–500 mg/kg in different food products. For non-alcoholic drinks, the maximum permitted level is 100 mg/L, whereas in alcoholic drinks, it is 200 mg/L. The maximum limit for soups and nutrition supplements is 50 mg/kg, for cheese, meat, and fish, is 100 mg/kg, for desert and flavored milk product, is 150 mg/kg, and for confectionary, is 300 mg/kg (EFSA Panel on Food Additives and Nutrient Sources 2009). According to CODEX Alimentarius (National Center for Biotechnology Information (NCBI) 2015), the maximum allowance limit for tartrazine is 50 mg/kg for soups. In Canada, the maximum permitted level in juice, marmalade, flavored milk, jelly, and sherbet is 300 mg/kg, single or combined with other synthetic food

coloring (Health Canada 2015). The maximum permitted used level for tartrazine has been set at 200 mg/kg of food in Thailand (Vachirapatama et al., 2008). However, there is no permitted limit for food colorant in Malaysia (Zailuddin et al., 2015).

Extraction method of tartrazine

An extraction method is needed prior to the detection step to remove all the impurities that may interrupt in the result. The selected method of extraction is important depending on the samples (Yamjala et al., 2015). Most common methods are liquid–liquid extraction (LLE), solid-phase extraction (SPE), membrane filtration, and cloud point extraction.

Liquid–liquid extraction (LLE)

LLE or solvent extraction is based on the separation of compounds according to their relative solubility in two different immiscible liquids such as water, ethanol, methanol, isopropyl alcohol, acetate, and ammonia (Yamjala et al., 2015). Tsai et al. (2015) have used acetonitrile for the extraction of synthetic dyes in chili powder and syrup-preserved fruits. The sample was extracted twice, centrifuged and filtered before injecting into the vial. Acetonitrile has been chosen because of its good extraction yield and less fat solubility, carbohydrate precipitation, and protein precipitation. Bento et al. (2015) have used methanol–ammonium hydroxide for extraction of the coloring in yogurt and milk drink. The methanol–ammonium hydroxide solution (8:2) is extracted synthetic food colorants but not natural colorants using the proposed method specify to synthetic color. Besides, Pávai et al. (2015) have used the green method of extraction where no hazardous chemical has used in sample preparation, only used deionized distilled water to dilute the sample.

Tang et al. (2014) have used LLE for extraction of synthetic food coloring in beverage, preserved fruit, candy, and gelatine. They only used water–ammonia aqueous solution as the solvent and the mixture of the sample, later the solvent has sonicated and diluted. Ma et al. (2015) have used methanol solvent for extraction of food additive in red wine. The mixture has degassed, centrifuged, acidified, and filtered prior to the detection steps.

Solid-phase extraction (SPE)

SPE is used sorbent such as C18, polyamide, gel permeation chromatography (GPC), and styrene-divinylbenzene polymer

and solvents to extract the azo dyes from food matrices. Selecting the proper solvents is essential for the synthetic colorants extraction which depending on the analytical structure (Yamjala et al., 2015). The SPE method is a simple and able to extract the colorants without contaminants. Before conducting SPE methods, the cartridges should be washed and preconditioning. Methanol and acetic acid are the common choice of conditioning (De Andrade et al., 2014, Bonan et al., 2013, Martin et al., 2016, Feng et al., 2011). Recently, Martin et al. (2016) adopted the SPE method to extract colorants from sugary and gummy confectionary, ice cream, and chocolate sweets. The SPE cartridges are preconditioned with acetic acid, and the colorants are eluted with ethanol–ammonia solution. Qi et al. (2015) have used *n*-hexane to eliminate fat from flour and meat foodstuffs. The methanol–ammonia–water solution is added to extract the samples. After that, the extracted samples are loaded into Strata-X-AW cartridges and eluted out with ethanol that contained ammonia–water.

SPE cartridges not only used for solid food matrices but can also be used for soft drinks. A study conducted by de Andrade et al. (2014) used Sep-Pack C18 cartridges to extract the colorants. The cartridges have precondition with isopropyl alcohol and acetic acid. The samples are flowed through the cartridge and the colorants are eluted with isopropyl alcohol. Yoshioka and Ichihashi (2007) have prepared the column by making slurry of polyamide and packed it into a column to extract 40 synthetic dyes in drinks and candies. The column has preconditioned with acetic acid and the colorants are eluted out with the ammonia–ethanol solution. Huang et al. (2002) have used a polyamide SPE column that has been preconditioned with methanol and water to extract colorants from milk samples. The sample has mixed with ethanol and acidified to pH 2. The samples are acidified as synthetic colorants adsorbed more strongly in the acidic condition (Yoshioka and Ichihashi, 2007). The mixture is centrifuged and flowed into the cartridges, and then eluted with 0.5% ammonia and methanol solution.

Khanavi et al. (2011) and Hajimahmoodi et al. (2013) have added polyamide sorbent into the treated sample to extract colorants from drink, syrup, candy, jelly gum, chocolate, colored rice, saffron, and gum. The mixtures are stirred vigorously and the sorbent is filtered out. The colorants are removed from the polyamide with alkaline-ammonia. For extraction of colorants from fish roe and caviar, Kirschbaum et al. (2006) have used polyamide with aqueous ammonia, sonicated, and centrifuged. The collected supernatant is defatted by three-fold treatment with *n*-hexane and acidified prior to addition of polyamide. Different from others, Gan et al. (2013) and Sorouraddin et al. (2011) have used white commercial wool yarn as the sorbent for coloring coated chocolate, commercial cakes, and soft drinks. The samples are diluted with distilled water, centrifuged, and mixed with acetic acid. The white wool yarn has been washed with detergent and water is added into the sample mixture and heated. After 1 hour, the colored yarn is taken out and washed with plenty of distilled water. The colorants are eluted out by putting the yarn in the ammonia solution and heated.

