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Rapid determination of formaldehyde and sulfur dioxide in food products and Chinese herbals

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Abstract

Simple, sensitive and rapid methods for the determination of formaldehyde and sulfur dioxide were developed. The formaldehyde determination is based on the reaction between formaldehyde and acetylacetone solution, producing yellow 3,5-diacetyl-l-1,4-dihydrolutidine. Sulfur dioxide was detected as the deoxidize of sulfurous acid by zinc in acidic medium, which produces sulfureted hydrogen that make lead acetate paper blackening due to lead sulfide formation. The detection limits were 0.8 μg mL^{-1} and 6.0 μg mL^{-1} for formaldehyde and sulfur dioxide, respectively. The linear range were 0.8–20.0 μg mL^{-1} for formaldehyde and 6.0–100.0 μg mL^{-1} for sulfur dioxide determination. The main advantages of the new analytical procedure are the low background level, high selectivity, and very little sample preparation for on-site analysis of formaldehyde and sulfur dioxide in food or Chinese herbal samples with reference color card for qualitative or semi-quantitative determination. The results from these methods correlated well with those obtained from the standard methods.

Keywords: Formaldehyde; Sulfur dioxide; Simple device; Rapid; Detection

1. Introduction

Formaldehyde is a flammable, colorless and readily polymerized gas at ambient temperature. The most common commercially available form is a 30–50% aqueous solution (WHO, 1989). Formaldehyde is also the most widespread carbonyl compound. It is sometimes added inappropriately in food processing for its preservative and bleaching effects such as dried foods, vermicelli, tripe and chicken paws, etc. However, this chemical also occurs naturally in the environment and can be found naturally in small amounts in a wide range of raw food, including fruit and vegetables (3.3–17.3 ppm), meat (5.7–20 ppm), milk and milk products (1.0–3.3 ppm), and fish (1.0–98 ppm) (WHO, 1989). The US Environmental Protection Agency (EPA) has established a maximum daily dose reference (RfD) of 0.2 mg/kg body weight per day for formaldehyde. At exposures increasingly greater than the RfD, the potential for adverse health effects increases (EPA, 1999).

Sulfur dioxide is a colorless gas with a pungent, suffocating odor. It is corrosive to organic materials and dissolves in water to form sulfurous acid, H_2SO_3. Sulfur dioxide is used in chemical manufacture as a refrigerant and food preservative. It is very effective in inhibiting the growth of some yeast and in preventing enzymatic browning reaction catalyzed by polyphenol oxidase. The proportion of the solid food supply likely to be treated with sulfites (WHO, 1999). In food processing, sulfur dioxide is widely used for fumigating, preserving, bleaching and steeping. Several Chinese herbs have relatively high levels of sulfur dioxide. This is the result of a processing method whereby herbs are spread on screens, underneath which is some heated sulfur. The sulfur fumes waft through the herb material and leave some residue. Treatment with sulfur is

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mostly carried out on those herbs that are moist or those that discolor significantly over time. However, the sulfur compounds resulting from this method of preserving the herb quality cause reactions in sulfite-sensitive individuals.

Formaldehyde or sulfur dioxide residue in the foods or Chinese herbals pose a threat to human health. The common effects of formaldehyde exposure are various symptoms caused by irritation of the mucosa in the eyes and upper airways (WHO, 1989). Exposure to high concentrations of formaldehyde for short period of time can constrict the bronchi and increase mucous flow, making breathing difficult. Consequently, the development of rapid methods for the determination of formaldehyde and sulfur dioxide in food is of special importance.

