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# Determination of Nine Preservatives in Food Samples by Solid Phase Extraction coupled with Capillary Electrophoresis

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An efficient method for sample treatment with solid phase extraction (SPE) and separation of components by capillary electrophoresis (CE) for simultaneous determination of nine preservatives in food samples was established. During the SPE, the effects of sample solution pH, extraction time, extraction temperature and salt dosage on extraction efficiency of nine preservatives were studied. The optimized parameters of SPE with 0.2 g hollow fiber in 10 mL sample solution were determined as follows: sample solution pH 5.5, extraction time 35 min, extraction temperature 45℃ and NaCl 1.5 g. During the CE separation, the effects of pH and ionic strength of buffer solution, additional additives and separation voltage on the separation efficiency of eight pair preservatives were investigated. The selected CE separation solution is phosphate buffer solution (pH=6.5) containing 45 mmol/L NaCl and 25 mmol/L cyclodextrin, and separation voltage is 17 kV. All of nine preservatives have good linear relationship. Their detection limits were 0.5 - 1.0 μg/kg. The recoveries of four food samples were 83.2% - 116.7%. The sensitive and accurate method can be used to determine preservatives in food samples rapidly.

**Keywords:** Preservative, Food sample, Solid phase extraction, Capillary electrophoresis

## 1. INTRODUCTION

Preservatives are a kind of food additives that can inhibit microbial activities and prevent food spoilage. During the production, storage and transportation of fruit juice, jam, pastry, soy sauce and sausage, there will inevitably be microbial invasion and reproduction. Food without preservatives is very easy to deteriorate. People eating deteriorated food will cause food poisoning, various gastrointestinal diseases and even death [1]. In order to make food have a certain shelf life, we must take certain measures to prevent microbial infection and reproduction. It has been proved that adding preservatives is one of the most economical, effective and simple ways to extend the shelf life of foods

[2-4]. Therefore, preservatives within the safe range are essential elements in many foods, which can effectively inhibit the growth of bacteria in food.

The commonly used food preservatives include benzoic acid (BA), sorbic acid (SA) [5], dehydroacetic acid (DHAA) [6], p-hydroxybenzoic acid esters [7], such as methyl ester (HBAME), ethyl ester (HBAEE) and propyl ester (HBAPE), and biological preservatives [8], such as natamycin (NMC), nisin (NS) and polylysine (PLS). The scope of use, quality standard and dosage of these preservatives are strictly regulated. The normal dose of preservatives in food will not cause any harm to human body. However, it is a pity and worry that many food production enterprises violate the rules, abuse food preservatives. Long term excessive intake of preservatives will cause certain damage to human health [9-12]. Taking BA, a widely used food preservative, as an example, BA and its sodium salt accumulation poisoning have been reported [13,14]. The European Community child protection group believes that it should not be used in children's food. Even as one of the internationally recognized safety preservatives, SA and potassium sorbate can affect the balance of human metabolism [15]. Therefore, the detection and monitoring of preservatives in food is of great significance.

The detection methods of food preservatives mainly include gas chromatography (GC) [16,17], GC- mass spectrometry (MS) [18], surface-enhanced raman spectroscopy [19], liquid chromatography (LC) [20,21], LC-MS [22], LC-ultraviolet (UV) [23,24], spectrophotometry [25] and luminescence sensing [26]. With the increasing coverage of food safety monitoring, the analysis task of food inspection is becoming more and more important. In the face of the requirements of mass samples and rapid analysis, the research and establishment of high-throughput detection methods for the simultaneous determination of multiple preservatives in different kinds of food become very important.

Capillary electrophoresis (CE) is a promising high-performance biochemical and medical separation method with short analysis time and less sample consumption. CE separation couple with end-column electrochemiluminescence (ECL) analysis (CE-ECL) have been widely studied and used to analyze various drugs [27–35], antibiotics [36-44] and pesticide residues [45-50] in different foods, pharmaceuticals, animals and plants. Most preservatives do not contain amino groups, which could not enhance chemiluminescence of ruthenium pyridine, so they cloud not be determined by ECL method. But most preservatives have UV absorption. It is a good attempt to separate and analyze them with CE-UV.

Sample pretreatment technology has great influence on the sensitivity, efficiency and reliability of analytical methods. Solid phase extraction (SPE) is an effective sample pretreatment technology, which integrates sampling, extraction and concentration, greatly speeding up the analysis and detection. Its significant technical advantages have been widely concerned by analysts in the environmental [51-53], food [54-56] and pharmaceutical industries [57,58]. In this paper, preservatives remaining in food samples, such as fruit juice, jam, cake, soy sauce and sausage, were purified and enriched by SPE method. Then nine preservatives, such as BA, SA, DHAA, HBAME, HBAEE, HBAPE, NMC, NS and PLS, were separated and detected simultaneously by CE-UV. Among the nine preservatives, BA, SA and DHAA are the most commonly used chemical preservatives. HBAME, HBAEE and HBAPE are p-hydroxybenzoic acid esters with similar structures. NMC, NS and PLS are biological preservatives with less toxicity. The results show that the present method is sensitive and reliable for the simultaneous determination of them in foods.

