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Article

Cationic Micellar Precipitation for Simultaneous Preconcentration of Benzimidazole Anthelmintics in Milk Samples by High-Performance Liquid Chromatography

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A method using micellar precipitation process using cationic surfactant cetyltrimethyl ammonium bromide at ambient temperature was investigated for simultaneous determination of benzimidazoles coupled with high-performance liquid chromatography (HPLC) analysis. The studied benzimidazoles was selected as model compounds including thiabendazole, albendazole, mebendazole, and fenbendazole. The experimental parameters affected the extraction efficiency, including the kind and concentration of salt, pH, concentration of CTAB, volume of 1-octanol, and the centrifugation extraction time, was optimized. The optimum extraction condition: 0.030 mol L^{-1} CTAB, 10% (m/v) NaCl, pH 4.0, 300 µL 1-octanol and centrifugation time 10 min. Under optimum conditions, enrichment factors of between 60 and 90 fold were obtained, leading to lower limit of detection in the range of 0.5-0.7 µg L^{-1}, depending on the analytes. The calibration range of the method was linear over the wide range of 0.5-1000 µg L^{-1}, with correlation coefficients more than 0.999. Finally, the proposed method was successfully applied in the analysis of benzimidazoles in milk samples.

Keywords: cationic micellar precipitation, HPLC, benzimidazoles, extraction

Introduction

Sample preparation is one of the most important and necessary step before instrumental analysis. Good sample preparation allows not only the analyte to be preconcentrated but also remove the other compounds present in the sample matrix. Conventional sample preparation methods have been used including liquid-liquid extraction (LLE)[1] and solid-phase extraction (SPE).[2,3] However, big disadvantages are the large quantities of solvent utilized and the multiple operation steps needed.[4] Recently, attention is being paid to the development of miniaturized, more efficient and environmentally friendly extraction techniques that could greatly reduce the toxic organic solvent consumption.[5] Alternative extraction approaches based on surfactant (e.g., non-ionic, anionic, and cationic), namely cloud-point extraction or micelle-mediated extraction have also been generally approved as a powerful method for sample preparation. Its major advantages are low cost, simple experimental procedures, high preconcentration factors, personal and environmental safety.[6-9] During the past years, cloud point extraction has become one of the most preferred preconcentration methodologies as "green technology" owing to its following unique characteristics: (*i*) it uses an inexpensive surfactant extractant; (*ii*) it generates less laboratory waste; and (*iii*) its surfactants are less toxic, not volatile and not inflammable, unlike organic solvents used in LLE.[7] It was used as a preconcentration method in simultaneous determination of various compounds (e.g., pesticides, antibiotics, pollutants, etc.) in different sample matrices.[10-14] This method has some disadvantages such as time-consuming, and requires a temperature control and centrifugation for phase separation.

Cationic micellar precipitation (CMP) has been introduced as a new extraction and pre-concentration method

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for the analysis of cadmium, cobalt and nickel in water samples.[6] In this method, the extractant is dispersed into the aqueous solution with precipitation solvent as dispersant. The use of precipitation solvent as an emulsifier solvent in CMP can accelerate the formation of the fine droplets of the extraction solvent in an aqueous sample solution, which increase the dispersion of the water immiscible phase into the aqueous phase, and enhance the mass transfer of the analytes from the aqueous phase to the organic phase.

Benzimidazole anthelmintic drugs are commonly used for prevention and treatment of parasitic infections in agriculture, aquaculture and veterinary practices.[15-17] However, there may be a concern that if withdrawal periods are not adhered to, or if products are administered to animals in unapproved applications, the levels may exceed maximum residue limits (MRLs) in foods.[15] The MRL values range from 10 to 5000 μg kg^{-1} depending on the compound and biological matrix.[18] Therefore, the development of sensitive, effective, and reliable analytical methods is still required to monitor these residues in food samples.