Formaldehyde has received a great attention in the past few years due to the toxic activity associated with respiratory tract. Several analytical methods for formaldehyde measurement have been reported such as fourier transform infrared absorption (FTIR), differential optical absorption spectroscopy (DOAS), laser-induced fluorescence spectroscopy (LIFS), tunable diode laser absorption spectroscopy (TDLAS) (Tang, Wang, Sheng, & Fu, 2005), fluorometric determination (Helaleh, Kumemura, Fujii, & Korenaga, 2001), gas chromatography (Dalene, Person, & Skarping, 1992), spectrophotometric detection (Feng, Fan, Wang, Chen, & Hu, 2004), enzymatic methods (Lazrus, Fong, & Lind, 1988), colorimetric methods (Muñoz, de Villena Rueda, & Díez, 1989) and oscilloscopic polarography (Zhang & Sun, 2003). Most frequently formaldehyde is determined by fluorometric, chromatographic and colorimetric methodologies. The fluorometric methods using the Hantzsch reaction are more applicable to trace determination of formaldehyde (Reche, Garrigós, Garrigós, & Jiménez, 2000), The techniques is specific, non-destructive and quantitative and allow the continuous detection, however, the requirement of large, complex, and expensive instrumentation makes the method not suitable for routine applications. Atmospheric levels of formaldehyde have been generated and most of this knowledge comes from measurements of the gas phase, for which sampling systems are impingers containing 2,4 dinitrophenylhydrazine (2,4-DNPH) which could reacted with carbonyls to form the corresponding hydrazone derivates, followed by quantification by gas chromatography (Dalene et al., 1992) or liquid chromatography (Sandner, Dott, & Hollender, 2001). The problem associated with this method is the interference of many carbonyls substances, including acetaldehyde, acetone. Colorimetric methods involve the pararosaniline (Muñoz et al., 1989), chromotropic acid (1,8-dihydroxy naphthalene-3,6-disulphonic acid) (Altshuller, Miller, & Sleva, 1961) and 3-methyl-2-benzothiazolonehydrazone (MBTH) (WHO, 1989). Although pararosaniline-based Schiff reaction has been used widely for formaldehyde determination, color development is relatively slow and sensitivity is limited and sulfur dioxide residue in food will influence the result (Muñoz et al., 1989). Chromotropic acid method usually quite rapid, however, its not only need oil of vitriol as the medium but also was interfered by acetaldehyde (Ahonen, Priha, & Äijälä, 1984). MBTH is expensive and color development is relatively slow and sensitivity is limited (Tang et al., 2005).

Traditional and official methods for sulfur dioxide determination in foods utilise the distillation of the samples under acidic condition and then titration, the methods were time consuming and laborious (P.R.C., 2003). In recent years, some new techniques, such as flow injection (Carballo, Dall'Orto, Balbo, & Rezzano, 2003), gas diffusion membrane (Segundo & Rangel, 2001) as well as a multisyringe flow system for the spectrophotometric determination of sulfur dioxide were reported (Segundo, Rangel, Cladera, & Cerdà, 2000). These methods proved to be more sensitive and selective, but most of them are either time-consuming or require rather expensive instrumentations, which prevent them from being applied widely in rapid determination of sulfur dioxide in food samples. Moreover, these methods require analytical skills and unsuitable for on-site determination of sulfur dioxide.

Although there are various methods mentioned for the determination of formaldehyde and sulfur dioxide, most methods require expensive apparatus or time-consuming. Therefore, it is necessary to establish a simple, rapid, inexpensive, specific, and sensitive analytical method for on-site analysis. This paper reported the development of new rapid methods for the determination of formaldehyde and sulfur dioxide without special skills or standard solutions. The formaldehyde determination is based on the reaction between formaldehyde and acetylacetone solution, producing yellow 3,5-diacetyl-l-1,4-dihydrolutidine. Sulfur dioxide is detected as the deoxidize to sulfurous acid by zinc in acidic medium, which produces sulfurated hydrogen that makes lead acetate paper darker due to the formation of lead sulfide. With the reference color card, the formaldehyde and sulfur dioxide in the samples could be quickly determined under the field conditions. The results obtained from the developed methods agreed well with those obtained by standard methods. The developed methods can be used as qualitative and semi-quantitative tools for on-site rapid screening of formaldehyde and sulfur dioxide in foods or Chinese herbals samples.

2. Experimental

2.1. Reagents and device

Analytical grade chemicals and doubly distilled water (DDW) were used in all experiments.

