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## A novel method for simultaneous analysis of tartrazine and indigo carmine by cloud point extraction using spectrophotometric technique

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### Abstract

Food dyes tartrazine (E-102) and indigo carmine (E132) are a food additive and generally used in cosmetics, foods, and pharmaceuticals. In this work, Brij 58 was used for the simultaneous spectrophotometric determination of tartrazine and indigo carmine in food samples and syrups after cloud point extraction (CPE). Some parameters such as extraction temperature and time, pH, centrifuge speed, surfactant concentration, and salt concentration were optimized. The limit of detection (LOD) of this method was  $0.0046 \mu\text{g mL}^{-1}$  for tartrazine and  $0.0213 \mu\text{g mL}^{-1}$  and for indigo carmine while the relative standard deviations (RSD) of tartrazine and indigo carmine (at low concentration levels  $0.04 \mu\text{g mL}^{-1}$ ) were 1.18% (n=5) and 1.41% (n=5), respectively. This novel CPE method can be used for the reproducible, selective and sensitive determination of tartrazine and indigo carmine in short time in routine analysis.

**Keywords:** Brij 58, Tartrazine (E-102), indigo carmine (E132), cloud point extraction (CPE), surfactant, spectrophotometric determination

### Introduction

Colorants added to foods are extremely used to enhance food color, provide more attractive, appetizing views to food, taste and flavor. Food dye tartrazine (trisodium;5-oxo-1-(4-sulfonatophenyl)-4-[(4-sulfonatophenyl)diazonyl]-4H-pyrazole-3-carboxylate) (E-102) is a substance that gives yellow color to foods and generally used in cosmetics, foods, and drugs. Nonetheless, despite the fact that tartrazine (TAR) is one of the synthetic colorants most likely present in widespread foodstuffs, they may lead to diarrhea, allergies and also could cause kidney problems and liver damage as well if they are consumed excessively (Vidal *et al.* 2018 and Nambiar *et al.* 2017) [33, 22].

TAR is also known as C.I. No. 19140, Food Drug & Cosmetic Yellow No. 5 and Food Yellow No. 4 with European Community (EC) number E 102. Acceptable daily dose of TAR is  $7.5 \mu\text{g g}^{-1}$  by Joint FAO/WHO Expert Committee on Food Additive (JECFA) (WHO, 2016) [35] in 1966 and EU Scientific Committee for Food (SCF) in 1975 and 1984 [4]. At the present time, there is great concern about the use of such additives, because of the number of risks (Zhu *et al.* 2014) [36]. Consuming orally of excessive amount of TAR by human can lead to unforeseen reactions such as blurred vision, rhinitis, migraine, purple skin patches, itching, asthma, intolerance, and eczema (Gupta *et al.* 2005) [13]. The high concentration of TAR in foods when combined with sodium benzoate may cause health complication such as hyperactivities in children (Rovina *et al.* 2017) [26].

Indigo carmine ((2E)-3-Oxo-2-(3-oxo-5-sulfonato-1,3-dihydro-2H-indol-2-ylidene)-5-indolinesulfonate de disodium) (E132) is approved by US FDA as FD & C Blue No.2. Acceptable daily dose of indigo carmine (IC), is  $0-5 \mu\text{g g}^{-1}$  (Otterstatter 1999) [24]. It has been reported by FAO and WHO that IC is extremely safe and biologically inert. On the other hand, current studies on genotoxicity, mutagenesis and metabolism of IC are not sufficient. Severe hypertension (Naitoh and Fox, 1994) [21] or hypotension (Graziano *et al.* 2005) [12] had been reported for IC. So, legislation of law, regulations and strict control over the use of these artificial colorants in alimentary products has become one of the important requirements. Therefore, it is obligated to make and monitor for the identification of TAR, IC and similar colorants in different samples.

