

Ionic Liquids- β -Cyclodextrin Polymer for Separation/Analysis Allura Red in Food Samples

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Abstract: Ionic liquids β -cyclodextrin polymer (mono-6-deoxy-6-(1-ethyl-imidazolium)- β -cyclodextrin iodide polymer, ILs- β -CDCP) was synthesized. A novel method based on ILs- β -CDCP solid phase extraction coupled with UV-visible spectrophotometry for the preconcentration / separation allura red (AR) was investigated. The results was shown that AR was adsorbed on ILs- β -CDCP and eluted with sodium dodecyl sulfate (SDS) (1%) rapidly. Under the optimum conditions, the preconcentration factor for AR was 27. The linear range, detection limit (DL), correlation coefficient (R) and relative standard deviation (RSD) were found to be 0.10-9.00 $\mu\text{g/mL}$, 5.2 $\mu\text{g/L}$, 0.9987 and 3.10% ($n=3$, $c=4.00 \mu\text{g/mL}$), respectively. The adsorption mechanism of ILs- β -CDCP on AR was studied through the FTIR analysis and the inclusion constant of ILs- β -CDCP-AR. This proposed method has been successfully applied to the determination of AR in food samples.

Keywords: Allura Red, Ionic Liquids- β -Cyclodextrin Polymer, Solid-Phase Extraction, UV-visible Spectrophotometry

1. Introduction

The advantages of solid phase extraction (SPE) was such as higher enrichment factor, better selectivity, easier operation, lower organic solvent consumption and little environment pollution [1]. The selection of SPE material (solid phase adsorbent) is one of the important factors, which mainly include fiber [2], silica gel [3], chelating resin [4], nano material [5], active carbon [6] and cyclodextrin cross-linking polymer (CDCP) [7]. CDCP as a solid-phase extraction adsorbent was widely used in separation/analysis of metal elements [8]; [9]; [10] and organic compounds [11]; [12]. Functionalized cyclodextrin has become a new hotspot in supramolecular field [13]. Various applications of functionalized cyclodextrins have been reported: biological-based chitosan grafted β -cyclodextrin-removal of benzoic acid [14]; Fe_3O_4 /cyclodextrin polymer nanocomposites-removal of heavy metals [15] and β -yclodextrin/ Fe_3O_4 modified glassy carbon electrode-tryptophan analysis [16]. The ionic liquid functionalized cyclodextrins for separation/analysis was

included (1) chiral stationary phases in HPLC [17]; (2) adsorbent of organic pollutants and heavy metals in sewage [18]; (3) adsorbent of specific ingredient in Chinese medicine [19]. However, ionic liquids functionalized β -cyclodextrin polymer as a solid phase extraction adsorbent for the determination of colorants in food samples seems to be lacking.

Synthetic colorants are common food additives used in food industry, due to their stable nature, bright colors and low price. Allura red (AR) (Figure 1) is one of the eleven synthetic colorants which are allowed to be used within a certain limit in food, such as ice cream, candies, pastries, drinks, jelly and hams [20]. The maximum usage of AR in candies and beverages are 0.3 g/kg and 0.1g/kg, respectively; which are strictly regulated by the hygienic standards for food additives of People's Republic of China [21]; [22].

The safety of excessive use of AR as food additives has been questioned, because of its potentially toxic and carcinogenic, especially harmful to intellectual development

of children [23]; [24]. Therefore, it is significant of accurate determination of AR for food safety.

The AR detection methods include high performance liquid chromatography [25]; [26]; [27], UV-vis spectrophotometry [28], oscillographic polarography [29], voltammetry [30]. Among these methods, UV-visible spectrophotometry has many advantages of lower-cost analysis, easier operation and better accuracy. However, the sensitivity of spectrophotometry is difficult to meet monitoring the lower concentration of AR in samples. So, in this work SPE (ILs- β -CDCP) coupled with UV-visible spectrophotometry was applied to analysis AR in food samples.

2. Experimental

2.1. Materials and Reagents

Centrifuge (Anke Scientific Instrument Factory, Shanghai), timing multifunctional oscillator (Guohua Co., Ltd., China), digital constant temperature water-bath (Guohua Co., Ltd., China) and PHS-25 meter (Shanghai Jinjke Co., Ltd., China) were used for the study.