Soylak and Cihan (2013) have used multiwalled carbon nanotubes to separate tartrazine compound from the food

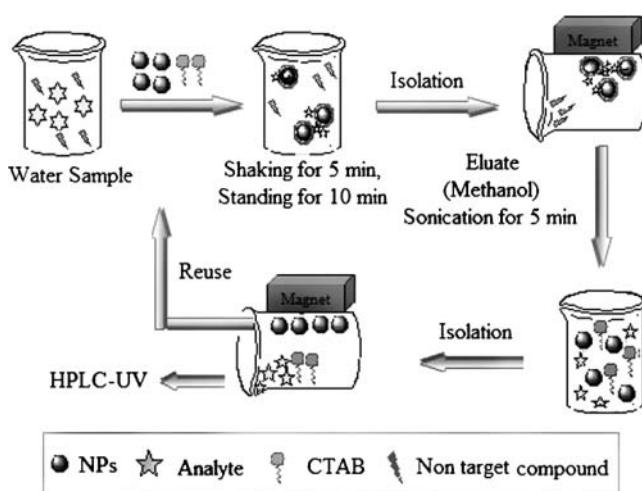


Figure 3. Schematic illustration of the extraction process by using mixed hemimicelle SPE based on CTAB-coated $\text{Fe}_3\text{O}_4/\text{SiO}_2$ NPs (Tavakoli et al., 2013).

matrices. The dye is extracted from tap water, powdered beverages, and in drug samples. The colorant is eluted with dimethylsulfoxide before analysis step. In addition, Wu et al. (2015) have used magnetic solid-phase extraction (MSPE) method for soft drinks, cocktails, solid beverages, ice cream, sugar-based and gelatine-based confection. The magnetic SPE method used Fe_3O_4 -poly (ionic liquid) core shells microspheres as sorbent. However, Tavakoli et al. (2013) have used the diverse hemimicelle solid-phase extraction (MHSPE) method, based on cetyltrimethylammonium bromide-coated (CTAB) $\text{Fe}_3\text{O}_4/\text{SiO}_2$ nanoparticle to extract synthetic colorants. For saffron rice, saffron dessert, honey cream, fruit juice-coated ice cream, and saffron ice cream, the samples are homogenized, diluted with water, and the pH adjusted to 9 with alkaline ammonia. Then, the samples are centrifuged and kept for the preconcentration process based on the MHSPE method. For liquid samples are filtered and used for extraction. NPs solution, CTAB solution, and alkaline food samples are added into a vial in sequence to preconcentrate the analytes. The extraction procedures of SPE based on CTAB-coated $\text{Fe}_3\text{O}_4/\text{SiO}_2$ NPs are shown in Figure 3.

Membrane filtration

A membrane filtration used a thin layer of semi-permeable substance to separate components of the samples when an external force has applied across the membrane with water as a diluent (Yamjala et al., 2015). Vidotti et al. (2006) have used the membrane filtration method to extract colorants from juice and gelatine. The samples are dissolved in water by heating, cooled, diluted to 50 mL with water, and filtered through a $0.45 \mu\text{m}$ membrane filter. Prado et al. (2006) have used filter membrane extraction to extract the colorants from alcoholic beverages. The samples are homogenized and degassed by mechanical agitation, and finally filtered with cellulose ester. Miniotti et al. (2007) have placed the samples in ultrasonic bath before the filtering the sample through a folded paper filter and the filtered sample brought up to 50 mL. The diluted sample then filtered through $0.45 \mu\text{m}$ disposable syringe filter before analysis. Serdar and Knežević (2009) have filtered the degassed and

diluted soft drink with a folded paper filter before the sample being filtered again with a 0.45 μm membrane filter.

Cloud point extraction

Cloud point extraction (CPE) is a green alternative technique for the LLE method, analyte preconcentration, and sample clean-up. Cloud point extraction commonly followed by the principles of “green chemistry” thus required small quantities of mildly toxic surfactants compared with toxic organic solvents. Besides, surfactants are not particularly volatile and non-flammable. The CPE method observed the clouding behavior when a solution containing a polyoxyethylene-type nonionic surfactant is heated and stirred before being allowed to settle. The liquid has separated into aqueous and surfactant-rich phases due to dehydrating the surfactant during the settling process (Nascimento et al., 2015; Sato et al., 2013). The volume of surfactant-rich phase is lesser; therefore, a high enrichment factor can be achieved. This improved the sensitivity of the analysis without further sample clean-up or evaporation steps (Xing and Chen, 2013).

Other extraction method

The promising purposes of the green chemistry have caused a key focus of the research efforts on the development of miniaturized, efficient, and environmentally moderate sample extraction procedures. Therefore, solid-phase micro-extraction (SPME) (Lord et al., 2007; Ouyang and Pawliszyn, 2008) and liquid-phase micro-extraction (LPME) (Jeannot and Cantwell, 1996; He and Lee, 1997) have been introduced as green chemistry techniques. Although SPME is a solvent-free extraction method, the SPME fiber used in this extraction method is high-priced, fragile, and short lifetime (Prosen and Zupancič-Kralj, 1999). The LPME method divided into two broad categories which are membrane-protected solvent and exposed solvent (Kokosa, 2013). Single-drop micro-extraction (SDME) and dispersive liquid-liquid micro-extraction (DLLME) are also green techniques used in the exposed solvent mode to extract food colorants (Rajabi et al., 2014; Rajabi et al., 2015). Matrix solid-phase dispersion (MSPD) is a good extraction method for the extraction of complex solid, semi-solid or viscous samples. It performed extraction and clean-up at the same time, which can even reduce the amount of solvent. Recently, MSPD has extracted synthetic colorants from meat and condiments (Llompart et al., 2013; Celeiro et al., 2014; Li et al., 2014). An aqueous two-phase system based on ionic liquid microextraction (IL-ATPS) is extracted five synthetic dyes including Tartrazine from soft drink, instant powdered drink, sugar-based and gelatine-based confectionary. The extraction agent used is 1-alkyl-3-methylimidazolium bromide and salt (Sha et al., 2015).

Antakli et al. (2015) have extracted the colorant from soft drinks by the ultrasound-assisted extraction (UAE) method. The soft drink sample is degassed using ultrasonication and the aliquot has taken for analysis. Shen et al. (2014) have extracted four artificial food colorants including tartrazine and three carotenoids by the UAE method. The sample is mixed with methanol and an ultrasonic probe is immersed in the mixture

to undergo ultrasonic-assisted extraction. Centrifugation has performed to separate the supernatant and the methanol extraction step has repeated at least three times.

Analytical method

Numerous analytical methods have been developed for identification and quantification of synthetic food colorant such as HPLC, TLC, spectrophotometry, CE and electrochemical techniques. The summary of analytical detection methods is available in Table 1.

Chromatography

High-performance liquid chromatography (HPLC)

HPLC has involved the separation of the sample compounds according to the interaction between the molecules with the packing particle of the column. HPLC generally coupled with UV-Vis, PDA, MS, and DAD detector and used different types of mobile phase according to the nature of the samples.