## 2. EXPERIMENTAL

### *2.1. Materials and drugs*

Standard substances of benzoic acid (BA), sorbic acid (SA), dehydroacetic acid (DHAA), p-hydroxybenzoic acid methyl ester (HBAME), p-hydroxybenzoic acid ethyl ester (HBAEE), p-hydroxybenzoic acid propyl ester (HBAPE), natamycin (NMC), nisin (NS) and polylysine (PLS) were purchased from National Institutes for Food and Drug Control (Beijing, China). Disodium hydrogen phosphate ($Na_2HPO_4$), sodium dihydrogen phosphate ($NaH_2PO_4$), sodium hydroxide (NaOH), hydrochloric acid (HCl), methanol, formic acid, methyl orthosilicate, sodium chloride (NaCl) and cyclodextrin were all of analytical reagent grade and were purchased from Beijing Chemical Factory (Beijing, China). Polypropylene hollow fiber (inner diameter 600 μm and micropore 0.3 μm) was purchased from Tianjin Film Technology Co., Ltd (Tianjin, China).

### *2.2. Apparatus and conditions*

CE-UV was performed on a MPI - B multi-parameter chemiluminescence analysis test system (Xi'an Remex analytical instruments Co., Ltd., Xi'an, China). Capillary (25μm x 40 cm) was rinsed respectively with 0.1 mol/L NaOH solution for 20 min, secondary distilled water for 10 min and running buffer for 15 min before use.

CE conditions: Separation voltage is 17 kV. The separation medium is composed of phosphate buffer solutions (PBS, pH 6.5) containing 45 mmol/L NaCl and 25 mmol/L cyclodextrin.

UV conditions: Detection wavelength is 275 nm.

### *2.3. Solid phase treatment*

The methyl orthosilicate was added into methanol solution, 30% hydrochloric acid was added under stirring condition, aged at room temperature for 24 h, and then the methyl orthosilicate was completely hydrolyzed to silica sol. The polypropylene hollow fiber with a length of 1 cm was completely immersed in silica sol. After ultrasonic vibration at room temperature for 40 min, and hollow fiber was removed from the sol and dried at 100 ℃ for standby.

### *2.4. Solid phase extraction*

Accurately weigh/measure 2.0 g /2 mL of homogeneous food samples such as fruit juice, jam, pastry, soy sauce and sausage, put it in a 50 mL centrifugal tube with cap, add 5 mL water, scroll for 1 min on the vortex oscillator, add 8 mL methanol, scroll for 1 min, place it on the ultrasonic oscillator for 20 min, scroll, put it in the ice water bath. Centrifuge at 5000 r/min for 5 min. Take 9 ml of supernatant intoanother 15 ml centrifuge tube with cap, add 1.5 g NaCl, vortex to make NaCl completely dissolved, adjust pH to 5.5 with 0.1 mol/L NaOH or 0.1 mol/L HCl, add distilled water to make the volume of sample solution 10 ml, raise the temperature to 45 ℃, completely immerse 0.2 g

hollow fiber in the solution, and extract by ultrasonic for 35 min. After the extraction, transfer the hollow fiber to a test tube, add 2.0 mL methanol (containing 15% acetonitrile) as eluate, and shake it with ultrasonic for 5 min. Blow dry the solution with nitrogen, add 0.5 mL methanol water solution (1:1) along the pipe wall to dissolve the analytes. After passing through 0.22 μm microporous membrane, the filtrate was ready for use.

## 3. RESULTS AND DISCUSSION

### *3.1. Optimization of SPE conditions*

SPE conditions, such as pH of sample solution, extraction time, extraction temperature and salt dosage, have significant influence on the collection of target analytes. In this part, the recoveries of nine preservatives were used as test indexes to study the influence of SPE conditions.

