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Separation of Seven Residues of Fluoroquinolones from Fish Samples using Solid Phase Microextraction and Electrophoresis and their detection by Electrochemiluminescence method

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A new method for the simultaneous determination of 7 fluoroquinolones (FQs) in fish was established based on purifing and enriching by solid phase microextraction, separating by capillary electrophoresis (CE) and detecting by column end electrochemiluminescence. The effects of pH value, extraction time, extraction temperature, ionic strength and eluate type on the extraction efficiency of 7 FQs were investigated. The effects of buffer additives and separation voltage on the separation efficiency of 7 FQs were investigated. The best purification and enrichment parameters of 10.0 ml extraction solution made from 2.0 g fish sample are: pH value of extraction system is 7, extraction time is 40 min, extraction temperature is 50 ℃, addition of NaCl is 2.0 g, and eluate is 2.0 ml methanol (containing 5% formic acid). The separation solution of CE is 0.02 mol/L phosphate buffer solution (pH=6, containing 18% cyclodextrin), and the separation voltage is 20 kV. All the 7 FQs have good linear relationship. Their detection limits were 0.4-0.8 μg/L. The recovery of fish samples was 89.2%-110.4%. 4 FQs were detected in two fish samples. This method has the advantages of small matrix effect, high sensitivity, simplicity and accuracy. It can be used for the detection of FQs in other fish or meat.

Keywords: Fluoroquinolones, Fish, Solid phase microextraction, Capillary electrophoresis, Electrochemiluminescence

1. INTRODUCTION

Fluoroquinolones (FQs) are a kind of synthetic antibiotics, which are widely used in the fields of animal husbandry and aquaculture due to their wide antibacterial, strong bactericidal and fast absorption [1, 2]. Our previous work focused on the detection conditions of electrochemiluminescence and pretreatment conditions of samples for ciprofloxacin (CIP), enrofloxacin (ENR), norfloxacin (NOR) and pefloxacin (PEF) [3]. Since the use of ofloxacin (OFL), fleroxacin (FLE) and lomefloxacin

(LOM) are also very common [4,5], it is very important to study the separation conditions and efficient sample processing technology for more FQs.

Numerous methods have been employed to analyze FQs residues, such as LC-MS [6-11], fluorescence [12-16], electrophoresis [17-20], LC-UV [21-23], spectrophotometry [24] and chemiluminescence [25]. Capillary electrophoresis (CE) is a promising high-performance biochemical and medical separation method with short analysis time and less sample consumption.It has good separation ability for molecules with similar structure. Electrochemiluminescence (ECL) based on tris (2, 2'-bipyridyl) ruthenium (II) ($Ru(bpy)_3^{2+}$) is an attractive analytical method for organic amines owing to its inherent high sensitivity, selectivity and stability. CE separation with end-column ECL detection (CE-ECL) have been widely studied and usedto analyze various drugs [26–34], antibiotics [35], enzymes [36], alkaloids [37–39], amines [40], hormones [41] and pesticide residues [42,43] in different foods, pharmaceuticals, animals and plants.

The residual FQs in fish is in trace level, so it is difficult to determine them. Sample pretreatment technology has great influence on the sensitivity, efficiency and reliability of analytical methods. Solid phase microextraction (SPME) is a new 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 [44-48], food [49-53] and pharmaceutical industries [54-58]. Hollow fiber (HF) with large surface area has been widely used [59-63]. Other functional stationary phases can be fixed on HF membrane template as reinforcer to make new HF composite materials [64-67].

In this paper, the analytical solution prepared from freshwater fish was treated with HF-SPME method. Then CIP, ENR, NOR, PEF, OFL, FLE and LOM were separated and detected simultaneously by CE-ECL. The results show that the present method is sensitive and reliable for the simultaneous determination of 7 residues of FQs in fish.

2. EXPERIMENTAL

2.1. Materials and drugs

Tris (2, 2'-bipyridyl) ruthenium (II) dichloride hexahydrate ($Ru(bpy)_3Cl_2 \cdot 6H_2O$) was purchased from Alfa Aesar (Johnson Matthey, USA). Disodium hydrogen phosphate (Na_2HPO_4), sodium dihydrogen phosphate (NaH_2PO_4), sodium hydroxide (NaOH), methanol, acetonitrile, formic acid, ethyl orthosilicate, polyvinylpyrrolidone, cyclodextrin, and iso-propyl alcoholwere all of analytical reagent gradeand 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). Standard substances of ciprofloxacin (CIP), enrofloxacin (ENR), norfloxacin (NOR), pefloxacin (PEF), ofloxacin (OFL), fleroxacin (FLE) and lomefloxacin (LOM) were purchased from National Institutes for Food and Drug Control (Beijing, China).