FTIR spectra were measured with a Bruker Tensor27 spectrometer (Bruker Company, Germany). Samples were pressed into KBr pellets and recorded at the frequencies from 4000 to 400 cm^{-1} with resolution of 4 cm^{-1} .

N, N-dimethylformamide, sodium hydroxide, hydrochloric acid, potassium iodide, methanol, acetonitrile, acetone, 1-ethylimidazole (Beijing Aipuxilong Biotechnology Co. Ltd., Beijing, China), β -CD (Xi'an Hongchang Pharmaceutical Corporation, China), and hexamethylene diisocyanate (Aladdin Reagent Corporation, China) were used in the experiment. The stock solution of 50.0 $\mu\text{g/mL}$ allura red was prepared by being directly dissolved in distilled water and kept in the dark all the time.

2.2. Synthesis and Characterization of ILs- β -CDCP

The synthetic procedure for ILs- β -CDCP were prepared according to the literature [31]; [32]; [33].

The mono-6-deoxy-6-(1-ethyl-imidazolium)- β -cyclodextrin iodide polymer (ILs- β -CDCP) was characterized by FTIR spectra of ILs- β -CDCP (IR/KBr, cm^{-1}) 3373, 2931, 2857, 1712, 1660, 1568, 1257, 1031. The peaks of 2931, 2857 cm^{-1} corresponded to ν_{as} and ν_{s} vibration of methylenes which were the characteristic groups of hexamethylene diisocyanate. The peaks of 1660, 1568, and 1257 cm^{-1} corresponded to the absorption band I, II and III of amide, which confirmed the formation of ILs- β -CDCP.

2.3. Determination Method

At room temperature, the sample solution after treatment and the pH buffer were added into a tube, and then distilled water was added to 40.0 mL. 0.1 g of ILs- β -CDCP was added into the tube. The mixture was shaken on the timing multifunctional oscillator for 20.0 min and then centrifuged. Sodium dodecyl sulfate (SDS) (1%) of 3.0 mL as elution was added into the used ILs- β -CDCP. The mixture was

ultrasonically vibrated at room temperature and then centrifuged to obtain supernatant solution. Supernatant solution was determined with UV-Vis spectrophotometer.

2.4. Sample Preparation

1.0 g Candy was weighed in a small beaker after ground into powder. The powder was dissolved in 30 mL distilled water at 60°C ultrasonically extracted for 30 min and then filtrated to obtain supernatant. The solution was transferred into a 100 mL volumetric flask and diluted to the mark with distilled water.

5.00 g beverage was transferred into a 100 mL volumetric flask and diluted to the mark with distilled water. The sample solution were kept in the dark at 4°C.

2.5. Determination of Inclusion Constant

The procedure of determination of inclusion constant was based on the literature (Wan et al., 2006).

3. Results and Discussion

3.1. Optimization of Adsorption

The factors affecting the adsorption process of AR such as pH, temperature and solution volume were studied and the adsorption behavior of ILs- β -CDCP on AR was compared with that of β -CDCP.

3.1.1. Effect of pH

As shown in Figure. 1, the adsorption efficiency of AR was varied with the pH. It could be concluded that (1) the adsorption efficiency of AR on ILs- β -CDCP (curve 2) was always higher than that on β -CDCP (curve 1); (2) the adsorption efficiency of AR on ILs- β -CDCP was above 95.0% when the pH was in the range of 4.0-7.0. It reached the highest value 98.6% when pH was 4.0.

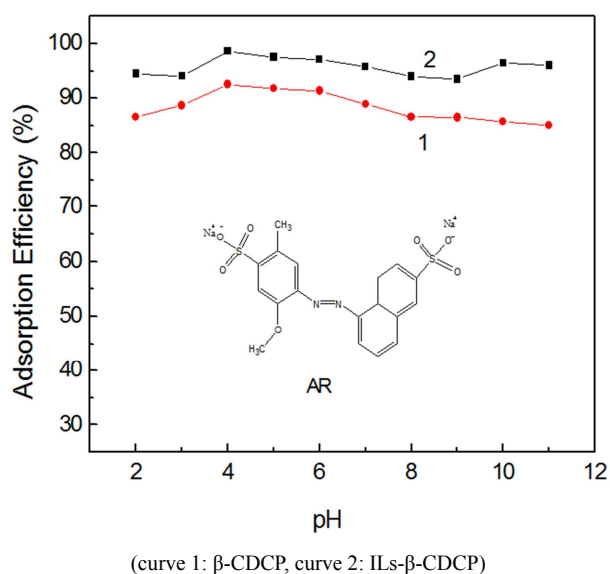


Figure 1. Effect of pH on adsorption efficiency ($c_0=4.00 \mu\text{g/mL}$).