Bento et al. (2015) have determined the presence of permitted azo dyes and non-azo dyes in yogurt and milk drink using HPLC coupled with PAD. In tartrazine, a limit of detection (LOD) and limit of quantification (LOQ) were found of 1.22 $\mu\text{g/L}$ and 3.71 $\mu\text{g/L}$, with the recovery rate of 98.1–106.6%. Tavakoli et al. (2013) have determined tartrazine by HPLC coupled with a UV-Vis detector. The samples were extracted using the mixed hemimicelle SPE method before being analyzed. The LOD and LOQ for tartrazine were found of 2.50 $\mu\text{g/L}$ and 8.33 $\mu\text{g/L}$, respectively. Bonan et al. (2013) have quantified the 17 colorant in beverages using HPLC-DAD and manage to recover 87.6% of tartrazine from the spiked sample. Feng et al. (2011) have reported that HPLC paired with electrospray ionization-tandem mass spectrometry (ESI-MS/MS) has improved in sensitivity and enhanced in the accuracy as compared to the traditional method. They screened among 40 food dyes in soft drinks and the LOD for tartrazine was found to be 0.5 mg/L and recovery percentage of 97.7%. Culzoni et al. (2009) have used HPLC coupled with the DAD detector and second-order algorithms for the analysis of three synthetic dyes in non-alcoholic beverages and the recovery values ranging between 97% and 105%. Besides, Vachirapatama et al. (2008) have analyzed seven synthetic dyes using the monolithic C18 column by HPLC. The LOD and LOQ for tartrazine were 1.92 $\mu\text{g/L}$ and 6.41 $\mu\text{g/L}$, respectively. García-Falcón and Simal-Gándara (2005) have used reverse-phase HPLC with UV-Vis detector to determine five synthetic food colors in soft drinks. The LOD and LOQ for tartrazine were 0.3 mg/L and 1.0 mg/L, respectively.

In an effort of reducing the use of hazardous chemical, the green chromatography method has developed. The C18 column with phosphate buffer and Triton X-100 (0.25%, v/v) aqueous solution have used as the mobile phase instead of organic solvent (Vidotti et al., 2006). In the presence of Triton X-100, the C18 column becomes more polar making the separation of the colorants possible. The LOD for tartrazine was found 0.125 mg/L (Vidotti et al., 2006). Vidotti et al. (2006) and Khanavi et al. (2011) have used the C8 column as the stationary phase with phosphate buffer and Triton X-100 as the

Table 1. Summary of analytical methods for identification of tartrazine in food products.

ANALYTICAL METHODS						
FOOD MATRIX	EXTRACTION	INSTRUMENT	DETECTION			
			MOBILE PHASE	LOD	RECOVERY	REFERENCES
Sugar and gummy confectionary, ice cream, and chocolate sweets	The prepared sample was loaded into SPE cartridge and eluted out with the ethanol-ammonia solution.	Liquid chromatography electrospray ionization tandem mass spectrometry	A: 40 mM ammonium acetate with 2.5% acetonitrile (pH 7.8) B: Acetonitrile	5 µg/kg	100.1–119.7%	(Martin et al., 2016)
Yogurt and milk drink	Methanol-ammonium hydroxide was added to the sample to extract the colorants.	HPLC-PAD	Mobile phase A: Ammonium acetate 1% Mobile phase B: Methanol:acetate (80:20)	1.22 µg/L	98.1–106.6%	(Bento et al., 2015)
Chilli powder and raisin	Acetonitrile was used as solvent. The sample was extracted twice, centrifuged and filtered before injected into the vial	LC-MS	A: Acetonitrile B: 20 mM ammonia acetate buffer with 1% acetic acid	N/A	91.2–96.1%	(Tsai et al., 2015)
Beverage, instant juice powder, and sugar-coated tablets	Carbonated beverage sample was degassed with slight boiling and diluted to 50 mL with water. Instant juice powder was dissolved and diluted to 50 mL with water. Sugar coated tablet sample was grinded and dissolved in water. The sample was then filtered and the step was repeated for 3 times. The supernatants were collected and diluted with water to 50 mL.	glassy carbon electrode modified with poly(L-phenylalanine) coupled with differential pulse voltammetry (DPV)	Phosphate-citrate buffer	10.7 µg/L	95.0–101.4% for beverage, 95.0–99.0% for instant juice powder, and 96.0–100.7% for sugar-coated tablets	(Chao and Ma, 2015)
Soft drinks, candy, juice powder and chewing gum	The candy sample was dissolved in distilled water and transferred to volumetric flask. Soft drink sample was degassed and the aliquot was taken for analysis. For chewing gum, the sample was dissolved in distilled water and centrifuged.	Spectrophotometric	—	0.12 g/mL	101.75–102.75%	(Antakli et al., 2015)
Soup powder, yogurt, sweet cream cheese, jams, sparkling tablet, and beverages	Deionized distilled water was used to dilute the sample and no hazardous chemical was used.	Cellophane test strip	—	N/A	N/A	(Pávai et al., 2015)
Soft drink, instant powdered drink, sugar-based and gelatine-based confectionary	Aqueous two-phase system based on ionic liquid microextraction (IL-ATPS) was used as extraction method. The extraction agent used was 1-alkyl-3-methylimidazolium bromide and salt.	HPLC-UV-Vis	A: 0.02 mol/L ammonium acetate aqueous solution (pH 4.5) B: Methanol	0.053 ng/mL	94.1–98.9%	(Sha et al., 2015)
Soft drinks, cocktails, solid beverages, ice cream, sugar-based and gelatine-based confection	Magnetic SPE method that used Fe ₃ O ₄ -poly (ionic liquid) core shells microspheres as sorbent was used for extraction steps. The diluted and filtered sample was flowed into MSPE system along with the sorbent to be extract.	HPLC	A: 0.02 mol/L ammonium acetate aqueous solution (pH 7.5) B: Methanol-acetonitrile (30:70, v/v)	5.6 ng/mL	N/A	(Wu et al., 2015)
Flour and meat foodstuffs	Methanol-ammonia-water solution was added to extract the sample and the sample extracts were loaded into Strata-X-AW cartridges and eluted out with ethanol that contained ammonia-water.	HPLC coupled with DAD and MS/MS	HPLC-DAD A: 20 mM ammonium acetate buffer B: Methanol HPLC-MS/MS A: Methanol	N/A	N/A	(Qi et al., 2015)