Because of wide spread use and possible health effects, it is desirable to monitor benzimidazoles in the food samples. Gas chromatography (GC) was also applied while extra derivatization step of residues to sufficiently volatilize was required.[19] Capillary electrophoresis (CE)[20] has also been applied in the separation of benzimidazole anthelmintic drugs, but suffers from low sensitivity because of small sample volumes injected and less sensitive detection systems that are employed. High performance liquid chromatography (HPLC) coupled with various detection systems, i.e., ultraviolet (UV),[3,21,22] fluorescence (FL),[23] mass spectrometry (MS)[24,25] and both UV and MS[26] have been accepted as popular and powerful tools for the analysis of benzimidazoles.

This work focuses on the development of the method for the extraction using cationic micellar precipitation (CMP) for benzimidazoles coupled with HPLC analysis. Four benzimidazoles (i.e., thiabendazole, albendazole, mebendazole, and fenbendazole) were selected as model compounds. CTAB were used as precipitating agents in extraction method. This is the first time for hyphenation of a simple CMP system with HPLC for benzimidazole applications.

Experimental

Chemicals and reagents

The standards of benzimidazoles of highest purity were purchased from Sigma-Aldrich including thiabendazole (Italy), mebendazole (USA), albendazole and fenbendazole (China). The chemical structures of the studied benzimidazoles evaluated here are shown in Table 1. The stock standard solutions of each benzimidazole (1000 mg L^{-1}) were prepared by dissolving each benzimidazole standard in 5% (v/v) formic acid/methanol. Methanol and acetonitrile (ACN) of HPLC grade were obtained from Merck (Germany). NaCl and anhydrous Na_2SO_4 were obtained from Ajax Finechem (New Zealand) and CH_3COONa was purchased from Carlo Erba (France). Cetyltrimethyl ammonium bromide (CTAB) was purchased from Calbiochem (Germany). CTAB was prepared in water before use. Acetic acid (glacial), formic acid and 1-octanol were obtained from Merck (Germany). Deionized water was obtained from RiOs™ Type I Simplicity 185 (Millipore Waters, USA) with the resistivity of 18.2 MΩ cm and was used throughout the experiments.

Chromatographic conditions

The HPLC system comprised a Waters 600 multisolvent delivery system, a Rheodyne injector with a sample loop of 20 μL, a Waters 996 photodiode array detector and were recorded at 296 nm. The Millennium software was used for data acquisition. The separation of benzimidazoles was carried out on an Atlantis dC18 column (4.6 × 150 mm, 5.0 μm) from Water (Ireland) with gradient elution using MeOH and 0.1% (v/v) acetic acid, a flow rate of 1.0 mL min^{-1} and detection at 296 nm. The gradient elution was performed as follows: 60% (v/v) MeOH (0-3 min), ramped to 95% (v/v) MeOH (3-10 min). After that, 100% (v/v) MeOH was held for 5 min to wash the excess surfactant from the system. Finally, 60% (v/v) MeOH was held (5 min) to equilibrate the column before the next run.

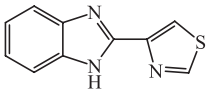
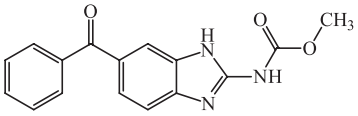
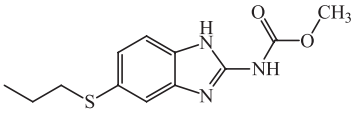
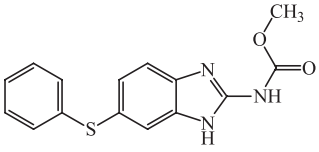
Cationic micellar precipitation (CMP) procedure

An aliquot of a standard (or sample solution) was mixed with NaCl (10%, m/v). After that, 1.0 mL of conc. HCl was added and shaken by hand for a minute. The sample solution was rapidly injected with 300 μL precipitation solvent (0.30 mol L^{-1} CTAB) and 300 μL extraction solvent (1-octanol) using 1-mL syringe. After that, the solution was centrifuged at 3,500 rpm for 10 min to complete the phase separation. The sediment phase floated on the top of the solution due to its density that is lower than that of water. The upper phase was collected and then directly injected into HPLC for analysis.