#### *3.1.1 pH of sample solution*

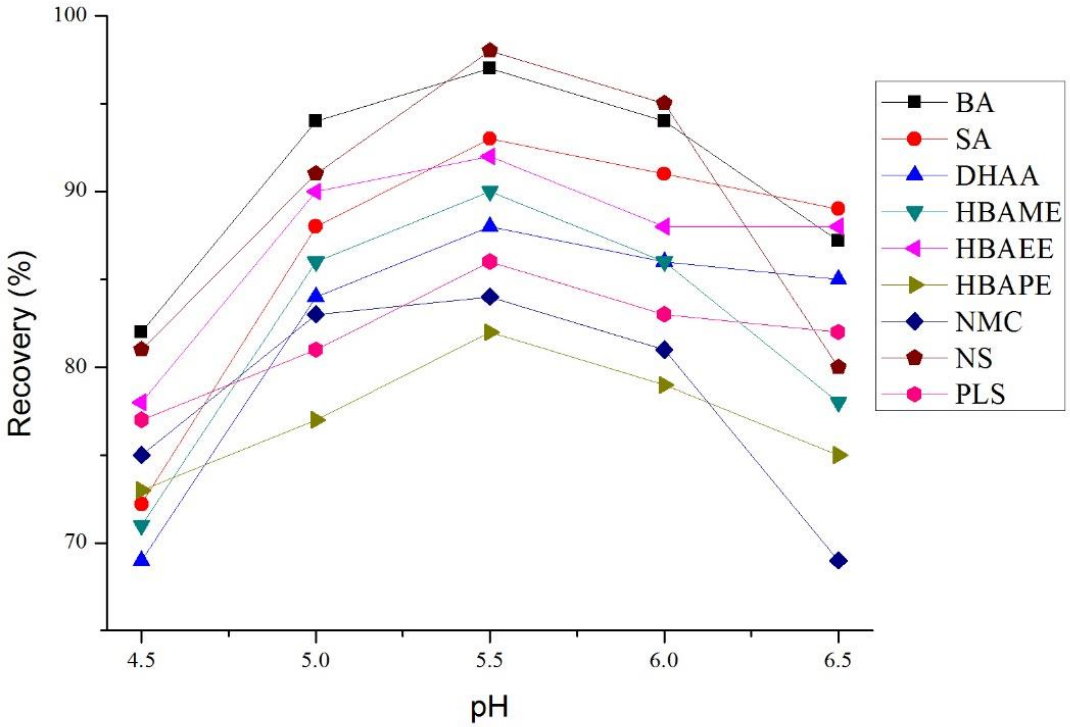


**Figure 1.** Effects of pH of 0.2 g hollow fiber in 10 mL sample solution on the recoveries of nine preservatives under extraction time 35 min, extraction temperature 45℃ and NaCl 1.5 g.

The effects of pH of the sample solution at 4.5, 5.0, 5.5, 6.0 and 6.5 on the recoveries of nine preservatives were investigated. The results in Figure 1 showed that the recoveries of nine

preservatives increased with the increase of pH value of sample solution from 4.5 to 5.5. When the pH of the sample solution was 5.5, the recoveries of nine preservatives almost reached the maximum. The recoveries of nine preservatives decreased with increasing pH of sample solution from 5.5. Therefore, we choose the pH of the sample solution to be 5.5. The pH values of sample solution used in SPE in literatures are mostly 5.0 to 6.5 [51-58], which is consistent with our conclusion.

### *3.1.2 Effect of extraction time*

The effect of different extraction time on the extraction efficiency of nine preservatives is shown in Figure 2. With the prolongation of extraction time, the extraction efficiency of nine preservatives increased continuously, and reached stability after 35 min. Some laboratories extract food ingredients, and the extraction time is more than 50 min [55]. There was no significant difference in extraction efficiency after 35 min for jam, cake, soy sauce and sausage in our experiments. So, we select the extraction time at 35 min.

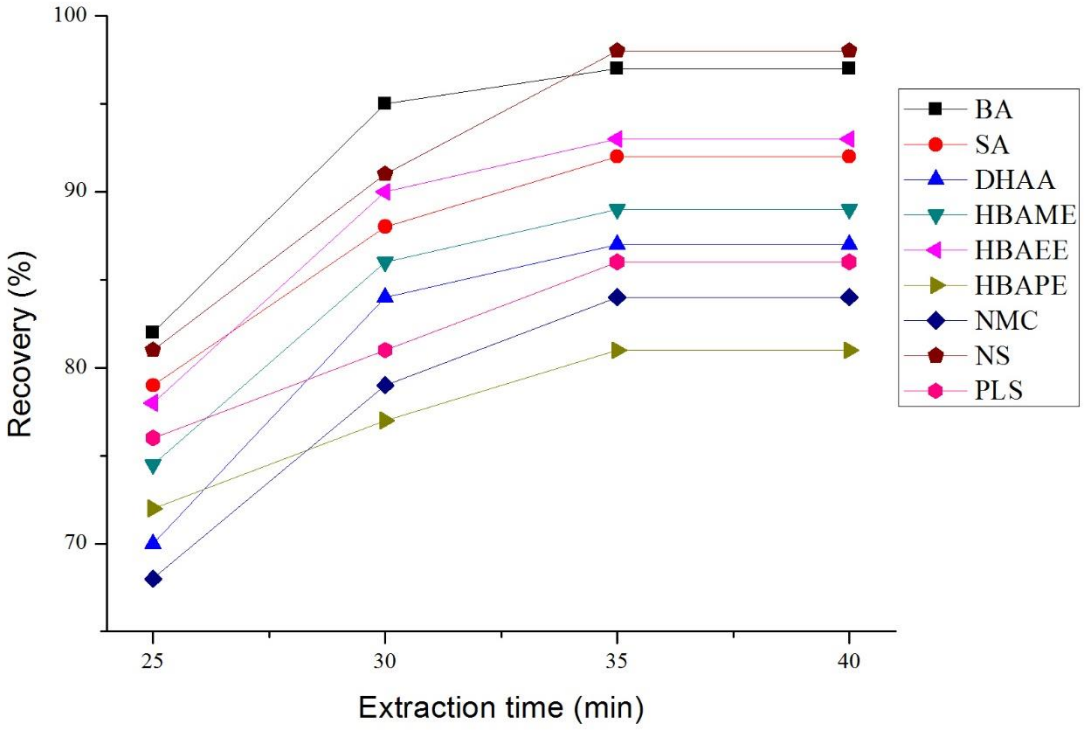


**Figure 2.** Effects of extraction time of 0.2 g hollow fiber in 10 mL sample solution on the recoveries of nine preservatives under pH 5.5, extraction temperature 45℃ and NaCl 1.5 g.

### *3.1.3 Effect of extraction temperature*

The effect of different extraction temperature on the extraction efficiency of nine preservatives

is shown in Figure 3. With the increase of extraction temperature, the extraction efficiency of nine preservatives increased first and then decreased, and reached the maximum at 45 ℃. In the literature, the extraction temperature is mostly about 50 ℃ [55-57], a few even 60 ℃ [58]. But in our experiment, when the temperature is over 50 ℃, the extraction efficiency decreases obviously, and the recovery rate of some components drops to less than 80%. Therefore, the best extraction temperature is 45 ℃.

