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Determining eight colorants in milk beverages by capillary electrophoresis

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Abstract

Milk beverages are popular because of their high nutritional value, and milk products that are enhanced with various fruit flavors are especially in high demand in Asia. Colorants are usually added to fruit flavored milk in order to increase its attraction and appearance, therefore, the detection and measurement of colorants in this type of beverage are relatively important for health issue reasons. Carminic acid, a natural colorant, along with tartrazine, Fast green FCF, Brilliant blue FCF, Allura Red AC, Indigo carmine, Sunset yellow FCF, and New coccine, which are seven different synthetic food colorants, are commonly used as food additives, therefore, this study would focus on the development of an analytical method for the detection of these common colorants in milk beverages. A high efficiency capillary electrophoresis separation method was finished by a pH 10.0 running buffer containing 7.0 m*M* β-cyclodextrin, and the eight colorants were separated with baseline resolution within 9 min. In order to reduce the matrix interference resulting from the constituents of milk, a suitable polyamide column solid-phase extraction (SPE) was also investigated for milk sample pretreatment. The combination of the simple SPE pretreatment and the fast separation method of capillary electrophoresis, was able to determine successfully without matrix interference the content of these colorant additives in commercial milk beverages. The recoveries of the eight food colorants in milk beverages were better than 85% and the detection limits were also lower than 0.5 μg/ml by the developed method.

Keywords: Milk; Food analysis; Color additives

1. Introduction

Milk is an excellent source of essential nutrients, including protein, fat, carbohydrate, vitamins and minerals. Advances in the food industry in the past century have allowed many dairy products to be made with a variety of flavors and colors in order to enhance their taste and visual aesthetics, and to promote sales.

Analytical methods for milk beverages that have been developed in recent years have been focused on the examination of proteins, antibiotics and mycotoxin residues, in addition to the effect of heat and pressure on milk treatment, and on the adulteration of milk [1–5]. In Asia, consumers usually prefer milk products that contain various fruit flavors, such as strawberry and apple, therefore, food colorants are commonly added in order to achieve their desired color appearance. Several food colorants, whether synthetic or natural, have been permitted to be used as food additives. The allowable amount of synthetic

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colorants, however, is strictly limited because of their potential toxic nature.

In order to detect the presence and level of these colorants in foodstuffs, many separation techniques have been developed. High-performance liquid chromatography (HPLC), including ion chromatography and ion-pair chromatography, has been widely employed in the separation of colorants with reliable results [6–8]. Recently, a technique known as capillary electrophoresis (CE) has become a valuable method for separating analytes (from large protein molecules to small ions) due to its many advantages which include high efficiency, low waste production, and fast separation [9–12]. Presently, CE is also employed as another major separation tool for the analysis of natural food pigments and synthetic food colorants [13–18]. A separation of colorants by CE can usually be completed in less time than by HPLC. The narrow internal diameter of the capillary tube, however, generally gives the CE technique a lower sample capacity and optical path, therefore, the detection limit in concentration is rather unsatisfactory. To date, most research using CE has focused on the detection of colorants in simple and clean samples, such as soft drinks and popsicles, while the analysis of milk beverages by this technique is still rare [19]. Milk's matrix, which has fat and protein as its major constituents, is rather complex, thus colorants that are present in milk beverages are impossible to analyze by CE without a pretreatment of the sample. Large protein and fat compounds in milk matrix can be removed by centrifugation, but other extraction procedures are also needed to further reduce the levels of small proteins, fats and carbohydrates. Liquid–liquid and solid-phase extractions are two popular pretreatments used for a variety of samples, however, the former method is more time consuming and it is hard to increase the concentration of colorants because the colorants are relatively soluble in the aqueous medium. Solid-phase extraction (SPE), on the other hand, has the functions of concentration and filtration simultaneously, which can help separate colorants from matrix when its material and operating condition are chosen carefully. The combination of SPE and CE separation could improve the problems owing to the lower sample capacity and the interference of the complex sample matrix. In this study, polyamide column SPE in combination with high efficiency CE separation were used to successfully detect seven common and US Food and Drugs Administration (FDA) approved colorants (tartrazine, Fast green FCF, Brilliant blue FCF, Allura red AC, Indigo carmine, Sunset yellow FCF, and New coccine), and one natural colorant (carminic acid) in beverages containing 50% fresh milk.

