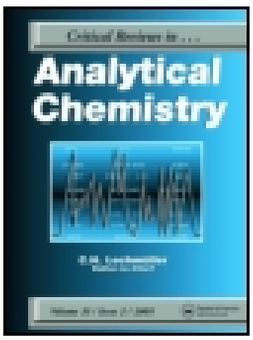

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A review of extraction and analytical methods for the determination of Tartrazine (E 102) in foodstuffs

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Abstract

Tartrazine is an azo food dye, orange-coloured and water soluble that usually used in foods, pharmaceuticals, cosmetics, and textiles. Tartrazine possess adverse health effect to human such as hyperactivity in children, allergy and asthma. Joint FAO/WHO Expert Committee on Food Additive (JECFA) and EU Scientific Committee for Food (SCF) standardized the acceptable daily intake (ADI) for Tartrazine is at 7.5 mg kg^{-1} body weight. Many researchers have been detected the presence of Tartrazine for monitoring the quality and safety of food products. In this review paper highlighted various detection and extraction methods of Tartrazine. Some of the analytical methods are available such as high performance liquid chromatography (HPLC), electrochemical sensor, thin-layer chromatography (TLC), spectrophotometry, capillary electrophoresis and liquid chromatography-tandem mass spectrometry (LC-MS). As extraction steps are discussed: liquid-liquid extraction (LLE), solid-phase extraction (SPE), membrane

filtration, cloud point extraction and other extraction method. Also, brief overview explained the synthesis process and metabolism of Tartrazine and the maximum permitted level in different countries. This review paper will give insight scenario on different extraction and analytical methods for determination of Tartrazine on healthy food among public attract attention on food safety and quality which can provide incalculable interest to food industry and government bodies.

Keywords

Tartrazine, Synthetic color, Extraction, Chromatography, Electrochemical Sensor.

1. Introduction

Food colorants are added to foods and beverages to restore the color lost during processing and to enhance the color of the food or to uniform the color of the final product. Food colorants have 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 as like as carmine or carminic acid extracted from insects. However, natural colorants are unstable towards pH, heat and light. Synthetic colorants are obtained from a chemical process and mostly classes are azo dyes, quinolone, xanthene, triarylmethanes and indigoid. Synthetic colorants are widely used due to their stability towards pH, heat and light, water soluble and lower production cost [1].

Tartrazine (E 102) is an azo dye, orange-colored, water-soluble powder commonly used in food products, drugs, cosmetics, and pharmaceuticals [2]. In food, Tartrazine used in soft drinks, juices, jellies, candies, cakes, cereal, soups, and other products [3]. It is known as Food Drug & Cosmetic (FD&C) Yellow No. 5, C.I. No. 19140 and Food Yellow No. 4 with European Community (EC) number E 102. High concentration of Tartrazine present in food may cause health complication such as hyperactivities in children when combined with sodium benzoate. The Acceptable Daily Intake (ADI) of Tartrazine has permitted at 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 [4]. The maximum level of Tartrazine in non-alcoholic beverages should not more than 0.01 g/mL [5].

Numerous analytical techniques have developed in determining Tartrazine including Spectrophotometry [6--8], Thin Layer Chromatography (TLC) [9,10], Capillary Electrophoresis

(CE) [11,12] and High Performance Liquid Chromatography [13--16]. Yamjala et al. [17] used the analytical methods in determining azo dyes in food products and Kaur and Gupta [18] have determined of synthetic food dyes and lakes using spectrophotometry determination methods. Rebane et al. [19] have analyzed the Sudan I-IV in various food matrices using LC-UV-Vis and LC-MS and determination of synthetic dyes by chromatographic methods [20]. Ahlström et al. [21] have developed the analytical methods for determination of banned azo dyes in consumer products. In my knowledge, there are no reviews previously summarized on the extraction and advance analytical techniques for determination of Tartrazine in food stuffs. This review paper is summarised on the available methods of extraction and analytical for determination of Tartrazine; it will worthwhile for food analysts and regulatory authorities to monitor and control the use of Tartrazine in food and beverage products.

2. Tartrazine (E 102)

Tartrazine (E 102) is an azo group food dye which characterized by N linkage (-N = N-) (**Figure 1)**, orange-coloured, and hydrophilic powder with molecular weight of 534.36 g/mol. This colorant first discovered in 1884. Tartrazine is essentially the 3-carboxy-5-hydroxy-1-(4'-sulfonatophenyl)-4-(4'-sulfonatophenylazo)-*H*-pyrazol-3-carboxylate [4] and 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 at 18 g/100 mL at 25°C, in propylene glycol is 7.0 g/100 mL at 25°C [22], ethanol is 0.8 mg/ml and in ethylene glycol monomethyl ether is at 20 mg/mL [23]. Melting point of Tartrazine is greater than 300°C and it can detect at wavelength of 425 nm in water [24]. Large concentration of Tartrazine in food may be harmful, and in a small portion of population, even a

small dose can give rise to health complication such as hyperactivities in children when combined with sodium benzoate.

2.1 Synthesis of Tartrazine

Tartrazine in foods are widely found in soft drinks, candies, jellies, jams, flavoured chips, cakes, ice cream, soups, sauces, and cereals [3] and non-food products found in cosmetics, medicine capsules, plastics, paint additives and textiles [25]. Tartrazine is synthesized by condensing phenylhydrazine-*p*-sulfonic acid with oxalacetic diethyl ester that will be paired with diazotized sulfanilic acid. The product ester will be hydrolysed with sodium hydroxide. The process is shown in **Figure 2**. Besides the mention 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 [26]. Additionally, Tartrazine can be converted to the similar aluminium lake by reacting aluminium oxide with the colouring matter under aqueous conditions. Undried aluminium oxide is prepared by reacting aluminium sulphate or aluminium chloride with sodium carbonate or sodium bicarbonate, or aqueous ammonia. Later the product is filtered, washed with water and dried [27].

2.2 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 azoreductases which will form aromatic amines [3]. These aromatic amines are oxidised to *N*-hydroxy derivative by P450 enzymes [29]. The carcinogenic activation mechanisms have reduction and cleavage of the azo dyes, oxidation of azo dyes and direct oxidation of the azo linkage to reactive electrophilic diazonium salt [30,31].

Tartrazine is metabolized by gastrointestinal microflora to sulphanilic acid and aminopyrorazalone. The compounds are cleaved to sulphanilic acid and α-amino-β-ketobutyric acid fragments by intermediary metabolism with release of carbon dioxide [4]. Excretion of 4-sulphophenylhydrazine metabolite labeled with sulphur-35 differed with the route of administration. After 48 hours of oral administration of this metabolite, 35% have excreted in urine and 49% in faeces. When specified intraperitoneal, 90% and 5% of the metabolites are excreted in urine and faeces, after 48 hours of administration. Through oral administration, 69% of the urinary radioactivity excreted is sulphanilic acid and 21% is 4-sulphophenylhydrazine. Through intraperitoneal administration, 9% of urinary radioactivity excreted is sulphanilic acid and 73% is 4-sulphophenylhydrazine. The outcome suggested the conversion of 4-sulphophenylhydrazine to sulphanilic acid occurs in the gut lumen.

3. Toxicology study of Tartrazine

Numerous studies have been shown on the health effect of Tartrazine that clinically significance is still unclear; however, intolerance reaction towards Tartrazine may happen. Health effects of Tartrazine are found as asthma, allergy and hyperactivity in children. Stenius and Lemola [32] have tested 140 asthmatics with acetylsalicyclic acid (ASA) with Tartrazine. They mentioned that one quarter of the patients found a positive reaction to one of the two tested agents and the most patients react are sensitive to ASA. Furthermore, a study conducted to 122 patients with 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 [33]. McCann et al. [34] have reported that an increased in hyperactivity has observed in three-year old and eight- to nine-year old children

when given two mixtures of four synthetic colours with sodium benzoate. About 166 atopic women with medical history of asthma, rhinitis, uticaria and with hypersensitivity to non-steroidal anti-flammatory agents have taken to test the ability of Tartrazine to cause hypersensitivity reactions. The results showed that Tartrazine has capable of provoking IgE and non-IgE dependent reactions in 6% of the volunteers from 99 volunteers who fulfilled all the requirements [35].

A first case study conducted by Trautlein and Mann [36], they are the firstly reported of systemic anaphylaxis secondary to a standard enema preparation. After the patient has given an enema containing Tartrazine and Sunset Yellow, the patient has developed asthma, uticaria and anaphylactic shock. The mechanisms are the immunoglobulin E mediated type hypersensitivity and the non-immonologic prostaglandin synthetase inhibition that are normally observed in allergic reactions to Tartrazine. Orchard and Varigos [37] 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 coloured cheese crisp and Tartrazine, have found positive. Besides, Bhatia [38] studied on food allergy or intolerance to psychotropic drug. Tartrazine is often times inculpated in causing allergic reaction. The total patients are given Tartrazine containing drug that is recorded. The results showed that 83 out of 2210 patients have found Tartrazine-containing drug developed allergy reaction and the symptoms lessen within 24 to 48 hours after the stopping the drug. No patients found allergic reaction toward non-Tartrazine-containing drug.