Milk sample analysis

The studied cow milk samples were commercial

Table 1. The chemical structures of the studied benzimidazoles

Analyte	Structure	Molar mass / (g mol^{-1})	Octanol/ water partition coefficient	pKa	Melting point / °C
Thiabendazole (TBZ)		201.2	2.2 (pH 5); 2.4 (pH 9)	4.64 at 25 °C	293 to 305
Mebendazole (MBZ)		295.2	2.8	8.44	288.5
Albendazole (ABZ)		265.3	1.57	6.90	208 to 210
Fenbendazole (FBZ)		299.3	3.85	–	233

UHT and pasteurized milk. The commercial milks were purchased from a supermarket in Mahasarakham province (Thailand). Milk samples were pretreated using the slightly modified procedure from our previous work.[16] Before analysis, a 5.00 mL of milk samples were mixed well with 0.2 g of anhydrous Na_2SO_4. After that, 1% (v/v) acetic acid in ACN (5.00 mL) was added and shaken vigorously by vortex agitation. Then, the homogenized milk samples were centrifuged at 3,500 rpm for 5 min for complete fat and protein precipitation. The supernatants were filtered through Whatman filter paper No. 1. The solutions were diluted with water to 10.00 mL in volumetric flasks. The 100 µL of acetic acid were added and centrifuged again to ensure complete fat and protein precipitation. Then, the clear solutions were subjected to cationic micellar precipitation procedure, and the extract phase was then analyzed by HPLC. For spiked samples, the samples were fortified with the target analytes at different concentrations (0.05, 0.10, and 0.50 mg L^{-1}) before fat and protein precipitation. All experiments were performed in triplicate.

Results and Discussion

Optimization of the cationic micellar precipitation procedure

To obtain the most favorable conditions of cationic micellar precipitation procedure, various parameters were studied including the kind and concentration of salt, pH, concentration of CTAB, volume of 1-octanol, and centrifugation time. The optimization was carried out on the aqueous solution containing 0.10 mg L^{-1} of each analytes. The extraction efficiency was evaluated in term of enrichment factor (EF), which was defined as the ratio between the analyte concentration in extraction solvent after and before the extraction process.[27] All experiments were operated at least in triplicate.

Generally, the addition of salt decreases the solubility of the analytes in aqueous samples and enhances their distribution into the organic phase.[28] Thus, addition of different electrolyte salts (NaCl, Na_2SO_4 and CH_3COONa) at 10% (m/v) was investigated to study the influence of ionic strength and the results were compared with that obtained from the process without salt addition. From the results (Figure 1), it is clearly seen that the addition of NaCl provided higher extraction efficiency in term of peak area of neonicotinoids. Consequently, the concentration of NaCl on the extraction efficiency of the target analyte were also studied within the range of 0-40% (m/v). The results in Figure 2 demonstrated an improvement of extraction efficiency for all analytes when 10% (m/v) NaCl was added, and the extraction efficiency decreased because sediment phase increased. Therefore, 10% (m/v) NaCl was selected.

Sample pH plays an important role in the extraction procedure because pH value determines the existing form of the analytes, and then the pH of the sample solution affects the extraction efficiency.[29] The effects of pH were studied in the range of 3-9 (data not shown). The conditions of all the other variables were kept constant: sample solution (10.00 mL), NaCl 10% (m/v), 300 µL of

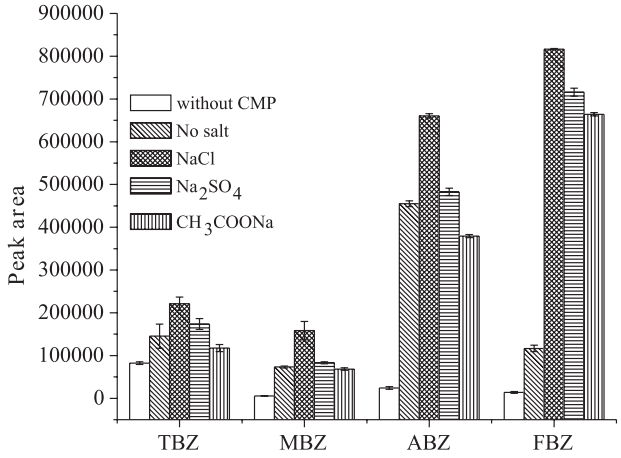


Figure 1. Effect of kind of salt on the CMP of benzimidazoles. The conditions of all the other variables kept constant: sample solution (10.00 mL), sample pH 4, 300 µL of extraction solvent, 0.30 mol L^{-1} CTAB surfactant, and 3,500 rpm for 10 min.

