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ScienceDirect
Food Chemistry 105 (2007) 340–345
Food Chemistry
www.elsevier.com/locate/foodchem

Analytical, Nutritional and Clinical Methods

Flow injection chemiluminescent detection of gallic acid in olive fruits

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Received 4 June 2006; received in revised form 11 November 2006; accepted 26 November 2006

Abstract

One economic and environment friendly flow injection chemiluminescent method for the determination of gallic acid was developed. It was based on the inhibited chemiluminescent emission of alkaline luminol–$KMnO_4$ system by gallic acid. The logarithm of the difference of chemiluminesent intensity of the alkaline luminol–$KMnO_4$ system in the absence of gallic acid from that in the presence of gallic acid was linear with the logarithm of the concentration of gallic acid in the range from 1.0×10^{-9} to 5.0×10^{-5} g ml^{-1} with a detection limit of 2.2×10^{-10} g ml^{-1}. The relative standard deviation of eleven determinations of 1.0×10^{-6} g ml^{-1} gallic acid was 1.7%. The method was successfully applied to the determination of gallic acid in olive fruits.

Keywords: Gallic acid; Inhibited chemiluminescence; Flow injection analysis; Luminol; Food analysis

1. Introduction

Developing analytical methods for the determination of polyphenols especially for individual polyphenol (e.g. gallic acid) continues to be a hot topic for analytical chemists (Ali, Maqsood, & Fahimuddin, 1988; Arce, Rios, & Valcarcel, 1998; Bianco & Savolainen, 1997; Dmitrienko, Medvedeva, Ivanov, Shpigun, & Zolotov, 2002; Gali, Perchellet, & Perchellet, 1991; Goto, Yoshida, Kiso, & Nagashima, 1996; Hayatsu, Arimoto, & Negishi, 1988; Malovana, Montelongo, Perez, & Rodriguez-Delgado, 2001; Ng, Lafontaine, & Harnois, 2000; Perchellet, Gali, Perchellet, Klish, & Armbrust, 1992; Rodriguez-Delgado, Malovana, Perez, Borges, & Garcia Montelongo, 2001; Sanchez-Moreno, Larrauri, & Saura-Calixto, 1998; Shahrzad & Bitsch, 1996; Shahrzad & Bitsch, 1998; Thies & Fischer, 1973; Tian, Zhang, Yang, & Ito, 2000; Zhu, Luo, Chen, Liu, & Li, 2005) in recent years. Gallic acid (2,3,4-trihydroxybenzoic acid) is one kind of natural phenolic compound widely existed in plants like olive fruits and has shown pharmacological properties, e.g. strong antimutagenic, anticarcinogenic, and antioxidant activities (Gali et al., 1991; Hayatsu et al., 1988; Perchellet et al., 1992; Sanchez-Moreno et al., 1998). Gallic acid thus has been deliberatively eliminated as an indicator of adulteration into fruit juices and alcoholic beverages with other polyphenolic compounds. Correspondingly, numerous methods including liquid chromatography (Bianco & Savolainen, 1997; Goto et al., 1996; Malovana et al., 2001; Rodriguez-Delgado et al., 2001; Shahrzad & Bitsch, 1996; Shahrzad & Bitsch, 1998; Tian et al., 2000), gas chromatography with mass spectrometric detection (Ng et al., 2000), capillary electrophoresis (Arce et al., 1998; Zhu et al., 2005), spectrometry (Dmitrienko et al., 2002; Thies & Fischer, 1973), and voltammetry (Ali et al., 1988) have been reported for the monitoring of gallic acid in food (e.g. plants, juices, tea, wines, alcoholic beverages), in wood, and in human plasma and urines. Unfortunately spectroscopic techniques are time-consuming and laborious; chromatographic techniques are slow and expensive and the complicated instruments are also required. Some

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doi:10.1016/j.foodchem.2006.11.061

methods among them such as HPLC and CE, are powerful separation tools, however, their linear ranges and detection limits reported prevent the determination of trace amount of polyphenols. Therefore it is hard to utilize these reported methods as an official way to monitor the content of gallic acid in the real samples.