4. Maximum Permitted Level of Tartrazine

According to European Parliament and Council Directive 94/36/EC, the maximum level of Tartrazine has permitted in the range of 50 to 500 mg/kg with different food products. For non-alcoholic drinks, the maximum permitted level is 100 mg/L whereas alcoholic drinks are 200 mg/L. The maximum limit for soups and nutrition supplements are 50 mg/kg, for cheese, meat and fish are 100 mg/kg, for desert and flavoured milk product are 150 mg/kg and confectionary is 300 mg/kg [4]. According to CODEX Alimentarious [39], the maximum allowance limit for Tartrazine is 50 mg/kg for soups. In Canada, the maximum permitted level in juice, marmalade, flavoured milk, jelly, and sherbet is at 300 mg/kg in single or combined with other synthetic food colouring [40]. The maximum permitted used level for Tartrazine has set at 200 mg/kg of food in Thailand [41]. However, there is no permitted limit for food colorant in Malaysia [42].

5. Extraction Method of Tartrazine

Extraction method has needed prior to the detection step to remove all the impurities that may interrupt the result. The selected method of extraction is important depending on the samples [17]. Most common methods are liquid-liquid extraction, solid phase extraction, membrane filtration and cloud point extraction.

5.1 Liquid-liquid extraction (LLE)

Liquid-liquid extraction (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 [17]. Tsai et al. [43] have used acetonitrile for extraction of synthetic dyes in chili powder and syrup preserved fruits. The sample was extracted twice, centrifuged and filtered before injected into the vial. Acetonitrile has chosen

because of its good extraction yield and less fat solubility, carbohydrate precipitation and protein precipitation. Bento et al. [13] have used methanol-ammonium hydroxide for extraction of the colouring in yogurt and milk drink. Methanol-ammonium hydroxide solution (8:2) is extracted synthetic food colorants but not natural colorants using the proposed method specify to synthetic colour. Besides, Pávai et al. [44] have used green method of extraction where no hazardous chemical has used in sample preparation, only used deionised distilled water to dilute the sample.

Tang et al. [9] have used LLE for extraction of synthetic food colouring 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. [45] 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.

5.2 Solid-phase extraction (SPE)

Solid phase extraction (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 [17]. SPE method is a simple and able to extract the colorants without contaminants. Before conducting SPE methods, the cartridges should be washed and precondition. Methanol and acetic acid are the common choice of conditioning [5,15,46,47]. Recently, Martin et al. [46] adopted 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. [48] have used *n*-hexane to eliminate fat from flour and meat foodstuffs. Methanol-ammonia-

water solution is added to extract the samples. After 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 also can be used for soft drinks. A study conducted by de Andrade et al. [5] and 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 [49] 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 ammonia-ethanol solution. Huang et al. [50] have used 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 acidic condition [49]. The mixture is centrifuged and flowed into the cartridges, then eluted with 0.5% ammonia and methanol solution.

Khanavi et al. [51] and Hajimahmoodi et al. [16] have added polyamide sorbent into the treated sample to extract colorants from drink, syrup, candy, jelly gum, chocolate, coloured 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. [52] 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. [53] and

Sorouraddin et al. [54] have used white commercial wool yarn as the sorbent for colouring 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 one hour, the coloured yarn is taken out and washed with plenty of distilled water. The colorants are eluted out by putting the yarn in ammonia solution and heated.

Soylak and Cihan [55] have used multiwalled carbon nanotubes to separate Tartrazine compound from the food 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. [56] have used magnetic solid-phase extraction (MSPE) method for soft drinks, cocktails, solid beverages, ice cream, sugar-based and gelatine-based confection. Magnetic SPE method used Fe_3O_4-poly (ionic liquid) core shells microspheres as sorbent. Whereas Tavakoli et al. [14] have used the diverse hemimicelle solid-phase extraction (MHSPE) method, based on catyltrimethylammonium bromide-coated (CTAB) Fe_3O_4/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 homogenised, diluted with water and the pH adjusted to 9 with alkaline ammonia. Then, the samples are centrifuged and kept for preconcentration process based on 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 Fe3O4/SiO2 NPs are shown in **Figure 3**.

5.3 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 [17]. Vidotti et al. [57] have used 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 µm membrane filter. Prado et al. [12] have used filter membrane extraction to extract the colorants from alcoholic beverages. The samples are homogenised and degassed by mechanical agitation, and finally filtered with cellulose ester. Minioti et al. [58] 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 µm disposable syringe filter before analysis. Serdar and Knežević [59] have filtered the degassed and diluted soft drink with folded paper filter before the sample being filtered again with 0.45 µm membrane filter.

5.4 Cloud Point extraction

Cloud point extraction (CPE) is a green alternative technique for LLE method, analyte preconcentration and sample clean-up. CPE 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. 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 dehydrate the surfactant during the settling process [60,61]. 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 [62].

5.5 Other Extraction Method

The promising purposes of the green chemistry have caused a key focus of the research efforts on the development of miniaturised, efficient, and environmentally moderate sample extraction procedures. Therefore, solid-phase micro-extraction (SPME) [63,64] and liquid-phase micro-extraction (LPME) [65,66] 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 [67]. LPME method divided into two broad categories which are membrane-protected solvent and exposed solvent [68]. 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 [69,70]. 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 [71--73]. 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 [74].

Antakli et al. [6] have extracted the colorant from soft drinks by ultrasound-assisted extraction (UAE) method. Soft drink sample is degassed using ultasonication and the aliquot has taken for analysis. Shen et al. [75] have extracted four artificial food colorants including

Tartrazine and three carotenoids by 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.

6. Analytical Method

Numerous analytical methods have been developed for identification and quantification of synthetic food colorant such as high performance liquid chromatography, thin-layer chromatography, spectrophotometry, capillary electrophoresis and electrochemical techniques. The summary of analytical detection methods is available in Table 1.

6.1 Chromatography

6.1.1 High Performance Liquid Chromatography (HPLC)

High performance liquid chromatography (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. [13] 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 µg/L and 3.71 µg/L, with the recovery rate of 98.1-106.6%. Tavakoli et al. [14] have determined Tartrazine by HPLC coupled with UV-Vis detector. The samples were extracted using mixed hemimicelle SPE method before being analysed. The LOD and LOQ for Tartrazine were found of 2.50 µg/L and 8.33 µg/L. Bonan et al. [15] 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. [47] 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. [76] have used HPLC coupled with DAD detector and second order algorithms for the analysis of three synthetic dyes in non-alcoholic beverages and the recovery values ranging between 97-105%. Besides, Vachirapatama et al. [41] have analysed seven synthetic dyes using monolithic C18 column by HPLC. The LOD and LOQ for Tartrazine were 1.92 µg/L and 6.41 µg/L, respectively. García-Falcón and Simal-Gándara [77] have used reverse-phase HPLC with UV-Vis detector to determine five synthetic food colours 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, green chromatography method has developed. C18 column with phosphate buffer and Triton X-100 (0.25%, v/v) aqueous solution has used as the mobile phase instead of organic solvent [57]. In the presence of Triton X-100, C18 column becomes more polar making the separation of the colorants possible. The LOD for Tartrazine was found 0.125 mg/L [57]. Vidotti et al. [57], Khanavi et al. [51] have used C8 column as the stationary phase with phosphate buffer and Triton X-100 as the 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 µg/mL. Hajimahmoodi et al. [16] used green chromatography method with C8 column and UV-Vis detector to analyze eight synthetic food colorants present in cookies, colour 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.

6.1.2 Thin-Layer Chromatography (TLC)

Thin-layer chromatography (TLC) is the method to compare the ratio of fronts (R_f) values and the extracted colours in relation to the standard for identifying the food colourant at the presence in samples [5]. Tang et al. [9] 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. [5] have used silica gel chromatography with a capillary to identify Sunset Yellow, Tartrazine, Amaranth and Brilliant Blue from the samples. Kartsova et al. [10] have used electromigration methods by using electroosmotic thin layer chromatography (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 mobile phase and the detection limit was found 100 µg/L. Gerasimov [78] 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%.

6.1.3 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 [17]. In recent year, Martin et al. [46] have analysed 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 µg/kg and 10 µg/kg, respectively. The recovery values range found between 100.1-119.7% when spiked with 10 µg/kg of standard Tartrazine. A study done by Tsai et al. [43] 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. [45] 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 mobile phase because it has better resolution compared to methanol. The detection limit was found at 0.3 µg/L and the recovery of 92.1-97.7%.

6.2 Capillary Electrophoresis (CE)

Capillary electrophoresis (CE) is an electrophoretic method where the analysis and the separation of the molecules are concluded in a capillary tube [17]. Česla et al. [11] have studied on the effect of pH and β-cyclodextrin additive to the background electrolyte on the separation of sulphonated 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 sulphonated azo dyes in capillaries coated with polyacralamide compared to non-coated fused silica capillaries and the addition of β-cyclodextrin to phosphate background electrolyte improved the separation selectivity.