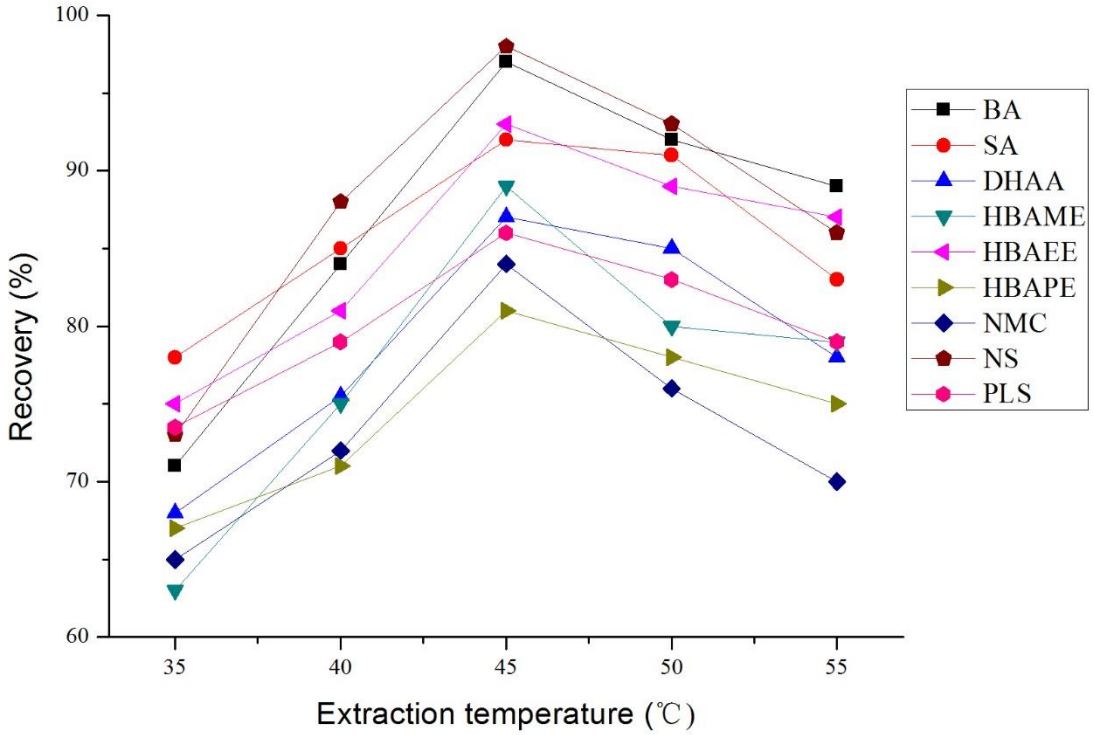


**Figure 3.** Effects of extraction temperature of 0.2 g hollow fiber in 10 mL sample solution on the recoveries of nine preservatives under pH 5.5, extraction time 35 min and NaCl 1.5 g.

*3.1.4 Effect of salt dosage*

Salt ions play a competitive role in aqueous solution. It can reduce the concentration of analyte in aqueous solution to a certain extent. With the increase of ionic strength of aqueous solution, the solubility of analyte in water decreases and the partition coefficient in hollow fiber increases, which can improve the extraction efficiency. However, with the increase of ionic strength, the viscosity and density of the solution increase, the mass transfer resistance increases, the mass transfer efficiency of analyte decreases, and the competitive adsorption is enhanced, which is not conducive to extraction. In this part, the effect of salt dosage on extraction efficiency was investigated for 0.2 g hollow fiber in 10 mL sample solution by adding different quality of NaCl. The results are shown in Figure 4. With the addition of NaCl, the ionic strength increased. The extraction efficiency of nine preservatives increased first and then decreased, and reached the maximum when NaCl was 1.5 g. It has been proved that adding a certain amount of salt can improve the effect of solid phase extraction [51-58]. However, the

type of solid phase and the amount of sample used in different work are not the same, so there is no comparability between them.