Thanks to the connection of chemiluminescence with flow injection analysis (FI-CL), it overcomes these drawbacks described above and exhibits wide linear range, high sensitivity, and nice selectivity (see reviews Townshend (1990)). The most outstanding advantage of FI-CL is possible to mix the samples and the reagents rapidly with a high degree of reproducibility. The amounts of samples required are always in microliter range and hence FI-CL reduces the cost of the sample analysis and also produces small amount of wastes. These advantages have been confirmed during the quantitive monitoring of DNA, amino acid, pharmaceutical, and polyphenol compounds (Ma et al., 2002; Townshend, 1990) in past years. Taking the detection of gallic acid as an example, Slawinska and Slawinski (1965) reported chemiluminesent detection of gallic acid by using potassium ferricyanide–hydrogen peroxide system. Lin and coworkers proposed the determination of gallic acid based on the electrochemically enhanced or inhibited CL emission of $Ru(bpy)_3^{2+}$–tri–*n*-propylamine system in pH 8.0 phosphate buffer solution (Lin, Li, Pang, & Cui, 2004) and luminol in alkaline solutions (Sun, Cui, Li, Li, & Lin, 2000; Sun, Cui, Lin, Li, & Zhao, 2000) by gallic acid. Ju et al. (Zhang, Zhou, & Ju, 2002) developed a FI-CL sensor for the detection of gallic acid based on the CL emission of the electrostatically immobilizing luminol and periodate on anion-exchange resins. Hydrogen peroxide–gallic acid systems enhanced by formaldehyde (Wang & Wang, 2005) and hexavalent chromium (Xie & Wang, 2005) also have been proposed for the monitoring of gallic acid in our laboratory.

Among various CL systems, the CL emission of the alkaline luminol–oxidation reagent systems and their applications have been used widely and investigated extensively (Ma et al., 2002; Townshend, 1990). In alkaline media, luminol can be oxidized by oxidation reagent (e.g. potassium ferricyanide, potassium dichromate) to generate CL emission, which can be enhanced or inhibited by some redox additives into the alkaline luminol solution (Ma et al., 2002; Townshend, 1990). Due to functional groups in gallic acid, it is promising to be oxidized easily and to inhibit the CL emission of the alkaline luminol system. Herein we attempted to develop a CL system for the determination of gallic acid with the help of flow injection analysis. The luminol–potassium permanganate system in the basic media has been adopted for the determination of paracetamol (Easwaramoorthy, Yu, & Huang, 2001) and doxycycline (Li, Duan, Chen, & Chen, 2004) in the literature. The acidic potassium permanganate was chosen as the oxidation reagent adding into the alkaline luminol system because the acidic potassium permanganate solution has advantages of stronger oxidation ability than alkaline one, high stability and low environmental pollution and cheap cost. In this paper we reported the inhibited CL emission of the alkaline luminol–$KMnO_4$ system by gallic acid, the optimum of experimental conditions for the establishment of FI-CL method for the detection of gallic acid, and the monitoring of gallic acid in olive fruits.

2. Experimental

2.1. Apparatus

The IFFL-D FI-CL system (Xi'an Ruike Electronic Ltd. Corporation, China) illustrated in our previous report (Wang, Wang, & Yang, 2007) consists of two peristaltic pumps and an eight-way injection valve. A PTFE tube (i.d. 0.75 mm) is used to connect all components in the flow system. The sample is injected into a carrier stream of redistilled water and the alkaline luminol solution *via* the eight-way injection valve. Three Y-shaped mixing elements positioned just after the eight-way valve are used for mixing the streams. The CL emission is amplified by a sensitive photomultiplier tube (PMT) operating at 750 V and measured with a detector under the control of a computer. The CL signal is collected using a home-made software.

A KQ-50 ultrasonic bath (Kunsan Ultrasonic Instrument Ltd. Corporation, China) was used to prepare real samples from olive fruits extracts.

2.2. Chemicals and solutions

Luminol (Fluka, America), gallic acid (Fluka, America), NaOH (Shanghai Chem. Co., China), potassium permanganate ($KMnO_4$) (Shanghai Chem. Co., China), sulfuric acid (H_2SO_4) (Shanghai Chem. Co., China), Sodium hydroxide (NaOH) (Shanghai Chem. Co., China) were used as received without further purification. All other chemicals were of analytical reagent grade. The redistilled water was used throughout the experiment.