Prado et al. [12] have used CE for determination of synthetic dyes in alcoholic beverages. The LOD and LOQ for Tartrazine were 2.0 μg/mL and 6.6 μg/mL, respectively. Additionally, Huang et al. [79] 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 μg/mL. The method found lower detection limits when compared with the conventional capillary electrophoresis method available. Huang et al. [50] have established analytical method based on CE for the detection of common colorants in milk beverages. High efficiency capillary electrophoresis separation methods for eight colors are separated by present of running buffer containing 7.0 mM β-cyclodextrin within 9 min. Polyamide column solid-phase extraction (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 μg/mL and 85%, respectively.

6.3 Spectrophotometry

Spectrophotometric techniques are simple, high sensitive, selectivity and less interference from the colored present in the foodstuffs. 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 [17]. Antakli et al. [6] 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. [7] have developed a simple kinetic spectrophotometric method based on silver nanoparticle (AgNPs) for determination of Tartrazine

in lemon, papaya-flavoured 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. [8] 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 oxidised in this method thus eliminating interference.

Altınöz and Toptan [80] have simultaneously detection of Tartrazine and Ponceau 4R in the wavelength range between 300-700 nm using spectrophotometric techniques. The linearity range was found 1.00-60.00 μg/mL for Tartrazine and 1.00-52.00 μg/mL for Ponceau 4R, respectively. Dinç et al. [81] 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 μg/mL. The procedures were proven by using synthetic ternary mixtures and successfully applied for simultaneous detection of synthetic colorants in soft drink products.

6.4 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 [82]. During the electrochemical process, Tartrazine takes place a one-electron and one-proton which is irreversible reaction on oxidation process. The mechanism of electrochemical oxidation of Tartrazine is shown in **Figure 5**. The working electrode reaction is based on Nernst equation by the mechanism involving an equal number of electrons and protons. Gan et al. [83] 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 electrochemical process.

Mercury-free electrodes have established for determination of Tartrazine synthetic azo dyes in foodstuff [53,82--85]. Previously, Gómez et al. [86] and López-de-Alba et al. [87] have introduced adsorptive stripping voltammetry by using hanging mercury drop electrode to determine the concentration of Tartrazine. Azo group (-N = N-) contain in Tartrazine is electrochemical active that able to reduce on electrode surface when predominately using mercury electrode. However, limitation of mercury electrode in electrochemical cell is containing high toxicity that may cause to environmental pollution and adverse health effects.

Recently, Rovina et al. [88] have developed a simple and rapid electrochemical sensor based on CHIT/CaONPs/MWCNTs with modified gold electrode for determination of Tartrazine in candy, jelly and soft drink. Under 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. [89] have simultaneously detected Tartrazine and Sunset Yellow based on modified glassy carbon electrode (GCE) with graphene oxide (GO) and multi-

walled carbon nanotubes (MWCNT). The MWCNT/GO/GCE showed 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 [90]. 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 [91] 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 8.3×10^{-10} M and 5.2×10^{-10} for Tartrazine and Sunset Yellow, respectively.

Chao and Ma [92] have used 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 μg/L with recovery ranging between 95.0-101.4% 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. [93] have

used 1-alyl-3-methyl imidazolium tetraflouroborate 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 behaviour toward the oxidation of Tartrazine and Sunset Yellow. The electrode succeeded in simultaneous determination of Tartrazine and Sunset Yellow in different food sample with LOD of 0.043 mg/L and recovery percentage of 94.2-101.3% for Tartrazine.

Ye et al. [82] 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 synthesising β-CD-PDDA-Gr as graphene (Gr) will reaggregate in water. The LOD for Tartrazine was found 1.43×10^{-8} mol/L and the recovery percentage was between 95.11% and 103.60%. Gan et al. [53] 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 97.41-102.10%.

A similar approach has developed a graphite with N,N-dimethylformamide to modify the surface of GCE via solvent evaporation, with LOD 1.5 μg L^{-1} [94]. Gan et al. [83] 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 μg/L for Tartrazine. In another effort, an electrochemical based on modified carbon paste electrode (CPE) coated silver wire electrode (sensor A), and Tartrazine-cetryltrimethyl 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 [95]. 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 sensing field [96--98].

6.5 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. [99] 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 0.03-5.0 μM and 5.0-20 μM with sensitivities of 0.72 μA/μM mm^2 and 0.24 μA/μM mm^2, respectively. The LOD was found of 8 nM, with recoveries for 88-108%. Jiang et al. [100] have developed 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 electro-chemical 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 [101].

6.6 Other Detection Method

Different from other detection methods, Pávai et al. [44] have developed a detection method using cellophane test strip to identify Tartrazine, Azorubine, Patent blue V, and natural colouring. The test was based on the colour change of the cellophane strip when immersed in the colour solution. This colour changes were because of the binding of the colour 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.

7. Conclusion

The awareness of the adverse health effects of Tartrazine has developed the detection analytical method in food stuffs. Different countries have different regulation 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. In 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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References

[1] Titus, A. M. M. *Chemistry of Food Additives and Preservatives*, John Wiley & Sons: West Sussex, 2013.

[2] Tanaka, T. Reproductive and neurobehavioural toxicity study of tartrazine administered to mice in the diet. *Food Chem. Toxicol.* **2006**, *44*, 179–187.

[3] Demirkol, O.; Zhang, X.; Ercal, N. Oxidative effects of Tartrazine (CAS No 1934-21-0) and New Coccin (CAS No. 2611-82-7) azo dyes on CHO cells. *J. Verbr. Lebensm.* **2012**, *7*, 229–236.

[4] EFSA Panel on Food Additives and Nutrient Sources. Scientific Opinion on the re-evaluation Tartrazine (E 102). *EFSA Journal* **2009**, *7(11)*, 1331.

[5] De Andrade, F. I.; Guedes, M. I. F.; Vieira, I. G. P.; Mendes, F. N. P.; Rodrigues, P. A. S.; Maia, C. S. C.; Ávilaa, M. M. M.; Ribeiroe, L. M. Determination of synthetic food dyes in commercial soft drinks by TLC and ion-pair HPLC. *Food Chem.* **2014**, *157*, 193–198.

[6] Antakli, S.; Nejem, L.; Katran, S. Simultaneous determination of Tartrazine and Brilliant Blue in foodstuffs by spectrometric method. *Int. J. Pharm. Pharm. Sci.* **2015**, *7(6)*, 214–218.

[7] Sahraei, R.; Farmany, A.; Mortazavi, S. S. A nanosilver-based spectrophotometry method for sensitive determination of tartrazine in food samples. *Food Chem.* **2013**, *138*, 1239–1242.

[8] Olgun, F. A. O.; Ozturk, B. D.; Apak, R. Determination of synthetic food colorants in water-soluble beverages individually by HPLC and totally by Ce(IV)-Oxidative Spectrophotometry. *Food Anal. Methods* **2012**, *5*, 1335–41.

[9] Tang, T. X.; Xu, X. J.; Wang, D. M.; Zhao, Z. M.; Zhu, L. P.; Yang, D. P. A rapid and green limit test method for five synthetic colorants in foods using Polyamide Thin-layer Chromatography. *Food Anal. Methods* **2014**, *8*, 459–66.

[10] Kartsova, L. A.; Alekseeva, A. V.; Khmel'nitskii, I. K.; Komissarchik, S. M.; Nyanikova, G. G.; Berezkin, V. G. Electromigration method in determination of synthetic food dyes. *J. Anal. Chem.* **2009**, *64(12*), 1264–69.

[11] Česla, P.; Fischer, J.; Tesařová, E.; Jandera, P.; Staněk, V. Effects of capillary coating and β-cyclodextrin additive to the background electrolyte on the separation of sulphonated azo dyes by capillary zone electrophoresis. *J. Chromatogr. A* **2007**, *1149*, 358–67.

[12] Prado, M. A.; Boas, L. F. V.; Bronze, M. R.; Godoy, H. T. Validation of methodology for simultaneous determination of synthetic dyes in alcoholic beverages by capillary electrophoresis. *J. Chromatogr. A* **2006**, *1136*, 231–236.

[13] Bento, W. D. A. S.; Lima, B. P.; Paim, A. P. S. Simultaneous determination of synthetic colorants in yogurt by HPLC. *Food Chem.* **2015**, *183*, 154–160.

[14] Tavakoli, M.; Shemirani, F.; Hajimahmoodi, M. Magnetic Mixed Hemimicelles Solid-Phase Extraction of Three Food Colorants from Real Samples. *Food Anal. Methods* **2013**, *7*, 100–108.

[15] Bonan, S.; Fedrizzi, G.; Menotta, S.; Elisabetta, C. Simultaneous determination of synthetic dyes in foodstuffs and beverages by high-performance liquid chromatography coupled with diode-array detector. *Dyes Pigm.* **2013**, *99*, 36–40.