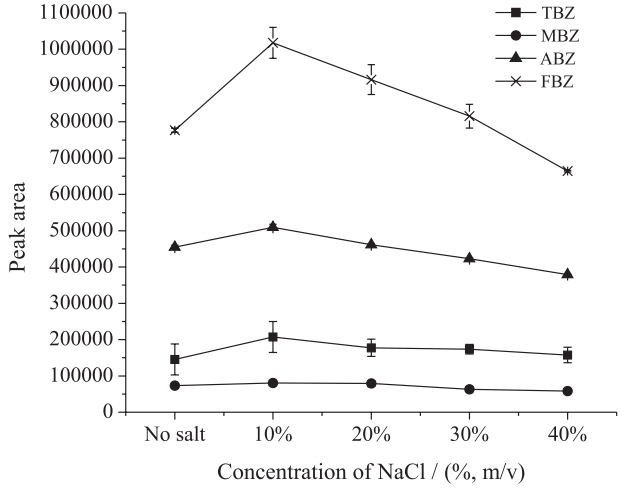


Figure 2. Effect of concentration of salt on the CMP of benzimidazoles. The conditions of all the other variables kept constant: sample solution (10.00 mL), sample pH 4, 300 µL of extraction solvent, 0.30 mol L^{-1} CTAB surfactant, and 3,500 rpm for 10 min.

extraction solvent, 0.030 mol L^{-1} CTAB surfactant, and 3,500 rpm for 10 min. It was found that the pH 4 provided high extraction efficiency in term of peak area. Therefore, pH 4 was selected in this study.

Surfactant could increase the dispersion of extraction solvent into the aqueous solution, thus improving the extraction efficiency. Different concentrations of CTAB were investigated in the range of 0.13-1.00 mol L^{-1}. The results are shown in Figure 3. It can be seen that, the peak area increased by increasing CTAB concentration up to 0.30 mol L^{-1}. Beyond this point, the peak areas of all benzimidazoles kept constant. It is noticed that the optimum CTAB concentration (0.30 mmol L^{-1}) is higher than its micellar concentration of CTAB (0.92 mmol L^{-1}). Therefore, 0.030 mol L^{-1} CTAB concentration was chosen as an optimum value.

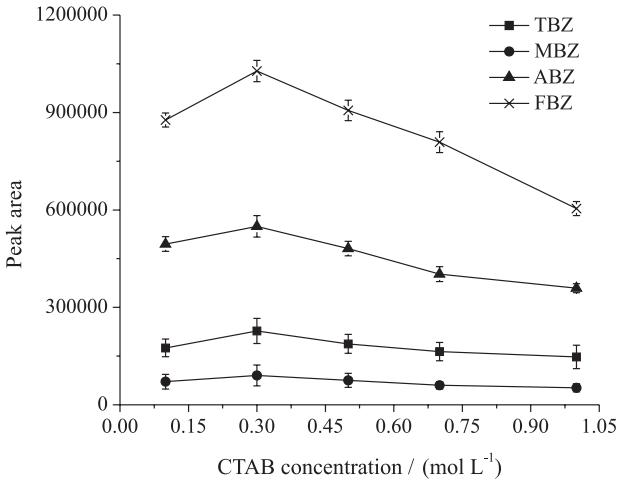


Figure 3. Effect of concentration of CTAB on the CMP of benzimidazoles. The conditions of all the other variables kept constant: sample solution (10.00 mL), sample pH 4, 300 µL of extraction solvent, and 3,500 rpm for 10 min.