The alkaline stock solution of luminol (1.0 mM) in 0.1 M NaOH was prepared from purchased luminol. The stock solution of gallic acid (0.1 mg ml^{-1}) was prepared by dissolving 0.0050 g gallic acid in the mixture of methanol and water with a volume ratio of 1:1. The stock solution of potassium permanganate (5.0 mM) was prepared by dissolving 0.0790 g potassium permanganate into a 100 ml flask with twice distilled water. The stock solution of sulfuric acid (0.1 M) was obtained by dilution of the concentrated sulfuric acid. The NaOH stock solution (2.0 M) was obtained by dissolving 8.0000 g NaOH in 100 ml redistilled water.

2.3. Procedure

The solutions of luminol, redistilled water, potassium permanganate, sulfuric acid were pumped continuously at 0.5, 0.5, 0.5, 0.5 ml min^{-1} into the mixing element by the peristaltic pump, respectively. The sample of gallic acid

was injected into the mixed stream of redistilled water and luminol by a 60 μl valve injector. Then the stream was merged into the mixed stream of potassium permanganate and sulfuric acid. The final stream was introduced into the flow CL cell and the full CL signal as a function of time was recorded automatically.

2.4. Determination of gallic acid in olive fruits

The olive fruit powder (1.0000 g) grounded from the frozen and dried olive fruits flesh with a mortar was extracted with a 60 ml 80% (v/v) methanol aqueous solution in an ultrasonic bath for 1 h and the precipitate was filtered. The supernatant was then diluted 500 times with redistilled water and the resulting solution was employed as sample solution for the monitoring of gallic acid in olive fruits.

3. Results and discussion

The CL of the alkaline luminol–$KMnO_4$ system was inspected using a static system. A series of experiments were then conducted for the optimum of experimental conditions including chemical variables (e.g. the concentration of luminol, of NaOH, of $KMnO_4$, of sulfuric acid) and physical ones (e.g. total flow rate, and sample loop volume).

Fig. 1 shows the dynamical profile of CL emission of the alkaline luminol–$KMnO_4$ system (baseline) and of inhibited CL emission of the alkaline luminol–$KMnO_4$ system by gallic acid (curve (a), (b), and (c)). The CL emission of the alkaline luminol–$KMnO_4$ system was very strong and stable, an indication of a fast CL reaction of the alkaline luminol with acidic potassium permanganate. However, after injection of gallic acid into the luminol–$KMnO_4$ mixed solution, the CL intensity (curve (a), (b), and (c)) decreased remarkably and reached the minimum after 12 s where negative peaks appeared and then returned to the baseline again after 18 s. Moreover, comparison of curve (a), (b), and (c) shows us that higher concentration of gallic acid led to stronger inhibited effect on the CL intensity. These facts indicate that gallic acid is an inhibiter towards the CL of the alkaline luminol–$KMnO_4$ system. It is well-known that generating CL emission in the alkaline luminol–oxidation reagent systems generally involves (i) reaction of oxidation reagent with luminol; (ii) formation of luminol free radical directly or indirectly and further oxidation of the formed luminol peroxide; (iii) decomposition of luminol peroxide into excited 3-aminophthalate and CL release of the excited of 3-aminophthalate (Townshend, 1990; Ma et al., 2002). After delivering gallic acid as a reduction additive into the luminol–$KMnO_4$ system, fewer luminol will react with oxidation reagent (acidic $KMnO_4$) since it also has to oxide gallic acid added and the amount of generated luminol peroxide which emits CL subsequently decreases. In other words, the inhibited CL emission in our system actually results from the decreased excited 3-aminophthalate caused by the reaction of gallic acid with acidic $KMnO_4$. Similar results in alkaline media are also reported in references (Easwaramoorthy et al., 2001; Li et al., 2004).

Since the inhibited CL emission of the alkaline luminol–$KMnO_4$ system showed variation with the concentration of gallic acid, the relationship of the inhibited CL intensity with the concentration of gallic acid is possibly to be utilized for the monitoring of gallic acid. In the following section the subtraction of the CL intensity, ΔI ($=I_0 - I$), of the alkaline luminol–$KMnO_4$ system without gallic acid (I_0) from that of the alkaline luminol–$KMnO_4$ system in the presence of gallic acid (I) was chosen as a quantitive parameter for the optimum of experimental conditions and for the setup of the calibration curves for the detection of gallic acid.