3.1.2. Effect of Adsorption Temperature and Time

The adsorption efficiency of AR on ILs- β -CDPC and β -CDPC at different temperatures (0.0-50.0°C) were studied. The adsorption efficiency of AR on ILs- β -CDPC was higher than that on β -CDPC and was always above 95% from 0.0°C to 50.0°C. The experiment was carried out at room temperature.

The adsorption process was completed within 20.0 min, and the adsorption efficiency remained almost stable (98.0%). 20.0 min as the adsorption time for allura red was adopted.

3.1.3. Effect of the Sample Volume

The adsorption efficiency of AR varied with the increase of sample volume. The amount of AR was fixed at 50.0 μ g and the volume of the sample solution increased from 10.0 mL to 100.0 mL. The adsorption efficiency of AR was above 98% from 10.0 to 30.0 mL and decreased slightly when the sample volume was greater than 30.0 mL. The adsorption efficiency of AR was still above 85% at sample volume of 80.0 mL and 83.2% when sample volume was 100.0 mL. So the largest sample volume allowed was 80.0 mL.

3.2. Adsorption Capacity

The adsorption capacity is defined as the maximum amount of AR adsorbed per gram of the polymer (ILs- β -CDPC). The adsorption capacity of AR on ILs- β -CDPC was studied (Figure. 2). When the concentration of AR was 50.0 μ g/mL (volume: 30 mL), the adsorption of AR for 0.1250g ILs- β -CDPC reached the maximum. The adsorption capacity for ILs- β -CDPC was calculated as 10.90 mg/g.

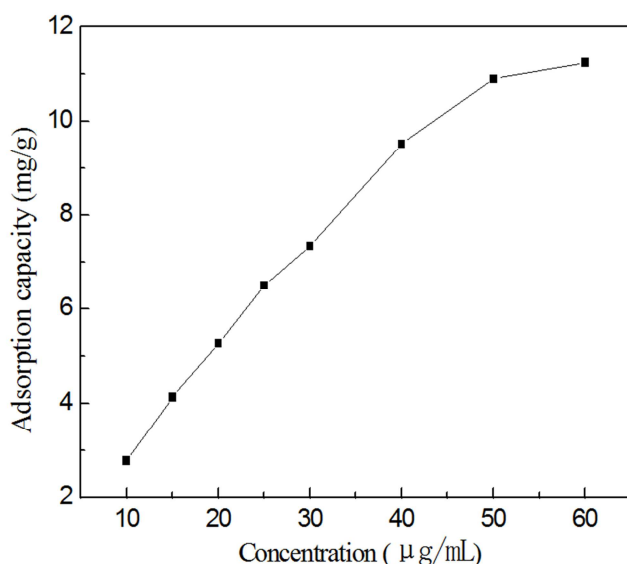


Figure 2. Adsorption capacity.

3.3. Optimization of Elution

Different eluants were investigated. The order of elution efficiency was 1.0% SDS (96.4%) > 75% ethanol-25% ammonia mixed solution (94%) > ethanol (61.4%) > 1mol/L HCl (58.3%) > methanol (48.5%). So 1.0% SDS was adopted as the eluent.

The elution efficiency of AR with 2.0-6.0 mL of SDS (1.0%) was studied. The elution efficiency of AR was above 95% from 3.0 to 6.0 mL. The preconcentration factor was 27 (the quotient of volume before absorption and after elution). And the optimum volume of SDS (1.0%) chosen for this work was 3.0 mL.

The elution process was completed within 10.0 min, and the elution efficiency did not change with a stable elution efficiency of 96.7% thereafter. The elution time of 10.0 min for allura red was adopted.