[16] Hajimahmoodi, M.; Afsharimanesh,M.; Moghaddam, G.;, Sadeghi, N.; Oveisi, M. R.; Jannat, B.; Pirhadi, E.; Zamani, M. F.; Kanan, H. Determination of eight synthetic dyes in

foodstuffs by green liquid chromatography. *Food Addit. Contam. Part A* **2013**, *30(5)*, 780–785.

[17] Yamjala, K.; Nainar, M. S.; Ramisetti, N. R.Methods for the analysis of azo dyes employed in food industry – A review. *Food Chem.* **2015**, *192*, 813–824.

[18] Kaur A. D.; Gupta, U. The review on spectrophotometric determination of synthetic food dyes and lakes. *Gazi Uni. J. Sci.* **2012**, *25(3)*, 579–88.

[19] Rebane, R.; Leito, I.; Yurchenko, S.; Herodes, K. A review of analytical techniques for determination of Sudan I-IV dyes in food matrixes. *J. Chromatogr. A* **2010**, *1217*, 2747–2757.

[20] Kucharska, M.; Grabka, J. A review of chromatographic methods for determination of synthetic dyes. *Talanta* **2010**, *80*, 1045–51.

[21] Ahlström, L.; Eskilsson, C. S.; Björklund, E. Determination of banned azo dyes in consumer goods. *Trends Anal. Chem.* **2005**, *24*, 49-56.

[22] Marmion, D. *Colorants for Food. In Kirk-Othmer Encyclopedia of Chemical Technology*, John Wiley & Sons: New York, 2001.

[23] Green, F. J. *The Sigma-Aldrich Handbook of Stains, Dyes, and Indicators*, Aldrich Chem Co.: Milwaukee, 1990.

[24] National Center for Biotechnology Information (NCBI). 2016. PubChem Compound Database: CID=164825 [Online]. https://pubchem.ncbi.nlm.nih.gov/compound/164825 (accessed Oct 21, 2016)

[25] Stolz, A. Basic and applied aspects in the microbial degradation of azo dyes. *Appl. Microbiol. Biotechnol.* **2001**, *56*, 69–80.

[26] Deshpande, S. S. *Handbook of Food Toxicology*, Marcel Dekker: New York, 2002.

[27] JECFA (Joint FAO/WHO Expert Committee on Food Additives). 63rd meeting of the Joint FAO/WHO Expert Committee on Food Additives. Aluminium Lakes of Colouring Matters, General Specifications. 2004.

[28] Weisz, A.; Ridge, C. D.; Roque, J. A.; Mazzola, E. P.; Ito, Y. Preparative separation of two subsidiary colours of FD&C Yellow No. 5 (Tartrazine) using spiral high-speed counter-current chromatography. *J. Chromatogr. A* **2014**, *1343*, 91–100.

[29] de Aragao Umbuzeiro, G.; Freeman, H. S.; Warren, S. H.; De Oliveira, D. P.; Terao, Y.; Watanabe, T.; Claxton, L. D. The contribution of azo dyes to the mutagenic activity of the Cristais River. *Chemoshere* **2005**, *60*, 55–64.

[30] Chen, H. Recent advances in azo dye degrading enzyme research. *Curr. Protein Pept. Sci.* **2006**, *7*, 101–111.

[31] Bardi, L.; Marzona, M. Factors Affecting the Complete Mineralization of Azo Dyes. In Biodegradation of Azo Dyes, Erkurt, H. A., Eds.; Springer-Verlag: Berlin, Heiderberg, 2010.

[32] Stenius, B. S. M.; Lemola, M. Hypersensitivity to acetylsalicyclic acid (ASA) and tartrazine in patients with asthma. *Clin. Exp. Allergy* **1976**, *6*, 119–29.

[33] Neuman, I.; Elian. R.; Nahum, H.; Shaked, P.; Creter, D. The danger of yellow dyes (Tartrazine) to allergic subjects. *Clin. Exp. Allergy* **1978**, *8*, 65–68.

[34] McCann, D.; Barrett, A.; Cooper, A.; Crumpler, D.; Dalen, L.; Grimshaw, K.; Kitchin, E.; Lok, K.; Porteous, L.; Prince, E.; Sonuga-Barke, E. Food additives and hyperactive

behaviour in 3-year-old and 8/9-year-old children in the community: a randomised, double-blinded, placebo-controlled trial. *The Lancet* **2007**, *370*, 1560–7.

[35] Caldas, L. Q. A.; Moreira, M. D.; Olej, B.; Pestana, S.; Morais, L.; Piumbini, R. Double-blind and crossed-over randomized placebo controlled study in atopic patients challenged with a yellow colorant Tartrazine. In: IV Congreso International da Sociedade Cubana de Toxicologia, 2007, Havana. Computer Disk of the Conference. Havana Cuba: Universidad de Havana, v.1. p. 49.

[36] Trautlein, J. J.; Mann, W. J. Anaphylactic shock caused by yellow dye (FD&C No. 5 and FD&C No. 6) in an enema (case report). *Annals of allergy* **1978**, *41*, 28–29.

[37] Orchard, D. C.; Varigos, G. A. Fixed drug eruption to Tartrazine. *Australas J. Dermatol.* **1997**, *38*, 212–214.

[38] Bhatia, M. S. Allergy to tartrazine in psychotropic drugs. *J. Clin. Psychiatry* **2000**, *61*, 473–476.

[39] CODEX Alimentarius. General Standard for Food Additives (CODEX STANDARD 192-1995); 2015.

[40] Health Canada. 2015. List of Permitted Food Additives [Online]. www.hc-sc.gc.ca/fn-an/securit/addit/list/3-colour-colour-eng.php

[41] Vachirapatama, N.; Mahajaroensiri, J.; Visessanguan, W. Identification and determination of seven synthetic dyes in foodstuffs and soft drinks on monolithic C18 column by High Performance Liquid Chromatography. *J. Food Drug. Anal.* **2008**, *16(5*), 77–82.

[42] Zailuddin, A.; Faridah, H. B.; Baharuddin, S. Investigation on the Presence of Sunset Yellow and Tartrazine in Commercial Beverages and Quantitation using Ion-Pair Formation and Extraction. *Jurnal Intelek* **2015**, *5*, 1-8.

[43] Tsai, C. F.; Kuo, C. H.; Shih, D. Y. C. Determination of 20 synthetic dyes in chili powders and syrup-preserved fruits by liquid chromatography/tandem mass spectrometry. *J. Food Drug Anal.* **2015**, *23*, 453–462.

[44] Pávai, M.; Szabó, T.; Paszternák, A. The potential use of cellophane test strips for the quick determination of food colours. *Cellulose* **2015**, *22*, 1883–1891.

[45] Ma, K.; Li, X. J.; Wang, H. F.; Zhao, M. Rapid and sensitive method for the determination of additives in red wine by Ultra-performance Liquid Chromatography Tandem Mass Spectrometry. *Food Anal. Methods* **2015**, *8*, 203–212.

[46] Martin, F.; Oberson, J. M.; Meschiari, M.; Munari, C. Determination of 18 water-soluble artificial dyes by LC-MS in selected matrices. *Food Chem.* **2016**, *197*, 1249–55.

[47] Feng, F.; Zhao, Y.; Yong, W.; Sun, L.; Jiang, G.; Chu, X. Highly sensitive and accurate screening of 40 dyes in soft drinks by liquid chromatography-electrospray tandem mass spectrometry. *J. Chromatogr. B* **2011**, *879*, 1813–1818.

[48] Qi, P.; Lin, Z.H.; Chen, G.Y.; Xiao, J.; Liang, Z.A.; Luo, L.N.; Zhou, J.; Zhang, X.W. Fast and simultaneous determination of eleven synthetic colour additives in flour and meat products by liquid chromatography coupled with diode-array detector and tandem mass spectrometry. *Food Chem.* **2015**, *181*, 101–110.

[49] Yoshioka, N.; Ichihashi, K. Determination of 40 synthetic food colors in drinks and candies by high-performance liquid chromatography using a short column with photodiode array detection. *Talanta* **2007**, *74*, 1408-1413.

[50] Huang, H. Y.; Shih, Y. C.; Chen, Y. C. Determining eight colorants in milk beverages by capillary electrophoresis. *J. Chromatogr. A* **2002**, *959*, 317–325.

[51] Khanavi, M.; Hajimahmoodi, M.; Ranjbar, A. M.; Oveisi, M. R.; Ardekani, M. R. S.; Mogaddam, G. Development of a green chromatographic method for simultaneous determination of food colorants. *Food Anal. Methods* **2011**, *5*, 408–415.

[52] Kirschbaum, J.; Krause, C.; Brückner, H. Liquid chromatographic quantification of synthetic colorants in fish roe and caviar. *Eur. Food Res. Technol.* **2006**, *222*, 572–574.

[53] Gan, T.; Sun, J.; Meng, W.; Song, L.; Zhang, Y. Electrochemical sensor based on graphene and mesoporous TiO2 for the simultaneous determination of trace colorant in food. *Food Chem.* **2013**, *141*, 3731–3737.