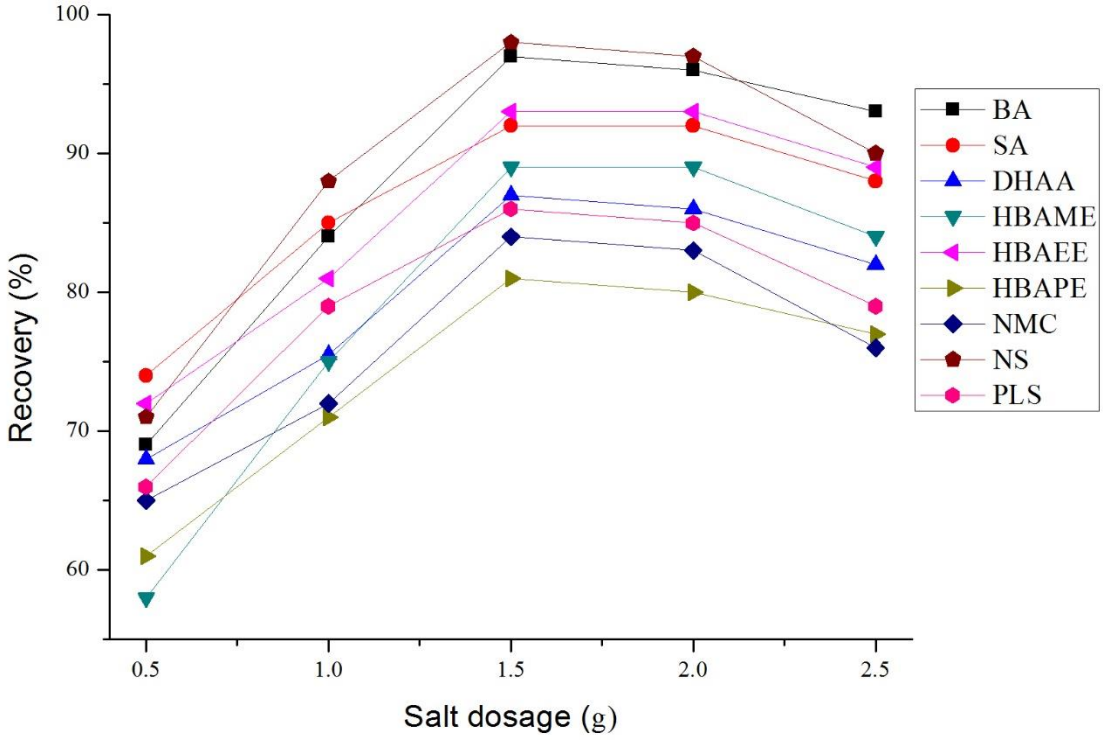


**Figure 4.** Effects of salt dosage for 0.2 g hollow fiber in 10 mL sample solution on the extraction efficiency under pH 5.5, extraction time 35 min and extraction temperature 45℃.

*3.2. Optimization of CE parameters*

The CE conditions, such as pH of separation PBS, ionic strength of PBS, separation voltage and additives, have great influence on the separation of target analytes. In this part, the resolutions of adjacent preservatives which is difficult to separate in electrophoretogram, such as SA/BA, BA/HBAME, HBAME/HBAEE, HBAEE/DHAA, DHAA/HBAPE, HBAPE/NMC, NMC/NS and NS/PLS, were used as test indexes to study the influence of different CE parameters. The resolution (R) is the ratio of the difference of retention time (t) between two adjacent components and the average width of peak base (Y) of two components. Its mathematical expression is as follows:

$$R = \frac{t_2 - t_1}{(Y_1 + Y_2) / 2}$$

It is generally believed that when the separation degree is not less than 1.5, the two components can be completely separated from the shape of the peaks.

Figure 5 shows the CE separation of nine preservatives in PBS at pH 6.0. Using the above expression of resolution, $R_{SA/BA}$ = 1.8, $R_{BA/HBAME}$ = 2.0, $R_{HBAME/HBAEE}$ = 1.1, $R_{HBAEE/DHAA}$ = 1.1, $R_{DHAA/HBAPE}$ = 1.0, $R_{HDAPE/NMC}$ = 2.3, $R_{NMC/NS}$ = 2.4 and $R_{NS/PLS}$ = 1.5 can be easily calculated, which

has been shown in Figure 6. The calculation process of other resolutions is similar to this one.