2.2. Apparatus

CE-ECL was performed on a MPI - B multi-parameter chemiluminescence analysis test system (Xi'an Remex analytical instruments Co., Ltd., Xi'an, China). Cyclic voltammetry and potentiostatic method were carried out in a three electrodes system with a platinum working electrode of 500 μm in diameter, an Ag/AgCl reference electrode of 300 μm in diameter and a platinum wire auxiliary electrode of 1 mm in diameter. 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.

2.3. Solutions preparation

$Ru(bpy)_3^{2+}$ solutions were prepared with $Ru(bpy)_3Cl_2 \cdot 6H_2O$ and secondary distilled water. Phosphate buffer solution (PBS) has good stability, easy preparation and wide pH range. Therefore, phosphate buffer solution is chosen as the experimental environment [30-38]. PBS was prepared with disodium hydrogen phosphate, sodium dihydrogen phosphate and secondary distilled water. NaOH solution was prepared with NaOH and secondary distilled water. Standard solutions of ciprofloxacin, enrofloxacin, norfloxacin, pefloxacin, ofloxacin, fleroxacin and lomefloxacin were prepared with their standard substances and secondary distilled water. All solutions used in the experiment must be filtered through a 0.22 μm cellulose acetate membrane.

2.4. Hollow fiber treatment

The ethyl orthosilicate was added into ethanol solution, 38% hydrochloric acid was added under stirring condition, aged at room temperature for 24 h, and then the ethyl 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 30 min, the hollow fiber was removed from the sol and dried at 120 ℃ for standby. This process is improved by referring to the literature [60,65].

2.5. Electrochemiluminescence detection conditions

Detection potential is 1.2 V (vs. Ag/AgCl). Concentration of $Ru(bpy)_3^{2+}$ is 6 mmol/L. Concentration of phosphate buffer solutions (PBS) is 40 mmol/L. pH of PBS is 6.5. The detailed process can be referred to our previous work [3].

2.6. Sample preparation and SPME

Fresh fish samples were processed, homogenized and stored at −20 °C. Accurately weigh 2.0 g of crushed homogeneous fish meat, put it in a 50 mL spiral cap centrifugal tube, add 2 mL water, scroll

for 1 minute on the vortex oscillator, add 8 mL mixed solution of acetonitrile and methanol (v/v=4/1), 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. The supernatant was removed 8 mL to another 20 mL screw cap centrifugal tube, add 2.0 ml 0.05 mol/l PBS (pH = 7) solution, 2.0 g NaCl, vortex to make NaCl completely dissolved, raise the temperature to 50 ℃, completely immerse the hollow fiber in the solution, and extract by ultrasonic for 40 min. After the extraction, transfer the hollow fiber to a test tube, add 2.0 ml methanol (containing 5% formic acid) as eluate, and shake it with ultrasonic for 5 min. Blow the solution dry with nitrogen, add 0.5 ml methanol water solution (1:1) along the pipe wall to dissolve the analyte, with even vortex. After passing through 0.22 μm microporous membrane, the filtrate was ready for use.

3. RESULTS AND DISCUSSION

3.1. Optimization of SPME conditions

3.1.1 Effect of pH value

The pH of the solid phase extraction system changes from 4 to 9 and the change of extraction efficiency is shown in Figure 1. With the increase of the pH of the extraction system, the extraction efficiency of the seven FQs shows a trend of "first increase and then decrease", and reaches the maximum at pH = 5-8. The maximum extraction efficiency of CIP and NOR appeared at pH = 6, PEF at pH = 5, and other four FQs at pH = 7. The extraction efficiency of CIP, NOR and PEF at pH = 7 was not significantly different from their maximum value. Therefore, pH 7 of sample solution is a better choice in this experiment. The pH values of SPME in literatures are mostly weak acid, weak base or neutral [44-58], which is consistent with our conclusion.