Fig. 2 shows the variation of ΔI against the concentration of luminol in the range from 50 μM to 1.0 mM. ΔI was comparatively weak when the concentration of luminol was lower than 50 μM. Increase in the concentration

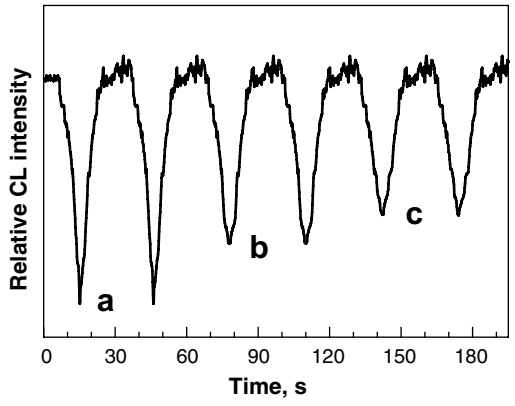

Fig. 1. The FI-CL intensity, I, of the luminol–$KMnO_4$ system as a function of time in the presence of gallic aicd with concentration of (a) 2.0 μg ml^{-1} (b) 1.0 μg ml^{-1} (c) 0.5 μg ml^{-1}. The concentration of luminol, $KMnO_4$ and H_2SO_4 were 0.25 mM, 50 μM, and 2.5 mM, respectively and the pH value of the alkaline luminol solution was 13.0.

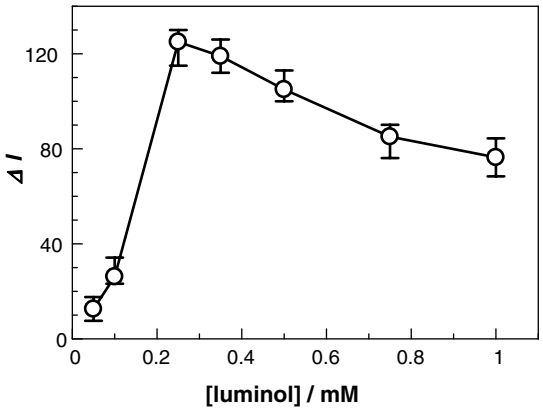

Fig. 2. Concentration dependence of luminol, [luminol], on the subtracted CL intensity, ΔI. The concentrations of $KMnO_4$, H_2SO_4, and gallic acid were 50 μM, 2.5 mM, and 1.0 μg ml^{-1}, respectively and the pH value of the alkaline luminol solution was 13.0.

of luminol resulted in enhancement of ΔI and ΔI reached the maximum value when the concentration of luminol was 0.25 mM. Larger concentration of luminol than 0.25 mM, on the contrary led to a decrease in ΔI. The concentration of luminol was then decided to be 0.25 mM in the following experiments.

It is well-known that the CL intensity of luminol system is affected by the pH values of the solutions that much (Ma et al., 2002; Townshend, 1990), the influence of pH values of the solution on ΔI was thus tested in the range from 11.3 to 14 and the corresponding result was shown in Fig. 3. The pH values of alkaline luminol solution were adjusted with 1.0 M NaOH solution. ΔI increased with an increase in pH value from 11.3 to 13 and then decreased when pH value was beyond 13. Therefore, the pH value of the luminol solution was adjusted to be 13.

Fig. 4 shows the variation of ΔI as a function of the concentration of potassium permanganate in the range from 10 μM to 0.1 mM. The maximum value of ΔI was approached when the concentration of potassium permanganate solution was 50 μM. Larger or smaller concentration of potassium permanganate than 50 μM led to weaker ΔI. This variation of CL intensity with the concentration of potassium permanganate presumably results from the molar ratio of the reaction of luminol with potassium permanganate (Ma et al., 2002; Townshend, 1990). Therefore, the concentration of potassium permanganate was selected as 50 μM.

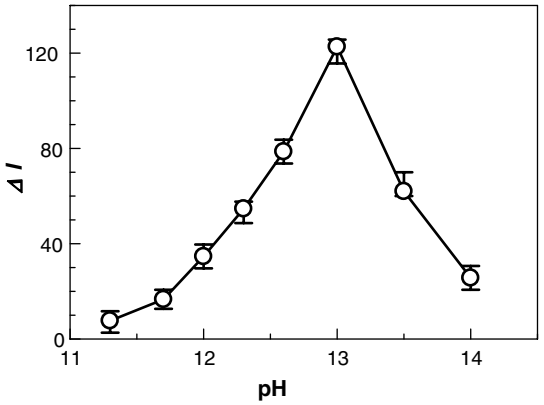

Fig. 3. The variation of pH values of luminol solution with the subtracted CL intensity, ΔI. The concentration of luminol, $KMnO_4$, H_2SO_4 and gallic acid were 0.25 mM, 50 μM, 2.5 mM, and 1.0 μg ml^{-1}, respectively.