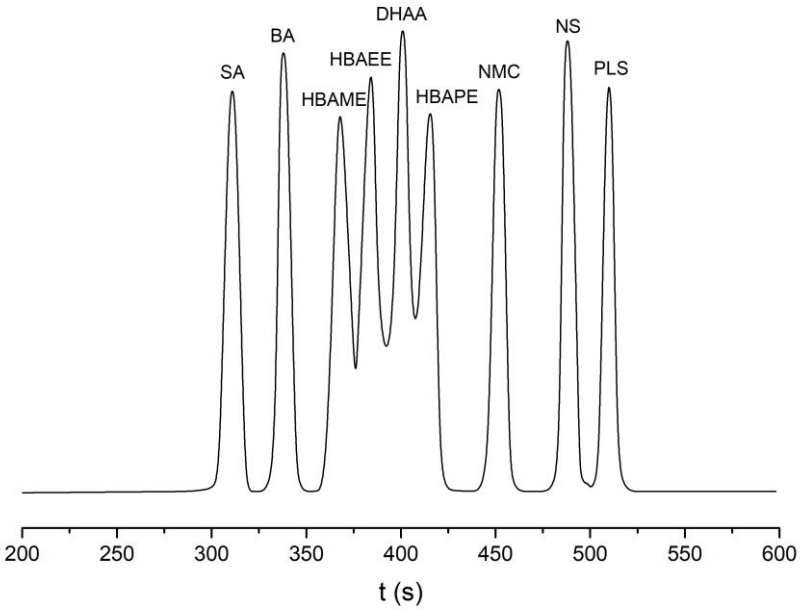


**Figure 5.** Electrophoretogram of nine preservatives in separation PBS (pH=6.0) at separation voltage 18 kV.

*3.2.1 pH of separation PBS*

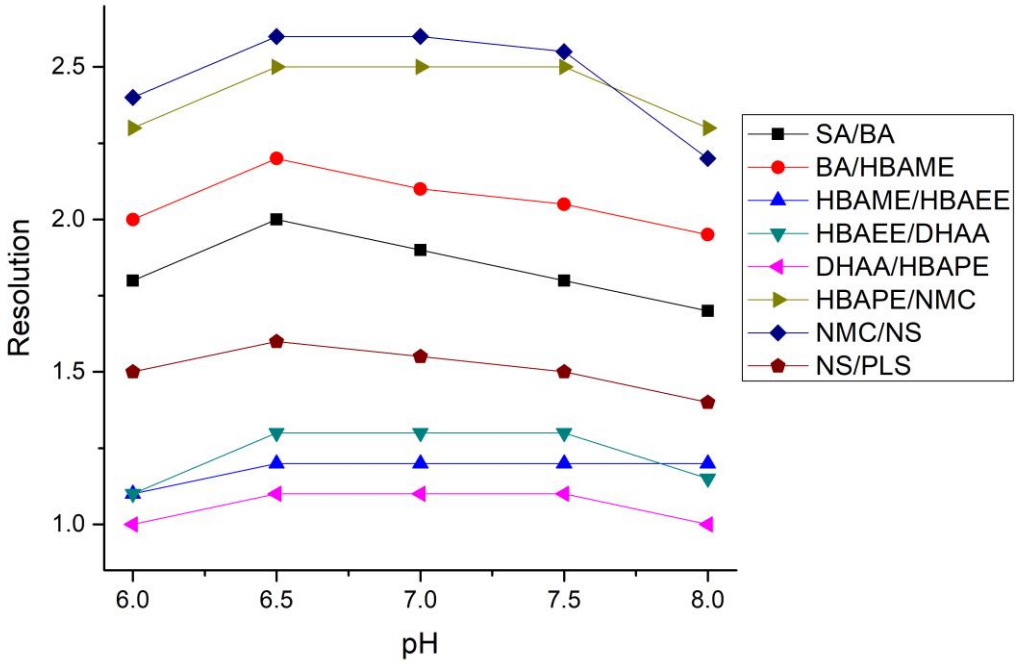


**Figure 6.** Effects of pH of separation PBS on the resolutions of 8 pairs of preservatives at separation voltage 18 kV.

The acidity of separation PBS is an important condition affecting the separation effect. In the literature work of PBS medium, the application range of pH value is mostly 6-8 [29-35,38,42-47]. Therefore, we mainly study the influence of pH in this range on the resolution. When the pH of the PBS changes from 6 to 8, the resolutions of 8 pairs of analytes are shown in Figure 6. With the increase of the pH of separation PBS, the resolutions of 8 pairs of analytes shows a trend of "first increase and then decrease", and reaches the maximum at pH = 6.5-7.5. At pH 6.5, the resolutions of 7 pairs of analytes reached the maximum value, so we chose 6.5 as the pH value of separation buffer solution.

*3.2.2 Ionic strength of separation PBS*

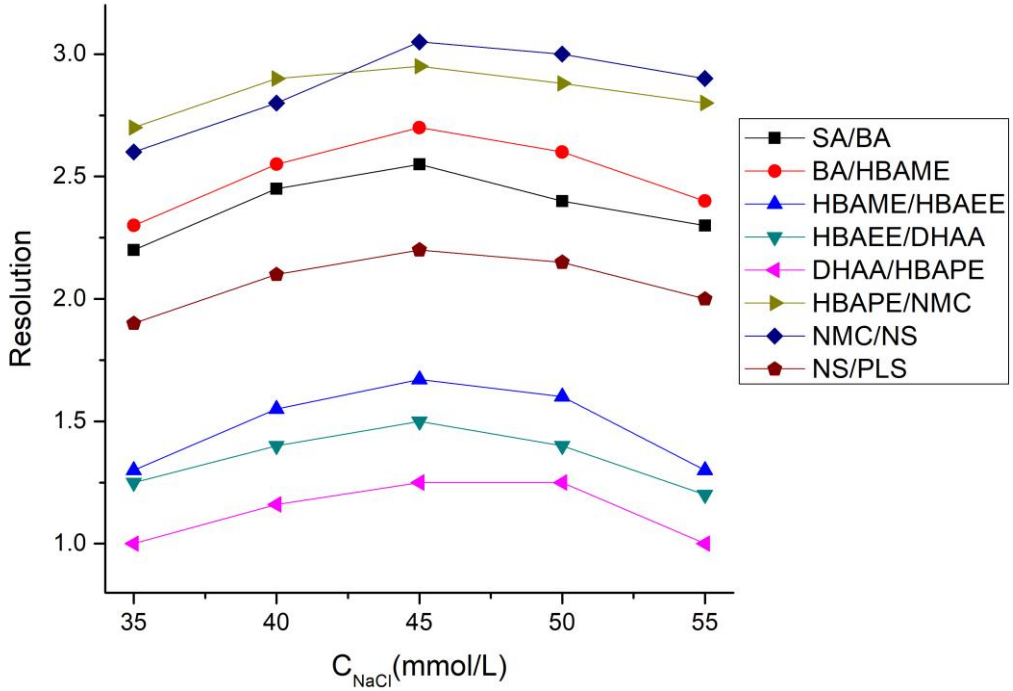


**Figure 7.** Effects of ionic strength of separation PBS (pH 6.5) on the resolutions of 8 pairs of preservatives at separation voltage 18 kV.

The ionic strength of separation PBS is another important condition affecting the separation effect. In this experiment, the effect of ionic strength on resolution was investigated by adding different concentration of NaCl. The results are shown in Figure 7. With the increase concentration of NaCl, the ionic strength of separation PBS increased. When the concentration of NaCl is 45 mmol/L, the resolutions of 8 pairs of analytes are relatively large. This conclusion is supported by many literatures [29-30,37,39-41]. NaCl, $Na_2SO_4$, KCl and $NH_4Cl$ can be used to change the ionic strength of buffer solution.