Saffron rice, saffron dessert, honey cream, fruit juice-coated ice cream, and saffron ice cream, beverages	Mixed hemimicelle solid-phase extraction (MHSP) method, based on cetyltrimethylammonium bromide-coated (CTAB) $\text{Fe}_3\text{O}_4/\text{SiO}_2$ nanoparticle was used for extraction of the colorants. NPs solution, CTAB solution and alkaline food sample was added into a vial in sequence to preconcentrate the analytes.	HPCL-UV-Vis	B: 5 mM ammonium acetate solution Mobile phase A: 0.1 M ammonium acetate solution (pH 6.7) Mobile phase B: Methanol-acetonitrile (50:50, v/v)	2.50 $\mu\text{g/L}$	N/A	(Tavakoli et al., 2013)
Beverages, gelatines, solid samples	Water-ammonia aqueous solution was used as the solvent and the mixture of the sample and solvent was sonicated and diluted.	Polyamide TLC method coupled with on-plate SPE and backlight-assisted detection	Ethanol (95%)-water-acetic acid solution (50:50:1)	4.19 ng	N/A	(Tang et al., 2014)
Soft drinks	Sep-Pack C18 cartridges was used to extract the colorants. The cartridges was precondition with isopropyl alcohol and acetic acid. The sample was flowed through the cartridge and the colorants were eluted with isopropyl alcohol.	Silica gel chromatography plate and ion-pair HPLC	Isopropyl alcohol and ammonium hydroxide	N/A	N/A	(De Andrade et al., 2014)
Red wine	Methanol was added as solvent for the extraction. The mixture was degassed, centrifuged, acidified and filtered prior to the detection steps	Ultra performance liquid chromatography coupled with electrospray ionization-tandem mass spectrometry (UPLC-ESI-MS/MS)	A: 10 mM acetate buffer solution B: Methanol/acetonitrile (9:1, v/v)	0.3 $\mu\text{g/L}$	92.1–97.7%	(Ma et al., 2015)
Soft drink	The sample was mixed with methanol and an ultrasonic probe was immersed in the mixture to undergo ultrasonic-assisted extraction. Centrifugation was done to separate the supernatant and the methanol extraction step was repeated three times.	HPCL-PAD	A: Ammonium acetate B: Methanol	10 ng/mL	N/A	(Shen et al., 2014)
Soft drink	The sample was transferred into 50-mL flask and adjusted for the volume with doubly distilled water.	1-allyl-3-methyl imidazolium tetrafluoroborate ionic liquid modified carbon-ceramic electrode	—	0.043 mg/L	94.2–101.3%	(Majidi et al., 2014)
Soft drink	No extraction procedure performed	Glassy carbon rotating disk electrode modified with β -cyclodextrin-coated poly (diallyl methyl ammonium chloride)-functionalized graphene (β -CD-PDDA-Gr) composite film	—	1.43×10^{-8} mol/L	95.11–103.60%	(Ye et al., 2013)
Candy, royal jellies, ice cream, solid custard jelly powder, juice powder, soft drink, coloring coated chocolate	Candy, royal jellies, ice cream, solid custard jelly powder and juice powder sample was dissolved and diluted to 100 mL and filtered with filter membrane. Soft drink sample was used directly. Coloring coated chocolate sample was added into water to dissolve the colored shell. The remaining solution was separated, centrifuged and diluted with CH_3COOH solution. White commercial wool yarn was added and	Graphene and mesoporous TiO_2 modified carbon paste electrode	—	8.0 nM	97.41–102.10%	(Gan et al., 2013)

(Continued on next page)

Table 1. (Continued).

ANALYTICAL METHODS						
FOOD MATRIX	EXTRACTION	DETECTION			RECOVERY	REFERENCES
		INSTRUMENT	MOBILE PHASE	LOD		
Beverages, solid food matrices	heated. The colorants was eluted out by heating the wool with NH ₃ . The samples were mixed with ethanol-water solution and centrifuged before being flowed into SPE cartridges. The cartridges were preconditioned with methanol and the colorants were eluted with methanol-ammonia solution.	HPLC-DAD with C8 column	Mobile phase A: Acetonitrile Mobile phase B: 100 mM sodium acetate buffer (pH 7)	N/A	87.6%	(Bonan et al., 2013)
Cookies, colored rice, saffron and fruit juice	Polyamide sorbent was added into the treated sample to extract the colorants. The colorants were removed from the polyamide with alkaline-ammonia.	HPLC-UV-Vis with C8 column	phosphate buffer and Triton X-100	0.04 mg/kg	98.1%	(Hajimahmoodi et al., 2013)
Tap water, powdered beverages and in drug samples	Multiwalled carbon nanotubes was used to separate Tartrazine compound from the food matrices. The colorant was eluted with dimethylsulfoxide before analysis step.	Spectrophotometry	Acetate buffer solution	3.4 μg/L	95%	(Soylak and Cihan, 2013)
Powdered gelatine, fruit syrup, candy	The gelatine sample was prepared in water and the aliquots was used. Fruit syrup and candy sample was mixed with double distilled water and filtered. The filtered sample was diluted and analyzed.	Silver nanoparticle (AgNPs) and spectrophotometry	Acetate-acetic acid buffer (pH 6)	0.3 ng/mL	97.0–100.3%	(Sahraei et al., 2013)
Beverage, powdered beverage	Sample was dissolved in deionized distilled water and degassed. The sample was filtered prior to analysis step	Ce(IV)-oxidative spectrophotometry	CERAC reagent	N/A	N/A	(Olgun et al., 2012)
Drink, syrup, candy, jelly, chocolate and gum	Polyamide sorbent was added into the treated sample to extract the colorants. The colorants were removed from the polyamide with alkaline-ammonia.	HPLC with C8 column	phosphate buffer and Triton X-100	0.05 μg/mL	N/A	(Khanavi et al., 2011)
Soft drink	The HLB cartridges were precondition with methanol and acidified water and the colorants were eluted out with methanol-ammonia solution.	LC-ESI-MS/MS	Mobile phase A: 20 mM Ammonium formate buffer containing 0.1% formic acid (v/v) Mobile phase B: Methanol-acetonitrile (7/3)	0.5 mg/L	97.7%	(Feng et al., 2011)
Coated chocolate, commercial cakes and soft drinks	White commercial wool yarn was used as the sorbent. The white wool yarn that had been washed with detergent and water was added into the sample mixture and heated. The colorants were eluted out by putting the yarn in ammonia solution and heated.	Multi-color light emitting diode based photocolormeter	A: NaH ₂ PO ₄ /Na ₂ HPO ₄ buffer (pH 6) B: Acetonitrile (35%)	N/A	N/A	(Sorouraddin et al., 2011)
Non-alcoholic beverages	No extraction procedure performed	HPLC-DAD and second order algorithms	N/A	N/A	97–105%	(Culzoni et al., 2009)
Stock solution	No extraction procedure performed	Electroosmotic thin layer chromatography and capillary zone electrophoresis	Methanol-2-propanol-ethyl acetate-water	100 μg/L	N/A	(Kartsova et al., 2009)
Soft drink		LC-DAD		N/A	96.9–97.1%	