An aqueous formaldehyde stock solution, 1000 μg mL^{-1}, was prepared by diluting 2.5 mL of 37% w/v stock formaldehyde solution to 1 L with DDW and was standardized by the thiosulfate–iodide method (Muñoz et al., 1989). Formaldehyde standard solutions (0.8–20.0 μg mL^{-1}) were prepared from the stock solution by appropriate dilution with DDW.

A 1000 μg mL^{-1} stock standard solution of sulfur dioxide was prepared by dissolving 0.5 g of sodium bisulfite in 500 mL water, the stock standard solution was standardized by iodimetric titration (P.R.C., 2003). Sulfur dioxide standard solutions (6.0–100.0 μg mL^{-1}) were prepared by appropriate dilution of the stock solution with DDW.

Acetylacetone solution was prepared by dissolving 15 g of ammonium acetate, 0.3 mL of acetic acid solution, and 0.2 mL of acetylacetone in 100 mL DDW. The detection limit that an analytical procedure may achieve greatly depends on the reagent blank quality. In this way, in order to improve the blank levels, the acetylacetone must be distilled to remove the impurity. This reagent is stable for at least 60 days if stored under refrigeration in the dark.

Absorbance was measured on a Cary 50 UV–Vis spectrophotometer (Varian).

Activated carbon granular (particle size: 0.5 mm; pH 6.0) from BeiFang Regent Plant (Tianjin, China), Activated carbon granular is a highly porous adsorbent material, produced by heating organic matter, such as coal, wood and coconut shell, in the absence of air, which is then crushed into granules. Activated carbon is carbon which has a slight electro-positive charge added to it, making it even more attractive to chemicals and impurities. It is mainly used in decolouring chemical raw materials and able to remove negative ions from the water such as chlorine, fluorides and dissolved organic solutes by absorption onto the activated carbon granular.

The device for the determination of formaldehyde was made with a 18 (diameter) × 75 (length) mm 5 mL injector and the injector was packed with 1 g activated carbon granular.

The device for determination of sulfur dioxide was made up of a 20 (diameter) × 80 (length) mm vitreous tube and the tube was sealed with a cap on which a wet lead acetate paper was suspended. The bottom of the tube was packed with 0.5 g zinc powder (purity: 99%; particle size: 6–9 μm; 100 mesh) from BeiFang Regent Plant (Tianjin, China).

2.2. Sample preparation

A total of samples of foods from different companies were purchased from retail stores in Tianjin, China. The medicinal samples used in this study were purchased from local drug stores (Tianjin, China).

Ten grams of sample was put into a beaker, and 50 mL of DDW was added. The mixture was shaken well and then stayed for 15 min. The supernatant filtered through a filter paper and the filtrate was measured according to the procedures described in the following section.

2.3. Procedure

2.3.1. Determination of formaldehyde

2.3.1.1. The standard acetylacetone method (P.R.C., 2002). The original acetylacetone solution was prepared by mixing of ammonium acetate (25 g), acetic acid (3.0 mL) and acetylacetone (0.4 mL). The volume was then completed to 100 mL with DDW.

For the determination of formaldehyde in food samples, the samples were accurately weighed (for solid foods) or pipetted (for liquid foods) into a bottle and soaked in water. The free formaldehyde was separated by steam distillation collecting 100 mL of distillate. Then 1 mL of acetylacetone solution was added to 5 mL of distillate. After mixing, the absorbance was measured at 435 nm with the reagent of acetylacetone solution as the reference.

2.3.1.2. The proposed rapid method. It would be essential to undertake a standard test whenever samples are to be analysed, under identical conditions for semi-quantitative measurements.

To establish a standard reference color card, a series of standard solutions at concentrations of 0.8, 2.0, 5.0, 7.0, 9.0, 15.0, 20.0 μg mL^{-1} were prepared. Then 2 mL of acetylacetone solution was added to 2 mL of standard solutions in a tube and the mixture was allowed to reacted for 6 min. A standard reference color card was prepared in correspondence to the concentration levels (Fig. 1a). The result formed a yellowish to yellow color in the concentration range of 0.8–20.0 μg mL^{-1}. Color intensity of concentrations exceeding 20.0 μg mL^{-1} did not facilitate visual comparison of formaldehyde concentrations.