*3.2.3 Additive in separation PBS*

The PBS is often used as separation medium for CE. However, in this experiment, HBAME, HBAEE, DHAA and HBAPE cannot be completely separated if only PBS is used (see Figure 5). Our in-depth study found that cyclodextrin has a great influence on their separation. Figure 8 shows the effect of different concentrations of cyclodextrin on the resolutions of 8 pairs of analytes. With the increase of concentrations of cyclodextrin in separation PBS, the resolutions of HBAME, HBAEE, DHAA and HBAPE showed different trends. When the concentration of cyclodextrin is 25 mmol/L, all of the resolutions of 8 pairs of analytes are not less than 1.5. The use of additives to improve the separation effect has been confirmed by many experiments [27,30,33,38-41,47]. Pyrrolidone, isopropanol, cyclodextrin and tween can be used as additives.

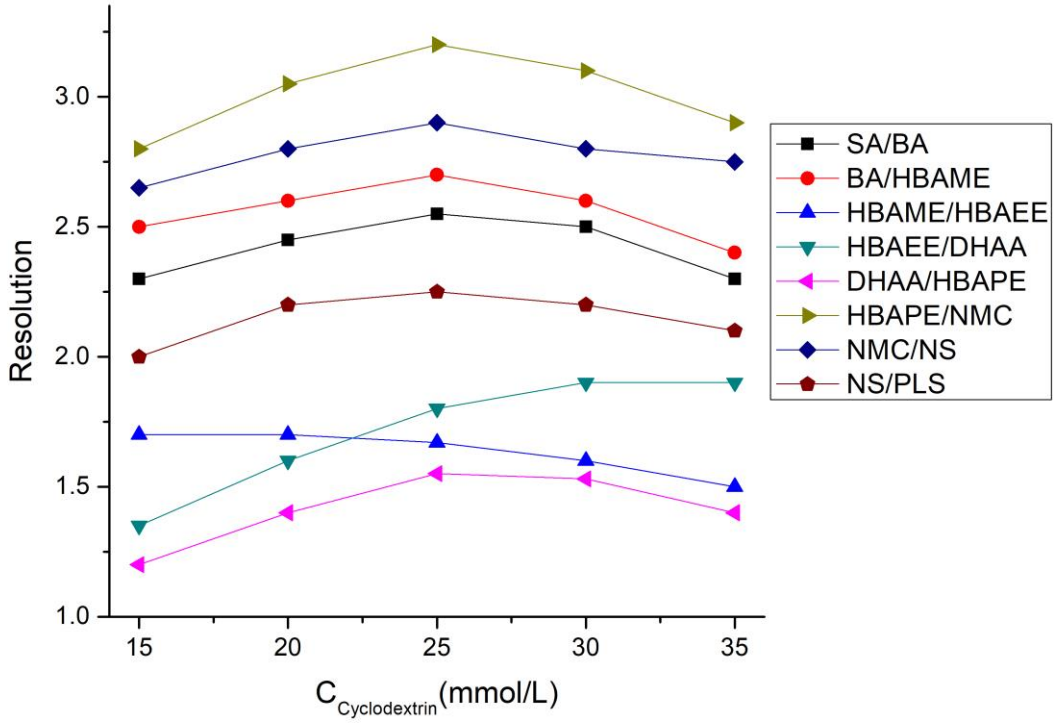


**Figure 8.** Effects of additive in separation PBS (pH 6.5) containing 45 mmol/L NaCl on the resolutions of 8 pairs of preservatives at separation voltage 18 kV.

*3.2.4 Selection of separation voltage*

The separation voltage affects the migration time of components, and then changes the resolution of components. In the literature work, the separation voltage of most experiments is below 20 kV [27-38,40-44], and few of them are over 20 kV [39,46]. In this experiment, we studied the effect of separation voltage on the separation degree of preservatives. The results are shown in Figure 9. As you can see, 17 kV is the best separation voltage in our experiments.