2. Experimental

2.1. Colorant standards

Tartrazine and Fast green FCF were purchased from Sigma (St Louis, MO, USA). Brilliant blue FCF, carminic acid, Allura red AC and Indigo carmine were obtained from TCI (Tokyo, Japan). Sunset yellow FCF was obtained from Aldrich (Milwaukee, WI, USA). New coccine was purchased from ACROS (NJ, USA). These standards were individually dissolved in deionized water at a stock concentration of 2 mg/ml.

2.2. Chemicals and extraction column

Disodium tetraborate, ammonia solution (25%), ethanol (absolute), sodium hydrogen phosphate, and disodium phosphate were bought from Merck (Darmstadt, Germany). β-cyclodextrin was obtained from Calbiochem (CA, USA). Sodium hydroxide and hydrochloric acid were obtained from Baker (NJ, USA). Methanol was bought from Pharmco (CT, USA). Polyamide cartridges (DPA-6S, 3 ml, 250 mg) used as the SPE column were purchased from Supelco (PA, USA).

2.3. Milk samples

Mixed fruit-, strawberry-, apple-, and blueberry-flavored milk samples which usually contain at least 50% fresh milk in addition to ingredients such as fruit juice, sugar, colorants and other additives were obtained from supermarkets in Taiwan.

2.4. Sample pretreatment

Milk samples, which were diluted with ethanol in the volume ratio of 1:1, were mixed for 10 min with a magnetic stir bar, then pH were adjusted to 2.0 if needed. The milk solutions were centrifuged for 60 min at 16 000 rpm, then the clear centrifuged liquids were applied to the polyamide SPE columns at the rate of approximately 0.5 ml/min. The polyamide column was conditioned prior to use by washing with methanol (2 ml) and deionized water (2 ml). After adding the centrifuged liquid, the extraction column was washed with deionized water (1 ml) followed with methanol (1 ml). The absorbed colorants were then eluted with 1 ml of eluting solution (0.5% ammonia solution mixed with methanol in the volume ratio of 1:1).

2.5. Running buffer for CE

A running buffer of pH 10.0 was prepared by adding 15 m*M* disodium tetraborate (borax) to 20 m*M* sodium hydroxide (NaOH) solution until the desired pH was achieved. β-cyclodextrin of different concentrations (1–10 m*M*) was added into the running buffer.

2.6. Apparatus and operating conditions for CE

All experiments were carried out with a Beckman Coulter (CA, USA) MDQ capillary electrophoresis system equipped with a photodiode-array detector. Beckman Coulter MDQ 32 Karat software was used for instrumental control and data analysis. Separations were carried out in a 50.2 cm (40 cm to detector)×50 μm I.D. uncoated fused-silica capillaries (Polymicro Technologies, Phoenix, AZ, USA). The capillaries were conditioned prior to separation by washing with 1 *M* sodium hydroxide (3 min), deionized water (5 min), then running buffer (5 min). Samples and standards were pressure-injected into the capillary column at 0.5 p.s.i. for 5 s (1 p.s.i.= 6894.76 Pa). Separations were carried out using an electrical voltage of 25 kV, and the temperature of the capillary was maintained at 25 °C while 200 nm was selected as the detection wavelength.

3. Result and discussion

3.1. Optimum colorant separation condition by CE

When the pH of the running buffer was below 8.0, all analytes had similar migration velocities and no single colorant was able to achieve baseline separation. By increasing the pH of the running buffer, differences in migration velocities also increased, and the shape of analytes' peaks became sharper and more symmetrical. The eight colorants had better separations in pH 10.0 buffer and the total separation was completed within 12 min, both Indigo carmine and Allura red AC, however, still had very close migration rates. According to previous research, cyclodextrins can influence the migration behavior of synthetic colorants by guest–host complex formation between colorants and cyclodextrins (CDs) [16]. The addition of α-, β- or γ-cyclodextrin, which each has a different interior cavity, into the running buffer of pH 10.0 resulted in a great improvement in the separation for the eight colorants with the β-form having most influence.