The effect of extraction solvent volume (1-octanol) were studied between 50 and 1000 µL (data not shown). It was found that, the extraction solvent volume of 50 µL was too low to provide three replicate measurements. Moreover, the 1-octanol more than 300 µL decreased the peak area of benzimidazoles. Therefore, 300 µL of 1-octanol was selected.

In general, centrifugation time is required to accelerate phase separation between two phases in extraction procedure; so the influence of centrifugation time on the peak area was examined. A series of centrifugation extraction times was studied in the range of 5-30 min at 3,500 rpm. The conditions of all the other variables were kept constant: sample solution (10.00 mL), sample pH 4, 300 µL of extraction solvent, and 0.30 mol L^{-1} CTAB surfactant. The extraction performance of benzimidazoles slightly increased with increasing the time up to 10 min and then kept constant, therefore 10 min was chosen to ensure the extraction performance of the proposed method.

Analytical performance of the method

Table 2 summarizes the analytical characteristics of the optimized method, including regression equation, linear range, limit of detection, limit of quantification and repeatability of the analytes determined after CMP-HPLC analysis. Linear equations in the range of 0.005-1 mg L^{-1} with high correlation coefficients ($r^2 > 0.999$) were obtained. The relative standard deviation (RSD) was determined using five solutions of the 0.002 mg L^{-1} of each benzimidazoles. The RSD values of the retention times and peak areas ranged 0.18-0.37% and 1.43-2.54%, respectively. The sensitivity was evaluated in term of LOD as concentration

Table 2. Analytical performance of the method

Benzimidazole	HPLC					CMP-HPLC					
	Linear range / (mg L^{-1})	LOD[a] / (mg L^{-1})	LOQ[b] / (mg L^{-1})	RSD[c] (n = 6) / %		Linear range / (mg L^{-1})	LOD / (mg L^{-1})	LOQ / (mg L^{-1})	RSD (n = 6) / %		EF[d]
				t_R	Peak area				t_R	Peak area	
TBZ	0.03-5	0.01	0.03	0.32	1.76	0.005-1	0.0005	0.001	0.25	1.77	60
MBZ	0.03-5	0.01	0.03	0.15	2.84	0.005-1	0.0005	0.001	0.37	1.65	60
ABZ	0.10-5	0.03	0.10	0.28	2.63	0.007-1	0.0007	0.002	0.28	2.54	90
FBZ	0.03-5	0.01	0.03	0.14	3.45	0.005-1	0.0005	0.001	0.18	1.43	60

HPLC: high-performance liquid chromatography; CMP-HPLC: cationic micellar precipitation-HPLC; LOD: limit of detection; LOQ: limit of quantification; RSD: relative standard deviation; EF: enrichment factor.

giving the signal-to-noise ratio of 3 (S/N = 3) and ranged between 0.0005-0.0007 mg L^{-1}. The LOQ (S/N = 10) ranged between 0.001 and 0.002 mg L^{-1}. The enhancement factors, calculated by comparing the slopes of the calibration graphs with and without preconcentration, were in the range of 60-90. The chromatograms obtained for the separation of benzimidazoles by direct HPLC injection (Figure 4) and CMP combined with HPLC (Figure 5) were compared.

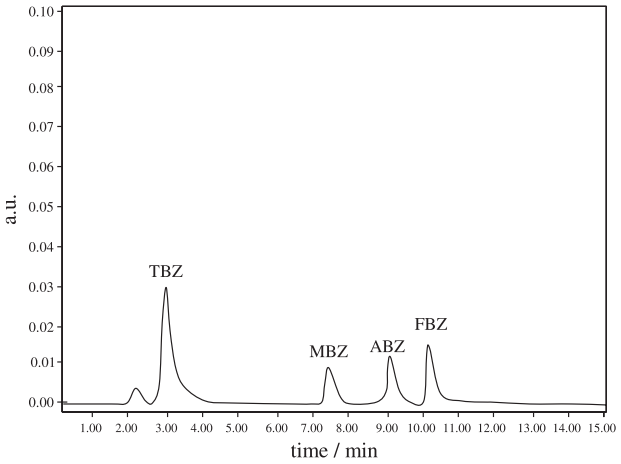


Figure 4. Chromatogram of the studied benzimidazoles obtained from benzimidazoles (2.00 mg L^{-1}) without pre-concentration (direct analysis).