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Different methods have been given in the literature for the determination of TAR and IC, such as chromatography (Yalcin and Seyhan 2013; Bento *et al.* 2015; Tavakoli *et al.* 2014; Bonan *et al.* 2013) [34, 4, 32, 7], electrophoresis (Prado *et al.* 2006; Cesla *et al.* 2007; Kuo *et al.* 1998) [25, 8, 19], thin layer chromatography (TLC) (Authority, E.F.S.A. 2014; Tang *et al.* 2015) [3, 30], electroanalytical methods (Mo *et al.* 1992; Nevado *et al.* 1997) [20, 23] and spectrophotometry (Berzas *et al.* 1999; Berzas *et al.* 1999; Sahraei *et al.* 2013; Antakli *et al.* 2015) [4, 5, 27, 1]. Among these analytical methods, spectrophotometry is the rapid, economical, simple and easy method.

The cloud point extraction (CPE) method, which is one of the sample preparation techniques used prior to instrumental analysis is moreover an alternative to the other separation and enrichment methods as sensitive, basic, inexpensive, environmentally sensitive, and non-polluting (Kuar and Gupta 2012; Heydari *et al.* 2015) [15, 18]. There is a certain level of temperature known as cloud point temperature, when the cloud point temperature is reached that level, the nonionic surfactants in the aqueous solution become cloudy. The analyte is then precipitated in a small volume by a surfactant. In addition, the CPE procedure may be referred to as temperature-dependent phase separation or micelle-mediated extraction (Saitho *et al.* 1195; Hinze and Pramauro 1993; Tani *et al.* 1997) [28, 16, 31]. Therefore, CPE can provide a large preconcentration factor and high recovery efficiency because the analyte dispersed in the original matrix volume are bound in a very small volume of micellar phase (Escaireira *et al.* 2009) [10]. CPE is a separation technique as well as preconcentration method.

Surfactants have been used for the determination of TAR, like Triton X-114 (Ateş *et al.* 2011) [2] and Triton X-100 (Nambiar *et al.* 2017) [2]. In addition, the ionic liquid, 1-butyl-3-methylimidazolium bromide ([C4MIM]Br) (Sha *et al.* 2014) [34], was used as a surfactant for in the determination of TAR. A similar situation exists for IC. 1-butyl-3-methylimidazolium hexafluorophosphate, BMIM.PF<sub>6</sub> (ionic liquid), was used as a surfactant for in the determination of IC (Fat'hi *et al.* 2012) [35]. In addition, chromatographic determination was performed for IC by using aqueous biphasic extraction with polyethylene glycol and ammonium sulfate [PEG-2000-(NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>] (Huddleston *et al.* 1998) [17]. As far as I am aware, there is no enrichment of TAR and IC with CPE method using Brij 58, in the literature.

Since yellow-colored TAR and blue-colored IC mixture create a green color, these dyes are used in the various samples for this purpose. For this reason, there is a need for the simultaneous determination of these two food colorants. Therefore, in this work, it was objected to develop a novel CPE method using Brij 58 for the first time for the simultaneous spectrophotometric determination of TAR and IC.

## Material and Methods

### Instrumentation

An Agilent Transport 60 UV spectrophotometer, (Stevens Creek Boulevard, Santa Clara, CA, USA), equipped at one cell (1.00 cm light-beam quartz cells) was used for measuring absorption spectra and absorbance. The samples were centrifuged with 4000 rpm by using Sigma, 1-6P (Osterode am Harz, Germany) centrifuge. pH measurements were carried out using Isolab Laborgeräte GmbH model *pH meter* (Bahnhofstrasse 10, D-97877 Wertheim Germany).