[54] Sorouraddin, M. H.; Rostami, A.; Saadati, M. A simple and portable multi-colour light emitting diode based photocolourimeter for the analysis of mixtures of five common food dyes. *Food Chem.* **2011**, *127*, 308–313.

[55] Soylak, M.; Cihan, Z. Solid-phase extraction of tartrazine on multiwalled carbon nanotubes for separation and enrichment. *Toxicol. Environ. Chem.* **2013**, *95(4)*, 559–566.

[56] Wu, H.; Gao, N.; Zhang, L.; Li, Y.; Shi, Y.; Du, L. Automated magnetic Solid-Phase Extraction for synthetic colorant determination. *Food Anal. Methods* **2015**, *9*, 614–623.

[57] Vidotti, E. C.; Costa, W. F.; Oliveira, C. C. Development of green chromatographic methods for determination of colorants in food samples. *Talanta* **2006**, *68*, 516–521.

[58] Minioti, K. S.; Sakellariou, C. F.; Thomaidis, N. S. Determination of 13 synthetic food colorants in water-soluble foods by reverse-phase high-performance liquid chromatography coupled with diode-array detector. *Anal. Chim. Acta* **2007**, *583*, 103–110.

[59] Serdar, M.; Knežević, Z. Simultaneous LC analysis of food dyes in soft drinks. *Chromatographia* **2009**, *70*, 1519–1521.

[60] Nascimento, C. F.; Rocha, D. L.; Rocha, F. R. A fast and environmental friendly analytical procedure for determination of melamine in milk exploiting fluorescence quenching. *Food Chem.* **2015**, *169*, 314–319.

[61] Sato, N.; Mori, M.; Itabashi, H. Cloud point extraction of Cu(II) using a mixture of triton X-100 and dithizone with a salting-out effect and its application to visual determination. *Talanta* **2013**,*117*, 376–381.

[62] Xing, W.; Chen, L. Micelle-mediated extraction and cloud point preconcentration of bergenin from *Ardisia japonica*. Sep. Purif. Technol. **2013**, *110*, 57–62.

[63] Lord, H. L.; Rajabi, M.; Safari, S.; Pawliszyn, J. A study of the performance characteristics of immunoaffinity solid phase microextraction probes for extraction of a range of benzodiazepines. *J. Pharm. Biomed. Anal.* **2007**, *44*, 506–519.

[64] Ouyang, G.; Pawliszyn, J. A critical review in calibration methods for solid-phase microextraction. *Anal. Chim. Acta* **2008**, *627*, 184–197.

[65] Jeannot, M. A.; Cantwell, F. F. Solvent microextraction into a single drop. *Anal. Chem.* **1996**, *68*, 2236–2240.

[66] He, Y.; Lee, H. K. Liquid-phase microextraction in a single drop of organic solvent by using a conventional microsyringe. *Anal. Chem.* **1997**, *69*, 4634–4640.

[67] Prosen, H.; Zupancˇicˇ-Kralj, L. Solid-phase microextraction. *Trends Anal. Chem.* **1999,** *18*, 272–282.

[68] Kokosa, J. M. Advances in solvent-microextraction techniques. *Trends Anal. Chem.* **2013**, *43*, 2–13.

[69] Rajabi, M.; Ghanbari, H.; Barfi, B; Asghari, A.; Haji-Esfandiari, S. Ionic liquid-based ultrasound-assisted surfactant-emulsified microextraction for simultaneous determination of three important flavouring compounds in plant extracts and urine samples. *Food Res. Int.* **2014**, *62*, 761–770.

[70] Rajabi, M.; Bazregar, M.; Daneshfar, A., Asghari, A. Comparison of air-agitated liquid–liquid microextraction and ultrasound-assisted emulsification microex- traction for polycyclic aromatic hydrocarbons determination in hookah water. *J. Sep. Sci.* **2015**, *38*, 2496–2502.

[71] Llompart, M.; Celeiro, M.; Lamas, J. P.; Sanchez-Prado, L.; Lores, M.; Garcia-Jares, C. Analysis of plasticizers and synthetic musks in cosmetic and personal care products by matrix solid-phase dispersion gas chromatography-mass spectrometry. *J. Chromatogr. A* **2013**, *1293*, 10–19.

[72] Celeiro, M.; Guerra, E.; Lamas, J. P.; Lores, M.; Garcia-Jares, C.; Llompart, M. Development of a multianalyte method based on micro-matrix-solid-phase dispersion for the analysis of fragrance allergens and preservatives in personal care products. *J. Chromatogr. A* **2014**, *1344*, 1–14.

[73] Li, H.; Sun, N.; Zhang, J.; Liang, S.; Sun, H. Development of a matrix solid phase dispersion-high performance liquid chromatography-tandem mass spectrometric method for

multiresidue analysis of 25 synthetic colorants in meat products. *Anal. Methods* **2014**, *6*, 537–547.

[74] Sha, O.; Zhu, X.; Feng, Y.; Ma, W. Aqeous two-phase based on ionic liquid liquid-liquid microextraction for simultaneous determination of five synthetic food colorants in different food samples by high-performance liquid chromatography. *Food Chem.* **2015**, *174*, 380–386.

[75] Shen, Y.; Zhang, X.; Prinyawiwatkul, W.; Xu, Z. Simultaneous determination of red and yellow artificial food colorants and carotenoid pigments in food products. *Food Chem.* **2014**, *157*, 553–558.

[76] Culzoni, M. J.; Schenone, A. V.; Llamas, N. E.; Garrido, M.; Di Nezio, M. S.; Band, B. S. F.; Goicoechea, H. C. Fast chromatographic method for the determination of dyes in beverages by using high performance liquid chromatography—Diode array detection data and second order algorithms. *J. Chromatogr. A* **2009**, *1216*, 7063–7070.

[77] García-Falcón, M. S.; Simal-Gándara, J. Determination of food dyes in soft drinks containing natural pigments by liquid chromatography with minimal clean-up. *Food Control* **2005**, *16*, 293–297.

[78] Gerasimov, A. V. Use of computer-assisted color treatment of chromatograms in the TLC determination of Tartrazine. *J. Anal. Chem.* **2003**, *58(3)*, 209–210.

[79] Huang, H.Y.; Chiu, C. W.; Sue, S. L.; Cheng, C. F. Analysis of food colorants by capillary electrophoresis with large-volume sample stacking. *J. Chromatogr. A* **2003**, *995*, 29–36.

[80] Altınöz, S.; Toptan, S. Determination of Tartrazine and Ponceau-4R in Various Food Samples by Vierordt's Method and Ratio Spectra First-Order Derivative UV Spectrophotometry. *J. Food Comp. Anal.* **2002,** *15*, 667–683.

[81] Dinç, E.; Baydan, E.; Kanbur, M.; Onur, F. Spectrophotometric multicomponent determination of sunset yellow, tartrazine and allura red in soft drink powder by double divisor-ratio spectra derivative, inverse least-squares and principal component regression methods. *Talanta* **2002**, *58*, 579–594.

[82] Ye, X.; Du, Y.; Lu, D.; Wang, C. Fabrication of β-cyclodextrin-coated poly (diallydimethylammonium chloride)-functionalized grapheme composite film modified glassy carbon-rotating disk electrode and its application for simultaneous electrochemical determination colorants of Sunset yellow and Tartrazine. *Anal. Chim. Acta* **2013**, *779*, 22–34.

[83] Gan, T.; Sun, J. Y.; Cao, S. Q.; Gao, F. X.; Zhang, Y. X.; Yang, Y. Q. One-step electrochemical approach for the preparation of grapheme wrapped-phosphotungstic acid hybrid and its application for simultaneous determination of sunset yellow and tartrazine. *Electrochim. Acta* **2012**, *74*, 151–157.

[84] Ghoreishi, S. M.; Behpour, M.; Golestaneh, M. Simultaneous determination of sunset yellow and tartrazine in soft drinks using gold nanoparticles carbon paste electrode. *Food Chem.* **2012**, *132*, 637–641.

[85] Zhang, W.; Liu, T.; Zheng, X.; Huang, W.; Wan, C. Surface-enhanced oxidation and detection of Sunset Yellow and Tartrazine using multi-walled carbon nanotubes film-modified electrode. *Colloids Surf. B: Biointerfaces* **2009**, *4*, 28–31.

[86] Gómez, M.; Arancibia, V.; Rojas, C.; Nagles, E. Adsorptive stripping voltammetric determination of tartrazine and sunset yellow in gelatins and soft drink powder in the presence of cetylpyridinium bromide. *Int. J. Electrochem. Sci.* **2012**, *7*, 7493–502.

[87] López-de-Alba, P. L.; López-Martinez, L.; De-León-Rodriguez, L. M. Simultaneous determination of synthetic dyes tartrazine, allura red and sunset yellow by differential pulse polarography and partial least squares. A multivariate calibration method. *Electroanal.* **2001**, *14(3)*, 197–205.