With the exception of carminic acid, the migration rates for all colorants increased noticeably with β-CD, and the best separation was achieved when the concentration of β-CD was 7.0 m*M* as the presence of β-CD could alter most colorants' mobilities by the introduction of guest–host interaction. The migration time of carminic acid was not influenced when β-CD was changed from the concentration of 0.0 to 7.0 m*M*, hence the interaction between carminic acid and β-CD was relatively weak.

The electropherogram of the eight food colorant standards with a borax–NaOH running buffer containing 7.0 m*M* β-CD and a pH of 10.0 is shown in Fig. 1. Baseline resolution was achieved for all analytes, and the relative standard deviations (RSDs) of migration time were in the range of 0.25–0.85% (Table 1). Quantitative analysis was carried out based on the peak area of each colorant. The correlation coefficients (*r*) of the calibration curves, which were constructed from triplicate measurements of each concentration in the region of 1–500 μg/ml for each colorant, were above 0.998 (Table 1), hence the linearity of the calibration curve was relatively good. Since all of the analytes had highly reproduc-

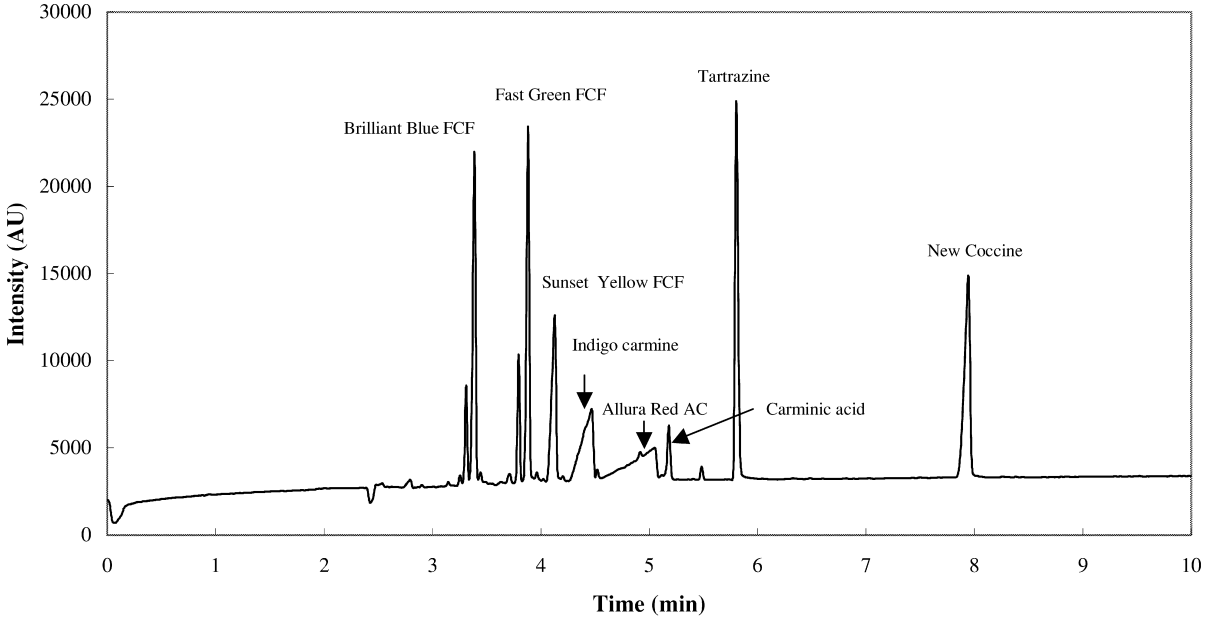

Fig. 1. The electropherogram of the eight food colorants. Separating conditions: borax–NaOH buffer (pH 10.0) containing 7.0 m*M* β-CD was used as running buffer. A voltage of 25 kV was applied to a fused-silica capillary tube of 50.2 cm×50 μm I.D. (40 cm from inlet end to the detection window).

ible migration times and good quantitative results, therefore the borax–NaOH running buffer of pH 10.0 that contained 7.0 m*M* β-CD was selected as the separation condition for milk beverage samples.