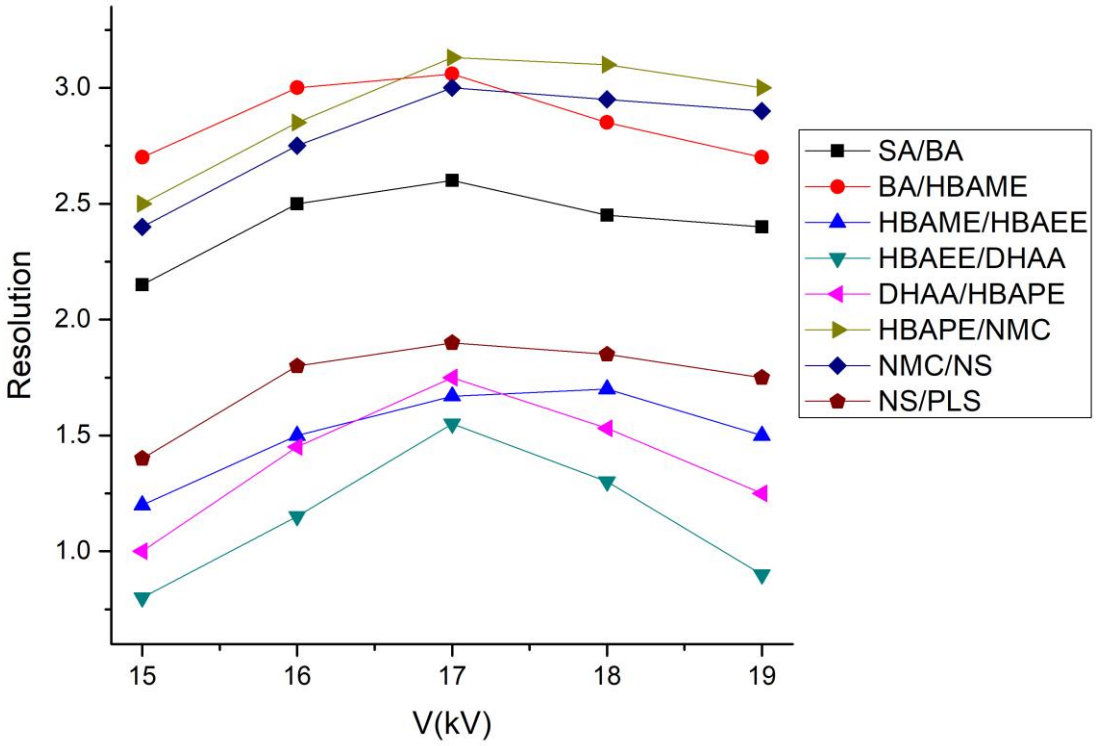


**Figure 9.** Effects of separation voltage on the resolutions of 8 pairs of preservatives in separation PBS (pH 6.5) containing 45 mmol/L NaCl and 25 mmol/L cyclodextrin.

*3.3 Methodology*

**Table 1.** Regression equation, linear range and detection limit of nine preservatives under selected SPE and CE conditions.

| Number | Preservative | Regression Equation | Linear Range/(μg/kg) | Detection Limit/(μg/kg) |
|---|---|---|---|---|
| 1 | BA | $I = 216.5C + 74.4$ | 1.2-1500 | 0.8 |
| 2 | SA | $I = 144.2C + 35.3$ | 1.5-1500 | 1.0 |
| 3 | DHAA | $I = 118.2C + 26.9$ | 1.2-1500 | 0.7 |
| 4 | HBAME | $I = 124.9C + 63.5$ | 0.8-1000 | 0.5 |
| 5 | HBAEE | $I = 173.4C + 52.6$ | 0.8-1000 | 0.5 |
| 6 | HBAPE | $I = 64.4C + 42.5$ | 0.8-1000 | 0.5 |
| 7 | NMC | $I = 77.4C + 53.8$ | 0.8-1000 | 0.5 |
| 8 | NS | $I = 270.1C + 52.1$ | 0.8-1000 | 0.5 |
| 9 | PLS | $I = 93.6C + 46.8$ | 1.5-1500 | 0.9 |

A method for simultaneous determination of nine preservatives was established by using the optimized extraction and separation conditions and UV detection. The optimized parameters of SPE with 0.2 g hollow fiber in 10 mL sample solution are sample solution pH 5.5, extraction time 35 min,

extraction temperature 45℃ and NaCl 1.5 g. The selected CE separation solution is PBS (pH=6.5) containing 45 mmol/L NaCl and 25 mmol/L cyclodextrin. The separation voltage is 17 kV. A series of preservative standard solutions were prepared and determined according to the experimental method. The linear relationship, linear range and detection limit of the method were investigated. The results were summarized in Table 1. Their detection limits are 0.5-1.0 μg/kg. This shows that the method is sensitive.

### *3.4 Sample analysis*

The residue and recovery of nine preservatives in jam, cake, soy sauce and sausage were studied. The recoveries of nine preservatives in them are 83.2% - 116.7%. Residual preservatives were detected in all four food samples, and the results are shown in Table 2.

**Table 2.** Analysis results of actual food samples under selected SPE and CE conditions.

| Antibiotics | Measured value (μg/kg) | | | | Added value | Recovery （%, n=9） | | | |
|---|---|---|---|---|---|---|---|---|---|
| | Jam | Cake | Soy sauce | Sausage | (μg/L) | Jam | Cake | Soy sauce | Sausage |
| BA | ND* | ND | 16.3 | 7.3 | 50 | 88.5 | 90.8 | 101.6 | 97.8 |
| SA | 6.3 | 5.5 | 3.3 | ND | 50 | 91.4 | 83.6 | 113.7 | 85.9 |
| DHAA | ND | 4.8 | 0.9 | 2.0 | 50 | 87.3 | 107.6 | 95.8 | 95.2 |
| HBAME | 1.4 | ND | ND | ND | 50 | 103.2 | 84.7 | 88.9 | 100.7 |
| HBAEE | 3.8 | ND | 4.1 | ND | 50 | 101.1 | 94.4 | 90.4 | 116.7 |
| HBAPE | 7.2 | ND | ND | ND | 50 | 93.2 | 103.5 | 86.4 | 97.7 |
| NMC | ND | 5.2 | ND | 3.6 | 50 | 83.2 | 89.8 | 103.9 | 106.1 |
| NS | ND | ND | ND | 4.2 | 50 | 108.2 | 85.8 | 93.2 | 99.1 |
| PLS | ND | 5.1 | ND | ND | 50 | 98.0 | 109.9 | 102.1 | 89.5 |

*Not detected

## 4. CONCLUSION

This work demonstrated a new analytical procedure for simultaneous determination of nine preservatives in food samples by SPE-CE-UV. The nine preservatives could be well separated and analyzed with high sensitivity, wide linear range, and good reproducibility.

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