Foodstuffs, soft drinks	The degassed and diluted soft drink was filtered with folded paper filter before the sample being filtered again with 0.45 μm membrane filter.	HPLC with monolithic C18 column	Tetrabutylammonium hydrogen sulfate, methanol and deionized water	(Serdar and Knezević, 2009)
Foodstuffs, soft drinks	Foodstuff sample was diluted to 25 mL in volumetric flask and soft drink was degassed and diluted. The sample was filtered before injected into HPLC.	HPLC with monolithic C18 column	Mobile phase A: Methanol-water (12%, v/v) Mobile phase B: 10 mM acetic acid	(Vachirapattana et al., 2008)
Stock solution Drinks and candies	No extraction procedure performed The column was prepared by making slurry of polyamide and packed it into a column. The column was preconditioned with acetic acid and the colorants were eluted out with ammonia-ethanol solution.	Capillary zone electrophoresis RP-HPLC-PAD	N/A	(Česla et al., 2007)
Fish roe and caviar	The sample was mixed with aqueous ammonia, sonicated and centrifuged. The collected supernatant was defatted and acidified prior to addition of polyamide. The sample was placed in an ultrasonic bath before the filtering the sample through a folded paper filter and the filtered sample was brought up to 50 mL. The diluted sample was then filtered through 0.45 μm disposable syringe filter before analysis.	RP-HPLC-DAD	A: 0.1 mol/L ammonium acetate aqueous solution (pH 6.70) B: Methanol-acetonitrile (50:50, v/v)	(Yoshioka and Ichihashi, 2007)
Beverage, alcoholic beverage, jam, sweets, sugar confectionary	The sample was homogenized and degassed by mechanical agitation. The sample was then filtered with cellulose ester.	RP-HPLC-DAD	A: 100 mM sodium acetate buffer (pH 7) B: Acetonitrile	(Kirschbaum et al., 2006)
Alcoholic beverages	The sample was transferred into amber glass bottles, sealed and stored under refrigerated condition at temperature below 4°C. Then, the sample was homogenized and degassed in an ultrasonic bath. The sample aliquots were injected directly into HPLC.	RP-HPLC-DAD	A: Ammonium acetate solution (pH 7) B: Methanol-acetonitrile (80:20, v/v)	(Miniotti et al., 2007)
Beverages	The sample was dissolved in water by heating, cooled, diluted to 50 mL with water and filtered through a 0.45 μm membrane filter.	Capillary electrophoresis HPLC-UV-Vis	Phosphate buffer with SDS	(Prado et al., 2006)
Juice and gelatine	No extraction procedure performed	HPLC with C18 column	Mobile phase A: Methanol Mobile phase B: 40 mM ammonium acetate aqueous solution (pH 5)	(García-Falcón and Simal-Gándara, 2005)
Stock solution	Polyamide column used was preconditioned with methanol and water. The sample was washed with deionized water and methanol and the colorant was eluted using ammonia solution-methanol.	Conversion of TLC plate into Windows 95/9 and the processing of the image received with Adobe Photoshop 5.0 program package CE separation method with large-volume sample stacking (LVSS)	phosphate and Triton X-100 aqueous solution	(Vidotti et al., 2006)
Soft drinks, jellies, and milk beverages	Polyamide SPE column that had been preconditioned with methanol and water was used. The sample mixture was then centrifuged and the supernatant was taken and flowed into the cartridges. The colorant was eluted with 0.5% ammonia and methanol solution.	Running buffer of sodium hydroxide and disodium tetraborate	N/A	(Huang et al., 2003)
Milk beverages		Capillary electrophoresis with borax-sodium hydronium buffer mixed with β -cyclodextrin	N/A	(Huang et al., 2002)

mobile phase in determination of eight synthetic dyes in drink, syrup, candy, jelly, chocolate, and gum. The used of column C8 is a better separation of the colorants. The LOD of tartrazine was found 0.05 $\mu\text{g/mL}$. Hajimahmoodi et al. (2013) used the green chromatography method with C8 column and the UV-Vis detector to analyze eight synthetic food colorants present in cookies, color rice, saffron, and fruit juice. The LOD, LOQ, and the recovery value for tartrazine were 0.04 mg/kg, 0.06 mg/kg and 98.1%, respectively.

Thin-layer chromatography (TLC)

TLC is the method to compare the ratio of fronts (R_f) values and the extracted colors in relation to the standard for identifying the food colorant present in samples (De Andrade et al., 2014). Tang et al. (2014) have developed a polyamide TLC method coupled with on-plate SPE and backlight-assisted detection to determine five synthetic colorants including tartrazine. The LOD of tartrazine was found 4.19 ng. Besides, de Andrade et al. (2014) have used silica gel chromatography with a capillary to identify Sunset Yellow, Tartrazine, Amaranth, and Brilliant Blue from the samples. Kartsova et al. (2009) have used electromigration methods by using electroosmotic TLC (EOTLC) to detect synthetic food dyes due to its ionogenic compounds. The schematic diagram of the facility for EOTLC is shown in Figure 4. Methanol-2-propanol-ethyl acetate-water is used as the mobile phase and the detection limit was found 100 $\mu\text{g/L}$. Gerasimov (2003) have developed a procedure which consist of conversion of TLC plate into Windows 95/98 and the processing of the image received with Adobe Photoshop 5.0 program package for the analysis of tartrazine. The average concentration of tartrazine was found 99.31%.

Liquid chromatography-tandem mass spectrometry (LC-MS)

Liquid chromatography coupled with mass spectrometers has high sensitivity, structural information on the basis of the molecular mass, and fragmentation pattern (Yamjala et al., 2015). In the recent year, Martin et al. (2016) have analyzed 18 water-soluble artificial dyes including tartrazine in sugar and gummy confectionary, ice cream, and chocolate sweets with LC-MS electrospray ionization. The LOD and LOQ for tartrazine were found 5 $\mu\text{g/kg}$ and 10 $\mu\text{g/kg}$, respectively.