3.2. The effect of pH on the colorants' recovery

All of the eight colorants except carminic acid have two or three sulfonic groups in their molecular

Table 1
Average migration times, correlation coefficients of calibration curves, and recoveries of eight colorants spiked in milk beverages by optimum sample pretreatment conditions

Colorant	Migration time[a] (min)	RSD (%)	Correlation coefficients[b] (*r*)	Recovery[c] (%)	RSD (%)
Brilliant blue FCF	3.39	0.25	0.9995	85.0	4.40
Fast green FCF	3.88	0.26	0.9998	108.4	3.56
Sunset yellow FCF	4.11	0.70	0.9986	107.4	2.38
Indigo carmine	4.36	0.41	0.9997	85.2	2.04
Allura red AC	4.72	0.85	0.9997	88.2	4.31
Carminic acid	5.18	0.39	0.9998	101.6	3.77
Tartrazine	5.74	0.31	0.9996	109.1	4.17
New coccine	7.91	0.77	0.9998	99.97	4.96

[a] Value are means of five inter-day replicates.

[b] The calibration curves constructed from triplicate measurements at each concentration in the region of 1–500 μg/ml.

[c] Values are means of triplicate determinations. The milk solution containing carminic acid colorant was adjusted to pH 2.0 at the beginning. After centrifuging at 16 000 rpm to remove the colloids, the SPE followed directly. For milk samples containing the other seven colorants, the sample can be centrifuged directly without any pH adjustment, then its pH value was reduced to 2.0 before SPE.

structures. The interaction between the colorants carrying negative charges and polar polyamide should be predominated by hydrogen bonding, dipole–dipole or London force when some colorants can be caught on polyamide columns at pH values higher than 7.0, i.e. no protonation on polyamide surface occurs under this situation. As the solution becomes more acidic, the polyamide would carry a positive charge due to an increase in fraction of the protonation of N-group at the polyamide. A more acidic solution would cause the polyamide surface to carry more positive charges thus enhancing the cation–anion attraction force.

In order to test the influence of the centrifuged solution's pH on the adsorption capacity of polyamide extraction column, the eight colorants were added to deionized water of various pH values then proceeded with SPE. The results indicated that for all colorants examined, the adsorption capacity increased with a decrease in pH. The highest adsorption capacity occurred at pH 2.0 where more than 1200 μg/ml of colorants could be treated, while the capacity was reduced to 480 μg/ml in the more basic solutions of pH 5.0 or 7.0. According to the results, the strong cation–anion attractive force was believed to be the major determining force for the adsorption behavior of the colorants on polyamide. Hence, in order to obtain higher recovery, the centrifuged solutions were adjusted to pH 2.0 before SPE.

Milk beverages on the market are usually composed of 50% fresh milk in addition to ingredients such as fruit juice, sugar, colorants or other additives in order enhance the taste. Milk contains many protein and fat molecules, thus milk beverages have plenty of colloidal particulates in the aqueous solution. In order to prevent the inner wall of the capillary tube from being clogged and adsorbing the fat and protein molecules in milk, these colloids need to be removed prior to colorant separation.

The usage of high speed centrifugation with various speeds and time were tested for the removal of colloidal particulates. It was determined that by centrifuging the samples at 16 000 rpm for 60 min, this was sufficient to remove the protein and fat molecules contained in most commercial milk products, thus these conditions were used in this study for the removal of these colloids.

After the centrifugation of commercial milk products, an adsorption phenomenon was observed in which some colorants were still adsorbed in the milk samples' precipitates. This observation, however, disappeared for most of the colorants when ethanol was added to milk samples prior to centrifugation. However, most carminic acid was nearly adsorbed on the precipitation of protein and fat, and the soluble fraction of carminic acid in the centrifuged liquid was very low even after the addition of ethanol. As the pH of the milk sample was lowered to 2 from the original 4.93 prior to centrifugation, the adsorption phenomenon was reduced. The binding between protein and carminic acid was inhibited probably due to the removal of ionic attractive force between carboxylic acid group in carminic acid and protein molecules in milk, since the pK_a value of carboxylic acid is approximately 5, and it is not almost dissociated at pH 2.0. On the other hand, changing the pH of milk samples containing the other seven colorants before centrifugation had a negative effect where adsorption had increased. This observation was likely due to an increase in formation of positive charges in protein molecules which in turn caused an enhancement of ionic attractive force.