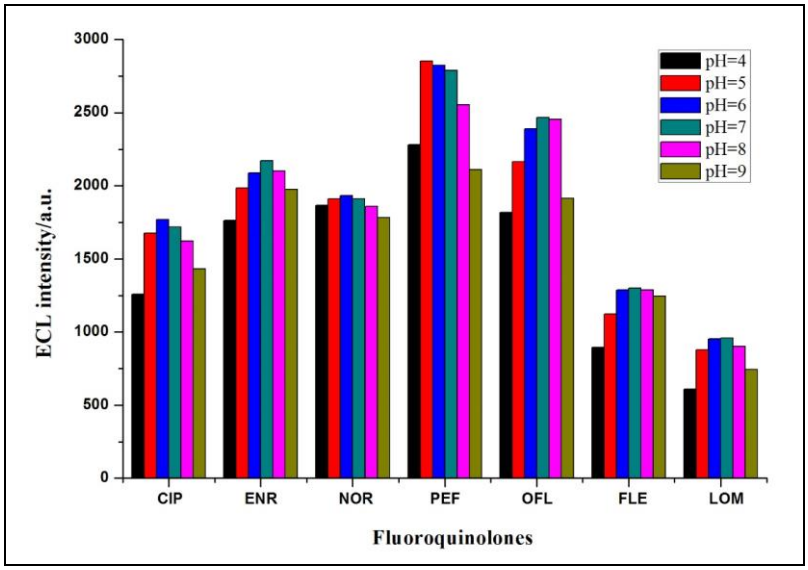

Figure 1. Effects of pH in samples on the extraction efficiency.

3.1.2 Effect of extraction time

The effect of different extraction time on the extraction efficiency of FQs is shown in Figure 2. With the prolongation of extraction time, the extraction efficiency of 7 FQs increased continuously, and reached stability after a certain time (30 min ~ 40 min). There was no significant difference in extraction efficiency after 40 min. It can be seen that the hollow fiber can be saturated after 40 min extraction of 7 FQs, and the longer extraction time has little effect on the extraction efficiency. Some people use hollow fiber to extract food ingredients, and the extraction time is more than 60 minutes [65], but it seems unnecessary in our experiments. So we determined that the extraction time was 40 minutes.

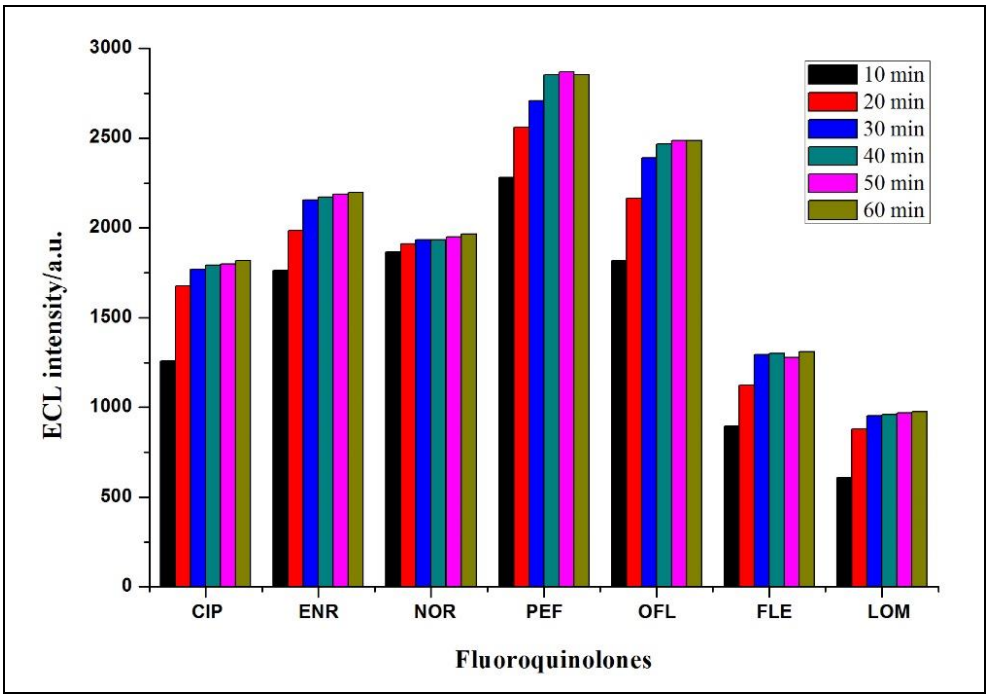

Figure 2. Effects of extraction time in samples on the extraction efficiency.

