



Sensitive and selective determination of gallic acid in green tea samples based on an electrochemical platform of poly(melamine) film



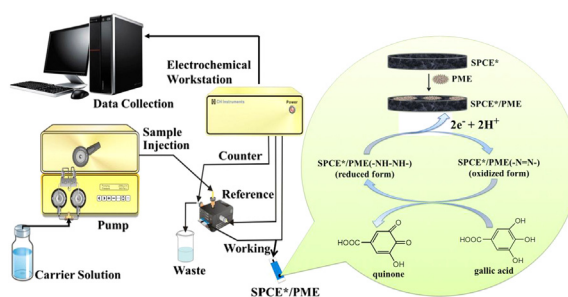
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HIGHLIGHTS

- A nitrogen-rich conducting polymer was used for electroanalysis of gallic acid.
- The sensor exhibits excellent electrochemical activity in both acidic and neutral media.
- Good analytical results in terms of low detection limit and wide linear range are obtained.
- The flow-injection amperometric assay is highly stable for continuous 57 replicates measurement (RSD = 3.9%).
- The assay shows good recovery for green tea samples.

GRAPHICAL ABSTRACT



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ABSTRACT

In this work, an electrochemical sensor coupled with an effective flow-injection amperometry (FIA) system is developed, targeting the determination of gallic acid (GA) in a mild neutral condition, in contrast to the existing electrochemical methods. The sensor is based on a thin electroactive poly(-melamine) film immobilized on a pre-anodized screen-printed carbon electrode (SPCE*/PME). The characteristics of the sensing surface are well-characterized by field emission scanning electron microscopy (FE-SEM), X-ray photoelectron spectroscopy (XPS) and surface water contact angle experiments. The proposed assay exhibits a wide linear response to GA in both pH 3 and pH 7.0 phosphate buffer solutions (PBS) under the optimized flow-injection amperometry. The detection limit (S/N = 3) is 0.076 μM and 0.21 μM in the pH 3 and pH 7 solutions, respectively. A relative standard deviation (RSD) of 3.9% is obtained for 57 successive measurements of 50 μM GA in pH 7 solutions. Interference studies indicate that some inorganic salts, catechol, caffeine and ascorbic acid do not interfere with the GA assay. The interference effects from some orthodiphenolic compounds are also investigated. The proposed method and a conventional Folin–Ciocalteu method are applied to detect GA in green tea samples using the standard addition method, and satisfactory spiked recoveries are obtained.

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1. Introduction

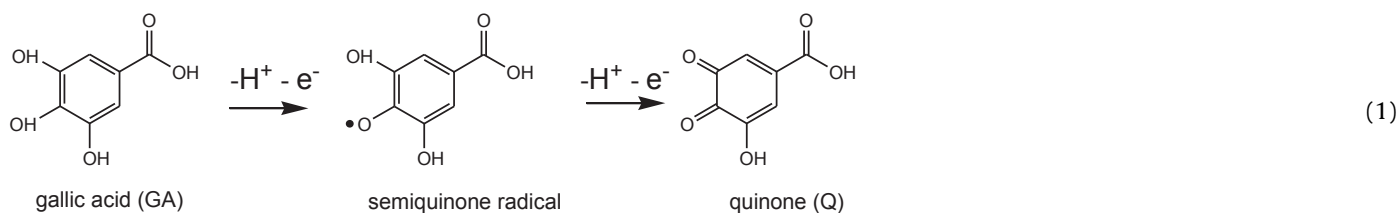
Gallic acid (3,4,5-trihydroxybenzoic acid, GA) and its structurally related compounds, such as ellagic acid (a cyclized dimer) and hydrolysable tannins (a type of polyphenol), are comprehensively

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present in green tea, black tea, grapes and plants [1]. Due to its powerful antioxidant and antiradical activity, GA has received considerable attention because of its multiple biological and pharmaceutical applications, such as anti-mutagenicity, anti-inflammation, antibacterial properties, antiviral properties, anti-cancer and its protection against cardiovascular diseases [1]. GA solution is generally accepted as a reference standard when determining the total polyphenolic content in plants by the Folin–Ciocalteu assay, and the resultant GA equivalents are used to indicate the antioxidant level of the plant extracts [2]. Owing to the bioactive and pharmacological importance of GA, the development of robust, sensitive and fast methods for GA determination of various types of samples is of great interest.