## Chemicals

Brij 58 (polyethylene glycol hexa decyl ether), TAR (trisodium;5-oxo-1-(4-sulfonatophenyl)-4-[(4-sulfonatophenyl)diazenyl]-4H-pyrazole-3-carboxylate), IC ((2E)-3-Oxo-2-(3-oxo-5-sulfonato-1,3-dihydro-2H-indol-2-ylidene)-5-indolinesulfonate de disodium), HCl, NaOH, CH<sub>3</sub>COOH, H<sub>3</sub>PO<sub>4</sub>, NaH<sub>2</sub>PO<sub>4</sub>, Na<sub>2</sub>HPO<sub>4</sub>, K<sub>2</sub>SO<sub>4</sub>, K<sub>2</sub>CO<sub>3</sub>, KCl, KI, anhydrous Na<sub>2</sub>SO<sub>4</sub>, Na<sub>2</sub>CO<sub>3</sub>, and NaCl were purchased from Sigma-Aldrich. All the chemicals are of analytical grade and were diluted using deionized water. For the analytical application, food samples and pharmaceutical preparations purchased from local markets and pharmacies were used.

### Procedure for preparation of solutions

To prepare 1000 mL, 0.10 mg mL<sup>-1</sup> stock solutions of TAR and IC, 100 mg of the dyes were weighed and dissolved in deionized water. Dilute working standard solutions are prepared daily from stock standard solutions. These solutions were stored in the dark at 4 °C. A 10% (w/v) Brij 58 solution was prepared by dissolving 10.0 g of the Brij 58 in 100 mL volumetric flask stirring.

5.0000 ± 0.0005 g of the powdered beverage drink was taken into a 500 mL volumetric flask and diluted with deionized water to the required volume. The appropriate amounts of this solution were then filtered to use for analysis. Appropriate quantities of food samples (fruit candy and fruit jelly) (5.0000 ± 0.0005 g) were dissolved in water, filtered, and completed to volume in 500-mL volumetric flasks. For complete dissolution of jelly samples, it was warmed for 5.0 min and used after filtration. Appropriate volumes were then taken from the sample solution for the application of the suggested extraction procedure for the spectrophotometric determination of the TAR and IC.

Ten milliliters Sitraks<sup>®</sup> syrup and Gayaben<sup>®</sup> syrup were transferred into a 100 mL volumetric flask. The required volumes were completed with deionized water. Afterwards, appropriate amount of this solutions were taken and used for the analysis.

### pH measure

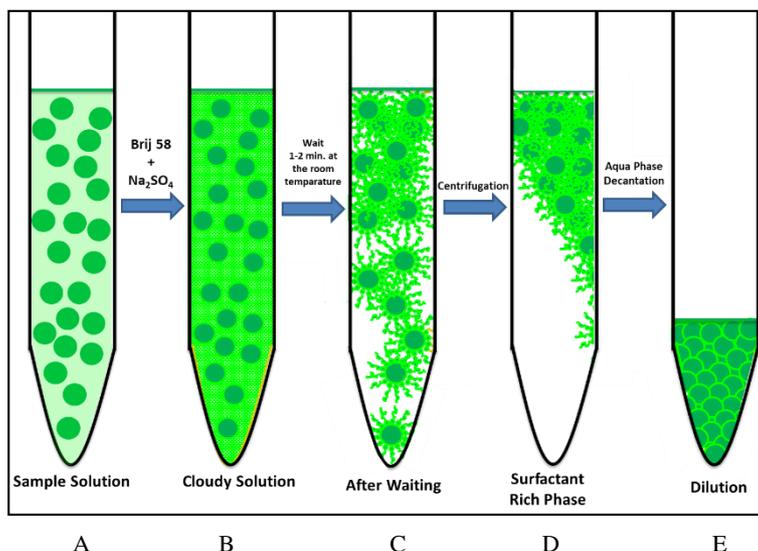
The pH is a significant parameter in the CPE procedure that change the coefficient of dispersion of the analyte between phases. Therefore, the pH effect on TAR and IC extraction efficiency was examined between pH 2 and 11. The H<sub>3</sub>O<sup>+</sup> ion concentration of the solution was adjusted to the desired value using HCl and NaOH and measured a pH meter. TAR and IC solutions were prepared according to the section of 'Developed CPE Method'.