[88] Kobun, R.; Siddiquee, S.; Shaarani, S. M. D. Sensitive Determination of Tartrazine (E 102) Based on Chitosan/Nanoparticles/MWCNTs Modified Gold Electrode in Food and Beverage Products. *Trans. Sci. Technol.* **2016**, *3(1-2)*, 176 -180

[89] Qiu, X.; Lu, L.; Leng, J.; Yu, Y.; Wang, W.; Jiang, M.; Bai, L. An enhanced electrochemical platform based on graphene oxide and multi-walled carbon nanotubes nanocomposite for sensitive determination of Sunset Yellow and Tartrazine. *Food Chem.* **2016**, *190*, 889–895.

[90] Zhang, Y.; Hu, L.; Liu, X.; Liu, B.; Wu, K. Highly-sensitive and rapid detection of ponceau 4R and tartrazine in drinks using alumina microfibers-based electrochemical sensor. *Food Chem.* **2015**, *166*, 352–357.

[91] Wang, M.; Zhao, J. Facile synthesis of Au supported on ionic liquid functionalized reduced graphene oxide for simultaneous determination of Sunset yellow and Tartrazine in drinks. *Sens. Actuators B: Chem.* **2015**, *216*, 578–85

[92] Chao, M.; Ma, X. Convenient Electrochemical Determination of Sunset Yellow and Tartrazine in Food Samples using a Poly(L-Phenylalanine)-Modified Glassy Carbon Electrode. *Food Anal. Methods* **2015**, *8*, 130–138.

[93] Majidi, M. R.; Pournaghi-Azar, M. H.; Baj, R. F. B.; Naseri, A. Formation of grapheme nanoplatelet-like structures on carbon-ceramic electrode surface: application for simultaneous determination of Sunset Yellow and Tartrazine in some food samples. *Ionics* **2014**, *21*, 863–875.

[94] Song, X. J.; Shi, Z.; Tan, X. H.; Zhang, S. H.; Liu, G. S.; Wu, K. B. One-step solvent exfoliation of graphite to produce a highly-sensitive electrochemical sensor for tartrazine. *Sens. Actuators B: Chem.* **2014**, *197*, 104–8.

[95] Shawish, H. M. A.; Ghalwa, N. A.; Saadeh, S. M.; Harazeen. Development of novel potentiometric sensors for determination of tartrazine dye concentration in foodstuff products. *Food Chem.* **2013**, *138*, 126–32.

[96] Shawish, M. H. A. A mercury(II) selective sensor based on N, N-bis(salicylaldehyde)-phenylenediamine as neutral carrier for potentiometric analysis in water samples. *J. Hazard. Mater.* **2009**, *167*, 602–608.

[97] Issa, M. Y.; Ibrahim, H.; Shehab, R. O. New copper(II)-selective chemically modified carbon paste electrode based etioporphyrin I dihydrobromide. *J. Electroanal. Chem.* **2012**, *666*, 11–18.

[98] Khaled, E.; Hassan, A. N. H.; Mohamed, G. G.; Seleim, A. A. Carbon paste and PVC electrodes for the flow injection potentiometric determination of dextromethorphan. *Talanta* **2010**, *81*, 510–515.

[99] Zhao, L. J.; Zeng, B. Z.; Zhao, F. Q. Electrochemical determination of tartrazine using a molecularly imprinted polymer – multiwalled carbon nanotubes - ionic liquid supported Pt nanoparticles composite film coated electrode. *Electrochim. Acta* **2014**, *146*, 611–617.

[100] Jiang, S.; Xu, J.; Xu, P.; Liu, L.; Chen, Y.; Qiao, C.; Yang, S.; Sha, Z.; Zhang, J. A novel molecularly imprinted sensor for direct Tartrazine detection. *Anal. Lett.* **2014**, *47*, 323–330.

[101] Geise, R. J.; Adams, J. M.; Barone, N. J. Electropolymerized films to prevent interferences and electrode fouling in biosensors. *Biosens. Bioelectron.* **1991**, *6*, 151–160.

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

FOOD MATRIX	ANALYTICAL METHODS					REFERENCES
	EXTRACTION	DETECTION				
		INSTRUMENT	MOBILE PHASE	LOD	RECOVERY	
Sugar and gummy confectionary, ice cream, and chocolate sweets	The prepared sample was loaded into SPE cartridge and eluted out with 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%	[46]
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% to 106.6%	[13]
Chilli	Acetonitrile was	LC-MS	A: Acetonitrile	N/A	91.2-	[43]

powder and raisin	used as solvent. The sample was extracted twice, centrifuged and filtered before injected into the vial		B: 20 mM ammonia acetate buffer with 1% acetic acid		96.1%	
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	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	[92]

	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.					
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	Spectrophotometric	-	0.12 g/mL	101.75-102.75%	[6]

	dissolved in distilled water and centrifuged.					
Soup powder, yogurt, sweet cream cheese, jams, sparkling tablet, and beverages	Deionised distilled water was used to dilute the sample and no hazardous chemical was used.	Cellophane test strip	-	N/A	N/A	[44]
Soft drink, instant powdered drink, sugar-	Aqueous two-phase system based on ionic liquid microextraction (IL-ATPS) was	HPLC-UV-Vis	A: 0.02 mol/L ammonium acetate aqueous solution (pH 4.5)	0.053 ng/mL	94.1-98.9%	[74]

based and gelatine-based confectionary	used as extraction method. The extraction agent used was 1-alkyl-3-methylimidazolium bromide and salt.		B: Methanol			
Soft drinks, cocktails, solid beverages, ice	Magnetic SPE method that used Fe_3O_4-poly (ionic liquid) core shells microspheres as sorbent was used	HPLC	A: 0.02 mol/L ammonium acetate aqueous solution (pH 7.5)	5.6 ng/mL	N/A	[56]

cream, sugar-based and gelatine-based confection	for extraction steps. The diluted and filtered sample was flowed into MSPE system along with the sorbent to be extract.		B: Methanol-acetonitrile (30:70, v/v)			
Flour and meat foodstuffs	Methanol-ammonia-water solution was added to extract the sample and the sample extracts were	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	[48]

	loaded into Strata-X-AW cartridges and eluted out with ethanol that contained ammonia-water.		B: 5 mM ammonium acetate solution			
Saffron rice, saffron dessert, honey cream,	Mixed hemimicelle solid-phase extraction (MHSPE) method, based on	HPLC-UV-Vis	Mobile phase A: 0.1 M ammonium acetate solution (pH 6.7)	2.50 µg/L	N/A	[14]

fruit juice-coated ice cream, and saffron ice cream, beverages	catyltrimethylammonium bromide-coated (CTAB) Fe_3O_4/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.		Mobile phase B: Methanol-acetonitrile (50:50, v/v)			
Beverages, gelatines, solid samples	Water-ammonia aqueous solution was used as the solvent and the mixture of the	Polyamide TLC method coupled with on-plate SPE and	Ethanol (95%)-water-acetic acid solution (50:50:1)	4.19 ng	N/A	[9]

	sample and solvent was sonicated and diluted.	backlight-assisted detection				
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	[5]
Red wine	Methanol was added as solvent for the extraction.	Ultra performance liquid	A: 10 mM acetate buffer solution	0.3 µg/L	92.1-97.7%	[45]

	The mixture was degassed, centrifuged, acidified and filtered prior to the detection steps	chromatography coupled with electrospray ionization-tandem mass spectrometry (UPLC-ESI-MS/MS)	B: Methanol/acetonitrile (9:1, v/v)			
	The sample was mixed with	HPLC-PAD	A: Ammonium acetate	10 ng/	N/A	[75]

	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.		B: Methanol	mL		
Soft drink	The sample was transferred into 50-mL flask and adjusted for the volume with doubly distilled	1-alyl-3-methyl imidazolium tetraflourobor ate ionic liquid	-	0.04 3 mg/ L	94.2-101.3%	[93]

	water.	modified carbon-ceramic electrode				
Soft drink	No extraction procedure performed	Glassy carbon rotating disk electrode modified with β-cyclodextrin-coated poly(diallylmethyl ammonium chloride)-functionalized graphene (β-CD-PDDA-Gr) composite film	-	1.43 × 10^{-8} mol/L	95.11% - 103.60%	[82]

Candy, royal jellies, ice cream, solid custard jelly powder, juice powder, soft drink, colouring 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. Colouring coated chocolate sample was added into water to dissolve the coloured shell. The remaining solution was separated, centrifuged and diluted with	Graphene and mesoporous TiO_2 modified carbon paste electrode	-	8.0 nM	97.41%-102.10%	[53]