3.3. The effect of ammonia on the colorants' recovery and separation

To elute the colorants adsorbed in the polyamide column, basic aqueous–methanol solutions ranging from pH 8.0 to 11.0 were used as eluents. Aqueous solution composed of ammonia was more effective than sodium hydroxide aqueous solution at every pH value. The results indicated that the eluting force should be primarily due to the ion-pair attractive force between colorants' anion and ammonium's cation, even though the de-protonation effect on the polyamide surface may be of minor influence.

The concentration effect of ammonia on eluting colorants was also considered, and the data showed that all the colorants examined could be washed out of the polyamide column with either 0.5 or 3% ammonia solution. However, a 3.0% aqueous ammonia–methanol solution caused more serious background peaks in electropherograms, therefore, 0.5% ammonia–methanol solution was chosen as the

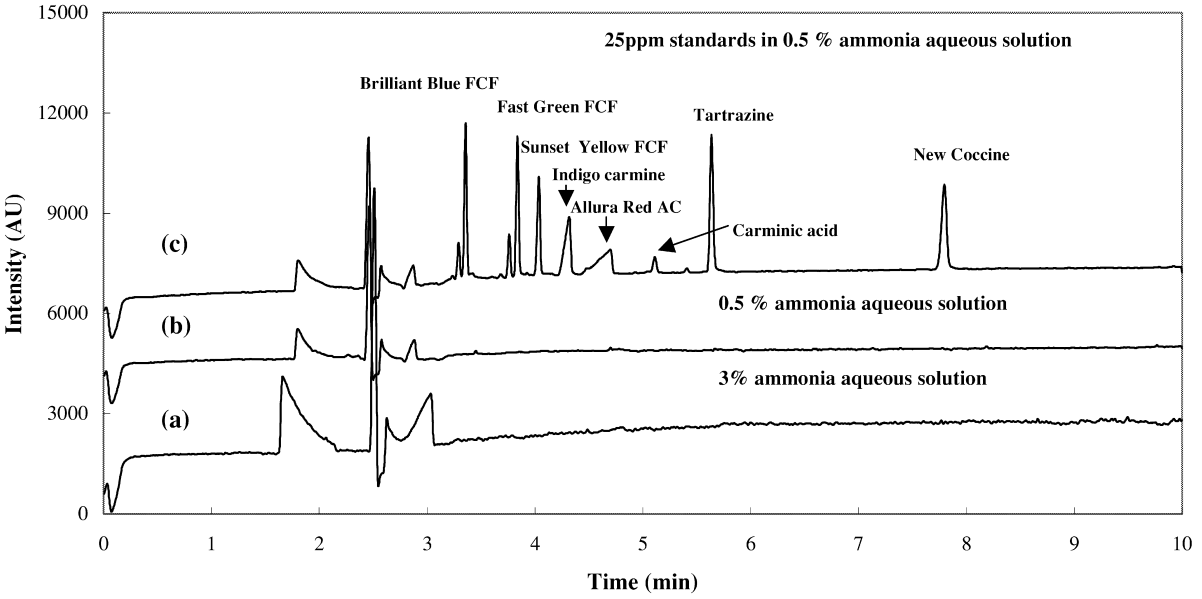

Fig. 2. The electropherograms of eight food colorants with different ammonia concentrations, (a) 3.0% ammonia–methanol solution, (b) 0.5% ammonia–methanol solution, and (c) 25 μg/ml standards in 0.5% ammonia–methanol solution. Other conditions as in Fig. 1.

elution solution in the SPE experiment since it had enough eluting ability and a lower interference in capillary electrophoresis separation (Fig. 2).