Various type and different concentration of the acidic media were used to investigate their effect on ΔI. The investigated media covered HCl, H_2SO_4, HNO_3, and H_3PO_4. The values of ΔI obtained in H_2SO_4 were bigger than those in other solutions. The investigated concentration of sulfuric acid was in the range from 1.0 to 10 mM. Enhancement of ΔI was noticed when the concentration of H_2SO_4 increased from 1.0 to 2.5 mM. When the concentration of H_2SO_4 was larger than 2.5 mM, ΔI decreased. Similarly with the variation of CL intensity with the concentration of potassium permanganate, the alteration of CL intensity with the concentration of sulfuric acid supports the effect of the molar ratio of the reaction reagent on the CL intensity in our system (Ma et al., 2002; Townshend, 1990). Therefore 2.5 mM H_2SO_4 solution was chosen.

The instrumental parameters such as the total flow rate and the volume of the sample loop were studied using the selected reaction conditions discussed above. The total flow rate was divided into the four flow streams. ΔI increased rapidly with increasing total flow rate up to 2.0 ml min^{-1} and increased at a slower rate up to 4.2 ml min^{-1}. However, the noise was excessive at total flow rate higher than 2.4 ml min^{-1}. A total flow rate of 2.0 ml min^{-1} was selected as a compromise of signal to noise ratio, sensitivity, and consumption of reagents. In the range from 30 to 90 μl, ΔI increased with an increase in the loop volumes to 60 μl, and then the signal became unstable and decreased with larger loop volumes, as illustrated in Fig. 5. Thus a loop volume of 60 μl was adopted.

Under the optimum experimental conditions mentioned above, the working curve for the detection of gallic acid was plotted by use of ΔI as a function of the concentration

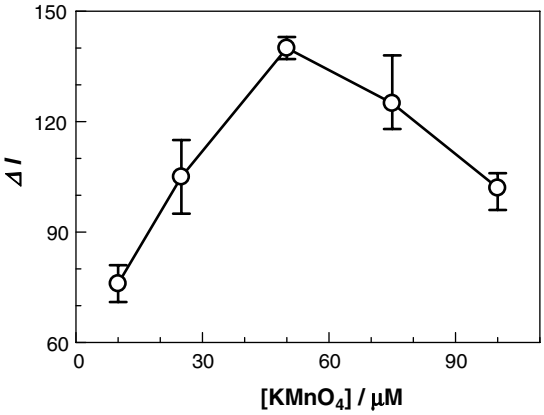

Fig. 4. Concentration effect of potassium permanganate, [$KMnO_4$], on the subtracted CL intensity, ΔI. The concentration of luminol, H_2SO_4 and gallic acid were 0.25 mM, 2.5 mM, and 1.0 μg ml^{-1}, respectively and the pH value of the alkaline luminol solution was 13.0.

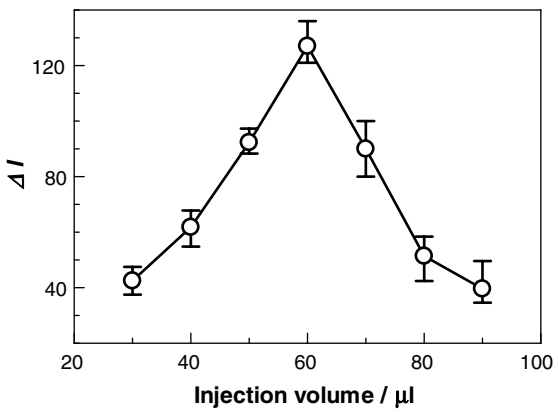

Fig. 5. Effect of injection volume on the subtracted CL intensity, ΔI. The concentrations of luminol, $KMnO_4$, H_2SO_4, and gallic acid were 0.25 mM, 50 μM, 2.5 mM, and 1.0 μg ml^{-1}, respectively and the pH value of the alkaline luminol solution was 13.0.