	CH_3COOH solution. White commercial wool yarn was added and heated. The colorants was eluted out by heating the wool with NH_3.					
Beverages, solid	The samples were mixed with	HPLC-DAD with C8	Mobile phase A: Acetonitrile	N/A	87.6%	[15]

food matrices	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.	column	Mobile phase B: 100 mM sodium acetate buffer (pH 7)			
Cookies, coloured rice, saffron and fruit juice	Polyamide sorbent was added into the treated sample to extract the colorants. The colorants were removed from the	HPLC-UV-Vis with C8 column	phosphate buffer and Triton X-100	0.04 mg/ kg	98.1%	[16]

	polyamide with alkaline-ammonia.					
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%	[55]
Powdered gelatine, fruit syrup, candy	The gelatine sample was prepared in water and the aliquots was used. Fruit syrup and candy sample was	Silver nanoparticle (AgNPs) and spectrophotometry	Acetate-acetic acid buffer (pH 6)	0.3 ng/mL	97.0%-100.3%	[7]

	mixed with double distilled water and filtered. The filtered sample was diluted and analysed.					
Beverage, powdered beverage	Sample was dissolved in deionised distilled water and degassed. The sample was filtered prior to analysis step	Ce(IV)-oxidative spectrophotometry	CERAC reagent	N/A	N/A	[8]
Drink, syrup, candy, jelly, chocolate and gum	Polyamide sorbent was added into the treated sample to extract the colorants. The colorants were	HPLC with C8 column	phosphate buffer and Triton X-100	0.05 µg/mL	N/A	[51]

	removed from the polyamide with alkaline-ammonia.					
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%.	[47]
Coated chocolate, commercial cakes	White commercial wool yarn was used as the sorbent. The	Multi-colour light emitting diode based photocolorim	A: NaH_2PO4/Na2 HPO4 buffer (pH 6)	N/A	N/A	[54]

and soft drinks	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.	eter	B: Acetonitrile (35%)			
Non-alcoholic beverages	No extraction procedure performed	HPLC-DAD and second order algorithms	N/A	N/A	97%-105%	[76]
Stock solution	No extraction procedure performed	Electroosmotic thin layer chromatography and	Methanol-2-propanol-ethyl acetate-water	100 µg/L	N/A	[10]

		capillary zone electrophoresis				
Soft drink	The degassed and diluted soft drink was filtered with folded paper filter before the sample being filtered again with 0.45 µm membrane filter.	LC-DAD	Tetrabutylammonium hydrogen sulphate, methanol and deionised water	N/A	96.9-97.1%	[59]
Foodstuffs, soft drinks	Foodstuff sample was diluted to 25 mL in volumetric flask and soft	HPLC with monolithic C18 column	Mobile phase A: Methanol-water (12%, v/v)	1.92 µg/L	N/A	[41]

	drink was degassed and diluted. The sample was filtered before injected into HPLC.		Mobile phase B: 10 mM acetic acid			
Stock solution	No extraction procedure performed	Capillary zone electrophoresis	N/A	N/A	N/A	[11]
Drinks and candies	The column was prepared by making slurry of polyamide and packed it into a column. The	RP-HPLC-PAD	A: 0.1 mol/L ammonium acetate aqueous solution (pH 6.70	0.040 µg/mL	89.5-93.9%	[49]

	column was preconditioned with acetic acid and the colorants were eluted out with ammonia-ethanol solution.		B: Methanol-acetonitrile (50:50, v/v)			
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	RP-HPLC-DAD	A: 100 mM sodium acetate buffer (pH 7) B: Acetonitrile	N/A	N/A	[52]

	polyamide.					
Beverage, alcoholic beverage,	The sample was placed in an ultrasonic bath	RP-HPLC-DAD	A: Ammonium acetate solution (pH 7)	1.87 µg/L	N/A	[58]

jam, sweets, sugar confectionary	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.		B: Methanol-acetonitrile (80:20, v/v)			
Alcoholic beverages	The sample was homogenised and degassed by mechanical agitation. The sample was then filtered with cellulose ester.	Capillary electrophoresis	Phosphate buffer with SDS	2.0 µg/mL	N/A	[12]

Beverages	The sample was transferred into amber glass bottles, sealed and stored under refrigerated condition at temperature below 4°C. Then, the sample was homogenised and degassed in an ultrasonic bath. The sample aliquots were injected directly into HPLC.	HPLC-UV-Vis	Mobile phase A: Methanol Mobile phase B: 40 mM ammonium acetate aqueous solution (pH 5)	0.3 mg/L	N/A	[77]
Juice and gelatine	The sample was dissolved in water by heating, cooled, diluted to 50 mL with water	HPLC with C18 column	phosphate and Triton X-100 aqueous solution	0.125 mg/L	N/A	[57]

	and filtered through a 0.45 µm membrane filter.					
Stock solution	No extraction procedure performed	Conversion of TLC plate into Windows 95/9 and the processing of the image received with Adobe Photoshop 5.0 program package	N/A	N/A	97.0-104.9%	[78]
Soft drinks, jellies, and milk beverages	Polyamide column used was preconditioned with methanol and water. The sample was washed with	CE separation method with large-volume sample stacking (LVSS)	Running buffer of sodium hydroxide and disodium tetraborate	0.003 µg/mL	N/A	[79]

	deionised water and methanol and the colorant was eluted using ammonia solution-methanol.					
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%	Capillary electrophores is with borax-sodium hydronium buffer mixed with β-cyclodextrin	N/A	N/A	109.1%	[50]

	ammonia and methanol solution.					

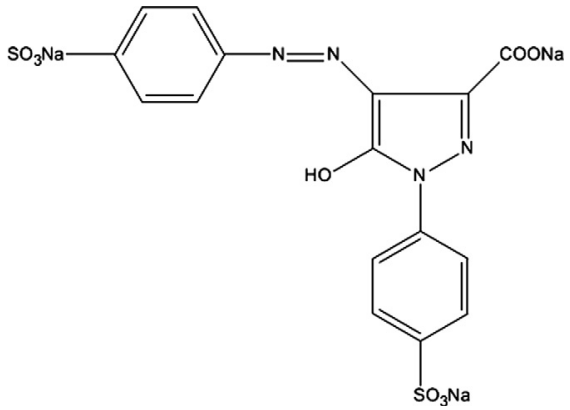


Figure 1. Chemical structure of Tartrazine

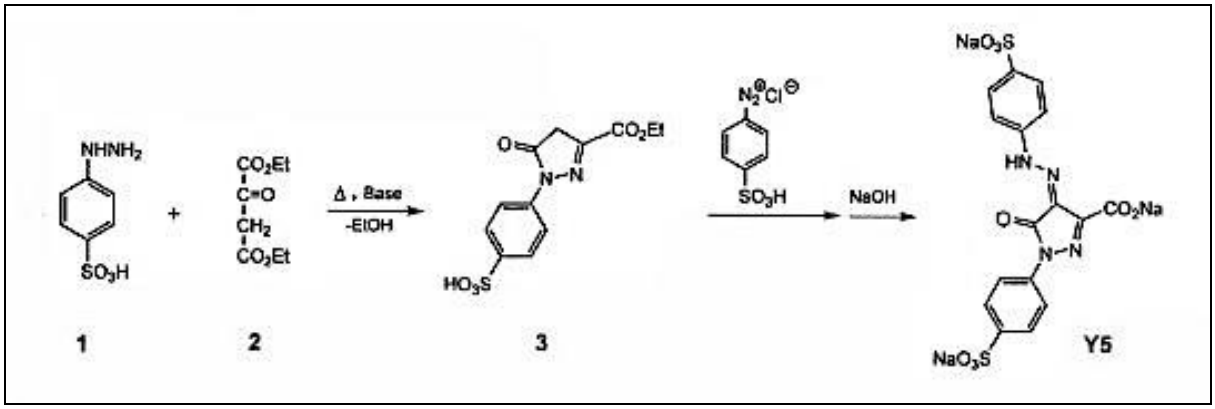


Figure 2. Condensation of (1) phenylhydrazine-p-sulfonic acid with (2) oxalacetic diethyl ester to form (Y5) Tartrazine [28].

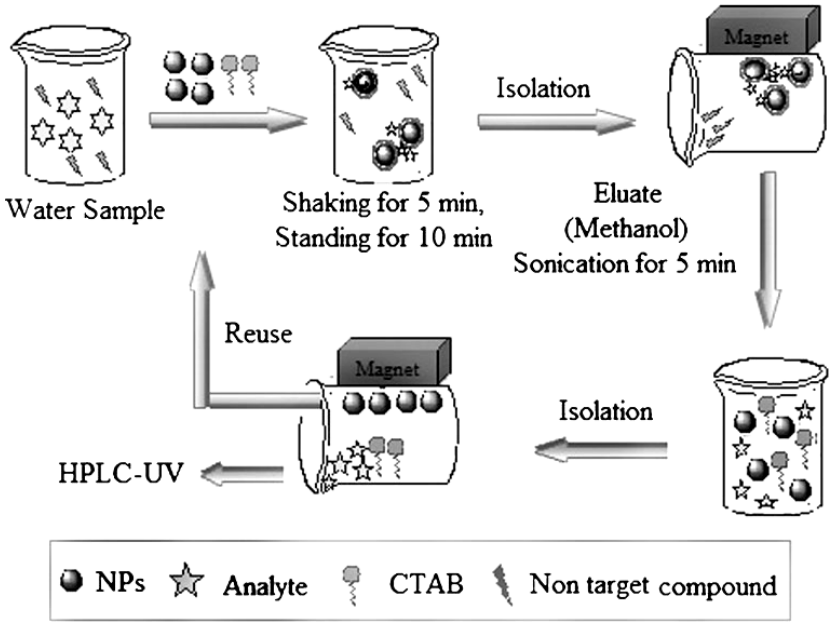


Figure 3. Schematic illustration of the extraction process by using mixed hemimicelle SPE based on CTAB-coated Fe3O4/SiO2 NPs [14].