3.4. Optimum sample pretreatment

To summarize the above discussion, sample pretreatment would be finished in the following steps. The milk solution containing carminic acid colorant, without anion form, was adjusted to pH 2.0 at the beginning in order to decrease the adsorption degree of the colorant on milk constituents and to increase the recoveries. After centrifugation at 16 000 rpm to remove the colloids, SPE was carried out directly. For milk samples containing the other seven colorants with existing anion form, the sample can be centrifuged directly without any pH adjustment, then its pH value was reduced to 2.0 before SPE to increase polyamide adsorption capacity. Table 1 lists the recoveries of the eight colorants in milk beverages by the sample treatment condition. The recoveries were above 85%, which indicated that the sample treatment condition was suitable for the colorants involved in real milk beverages.

3.5. Real sample separation

In the milk beverage market of Taiwan, the mixed fruit- and apple-flavored milk are preferred by consumers, and yellow colorants are usually used to adjust the liquid milk to a yellow color similar to that of the fruit juice in appearance. Milk beverages with mixed fruit, apple and mango flavors were treated according to the optimum pretreatment conditions, and the analytical results from capillary electrophoresis are shown in Fig. 3. Tartrazine was the major colorant additive in the mixed fruit flavored milk beverages, and its levels were approximately 47.1 μg/ml.

Both tartrazine, and Sunset yellow FCF were found in milk beverage samples that were flavored with apple or mango, and after quantitative analysis of the peak areas, the contents were determined to be 12.6 and 27.7 μg/ml for tartrazine, and 7.6 and 10.6 μg/ml for Sunset yellow FCF, respectively. According to Fig. 3, the sample treatment was suitable for these types of milk beverages since no matrix interferences appeared in their electropherograms.

Strawberry-flavored milk and yogurt are also common on the market, and in this case, red

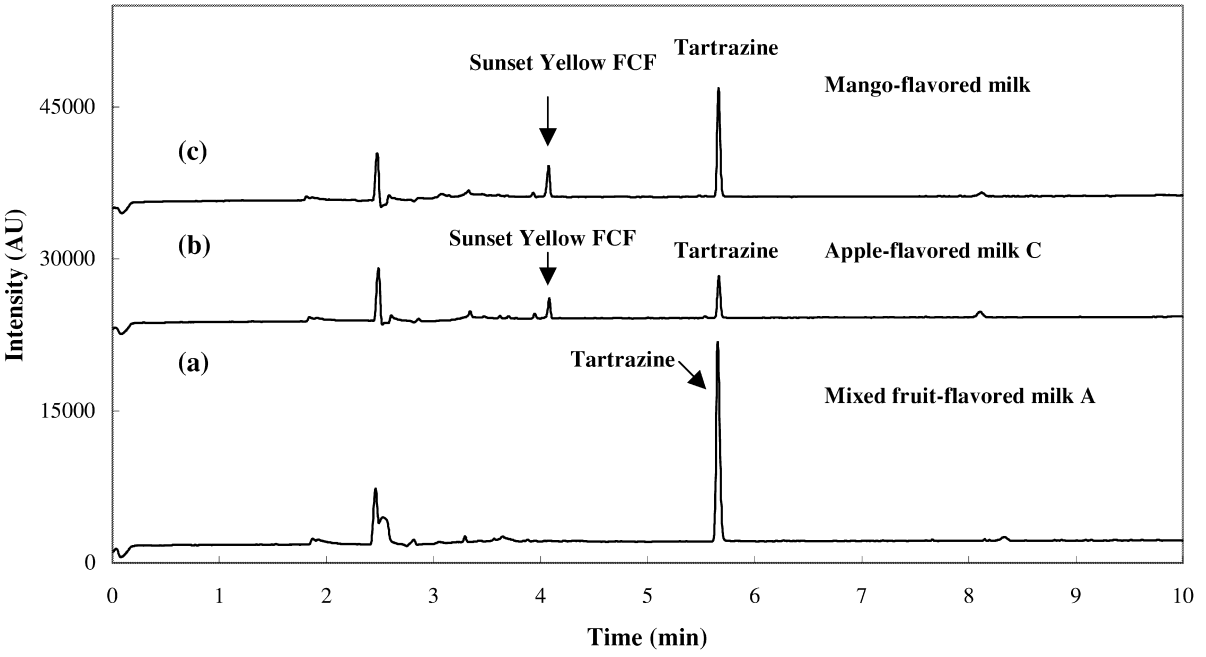

Fig. 3. The electropherograms of three flavored milk beverages. (a) Mixed fruit-flavored milk A, (b) apple-flavored milk C, and (c) mango-flavored milk. Tartrazine or/and Sunset yellow FCF were found in the products. The sample was directly centrifuged without adjusting the pH, followed by polyamide column extraction after lowering the pH to 2.0. Other conditions as in Fig. 1.