3.1.3 Effect of extraction temperature

This experiment investigated the effect of different extraction temperature on the extraction efficiency of FQs, as shown in Figure 3. With the increase of extraction temperature, the extraction efficiency of 7 FQs substances increased first and then decreased, and reached the maximum at 50-60 ℃. When hollow fiber is used as solid phase extraction agent, higher temperature is not conducive to maintain its shape [61]. Most of the operating temperatures in the literature are less than 60 ℃. So we chose 50 ℃ as our extraction temperature.

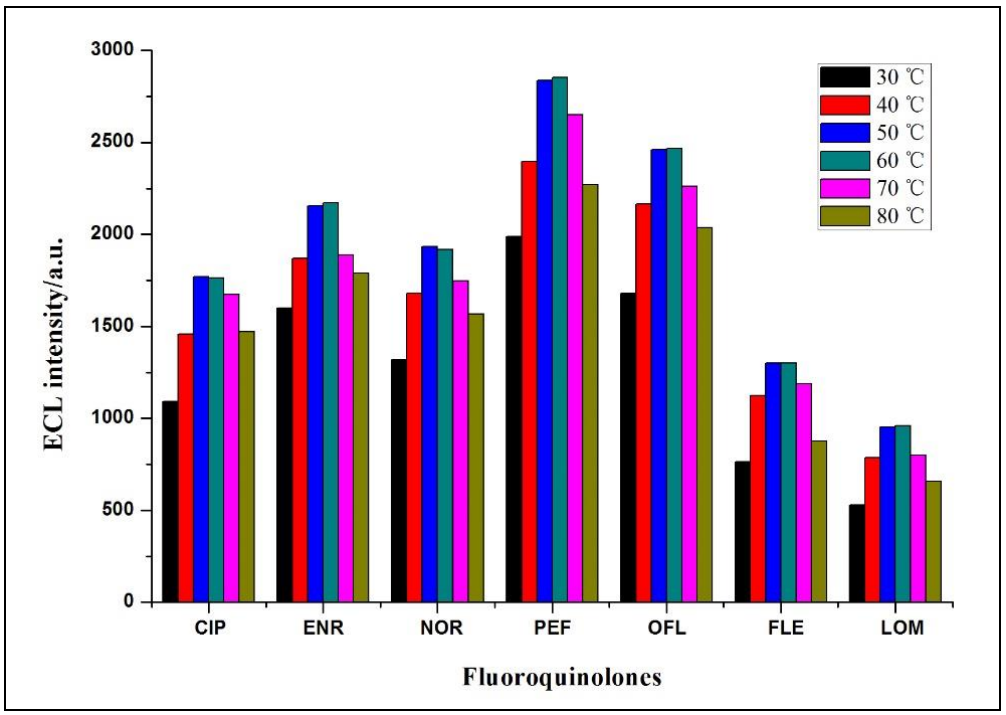

Figure 3. Effects of extraction temperature in samples on the extraction efficiency.

3.1.4 Effect of ionic strength

In the process of SPME, the ion strength of the matrix can be changed by adding appropriate inorganic salts, such as NaCl, Na_2SO_4 or $(NH_4)_2SO_4$, so as to reduce the affinity between the target analyte and the matrix and improve the extraction efficiency [45,50]. In this experiment, the effect of ionic strength on extraction efficiency was investigated 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 7 FQs increased first and then decreased, and reached the maximum when NaCl was 2.0 g.

Due to the salting out effect, salt ions play a competitive role in the aqueous solution, which leads to the decrease of the dissolved organic matter concentration in the aqueous solution to a certain extent. Therefore, the salting out effect increases with the increase of ionic strength. The solubility of analyte in water decreased and the partition coefficient of analyte in fiber increased. Thus, the extraction efficiency can be improved. When NaCl increased from 0.5 g to 2.0 g, the extraction efficiency of 7 FQs increased. However, with the increase of ionic strength, the viscosity and density of the solution increase. With the increase of mass transfer resistance, mass transfer efficiency of analyte will be reduced, and competitive adsorption will be strengthened, which is not conducive to extraction. Therefore, the extraction efficiency of 7 FQs began to decrease since NaCl was 2.0 g. It shows that there are mass transfer resistance and competitive adsorption. The mass of NaCl added into the working solution selected in this experiment is 2.0 g. This result is consistent with many literatures [50-55].