For determination of formaldehyde in the samples, 2 mL of acetylacetone solution was added to 2 mL of real sample solution in the tube and shaken well. For semi-quantitative detection of formaldehyde levels, the color of the final solution was compared to the standard reference color card after 6 min.

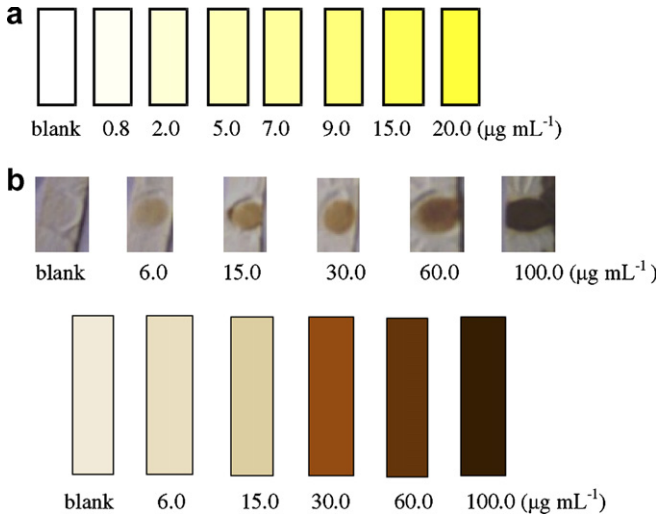

Fig. 1. (a) Reference color card for formaldehyde. (b) The result of determination of sulfur dioxide and the reference color card of sulfur dioxide.

The simple device was packed with active carbon granular in order to prevent the color interference of sample solution in the experiment. Therefore, the sample aqueous solution was filtrated through the device if the sample solution was colorful.

2.3.2. Determination of sulfur dioxide

It would be essential to undertake a standard test whenever samples are to be analysed, under identical conditions for semi-quantitative measurements.

To establish a standard reference color card, after 0.5 g zinc powder was placed in the bottom of the tube, 10 mL standard solutions and 2 mL of 7.0 mol L^{-1} hydrochloric acid were added. The tube was sealed with a cap on which a wet lead acetate paper was suspended. The mixture was allowed to react for 3 min. A standard reference color card was prepared in correspondence to the concentration levels (6.0, 15.0, 30.0, 60.0, 100.0 μg mL^{-1}) (Fig. 1b). The result formed a light-brown to brown color in the concentration range of 6.0–100.0 μg mL^{-1}. Color intensity of concentrations exceeding 100.0 μg mL^{-1} did not facilitate visual comparison of sulfur dioxide concentrations.

To detect the sulfur dioxide concentration in samples, 10 mL of sample filtrate and 2 mL of hydrochloric acid were added to the tube in which 0.5 g zinc powder was added previously. The samples were mixed well and sealed with the cap quickly. For semi-quantitative detection of sulfur dioxide levels, the color of the final solution was compared to the standard reference color card after 3 min.

2.3.2.1. Recovery study of the developed method. This method has been used to analyze acanthopanax bark (*Cortex acanthopanacis*) and liquorice (*Glycyrrhiza uralensis*) sample, which spiked three different levels, that is 6 μg mL^{-1}, 15 μg mL^{-1}, 30 μg mL^{-1}. At each concentration, three measurements were performed. Then the sulfur dioxide content was measured by the developed procedure. As it was observed in the Fig. 2, the color of the sample of acanthopanax bark (below LOD) after spiked three levels was similar to the not spiked color and the color of the sample liquorice (6.0 μg mL^{-1}) was in the range of 10–15, 20–25, 30–35 μg mL^{-1}, respectively after spiked 6 μg mL^{-1}, 15 μg mL^{-1}, 30 μg mL^{-1}. Therefore, the results showed that the recoveries for all the investigated samples were satisfactory by the developed method.

3. Results and discussion

3.1. Formaldehyde

3.1.1. Influence of concentration of acetylacetone on determination of formaldehyde

Acetylacetone was changed to brown when exposed to light. In traditional method, the result was not affected when the solution was changed due to the absorbance being measured with the reagent of acetylacetone solution (0.4%) as the reference. However, the method developed in this study was observed visually. For this reason, a method was needed to decrease background color and improve the sensitivity of the test.