colorants are usually added to achieve the desired appearance. The study of red colorants here indicated that there are different adsorption phenomena for these compounds in the centrifugation procedure. After centrifugation at 16 000 rpm, carminic acid was nearly adsorbed on the precipitation of protein and fat, however, no obvious adsorption was observed for New coccine. After properly adjusting the pH of the milk sample, the adsorption of carminic acid disappeared. Fig. 4 shows the electropherograms of commercially available strawberry-flavored milk and strawberry-flavored yogurts, and either New coccine or carminic acid was detected in these samples, respectively. These results once again indicated that the pretreatment of the samples was adequate to concentrate the colorants and to remove the matrix interference for yogurt samples with high viscosity and high bacterial content.

Blueberry-flavored milk beverages are also popular on the milk market, and in this case, both red and blue colorants are usually added in order to obtain their attractive appearances. During the SPE of the clear centrifuged liquid that was maintained at pH 2.0, red colors were caught by the polyamide column, but the liquids which carried blue colors flowed through the column and were obviously lost. After washing out the red colorant from polyamide, the solution carrying the unknown blue color was then again put through the same polyamide column that had been used in extracting the red color from the same sample. The blue color can now be trapped in the polyamide resin and was also eluted successful by 0.5% ammonia–methanol solution. This phenomenon may be due to New coccine and other constituents in milk beverages having higher competition ability for active adsorption sites of polyamide than Brilliant blue FCF, while the adsorption sites that polyamide can provide were not enough for all colorants in milk sample, hence leaving Brilliant blue FCF in the centrifuged liquid. In this situation, repeating the same SPE procedure to trap two colorants or choosing another SPE column with high polyamide loading was suggested. The two eluted solutions above were then combined and analyzed by CE (Fig. 4), and the electropherogram was still relatively satisfactory without any sign of interfer-