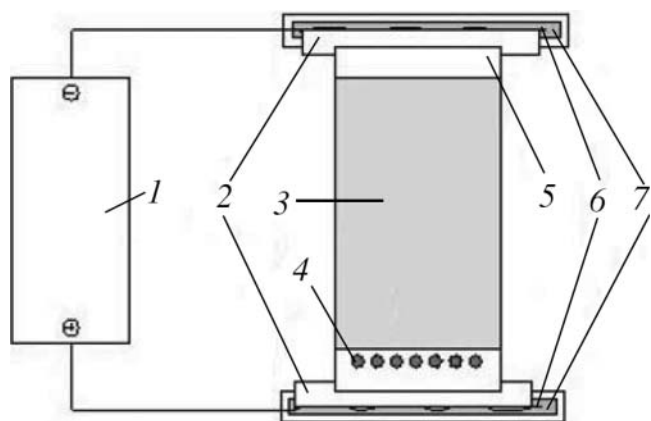


Figure 4. The schematic diagram of EOTLC: (1) voltage supply source, (2) filter paper, (3) polymer film, (4) analytes, (5) TLC plate, (6) electrodes, (7) the mobile phase reservoir (Kartsova et al., 2009).

The recovery values range found between 100.1% and 119.7% when spiked with 10 $\mu\text{g/kg}$ of standard tartrazine. A study done by Tsai et al. (2015) in detecting 20 food dyes including tartrazine in chilli powder and raisin are detected by using LC-MS. The recovery percentages for chili powder and raisin were 91.2–92.5% and 94.9–96.1%, respectively. Ma et al. (2015) have addressed ultra-performance liquid chromatography coupled with electrospray ionization-tandem mass spectrometry (UPLC-ESI-MS/MS) for quantification of synthetic colorants in red wine. A mixture of acetonitrile/methanol-ammonium acetate buffer solution is used as the mobile phase because it has better resolution compared to methanol. The detection limit was found at 0.3 $\mu\text{g/L}$ and the recovery of 92.1–97.7%.

Capillary electrophoresis (CE)

CE is an electrophoretic method where the analysis and the separation of the molecules are concluded in a capillary tube (Yamjala et al., 2015). Česla et al. (2007) have studied on the effect of pH and β -cyclodextrin additive to the background electrolyte on the separation of sulfonated azo dyes by using capillary zone electrophoresis (CZE). It has found that the effect of the working conditions is significantly different between non-coated fused silica capillaries and capillaries coated with polyacrylamide. Phosphate and borate background electrolyte have improved the separation efficiency and separation time of the sulfonated azo dyes in capillaries coated with polyacrylamide compared to non-coated fused silica capillaries and the addition of β -cyclodextrin to phosphate background electrolyte improved the separation selectivity.

Prado et al. (2006) have used CE for determination of synthetic dyes in alcoholic beverages. The LOD and LOQ for tartrazine were 2.0 $\mu\text{g/mL}$ and 6.6 $\mu\text{g/mL}$, respectively. Additionally, Huang et al. (2003) have developed a technique combining an on-capillary concentration method known as large-volume sample stacking and high-efficiency CE separation to analyze and detect colorants in soft drinks, jellies, and milk beverages. The method is successfully determining concentration of food colorants as low as 0.1–0.5 $\mu\text{g/mL}$. The method found lower detection limits when compared with the conventional CE method available. Huang et al. (2002) have established an analytical method based on CE for the detection of common colorants in milk beverages. High-efficiency CE separation methods for eight colors are separated in the presence of running buffer containing 7.0 mM β -cyclodextrin within 9 min. Polyamide column SPE used in order to reduce the matrix effect from the milk sample. Simple SPE pretreatment and fast separation method of CE is successfully able to determine food colorants without matrix interference in commercial milk beverages. The detection limits and recovery values were of 0.5 $\mu\text{g/mL}$ and 85%, respectively.

Spectrophotometry

Spectrophotometric techniques are simple, high sensitive, selectivity, and less interference from the colored present in the foodstuffs. The spectrophotometry method is one of the qualitative analytical methods used in determining azo dyes as these dyes have highlighted absorbing species in the visible region

(Yamjala et al., 2015). Antakli et al. (2015) have successfully determined of tartrazine and Brilliant Blue in foodstuffs using spectrophotometric. The LOD and LOQ were of 0.12 g/mL and 0.35 g/mL, respectively. Besides, Sahraei et al. (2013) have developed a simple kinetic spectrophotometric method based on silver nanoparticle (AgNPs) for determination of tartrazine in lemon, papaya-flavored gelatin, candy, and fruit syrup with satisfactory results. The detection limit was found of 0.3 ng/mL with relative standard deviation (RSD) of 0.98% ($n = 10$). Olgun et al. (2012) have used Ce(IV)-oxidative spectrophotometry. The colorants are determined by constructing their calibration curves as Ce(IV) absorbance at 320 nm versus colorant concentrations and calculating their indirect absorptivity with respect to their Ce(IV) reducing power from the slopes of the lines. The total content of tartrazine by this method was found at $20.12 \pm 0.55 \times 10^{-5}$ mol/L. Compounds that are not food colorant such as simple sugars and citric acid did not oxidized in this method thus eliminating interference.

Altınöz and Toptan (2002) have simultaneously detection of tartrazine and Ponceau 4R in the wavelength range between 300 and 700 nm using spectrophotometric techniques. The linearity range was found 1.00–60.00 $\mu\text{g/mL}$ for tartrazine and 1.00–52.00 $\mu\text{g/mL}$ for Ponceau 4R, respectively. Dinç et al. (2002) have established double divisor-ratio spectra derivative, classical least squares, and principal component regression for the spectrophotometric multicomponent determination of soft drink powders and synthetic mixtures. The graphical method showed the linear determination ranges for tartrazine was 4–18 $\mu\text{g/mL}$. The procedures were proven by using synthetic ternary mixtures and successfully applied for simultaneous detection of synthetic colorants in soft drink products.

Electrochemical sensor

Electrochemical sensing has a strong promise tool due to rapid, high sensitive and selective, miniaturized, and relatively low cost detection platforms. Synthetic color-based electrochemical sensing relies upon the oxidation–reduction of electro-analyte as monitored by linear sweep voltammetry (LSV), square wave voltammetry (SWV), differential pulse voltammetry (DPV), or conductivity. Electrochemical method-based determination of tartrazine is developed with the different chemically modified electrodes (Ye et al., 2013). During the electrochemical process, tartrazine takes place in a one-electron and one-proton which is an irreversible reaction on the oxidation process. The mechanism of electrochemical oxidation of tartrazine is shown in Figure 5. The working electrode reaction is based on the Nernst equation by the mechanism involving an equal number of electrons and protons. Gan et al. (2012) have electrochemically

determined tartrazine based on the direct oxidation of phenolic hydroxyl group. The result showed oxidation of phenolic hydroxyl group transfers one electron and one proton during the electrochemical process.