3.4. Evaluation of Interferents

With a relative error of less than $\pm 5\%$, the influence of some interferents that food samples contain on the determination of AR was studied and the Tolerance ratio (w_{AR}/w_i) was follow: Mg^{2+} , Ca^{2+} , Na^+ , K^+ , NO_3^- , Cl^- 500; SO_4^{2-} , Zn^{2+} 750; Sodium citrate, Glucose 1000; Lemon Yellow 3; Sunset Yellow, Brilliant Yellow, Carminum 2. The results indicated that the majority of these substances in samples had no remarkable interference on the AR determination.

3.5. Analytical Performance of the Method

Under optimum conditions described above, ILs- β -CDPC showed a linear calibration curve within the concentration range of 0.1-9.0 μ g/mL. The equations of calibration graph was $A=0.5123c+0.0213$ (μ g/mL) with a correlation coefficient of 0.9987. The limit of detection (LOD) was 5.2 ng/mL. The relative standard deviation was 3.10% ($n=3$, $c=4.0$ μ g/mL). The enhancement factor, defined as the quotient of volume before absorption and after elution, was 27.

3.6. Determination of Allura Red in Food Samples

The proposed method was applied to determine the amount of AR in certain brands of candy and beverage. The amount of AR in candy and beverage measure up to the national standard. To further verify the viability of the method, recovery experiments were carried out, the results were satisfactory (Table 1).

Table 1. The recoveries of allura red in candy samples and beverages ($n=3$).

Sample	Added (mg/g)	Found (mg/g)	Recovery (%)
Candy	0.00	0.24	—
	0.04	0.26	90.0
	0.08	0.32	95.0
	0.13	0.35	90.3
Beverages	0.00	0.08	—
	0.03	0.11	110.7
	0.10	0.16	104.0
	0.15	0.25	107.1

3.7. Comparison of the Proposed Method with Relevant Literature

The linear range and the limit of detection for the analysis

of AR in real samples obtained by the reported methods, such as high performance liquid chromatography, solid phase extraction coupled with high performance liquid chromatography, cloud point extraction coupled with spectrophotometry and electrochemical method. Compared with other reported methods, the method adopted in the present work obviously had a satisfactory linear range and limit of detection.

3.8. The Adsorption Mechanism of ILs- β -CDCP

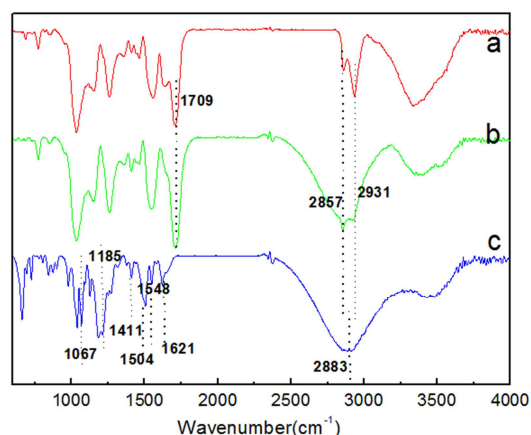
The key point for adsorption mechanism of ILs- β -CDCP was the inclusion effect between the cyclodextrin cavity (ILs- β -CDCP) and AR. In this work, the inclusion effect was studied through FTIR analysis and the inclusion constant.

3.8.1. FTIR Analysis

Figure 3 showed the FTIR spectra of ILs- β -CDCP (curve a), ILs- β -CDCP-AR inclusion complex (curve b) and AR (curve c). Two points were concluded as follows:

(1) Compared with ILs- β -CDCP (curve a), the peaks at 2931 and 2857 cm^{-1} corresponding to the stretching vibration of C-H ($\nu_{\text{C-H}}$) in methylene was a slight changed in shape in ILs- β -CDCP-AR inclusion complex (curve b). This was because the peak at 2883 cm^{-1} of AR (curve c) overlaid in the stretching vibration of C-H ($\nu_{\text{C-H}}$) at the peaks of 2931 and 2857 cm^{-1} , which broadened the original peak at 2931 and 2857 cm^{-1} . It confirmed the formation of ILs- β -CDCP-AR inclusion complex; It was the peak at 2883 cm^{-1} which made spectral signature of ILs- β -CDCP-AR inclusion complex (curve b) different from ILs- β -CDCP (curve a).