### Developed CPE method

The method was applied to 8 mL of standard TAR and IC solutions transferred to a 15 mL screw cap centrifuge tube (Figure 1 a). This solution was buffered to pH ~5.5 (5-6) with CH<sub>3</sub>COOH and NaOH. Brij 58 (0.75 mL, 10% (w/v)), Na<sub>2</sub>SO<sub>4</sub> (2.0050 ± 0.0005 g) was added and diluted to 10 mL with deionized water (Figure 1 b). This mixture is stirred to dissolve. The mixture left at 25 °C (~the room temperature) for 1-2 minutes until cloudy phenomena occurred (Figure 1 c). The mixture was centrifuged at 4000 rpm for 5 minutes for phase separation (Figure 1 d). The surfactant-rich phase was separated, and it was amassed on the top of the tube (Figure 1 e).

Eventually, the surfactant-rich phase remaining by removal of the aqueous phase was diluted with water up to 1 mL. The signals of analytes were measured by UV-visible spectrophotometry against blank at 425 nm for TAR and 610 nm for IC. The experiment was performed five (5) time in

each as replicate. The developed method was applied to the powdered beverage, fruit candy, fruit jelly and two drugs as explained in 'Procedure for Preparation of Standard and Sample Solutions' section.

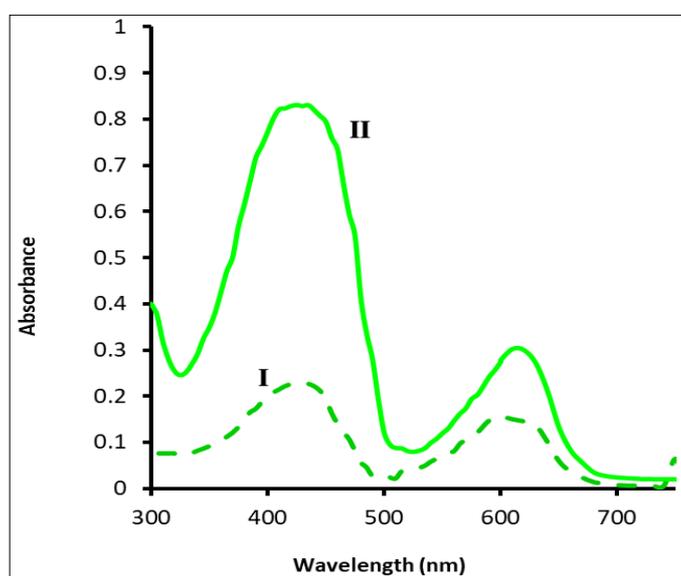


**Fig 1:** a- Sample solution, b- Mixture after Brij 58 + Na<sub>2</sub>SO<sub>4</sub> added, c- After incubation, d- After centrifugation, e- After decantation of 1 mL of water-soluble surfactant-rich phase

### Result and Discussion

The maximum absorbances at 425 nm and 610 nm were observed for 0.04-2.50  $\mu\text{g mL}^{-1}$  TAR standard solution and 0.5-2.50  $\mu\text{g mL}^{-1}$  for IC standard solution, respectively (Figure 2 I). There was no significant change in the maximum wavelengths at maximum absorbances of the standard TAR and IC solutions by adding the surfactant (Brij 58) to the

medium (Figure 2 II). Therefore, all measurements of the quantity of light absorbed by analytes were made at these wavelengths. The effects of the major variables such as the medium H<sub>3</sub>O<sup>+</sup> concentration, Brij 58 concentration, Na<sub>2</sub>SO<sub>4</sub> concentration, incubation time and temperature, were optimized to achieve recovery and high sensitivity.



**Fig 2:** Absorption spectra for 2.5  $\mu\text{g mL}^{-1}$  TAR and 2.5  $\mu\text{g mL}^{-1}$  IC, I- without extraction, II- after CPE

### The effect of pH

pH indicates the intensity of basic or acidic character at a given temperature. Measurement of pH is one of the most important parameters that is required to be optimized in determining TAR and IC within desired range. (Figure 3a)

showed that the highest analyte signal was observed at pH 5-6 (slightly acidic). Therefore, all the experiments were achieved at pH ~5.5 (5-6) and also the absorbances of TAR and IC were measured at 425 nm and 610 nm as well.