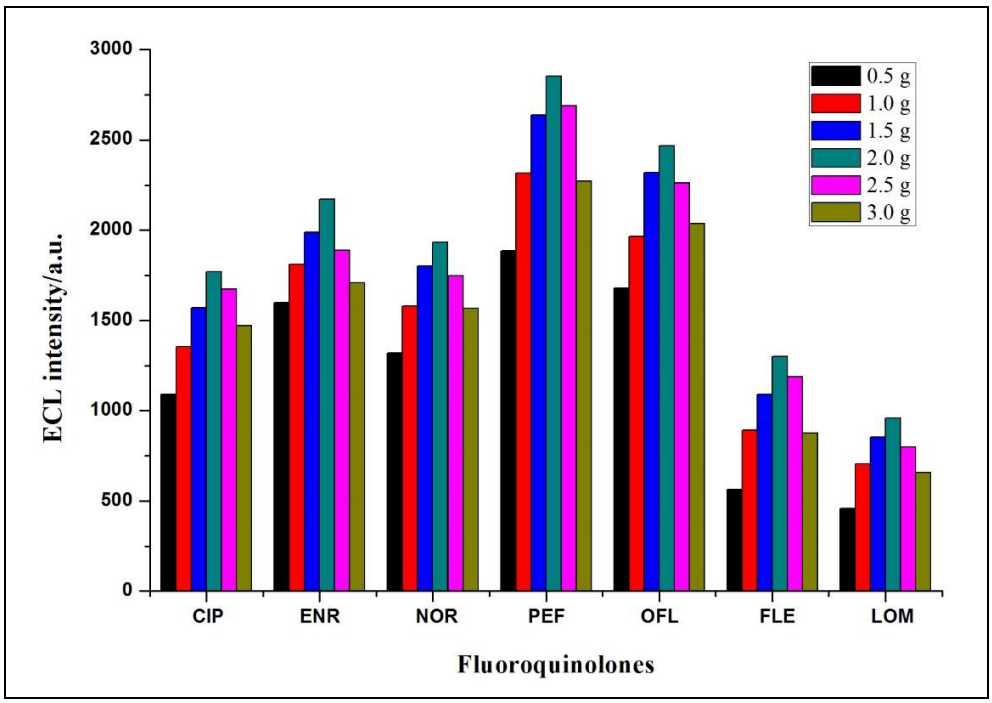

Figure 4. Effects of salt addition in samples on the extraction efficiency.

3.1.5 Effect of eluate type

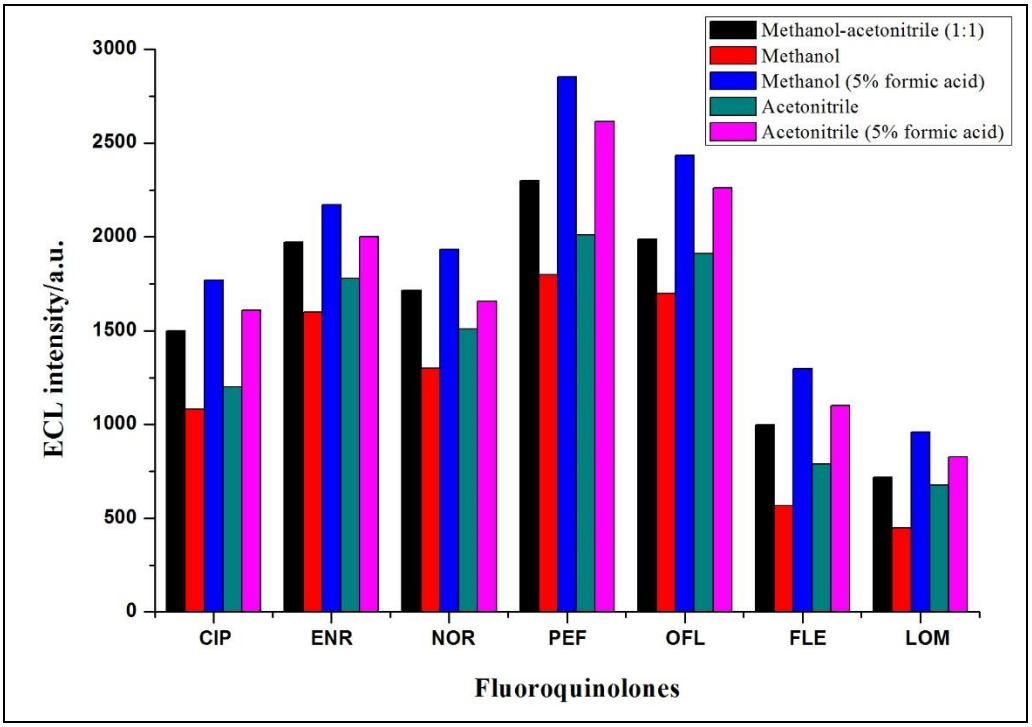

Figure 5. Effects of salt addition in samples on the extraction efficiency.