The absorbance of background was measured at 435 nm with DDW as the reference at room temperature. The curve of the effect of concentration of acetylacetone was shown in Fig. 3. It could be seen that the background absorbance was lower than 0.05 when the volume of acetylacetone from 0.1 to 0.2 mL and the background level increase with the volume of acetylacetone over 0.2 mL. When the absorbance lower than 0.1, the difference of the color development was undistinguishable by eyes. At the same time, the detection limit for the rapid method was 0.8 μg mL^{-1} as the concentration of acetylacetone at 0.2%. Therefore, 0.2% acetylacetone was chosen for the analysis.

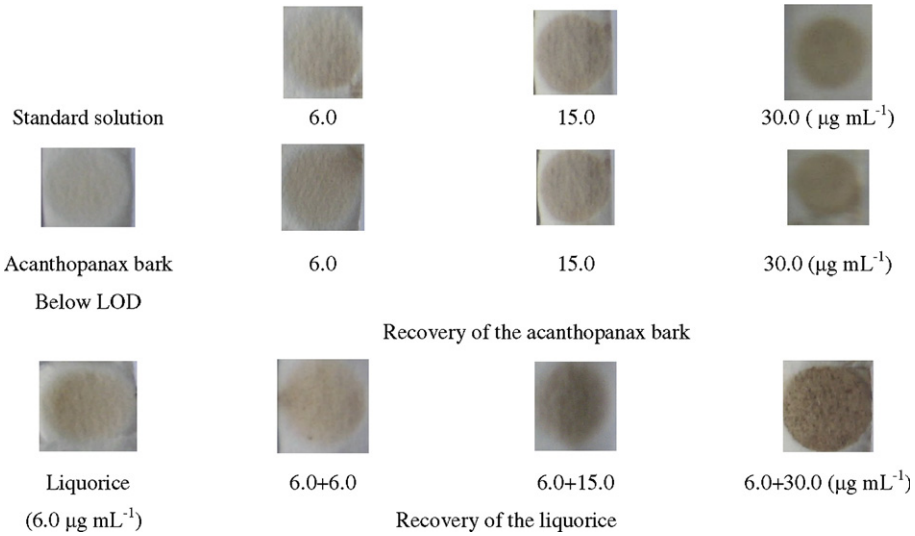

Fig. 2. The recovery of sample on determination of sulfur dioxide.

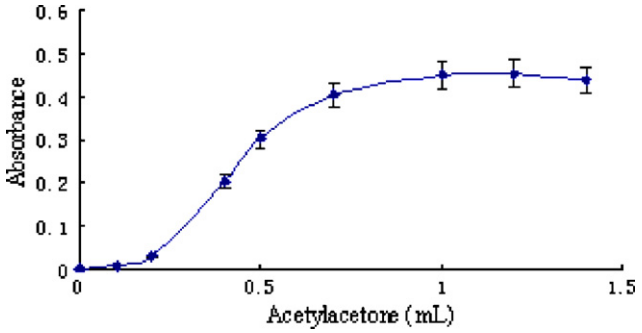

Fig. 3. Effect of concentration of acetylacetone on the determination of formaldehyde. The values are means for triplicate determinations; the error bars indicate standard deviations.

3.1.2. Effect of temperature on color development

The effect of temperature on the absorbance was checked in the range of 10–30 °C. The experiments were conducted under the different temperatures with the same concentration of formaldehyde (0.8 μg mL^{-1}). The result demonstrated that the absorbance lower than 0.1 when the temperature in the range of 10–15 °C, while the temperature in the range of 15–30 °C, the absorbance increased dramatically. Thus, the method should be carried out at temperature greater than 15 °C.

It was also found the reaction rate was temperature-dependent. At 15 °C, it required 6 min to reach the best effect. It only needed 5 min at 25 °C, while at 30 °C, only 4 min was required.