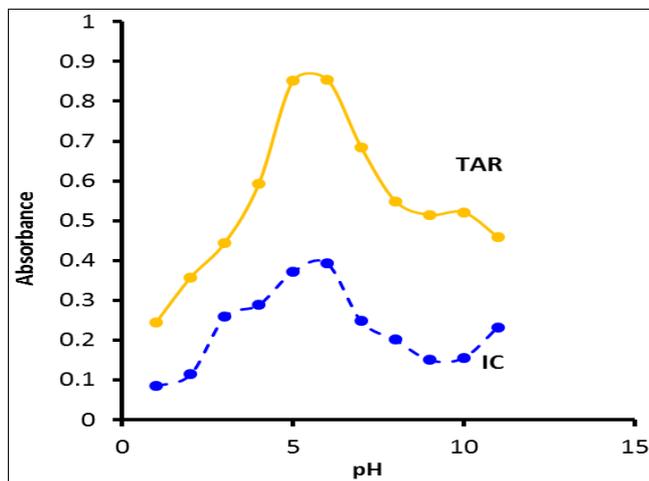


Fig 3(A)

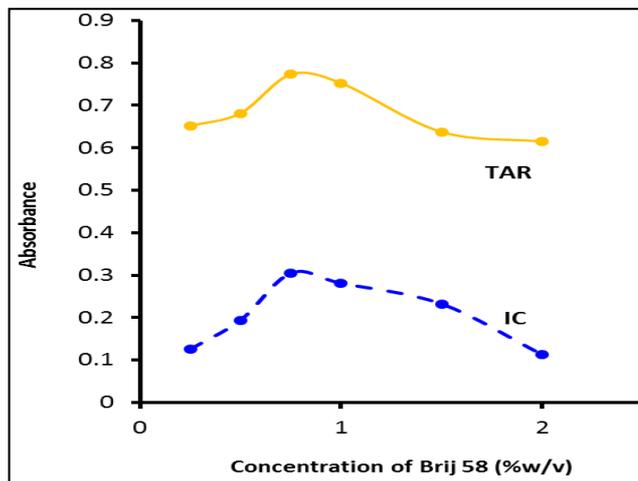
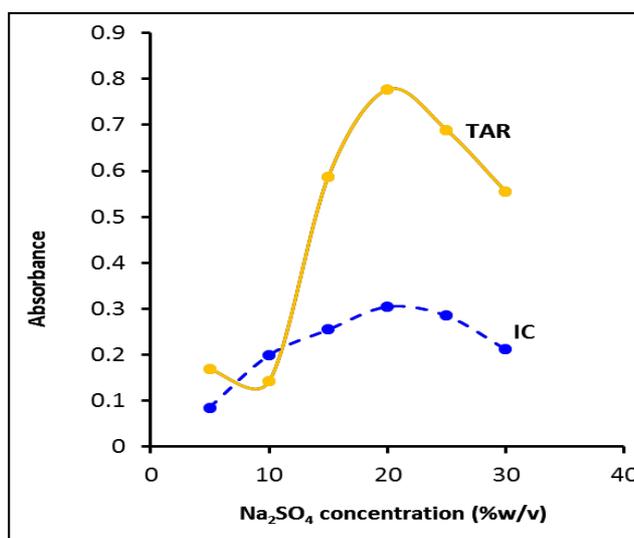


Fig 3(B)



**Fig 3(A-C):** The optimization parameters of the proposed preconcentration method. Conditions:  $2.5 \mu\text{g mL}^{-1}$  TAR and  $2.5 \mu\text{g mL}^{-1}$  IC; pH  $\square$  5.5 (5-6); 0.75% (w/v) Brij 58; 2.00 g Na<sub>2</sub>SO<sub>4</sub>; incubation at 25 °C.