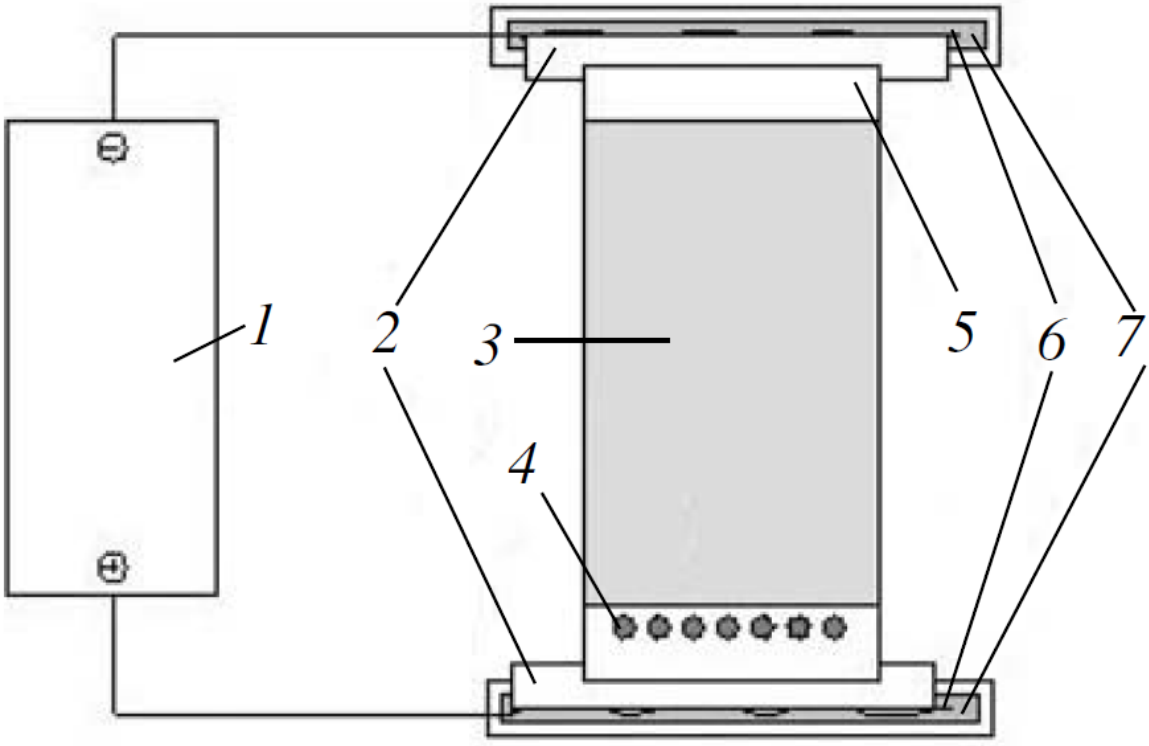
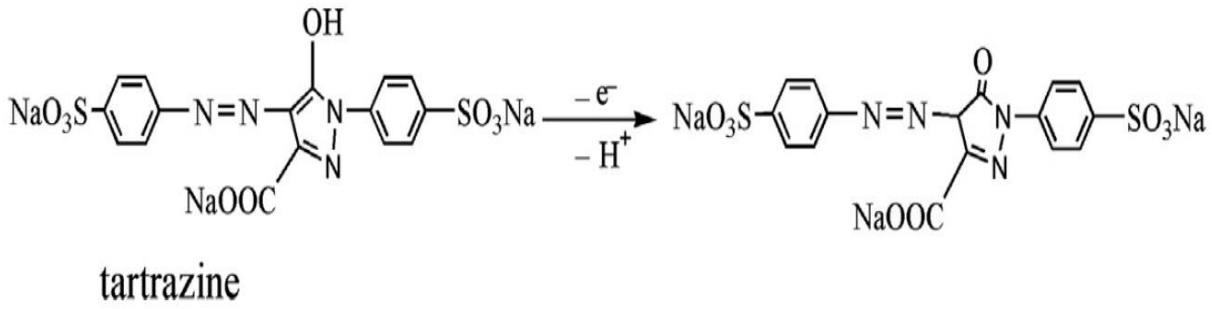


Figure 4. The schematic diagram of EOTLC; (1) voltage supply source, (2) filter paper, (3) polymer film, (4) analytes, (5) TLC plate, (6) electrodes, (7) the mobile phase reservoir [10].



tartrazine

Figure 5. The mechanism for the electrochemical processes of Tartrazine [89].

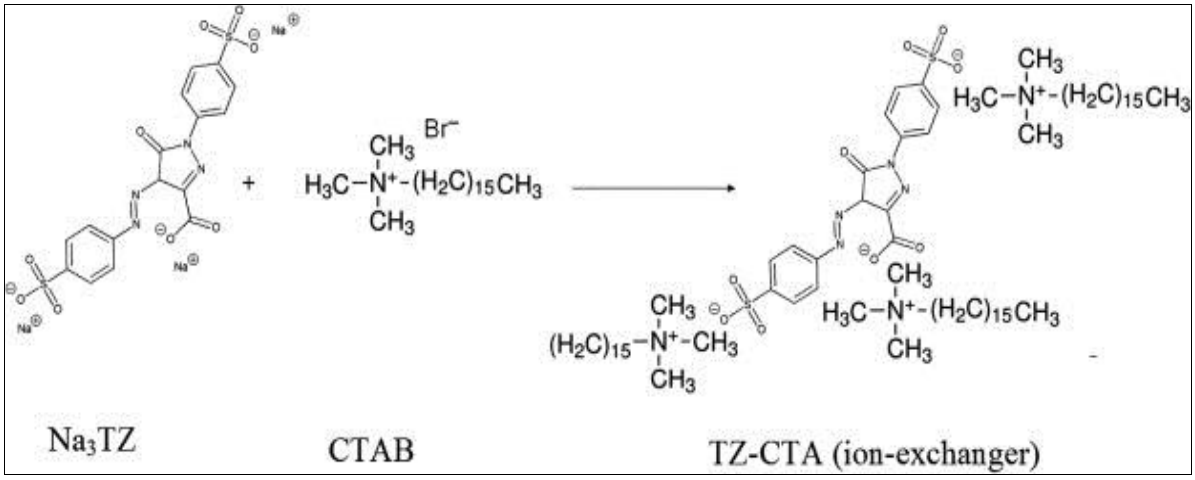


Figure 6. The mechanism reaction between Na3TZ and CTAB [95].

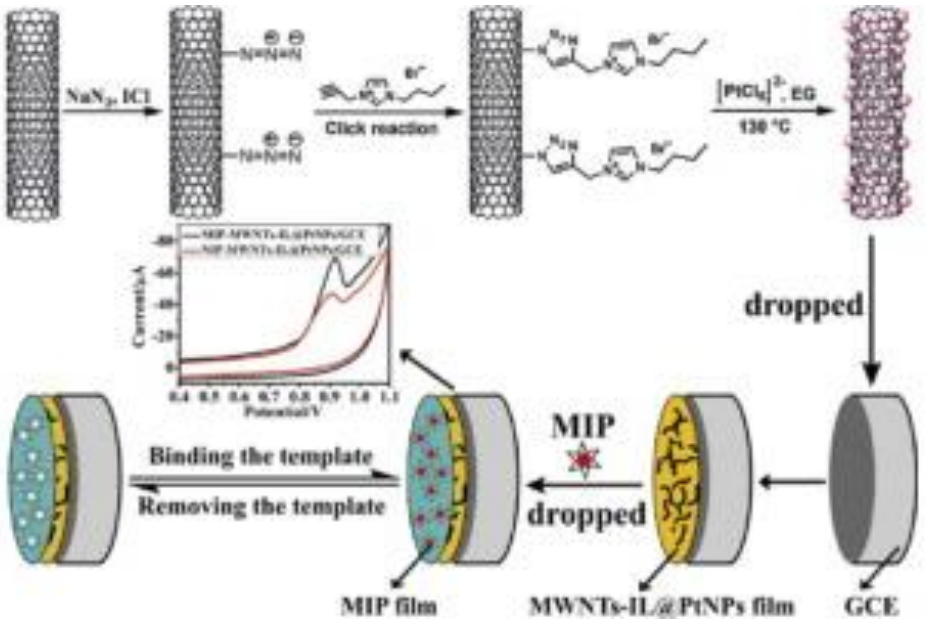


Figure 7. Schematic of MIP based on MIP/MWCNTs-IL-PtNPs/GCE composite film for detection of Tartrazine [99].