Mercury-free electrodes have established for determination of Tartrazine synthetic azo dyes in foodstuff (Gan et al., 2013, Ye et al., 2013, Gan et al., 2012, Ghoreishi et al., 2012, Zhang et al., 2009). Previously, Gómez et al. (2012) and López-de-Alba et al. (2001) have introduced adsorptive stripping voltammetry by using hanging mercury drop electrode to determine the concentration of tartrazine. The azo group ($-\text{N}=\text{N}-$) contained in tartrazine is electrochemical active that is able to reduce on the electrode surface when predominately using the mercury electrode. However, limitation of mercury electrode in the electrochemical cell is containing high toxicity that may cause to environmental pollution and adverse health effects.

Recently, Rovina et al. (2016) have developed a simple and rapid electrochemical sensor based on CHIT/CaONPs/MWCNTs with a modified gold electrode for determination of tartrazine in candy, jelly, and soft drink. Under the optimal conditions, the DPV was detected with different concentrations of tartrazine in the range of 0.1 to 10 ppm. The detection limit calculated was 0.9 ppm, which was lower than traditional methods. A linear coefficient found of 0.99354 and the recovery rate of 93.2 – 96.6%. Qiu et al. (2016) have simultaneously detected tartrazine and sunset yellow based on the modified glassy carbon electrode (GCE) with graphene oxide (GO) and multi-walled carbon nanotubes (MWCNT). The MWCNT/GO/GCE showed a strong enhancement effect, and greatly increased the oxidation current signal of food colorants compared with bare GCE. The combination of nanomaterials exhibited good signal amplification, excellent electronic, and antifouling. Under optimal condition, LOD was found of 0.01 μM for tartrazine and 0.025 μM for sunset yellow using the modified electrode. Finally, the proposed method is successfully applied in orange juice with satisfactory results. In another effort, sensitive, rapid and simple electrochemical based on alumina microfibers has developed for simultaneously detection of tartrazine and Ponceau 4R (Zhang et al., 2015). Alumina microfibers showed high accumulation efficiency to food colorants and increase of oxidation signals due to highly porous and large surface area. The LOD were found 0.8 and 2.0 nM for Ponceau 4R and tartrazine, respectively. Wang and Zhao (2015) used ionic liquid of 1-allyl-3-methylimidazolium chloride (AMIM-Cl) to functionalize graphene oxide (GO) and mixed to gold nanoparticle (ILRGO-Au). The ILRGO-Au composites exhibited an improved conductivity, increase the specific surface area, as well as accelerate electron transfer for simultaneous determination of tartrazine and sunset yellow in beverages. The LOD was found of $8.3 \times$

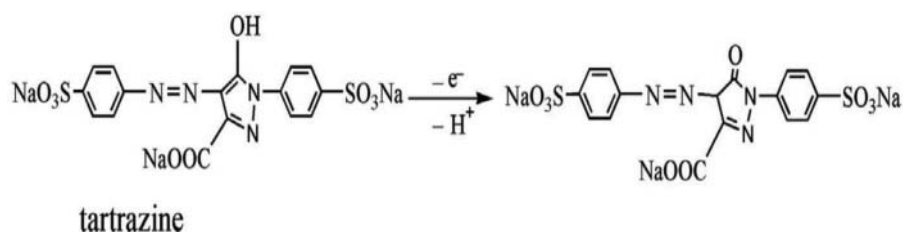


Figure 5. The mechanism for the electrochemical processes of Tartrazine (Qiu et al., 2016).

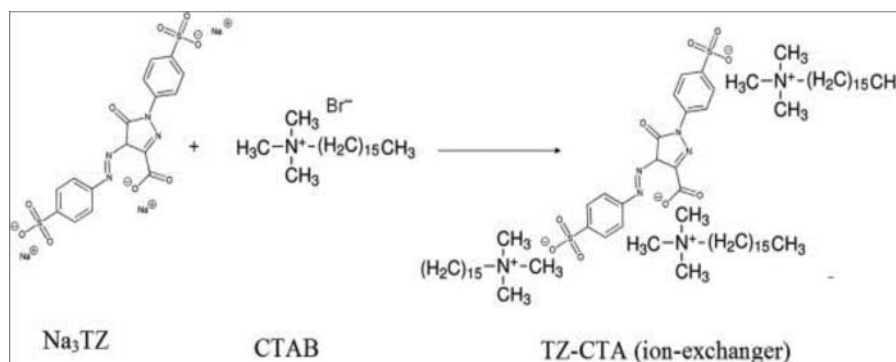


Figure 6. The mechanism reaction between Na_3TZ and CTAB (Shawish et al., 2013).

10^{-10} M and 5.2×10^{-10} for tartrazine and sunset yellow, respectively.

Chao and Ma (2015) have used a glassy carbon electrode modified with poly(L-phenylalanine) (PLPA/GCE) coupled with differential pulse voltammetry (DPV) for tartrazine determination in various food samples such as beverage, instant juice powder, and sugar-coated tablets. The LOD for tartrazine was found $10.7 \mu\text{g/L}$ with recovery ranging between 95.0% and 101.4% for beverage, 95.0–99.0% for instant juice powder, and 96.0–100.7% for sugar-coated tablets. They never found any significant different in the recovery result in comparison with HPLC. Majidi et al. (2014) have used 1-allyl-3-methyl imidazolium tetrafluoroborate ionic liquid modified carbon-ceramic electrode for simultaneous detection of tartrazine. The deposition of ionic liquid on the surface of the carbon-ceramic electrode is resulting in the development of graphene nanoplatelet-like structure on its surface. The modified electrode showed electrocatalytic behavior toward the oxidation of tartrazine and sunset yellow. The electrode succeeded in simultaneous determination of tartrazine and sunset yellow in different food samples with LOD of 0.043 mg/L and recovery percentage of 94.2–101.3% for tartrazine.

Ye et al. (2013) have used glassy carbon rotating disk electrode modified with β -cyclodextrin-coated poly (diallylmethylammonium chloride)-functionalized graphene (β -CD-PDDA-Gr) composite film for determination of tartrazine. L-ascorbic acid was used as the reducing agent when synthesizing β -CD-PDDA-Gr as graphene (Gr) will reaggregate in water. The LOD for tartrazine was found to be $1.43 \times 10^{-8} \text{ mol/L}$ and the recovery percentage was between 95.11% and 103.60%. Gan et al. (2013) have determined the simultaneous electrocatalytic oxidation of tartrazine and sunset yellow by using graphene and mesoporous TiO_2 -modified carbon paste electrode. The detection limit was 8.0 nM and the recovery rate on the real samples from 97.41% to 102.10%.