#### The effect of nonionic surfactant concentration

The surfactant concentration is a significant factor in the CPE process. For this reason, the effect of concentration of nonionic surfactant (Brij 58) on the absorbances of TAR and IC was well-studied. The increase of the concentration of Brij 58 up to 0.75% (w/v) has been raised up the absorbance. However, absorbance decreased at concentrations higher than 0.75 (w/v). Therefore, the concentration of Brij 58 at 0.75% (w/v) was chosen as the concentration that maximizes the absorption of TAR and IC as shown in Figure 3b.

#### Effect of Na<sub>2</sub>SO<sub>4</sub>

It is important to optimize the salt concentration, which is one of the significant parameters that increases the extraction efficiency. Salt helps phase separation by increasing the mass transfer of the analyte between phases and reducing the CPT (Cloud Point Temperature). Therefore, the generally used salts such as Na<sub>2</sub>SO<sub>4</sub>, Na<sub>2</sub>CO<sub>3</sub>, NaCl, KI, KCl, K<sub>2</sub>SO<sub>4</sub> and K<sub>2</sub>CO<sub>3</sub>, was tried and their effects on the extraction process was examined. The best result was obtained with sodium sulfate. Thus, the effect of concentration of Na<sub>2</sub>SO<sub>4</sub> on the absorbances of TAR and IC was examined. The results are given in Figure 3c. As can be seen in this figure, the absorbance decreased at concentrations greater than 0.75% (w/v) concentration of sodium sulfate. Therefore, 0.75% (w/v) Brij 58 was used throughout the work.

#### The effect of other experimental factors

The incubation time and equilibrium temperature are two other significant parameters in this procedure. Because, these parameters are necessary to increase extraction efficiency and to achieve easy phase separation. The studied temperature range from room temperature to 70 °C and the maximum extraction efficiency was observed at room temperature. In fact, the extraction efficiency at 60 °C for TAR increased slightly. However, at temperatures above 30 °C, the extraction efficiency for the IC is reduced. Moreover, when the TAR and IC are analyzed together, the green color of the solution becomes yellow and light brown. The incubation time in the range of 0-40 min was optimized. The maximum absorbance was observed at 1-2 min. The forward studies were carried out at these optimum conditions.

#### Effect of interfering ions and food dyes

In this work, the effects of potential ions for the determination of  $2.50 \mu\text{g mL}^{-1}$  TAR and IC were examined using the developed method. The tolerance limit was investigated by considering the amount of foreign species which caused the change in absorbance to be less than  $\pm 5\%$ . The results are given in Table 1.

**Table 1:** The effect of foreign ions on the determination of TAR and IC in the optimum conditions,  $C_{TAR} = 2.5 \text{ mg L}^{-1}$   $C_{IC} = 2.5 \text{ mg L}^{-1}$ 

Foreign ions (I)	Tolerance limits (TAR: I)	Tolerance limits (IC: I)
$\text{Na}^+, \text{K}^+, \text{Mg}^{2+}$	1:1000	1:1000
$\text{Fe}^{3+}, \text{Al}^{3+}, \text{Hg}^{2+}, \text{Mn}^{2+}, \text{Pb}^{2+}, \text{Be}^{2+}, \text{Co}^{2+}, \text{Zn}^{2+}, \text{Ni}^{2+}, \text{Cd}^{2+}, \text{Sr}^{2+}, \text{Sn}^{2+}, \text{Si}^{2+}$	1:200	1:100
$\text{NO}_3^-, \text{SO}_4^{2-}, \text{F}^-, \text{Cl}^-, \text{Br}^-, \text{I}^-$	1:2000	1:2500
$\text{SCN}^-, \text{B}_4\text{O}_7^{2-}, \text{HPO}_4^{2-}, \text{AsO}_4^-$	1:100	1:80