Methanol and acetonitrile are the most commonly used eluents in the literature [39,41,42,49,53]. Sometimes a small amount of formic acid is added to change the polarity of methanol and acetonitrile [40,45,50,55]. In this experiment, the effects of five eluents, (a) methanol acetonitrile (1:1), (b) methanol, (c) methanol (containing 5% formic acid), (d) acetonitrile, and (e) acetonitrile (containing 5% formic acid), on the resolution efficiency of FQs were studied. The results are shown in Figure 5. The extraction efficiency of 7 FQs was the highest when methanol (containing 5% formic acid) was used. The extraction efficiency of methanol and acetonitrile containing 5% formic acid was higher than that of them alone, which indicated that the protons provided by 5% formic acid had a great influence on 7 FQs. Therefore, methanol (containing 5% formic acid) was selected as the eluent.

3.2. Optimization of separation parameters

3.2.1 Selection of separation additive

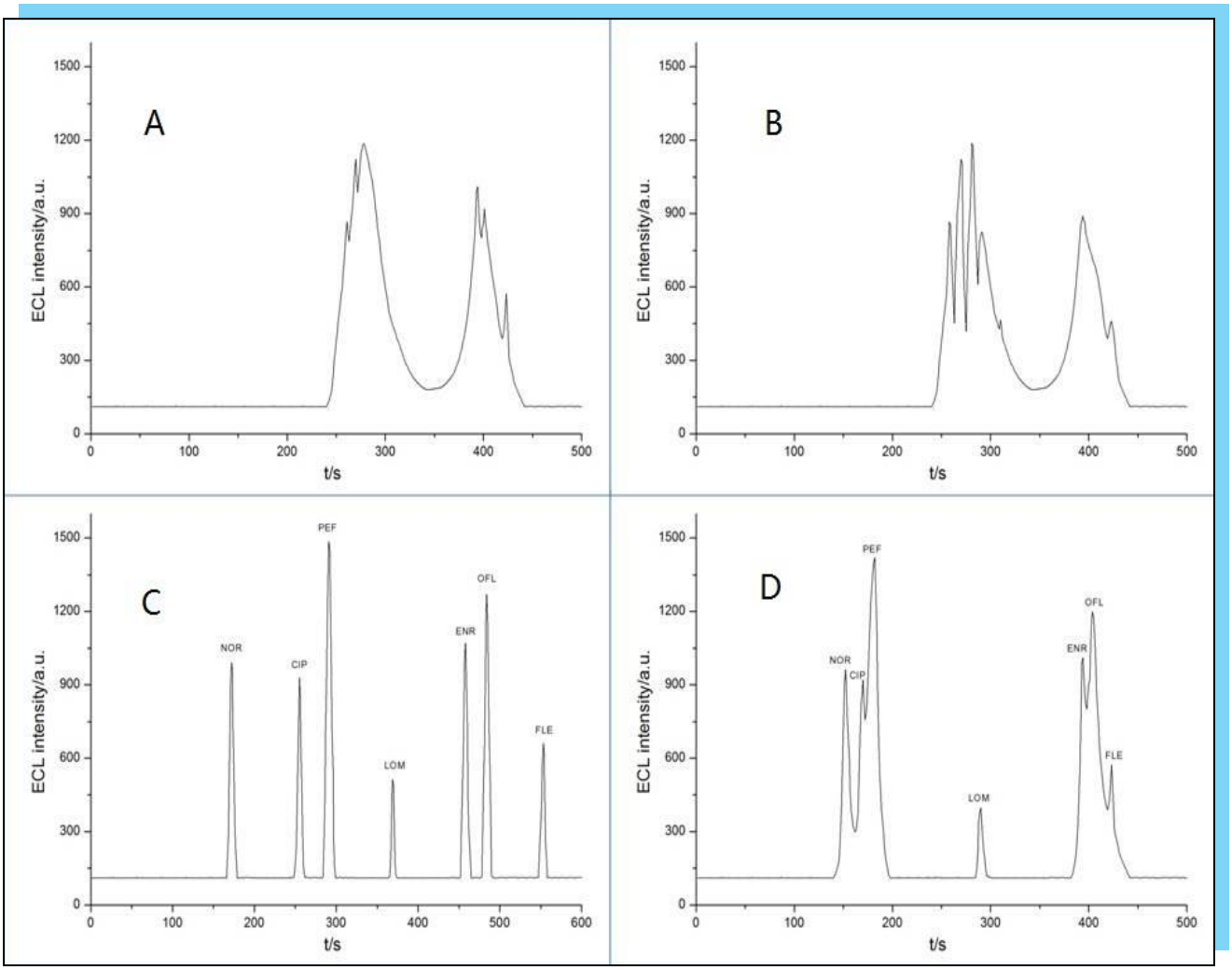

Figure 6. Effects of additive in PBS on separation efficiency.

PBS is commonly used in capillary electrophoresis. However, the separation effect of 7 FQs is very poor when PBS is only used in this experiment, as shown in Figure 6A. Sometimes, the unexpected effect can be obtained by adding proper amount of additives into PBS. Isopropanol, cyclodextrin and polyvinylpyrrolidone are commonly used separation additives in literature [36,40]. Their effects on separation were studied in this experiment. Different concentrations of isopropanol have little effect on their separation, as shown in Figure 6B. 18% of cyclodextrin can achieve good separation effect, as shown in Figure 6C. Different concentrations of polyvinylpyrrolidone have a certain positive effect, but did not get the expected effect. Figure 6D is an experimental result of 20% addition.