Application to real samples

To evaluate the applicability and accuracy of the developed CMP method in real milk sample determination, each sample was pre-treated as described in Milk sample analysis section, and then extracted using the CMP procedure (see Cationic micellar precipitation (CMP) procedure section) before analysis by HPLC. The results are summarized in Table 3. It was observed that some benzimidazoles was found in the studied pasteurized milk samples. However, the amounts of benzimidazoles found in the pasteurized milk samples were lower than the maximum residue limits (MRLs) established by EU (liver tissue, 100-1000 μg kg^{-1}; kidney and muscle, 50-500 μg kg^{-1}; fat 50-200 μg kg^{-1}, milk 10-100 μg kg^{-1}).

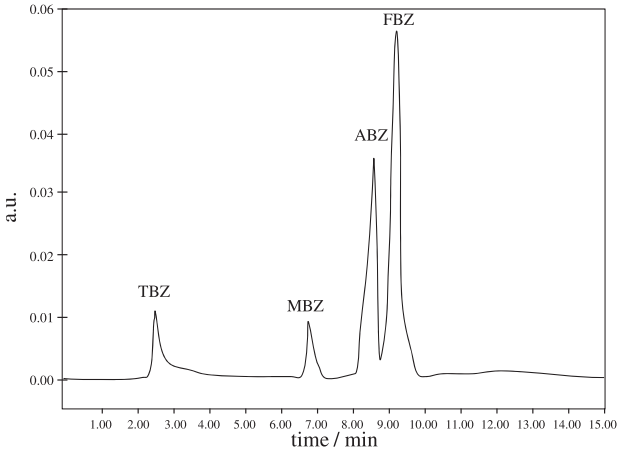


Figure 5. Chromatogram of the studied benzimidazoles (0.10 mg L^{-1}) with preconcentration using CMP condition: 0.30 mol L^{-1} CTAB, 10% (m/v) NaCl, pH 4.0, 300 μL 1-octanol and centrifugation time 10 min.

The recovery experiments were carried out to investigate the method accuracy and precision. The samples were spiked with standard benzimidazoles at three concentration levels of 0.05, 0.10 and 0.50 mg L^{-1} before analysis by the whole analytical processes proposed. As listed in Table 4, the recoveries were observed in the range of 87-106% for pasteurized milk samples. Figures 6 and 7 show the typical chromatograms of the studied samples.

Conclusions

The proposed method gives a precise, sensitive and selective cationic micellar precipitation (CMP) procedure for the simultaneous preconcentration and determination of benzimidazoles coupled with HPLC system. The extraction was performed at ambient temperature in the absence of any organic dispersive solvent and showed reliability with well suited analytical detection range for application in milk samples. CMP for benzimidazoles provides high

Table 3. Analysis of benzimidazole anthelmintics in real samples

Sample	Amount found ± SD (n = 3) / (mg L^{-1})			
	Thiabendazole	Mebendazole	Albendazole	Fenbendazole
Pasteurized milk 01	0.04 ± 0.01	–	1.72 ± 0.13	–
Pasteurized milk 02	0.05 ± 0.02	0.03 ± 0.01	–	1.45 ± 0.35
Pasteurized milk 03	0.08 ± 0.02	–	1.56 ± 0.25	–
Pasteurized milk 04	0.04 ± 0.01	–	–	0.88 ± 0.05
UHT milk 01	–	–	–	–

SD: standard deviation; –: not detected.