3.1.3. Interference study and stability of the acetylacetone solution

The effect of various potential interferences concurrently present in the sample on the determination of formaldehyde was investigated. The effect of other carbonyl compounds such as glyoxal, benzophenone, acetone, and phenol do not interfere under the conditions employed in this study even at 10,000-fold higher concentration than formaldehyde concentration (20 μg mL^{-1}). Other carbohydrates such as glucose, fructose and sucrose do not interfere under the conditions given. This result indicated that the developed method was specific for formaldehyde.

The stability of the acetylacetone solution was also studied. It was found the solution was stable for 60 days if stored under refrigeration in the dark. Calibration curves were measured every week with the freshly-made formaldehyde working solution.

3.1.4. Effect of activated carbon granular on determination of formaldehyde

It was reported that active carbon could absorb the formaldehyde (Zhang, Fang, & Song, 2003). In order to test the effect of adsorption, two methods were studied in this paper. The first one was to pass 0.8 μg mL^{-1} formaldehyde standard solution through the simple device which packed 1 g active carbon powder at 15 °C. The second method was to test the formaldehyde without passing through the device. The results demonstrated that the active carbon had no influence on the determination of formaldehyde.

3.1.5. Application to real food samples

The developed method was applied to the determination of formaldehyde in real foods. The results obtained for 12 samples were shown in Table 1. The accuracy of the developed method was evaluated by comparing with the results from the standard photometric method (P.R.C., 2002). It could be seen that the results from the developed method correlated well with the ones from the standard method. Therefore, the results demonstrated that the developed method was suitable for the rapid determination of formaldehyde in foods.

3.2. Sulfur dioxide

3.2.1. Effect of acidity

Hydrochloric acid plays an important role in the determination of sulfur dioxide as sulfur dioxide is deoxidized to sulfurous acid by hydrochloric acid. The effect of hydrochloric acid concentration on the determination of sulfur dioxide (30 μg mL^{-1}) was tested. As shown in Fig. 4a, the color of the card became darker with increasing HCl concentration. When hydrochloric acid concentration was over 6.0 mol L^{-1}, the maximum color development was reached. Thus, 7.0 mol L^{-1} hydrochloric acid was applied in the study.

3.2.2. Effect of the zinc powder on the color development

The effect of the weight of zinc powder in the reaction vessel on the determination of sulfur dioxide was also studied. The results showed that with decreasing the amount of zinc powder from 3.0 to 0.5 g, the sensitivity of the developed method did not change (Fig. 4b). Thus, 0.5 g of zinc powder was selected throughout the study.

Table 1
Determination of formaldehyde in real samples

Sample	The result of rapid method (μg mL^{-1})	The result of standard method[a] (μg mL^{-1})
Mushroom	15–20	15.90 ± 0.22
Bambooshoot	>20	25.78 ± 0.13
Sleeve-fish	<0.8	Below LOD[b]
Shrimp	>20	100.10 ± 0.42
Sea cucumber	7–9	5.52 ± 0.11
Cuttlefish	>20	19.03 ± 0.31
Jellyfish	<0.8	Below LOD
Melon seeds	>20	27.34 ± 0.10
Flour	>20	33.46 ± 0.10
Bean curd	2–5	5.12 ± 0.21
Vermicelli	<0.8	Below LOD
Confect	<0.8	Below LOD

[a] Values are expressed as means (standard deviation) of five determinations.
[b] The limit of detection.

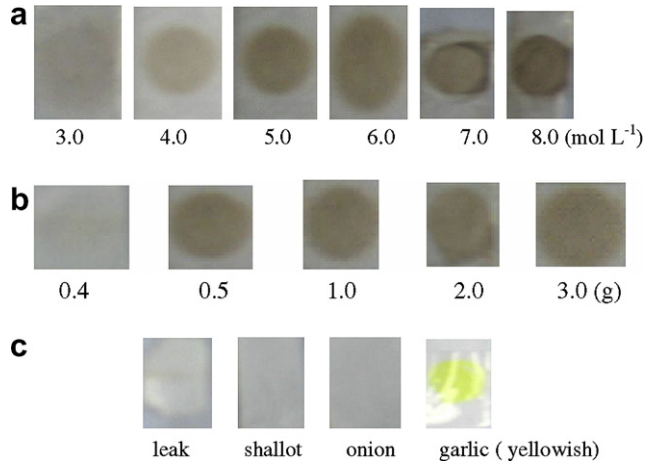

Fig. 4. (a) Effect of concentration of hydrochloric acid on determination of sulfur dioxide. (b) Effect of concentration of zinc powder on determination of sulfur dioxide. (c) The effect of matrices on determination of sulfur dioxide.