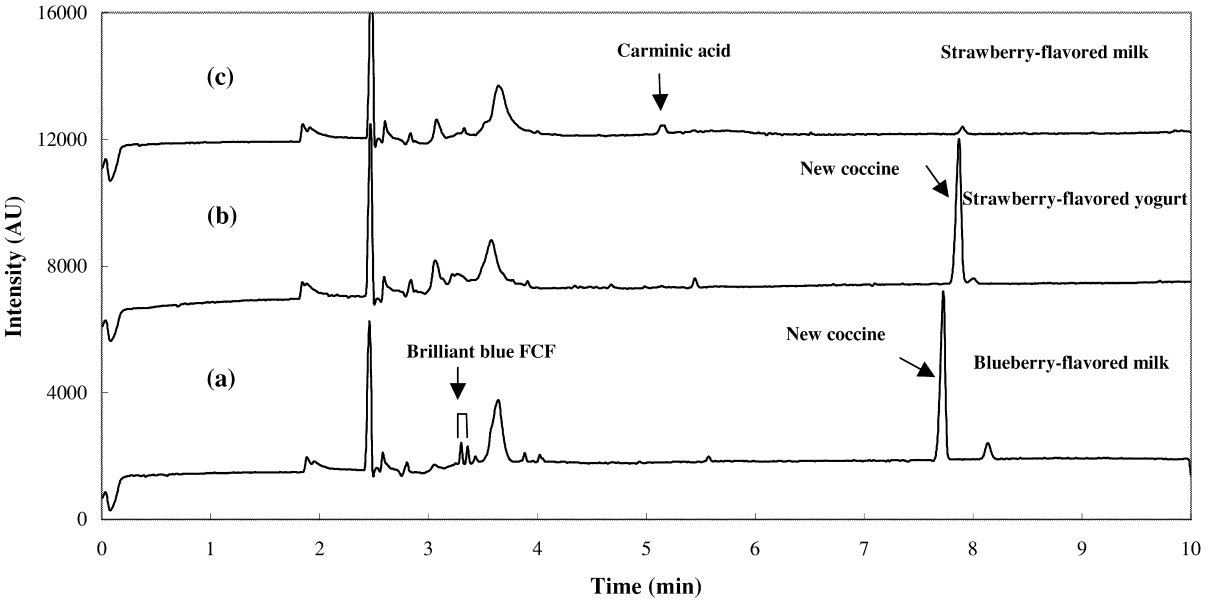

Fig. 4. The electropherograms of commercially available blueberry-flavored and strawberry-flavored milk beverages. (a) Blueberry-flavored milk, (b) strawberry-flavored yogurt, and (c) strawberry-flavored milk. New coccine, Brilliant blue FCF, or carminic acid were found in the products. The samples that contained New coccine were directly centrifuged without a pH change, while the sample that contained carminic acid was adjusted to pH 2.0 prior to centrifugation. Other conditions as in Fig. 1.

ence. The source of red color was identified as New coccine and the blue color was caused by Brilliant blue FCF, and the contents were 10.2 and 27.7 μg/ml, respectively.

3.6. Determining of colorants in commercial milk beverages

Table 2 summarizes the content of colorants in several milk beverages discussed above. The relative standard deviations of the quantitative results were in the range of 0.1–5% with triplicate measurements. The detection limits for the methods were in the range of 0.05–0.40 μg/ml based on S/N ratio of 3. The results reflect that there is a need for the development of an extraction method for determining lower amount of colorants in samples.

4. Conclusion

In this paper, a method for analyzing eight food colorants commonly used in milk beverages was developed using a simple pretreatment method plus a fast separation method based on combined SPE and capillary electrophoresis techniques. A polyamide column was chosen as the SPE stationary phase, and the extraction recoveries were more than 85% for all eight colorants by suitably adjusting the pH value of the sample solution. The CE separation conditions were optimum with a borax–NaOH buffer containing 7.0 mM β-cyclodextrin, where all eight colorants were completely separated within 9.0 min and the resolutions were more than 1.5. The proposed method can decrease effectively the matrix interferences resulting from milk colloidal particulates and increase the detection ability of CE resulting from low sample capacity and narrow optical pathlengths. With milk beverages containing special flavors becoming more popular, analytical methods for colorants involved in milk matrix are important and worth developing. The content of food colorants needs to be monitored because of consumer health and quality control issues in food production. This study demonstrated that the use of a combination of a simple SPE and of the high efficiency separation of CE, was successful in detecting food colorants involved in some popular flavored milk beverages.

Table 2
Contents of colorants determined in commercial milk beverages

Milk sample	Colorant	Concentration[a]	
		μg/ml	RSD (%)
Mixed fruit-flavored milk A	Tatrazine	47.1	0.90
Mixed fruit-flavored milk B	Tatrazine	23.3	0.91
Apple-flavored milk A	Tatrazine	13.4	0.89
	Sunset yellow FCF	6.5	0.11
Apple-flavored milk B	Tatrazine,	17.3	1.23
	Sunset yellow FCF	6.3	2.24
Apple-flavored milk C	Tatrazine,	12.6	1.41
	Sunset yellow FCF	7.6	3.08
Mango-flavored milk	Tatrazine,	27.7	0.51
	Sunset yellow FCF	10.6	1.33
Blueberry-flavored milk A	New coccine,	27.7	1.28
	Brilliant blue FCF	10.2	4.87
Blueberry-flavored milk B	New coccine,	20.42	3.67
	Brilliant blue FCF	4.17	2.71
Strawberry-flavored milk	Carminic acid	25.2	1.12
Strawberry-flavored yogurt A	New coccine	6.5	4.88
Strawberry-flavored yogurt B	Carminic acid	15.9	1.33
Strawberry-flavored yogurt C	Carminic acid	11.35	0.62

[a] Values are means of triplicate determinations.

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