Table 1
Determination results of gallic acid in olive fruits

Sample	Found ($\mu g\ ml^{-1}$)		RSD (%)	Added ($10^{-7}\ g\ ml^{-1}$)	Recovered ($10^{-7}\ g\ ml^{-1}$)	Recovery (%)
	Proposed method	Spectrophotometry				
1	1.32	1.31	1.3	1.30	1.34	103.1
2	1.29	1.28	0.8	1.30	1.35	103.8
3	1.35	1.33	1.4	1.30	1.27	97.7
4	1.33	1.32	2.1	1.30	1.23	94.6

of gallic acid. The logarithm of ΔI varied linearly with the logarithm of concentration of gallic acid (c) in the range from 1.0×10^{-9} to $5.0 \times 10^{-5}\ g\ ml^{-1}$ with a regression equation of $\log \Delta I = 6.9318 + 0.71953 \log c$. The detection limit was $2.2 \times 10^{-10}\ g\ ml^{-1}$ calculated from the IUPAC recommendations (3σ). The average value of relative standard deviations (RSD) for 11 injections with $1.0\ \mu g\ ml^{-1}$ gallic acid was 1.7%. The sample throughput was 120 injections h^{-1}. The reproducibility was studied by analyzing 10 identical solutions of gallic acid solution ($1.0\ \mu g\ ml^{-1}$) on the sequential days and each day five injections were detected. The average value of RSDs for these detections was 2.3%.

The effect of foreign species on the determination of $1.0\ \mu g\ ml^{-1}$ gallic acid was studied. No effect was noticed when the mass concentration ratios of the foreign species to the sample were more than 1000 for K^+, Na^+, NH_4^+, Ca^{2+}, Cl^-, glucose, sorbinose, ethanol, methanol, glycol, isopropanol, 1-butanol, acetone, 500 for citric acid, benzene carboxylic acid, PO_4^{3-}, 200 for nitrophenol, 2,4-dinitrophenol, salicylic acid, ascorbic acid, 100 for tartaric acid, 50 for phenol, 5-sulfosalicylic acid, oxalic acid, 20 for Hg^{2+}, Fe^{3+}, 10 for Cd^{2+}, Zn^{2+}, 2 for pyrogallol, 1 for resorcinol, 0.5 for Mn^{2+}, 0.2 for tannic acid, phloroglucinol, respectively.

The proposed method was utilized for the determination of gallic acid in olive fruits which has been treated as described in experimental section. Table 1 shows the detection results compared with those obtained *via* spectrophotometry. The average content of gallic acid was $1.32 \times 10^{-7}\ g\ ml^{-1}$, which is in agreement with that obtained from spectrophotometry and the RSDs for these determinations were in the range from 0.8% to 2.1%. Recovery experiments were also performed by adding $0.13\ \mu g\ ml^{-1}$ into the sample solutions and the recovery was in the range from 94.6% to 103.8%. The RSD of the detection of these samples and the recovery experiment indicate that this FI-CL method is sensitive and accurate for the concentration detection of gallic acid in fruit samples. Comparison of this method with the published methods (Ali et al., 1988; Arce et al., 1998; Bianco & Savolainen, 1997; Dmitrienko et al., 2002; Goto et al., 1996; Malovana et al., 2001; Ng et al., 2000; Rodriguez-Delgado et al., 2001; Shahrzad & Bitsch, 1996, 1998; Thies & Fischer, 1973; Tian et al., 2000; Zhu et al., 2005), the main advantage of the FI-CL method proposed here is that it is more economic and more environment friendly for the monitoring of gallic acid.

4. Conclusion

Acidic potassium permanganate was adopted as oxidation reagent to the alkaline luminol system to generate chemiluminesence emission. The resulting chemiluminesence emission can be inhibited by gallic acid. The logarithm of the subtractions of the chemiluminesence intensity of the alkaline luminol and the acidic potassium permanganate system without gallic acid from that in the presence of gallic acid were linear with the logarithm of the concentration of gallic acid in the range from 1.0×10^{-9} to $5.0 \times 10^{-5}\ g\ ml^{-1}$ with a detection limit of $2.2 \times 10^{-10}\ g\ ml^{-1}$. Based on this calibration curve, the content of gallic acid in olive fruits was determined successfully. This proposed method is rapid, sensitive, economic, environment friendly and has the potential to be adopted as an official quantitative method for the monitoring of gallic acid in food, wood, and urine in future.

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