3.2.3. Matrix study

The effect of potential interferences in the samples containing rich sulphur matrices such as leek, shallot, garlic, onion was investigated for the developed method. The results shown in Fig. 4c indicated that no matrix effect was observed in leek, shallot and onion samples, however, the color of garlic was yellowish. To evaluate the usefulness of the developed method for garlic, the real sample spiked at level of 6.0 μg mL^{-1} was tested, the result demonstrated the yellowish color did not interfere the determination. Therefore, the effect of the yellowish color can be negligible and the proposed method was applied successfully to the determination garlic without any pretreatment. It could be concluded that the developed method was applicable to these samples.

It was also found that carbohydrates such as glucose, fructose and sucrose starch did not interfere the determination of sulfur dioxide in this study. Therefore, the developed method was specific for the sulfur dioxide determination.

3.2.4. Application to real samples

The samples were treated as described in the section of sample preparation. The concentrations of sulfur dioxide in food samples were determined by the developed methods and then compared to the results obtained by the standard iodin titration method (P.R.C., 2003). The developed rapid method was also applied to herbal samples, including liquorice (*Glycyrrhiza uralensis*), white peony, acanthopanax bark (*cortex acanthopanacis*), ophiopogon root (*radix ophiopogonis*) etc., the results were shown in Table 2. All of these investigated samples were among the typical food that might be processed with sulfur dioxide. The results from the developed method were in good agreement with the results of the standard method.

4. Conclusions

The methods developed in this paper are simple and reliable for the determination of formaldehyde and sulfur dioxide. With a reference color card, the developed methods can be used as a qualitative or semi-quantitative method for on-site analysis of samples containing formaldehyde and sulfur dioxide. The developed method as a new field screening tools was used to test samples at the point of sale for the supervisor of the government or the consumer. Moreover, the present method is sensitive, time saving, require no complicated instruments and consumes a small amount of reagent, and the reagent itself is of low toxicity. This method is applied with success to the analysis

Table 2
Determination of sulfur dioxide in real samples

Sample	The result of rapid method (μg mL^{-1})	The result of standard method[a] (μg mL^{-1})
Citron Daylily (*Hemerocallis citrina* Baroni)	⩾100	15900 ± 0.20
Lotus Seed (*Semen nelumbinis*)	15–30	20.89 ± 0.13
Gelly fungi (*Tremella fuciformis* Berk.)	⩾100	180.45 ± 0.14
Winter mushroom (*Flammulina velutipes*)	6–15	11.76 ± 0.21
Liquorice (*Glycyrrhiza uralensis*)	6–15	6.0 ± 0.11
Cashew (*Anacardium occidentale* L.)	<6	Below LOD[b]
Vermicelli	<6	Below LOD
Confect	<6	Below LOD
White sugar	6–15	12.65 ± 0.23
Bambooshoot	⩾100	93.36 ± 0.15
Mushroom	30–60	48.97 ± 0.10
Whitepeony (*Paeonia lactiflora* Pall.)	60–100	70.54 ± 0.28
Acanthopanax bark (*Cortex acanthopanacis*)	<6	Below LOD
Ophiopogon root (*Radix ophiopogonis*)	15–30	12.38 ± 0.32
Haw (*Aloe vera* L. var. *chinensis*)	30–60	38.90 ± 0.13
Panax	⩾100	355.64 ± 0.25
Peppermint (*Mentha x piperita* L.)	<6	Below LOD

[a] Values are expressed as means (standard deviation) of five determinations.
[b] The limit of detection.

of formaldehyde or sulfur dioxide in the foods or Chinese herbals and the results agree well with those obtained by the standard method.

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