(2) The peaks at 1411, 1504, 1548 and 1621 cm^{-1} corresponded to the skeletal vibration of benzene ring ($\delta_{\text{C=C}}$) of allura red, the peak at 1067, 1185 cm^{-1} corresponded to sulfonic acid group of AR; these above characteristic absorption peaks of AR disappeared in the spectra of ILs- β -CDCP-allura red, which illustrated the aromatic ring and sulfonic acid group of allura red were included in the hydrophobic cavity of ILs- β -CDCP.



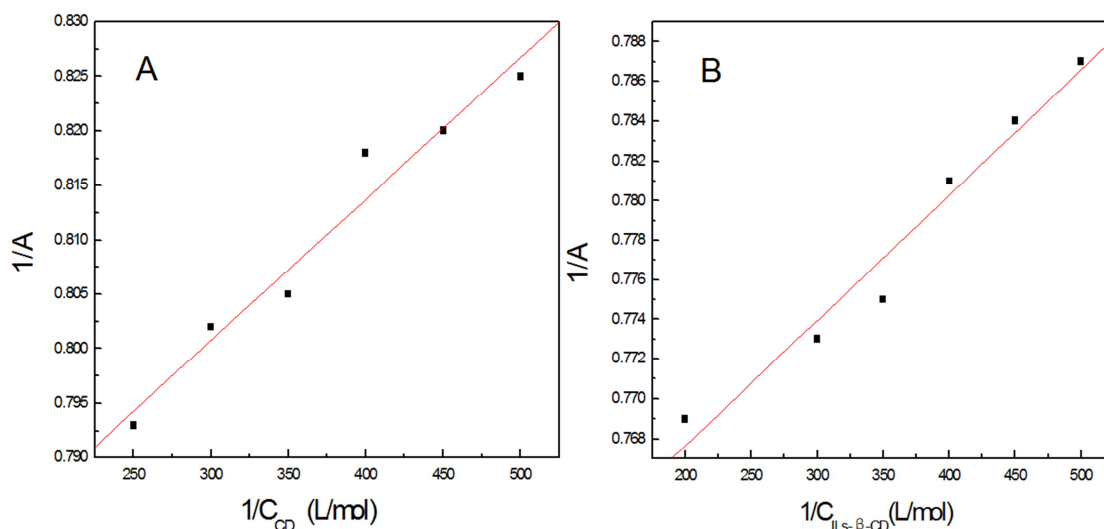
(a) ILs- β -CDCP, (b) ILs- β -CDCP-allura red inclusion complex; (c) allura red

Figure 3. FTIR spectra.

3.8.2. Inclusion Constant

The inclusion constant K is a significant parameter which presents inclusion properties of host-guest molecules. The higher K was, the more stable inclusion complex was. The inclusion constant was calculated according to Benesi-Hildebrand-equation (double reciprocal plot) (Wan et al., 2006). In this paper, the inclusion constants of the monomers of two kinds of polymers (β -CD and ILs- β -CD) and AR were obtained.

The double reciprocal plots of the β -CD-AR inclusion complex and ILs- β -CD-AR inclusion complex were shown in Figure 4. The two double reciprocal plots showed a good linearity with correlation coefficients of 0.9806 for β -CD and 0.9827 for ILs- β -CD. It could be concluded that both β -CD and ILs- β -CD form the inclusion complexes with allura red at the ratio of 1:1. The inclusion constant (K) of ILs- β -CD-AR inclusion complex was 1.19×10^4 L/mol, which was higher than 5.87×10^3 L/mol for β -CD-AR. It indicated that the inclusion ability of ILs- β -CD towards AR was stronger than that of β -CD. It was the reason why adsorption efficiency of ILs- β -CDCP was better than that of β -CD.



(A) β -CD-allura red inclusion complex; (B) ILs- β -CD-allura red inclusion complex

Figure 4. Double reciprocal plot.

4. Conclusion

In this work, ILs- β -CDCP was synthesized as solid phase extraction material to pre-concentrate / separate AR in food samples. The results by using *FTIR analysis and* inclusion constant showed that the adsorption efficiency of ILs- β -CDCP was better than that of β -CDCP. The proposed method for the analysis of allura red in food samples was proved to be satisfactory.

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