A similar approach has developed a graphite with N,N-dimethylformamide to modify the surface of GCE via solvent evaporation, with LOD $1.5 \mu\text{g L}^{-1}$ (Song et al., 2014). Gan et al. (2012) have demonstrated a one-step and effective electrochemical method based on graphene (GN) layer-wrapped phosphotungstic acid (PTA) hybrid on the surface of GCE (GN/PTA/GCE). The GN acted as an electron transfer mediator during the oxidation process of tartrazine. The GN/PTA/GCE exhibited high sensitivity and selectivity for the simultaneous determination of sunset yellow and tartrazine. The modified electrode

showed well-defined oxidation peaks in DPV with detection limit was found to be $30.0 \mu\text{g/L}$ for tartrazine. In another effort, an electrochemical based on the modified carbon paste electrode (CPE)-coated silver wire electrode (sensor A), and tartrazine-cetyltrimethyl ammoniumbromide CTAB (TZ-CTA) as a chemical modifier (sensor B) has developed for the detection of tartrazine. The mechanisms of Na_3TZ and CTAB have shown in Figure 6. Based on the results, the LOD was found of 3.2×10^{-7} M for sensor A and 5.5×10^{-8} M for sensor B (Shawish et al., 2013). The CPE offer unique properties such as requiring simple preparation procedure, good reproducibility and repeatability, chemical inertness, robustness, low ohmic resistance, and stable which appropriate to apply in the sensing field (Shawish, 2009, Issa et al., 2012, Khaled et al., 2010).

Molecular imprinted polymer (MIP)

Recently, a rapid and high sensitivity has established for direct Tartrazine detection in foodstuff based on molecularly imprinted polymer (MIP). The method found chemical stability, inexpensive, and easy to operate. Zhao et al. (2014) have presented MIP based on modified GCE with multiwalled carbon nanotubes-ionic liquid and platinum nanoparticles composite film (MIP/MWCNTs-IL-PtNPs/GCE). The mechanism of MIP/MWCNTs-IL-PtNPs/GCE exhibits effective analytical performance during electrochemical oxidation of tartrazine as shown in Figure 7. The oxidation peak current was linear to tartrazine concentration range between $0.03\text{--}5.0 \mu\text{M}$ and $5.0\text{--}20 \mu\text{M}$ with sensitivities of $0.72 \mu\text{A}/\mu\text{M mm}^2$ and $0.24 \mu\text{A}/\mu\text{M mm}^2$, respectively. The LOD was found of 8 nM , with recoveries for 88–108%. Jiang et al. (2014) have developed the MIP polypyrrole sensor that is specifically bound to tartrazine quickly without sample pretreatment. Polypyrrole is suitable as MIP material due to high density of polypyrrole film can deposit on the surface of working electrode through electrochemical polymerization. The MIP-polypyrrole showed a linear relationship from 1 to 10 nM , and the LOD was achieved of 1 nM . The skeleton of polypyrrole film can form oxygen that contains groups such as carboxylic acid and carbonyl after oxidation during electrochemical process. The groups provide negative charges in the polymer skeleton to improve the selectivity of the sensor (Geise et al., 1991).

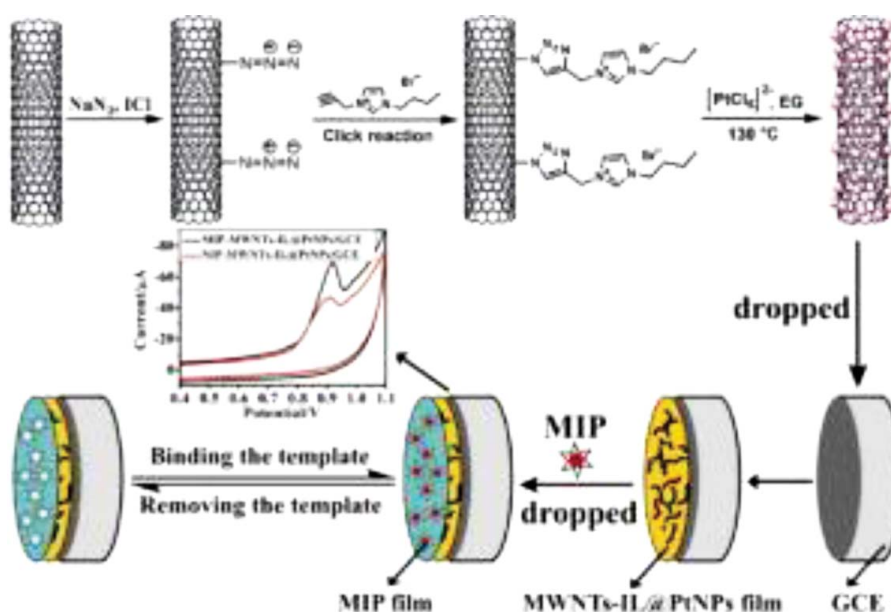


Figure 7. Schematic of MIP based on MIP/MWCNTs-IL-PtNPs/GCE composite film for detection of Tartrazine (Zhao et al., 2014).

Other detection method

Different from other detection methods, Pávai et al. (2015) have developed a detection method using the cellophane test strip to identify tartrazine, azorubine, Patent blue V, and natural coloring. The test was based on the color change of the cellophane strip when immersed in the color solution. This color changes were because of the binding of the color molecule with the cellophane strip. The characterization has completed by UV-Vis spectrophotometry at wavelength between 300 and 800 nm. The developed method was qualitative, sensitive, and useful for testing adulterated food products with synthetic colorants or for in situ tests at catering and mobile vendor.

Conclusion

The awareness of the adverse health effects of tartrazine has developed the detection analytical method in food stuffs. Different countries have different regulations on the maximum permitted level of tartrazine in food products. Diverse extraction methods are summarized for sample preparation such as liquid-liquid, solid-phase, and filter membrane extraction; thus, it is important to reduce and eliminate interference during the analysis in real samples. Appropriate extraction steps provide high sensitivity and selectivity in analytical methods. This review paper has highlighted insight scenario on different extraction and analytical methods for determination of tartrazine on healthy food among public interest on food safety and quality which can provide incalculable awareness to food industry and government bodies.

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