3.2.2 Selection of separation voltage

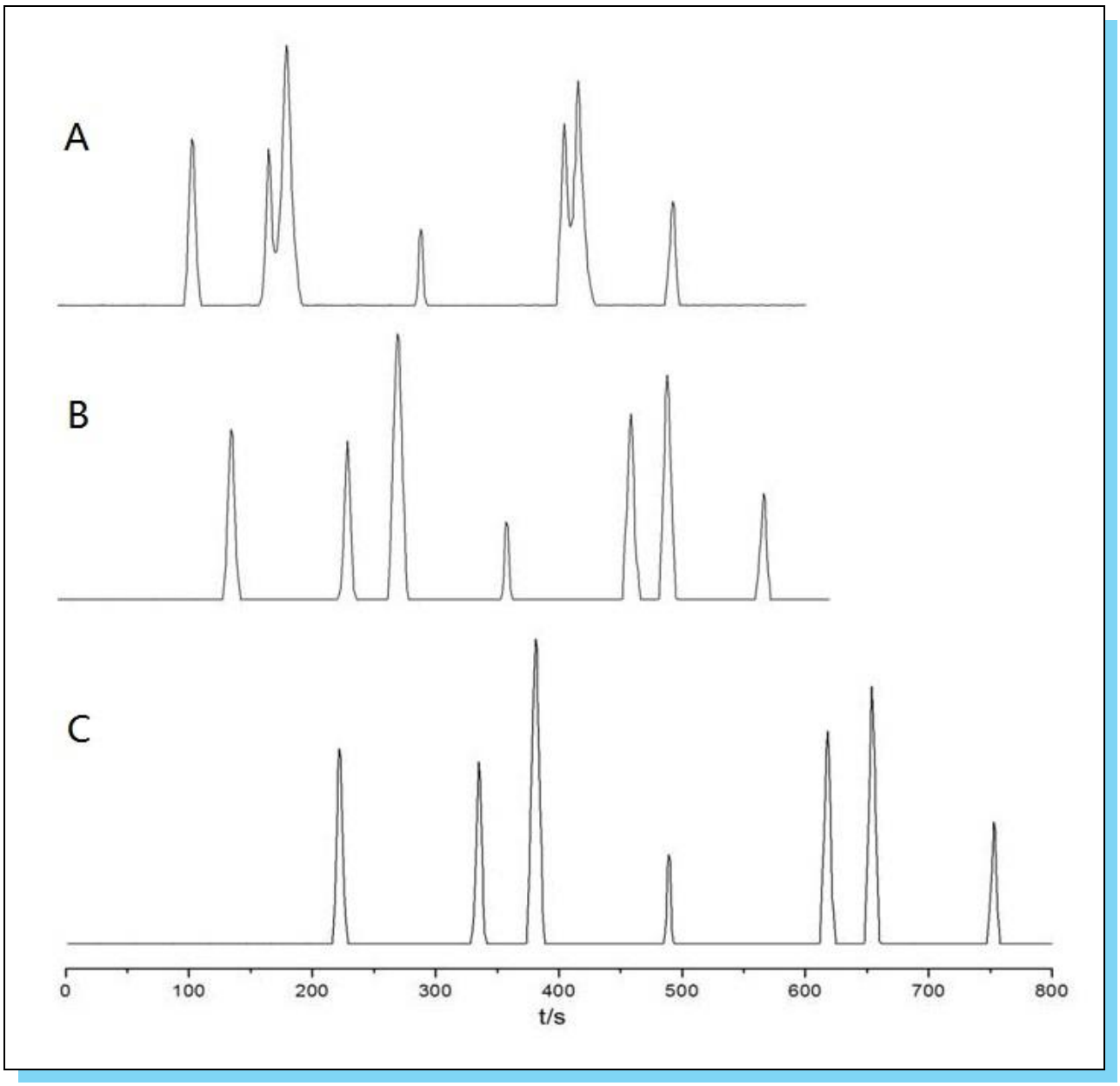

Figure 7. Effects of separation voltage on separation efficiency.

The influence of the separation voltage on separation efficiency of 7 FGs was investigated. The separation voltage affects the migration time and resolution. In this experiment, the separation conditions of 7 FQs under the separation voltage of 15, 20 and 25 kV are investigated (see Figure 7). The results show that with the increase of separation voltage, the migration time of each component is gradually shortened, but the separation degree is decreased. When the voltage is 15 kV and 20 kV, all the 7 FQs are separated by baseline, but the migration time of the 7 FQs is shorter (no more than 10 min) and the separation efficiency is higher when the voltage is 20 kV. When the voltage reaches 25 kV, the four components cannot be separated completely. 20 kV is the best separation voltage. The separation voltage in the literature is mostly between 18-25 kV [32-41].

3.3 Methodology

A series of FQs standard solutions were prepared and determined according to the experimental method. The linear relationship, linear range and detection limit of the method were investigated with the mass concentration as the abscissa and the ECL intensity as the ordinate. The results were summarized in Table 1.

Table 1. Regression equation, linear range and detection limit of 7 FQs.