Table 4. Recoveries of the studied benzimidazoles in spiked samples

Sample	Spiked / (mg L^{-1})	Thiabendazole		Mebendazole		Albendazole		Fenbendazole	
		RR / %	RSD / %	RR / %	RSD / %	RR / %	RSD / %	RR / %	RSD / %
Pasteurized milk 01	0.05	90	1.2	93	1.7	97	2.4	95	1.3
	0.10	98	2.7	106	2.2	99	1.7	95	1.8
	0.50	95	2.4	101	2.4	88	1.5	97	2.1
Pasteurized milk 02	0.05	105	1.2	94	2.5	93	1.7	89	2.2
	0.10	99	1.6	107	1.8	95	1.8	102	2.5
	0.50	97	2.3	98	1.9	92	1.8	96	2.4
Pasteurized milk 03	0.05	93	2.8	88	2.6	94	2.5	97	2.7
	0.10	89	2.8	93	2.2	102	2.6	99	1.6
	0.50	98	2.5	105	2.5	108	1.9	93	3.1
Pasteurized milk 04	0.05	90	2.7	100	1.9	105	1.9	98	1.8
	0.10	92	1.6	98	1.8	106	1.6	98	2.5
	0.50	93	1.9	99	1.9	98	1.8	87	2.7
UHT milk 01	0.05	91	1.7	105	1.7	98	2.1	95	2.3
	0.10	95	2.1	101	2.2	94	2.5	98	1.9
	0.50	93	2.4	102	2.1	97	2.7	96	1.6

RR: relative recovery (on average, n = 3); RSD: relative standard deviation.

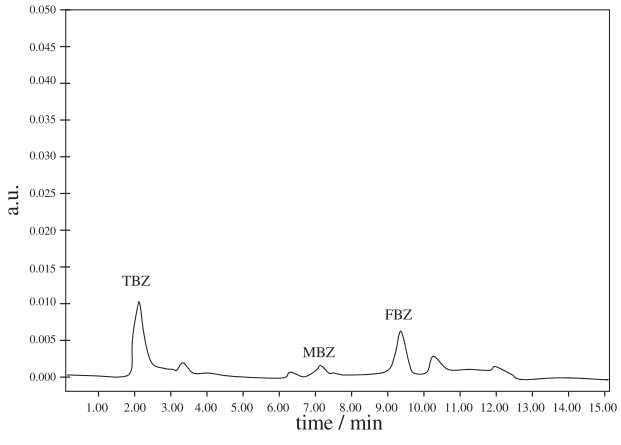


Figure 6. Chromatogram of pasteurized milk 02 by CMP method. The extraction conditions: sample solution (10.00 mL), sample pH 4, 300 µL of extraction solvent, 0.30 mol L^{-1} CTAB surfactant, and 3,500 rpm for 10 min.

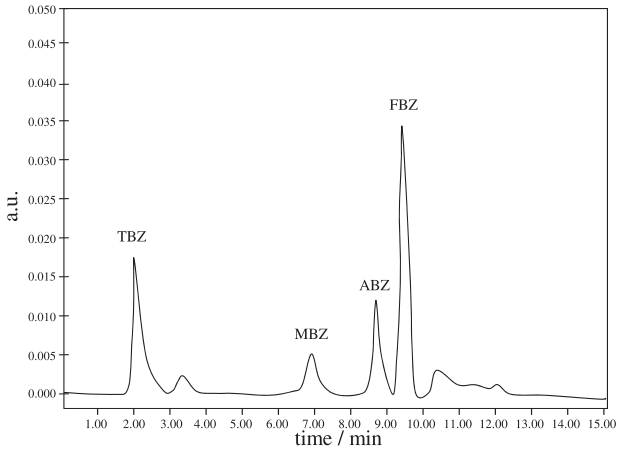


Figure 7. Chromatogram of pasteurized milk 02 with spiked benzimidazole standard (0.05 mg L^{-1} each) by CMP method. The extraction conditions: sample solution (10.00 mL), sample pH 4, 300 µL of extraction solvent, 0.30 mol L^{-1} CTAB surfactant, and 3,500 rpm for 10 min.

efficacy for extraction with the obtained enrichment factor ranging from 60 to 90. To the best of our knowledge this extraction technique has not been reported in the literature and it is being used for the first time for the analysis of benzimidazoles coupled with HPLC.

Acknowledgments

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