Several techniques have been used to investigate the determination of GA and its derivatives, such as high performance liquid chromatography [3–5], ultrahigh performance liquid chromatography [6,7], flow injection chemiluminescent analysis [8,9], reverse flow spectrophotometry [10], photoelectrochemical platform [11] and electrochemical techniques [12–24]. Electroanalytical methods have certain advantages over other analytical methods, such as the low cost of instrumentation, high sensitivity, high accuracy, wide dynamic range and fast response time [13,17]. Direct electrochemical oxidation of electroactive GA is facile at glassy carbon electrodes [12,13], indium–tin oxide electrodes [13], and Pt electrodes [14]. The detailed ECEC–first order mechanisms were explored by Abdel-Hamid et al., and are summarized as Equation (1) [12].



In acidic solutions (pH 2.5), the 4-hydroxyl groups of GA are oxidized/deprotonated irreversibly at about 0.4–0.6 V to form semiquinone radicals, followed by irreversible oxidation/deprotonation to form quinone (Q) at about 0.8–0.9 V at carbon electrodes [12,13]. The oxidation of GA has also been reported in acidic solutions at electrodes modified with Zn/Al-layered double hydroxide film [16], SiO₂ nanoparticles [17], multi-walled carbon nanotubes [18,19], polyepinephrine [20], polyethyleneimine-functionalized graphene oxide [13], dinuclear copper (II) octaazamacrocyclic complex [21], thionine/nickel hexacyanoferrate composites [22] and boron-doped graphene [23]. Generally, acidic solutions (pH 2–3) are used as the optimal electrolytes for analysis of GA using an electrochemical oxidation approach [16–21]. According to the study of Ghoreishi et al., the peak current for GA oxidation decreased with an increasing pH, and the current response vanished in pH > 5 [19]. Abdel-Hamid et al., also reported finding no defined peaks for GA oxidation in solutions of pH > 6.22 [12]. A tyrosinase-gold nanoparticle biosensor has been used to detect GA in pH 7.4, where an electrochemical reduction method was employed [24]. Only a few studies have investigated the analysis of GA using non-acidic working media [14,22]. Narayanan et al. investigated the electrocatalytic oxidation of GA in NaNO₃

electrolyte solutions, where a bimediator-modified electrode based on polythionine and nickel hexacyanoferrate was employed [22]. Kunitake et al. reported the electrochemical oxidation of GA with an indium tin oxide electrode in a water–hexane microemulsion solution [14]. Another aspect of the electrochemical oxidation of GA is the serious surface fouling accompanied by the adsorption of oxidized products. This typically occurs when determining the phenol derivatives by means of electrochemical oxidation, and the same electrode cannot be used for the next measurement [25]. Recently, a synthesized copper complex was used to modify a graphite electrode for GA determination, and the surface fouling problems were significantly reduced [21]. The above electrochemical tools are effective for GA determination; however, the electrode preparation is tedious and the use of acidic media or nonaqueous solvents is not environmentally friendly. Therefore, it is worth investigating the use of an alternative platform and a mild aqueous solution for the continuous determination of GA.

Poly(melamine) film (PME) is a new type of conducting polymer, which has attracted considerable attention for analytical purposes because of its high stability and abundant nitrogen functional groups [26–28]. PME was first fabricated onto a glassy carbon electrode by Liu et al. for the electrochemical detection of guanine, adenine and epinephrine [26]. In 2014, Goyal et al. used edge plane pyrolytic graphite as an electrode substrate for PME modification to develop a serotonin sensor [27]. PME-modified screen-printed carbon electrodes have been applied for the electrochemical detection of free chlorine [28]. PME is also believed to provide

binding sites for the accumulation of GA. Thus, the objective of the present study was to investigate the electrochemical detection of GA using PME-modified electrodes, the aim being to detect GA efficiently and to solve the problems described above. The results indicated that GA could be detected in aqueous solutions over a wide pH range using PME-modified electrodes and a flow-injection system. After optimizing the experimental conditions, such as PME amount, pH values of the aqueous solutions, hydrodynamic flow rates and applied electrode potentials, we developed a simple platform for the rapid determination of GA over a wide linear range and low detection limit in both acidic and neutral media. To validate the proposed assay, the determination of the total GA content in the green tea samples was performed and compared with the Folin–Ciocalteu assay, and satisfactory recoveries were obtained.

2. Experimental

2.1. Reagents

Melamine (ME, Sigma–Aldrich), gallic acid (GA, Acros Organics, USA) and all other compounds (Acros Organics, USA) were of the highest analytical grade available and were used without further purification. Folin–Ciocalteu reagent (2 N) was obtained from Sigma–Aldrich. All solutions were prepared using deionized water

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