FQs	Regression Equation	Linear Range/(μg/L)	Detection Limit/(μg/L)
CIP	I = 168.2C+ 89.1	1.4-2000	0.8
ENR	I = 212.5C+ 47.8	0.8-1200	0.5
NOR	I = 188.6C+48.1	1.2-1500	0.6
PEF	I = 273.7C+ 117.2	0.7-1000	0.4
OFL	I = 239.1C+ 75.9	0.9-1100	0.5
FLE	I = 127.8C+ 21.3	1.0-1200	0.5
LOM	I = 89.4C+ 66.5	0.8-1000	0.4

3.4 Sample analysis

The residue and recovery of FQs in three kinds of fresh water fish samples were studied. The recoveries of 7 FQs in the actual samples are 89.2% - 110.4%. Two samples have detected FQs residues, and the results are shown in Table 2.

Table 2. Analysis results of actual samples.

FQs	Measured value (μg/L)			Added value (μg/L)	Recovery（%, n=7）		
	Sample1	Sample2	Sample3		Sample1	Sample2	Sample3
CIP	ND	2.3	ND	50	92.3	106.8	94.8
ENR	ND	ND	ND	50	89.7	110.0	91.1

NOR	3.1	ND	ND	50	102.5	92.9	107.7
PEF	ND	3.4	ND	50	97.6	98.4	110.4
OFL	ND	ND	ND	50	107.2	103.3	89.9
FLE	4.8	ND	ND	50	98.0	89.2	103.9
LOM	ND	ND	ND	50	95.2	109.1	99.5

4. CONCLUSION

This work demonstrated a new analytical procedure for simultaneous determination of CIP, ENR, NOR, PEF, OFL, FLE and LOM by HF-SPME-CE–ECL. The 7 FQs could be well separated within 10 min with high sensitivity, wide linear range, and good reproducibility. The method can be used to directly simultaneously detect CIP, ENR, NOR, PEF, OFL, FLE and LOM in fish sample.

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