### Analytical performance

Correlation graphs of increasing concentrations of TAR and IC versus the measured absorbance were obtained at the optimal conditions. Dynamic range were obtained between  $0.04\text{-}2.50 \mu\text{g mL}^{-1}$  and  $0.50\text{-}2.50 \mu\text{g mL}^{-1}$  with equations of  $A = 0.3572C (\mu\text{g mL}^{-1}) + 0.0027$  ( $R^2=0.9997$ ) and  $A = 0.0992C (\mu\text{g mL}^{-1}) + 0.0526$  ( $R^2=0.9997$ ) for TAR and IC, respectively. The limits of detection were  $0.0046 \mu\text{g mL}^{-1}$  TAR and  $0.0213 \mu\text{g mL}^{-1}$  IC, respectively. As can be seen from LOD, the developed method has low LOD and can be used trace analysis of these colorants in different syrups and food samples.

### Determination of TAR and IC in syrups and food samples

This method was developed for the spectrophotometric determination of TAR and IC. It was applied to a different

drug and food samples (powder drink, fruit sugar and fruit jam). TAR and IC analysis in the samples were performed as indicated in the 'Recommended CPE Method' explained in the experimental section. The recovery of TAR and IC from spiked real samples ranged from 98.17-104.92%. This shows that the applicability of the proposed method for the simultaneous CPE of TAR and IC from syrups and food samples (Table 2). The acceptable Daily Intake was given as  $0.0\text{-}7.5 \mu\text{g g}^{-1}$  and  $0.0\text{-}5.0 \mu\text{g g}^{-1}$  body weight by World Health Organization (WHO) for TAR and IC, respectively (WHO 2016) [35]. Therefore, the results indicates that the proposed CPE method can be used accurately for spectrophotometric determination of TAR and IC in syrups and food samples.

**Table 2:** Determination of TAR and IC in food samples and syrups after the proposed CPE

Sample	Added, TAR ( $\mu\text{g mL}^{-1}$ )	Added, IC ( $\mu\text{g mL}^{-1}$ )	Found, TAR $\pm$ CL <sup>a</sup> ( $\mu\text{g mL}^{-1}$ )	Found, IC $\pm$ CL <sup>a</sup> ( $\mu\text{g mL}^{-1}$ )	Recovery <sup>b</sup> , % TAR	Recovery <sup>b</sup> , % IC
<b>Foods</b>						
<b>Powdered beverage</b>	-	-	$1.04 \pm 0.04$	-	-	-
	0.50	0.50	$1.56 \pm 0.03$	$0.51 \pm 0.01$	99.10	102.46
	1.00	1.00	$2.06 \pm 0.05$	$0.98 \pm 0.02$	98.26	98.17
<b>Fruit candy</b>	-	-	$0.60 \pm 0.01$	$0.32 \pm 0.04$	-	-
	0.50	0.50	$1.10 \pm 0.01$	$0.81 \pm 0.03$	100.78	98.34
	1.00	1.00	$1.59 \pm 0.04$	$1.34 \pm 0.05$	99.10	101.71
<b>Fruit jelly</b>	-	-	$1.23 \pm 0.03$	$0.47 \pm 0.02$	-	-
	0.50	0.50	$1.73 \pm 0.03$	$0.99 \pm 0.01$	99.10	104.92
	1.00	1.00	$2.23 \pm 0.05$	$1.46 \pm 0.03$	100.50	99.35
<b>Pharmaceutical preparations</b>						
<b>Sitraks<sup>®</sup></b>	-	-	$0.32 \pm 0.01$	-	-	-
	0.50	0.50	$0.82 \pm 0.01$	$0.49 \pm 0.02$	98.88	98.28
	1.00	1.00	$1.32 \pm 0.03$	$0.99 \pm 0.01$	99.36	99.47
<b>Gayaben<sup>®</sup></b>	-	-	$0.72 \pm 0.04$	$0.62 \pm 0.02$	-	-
	0.50	0.50	$1.23 \pm 0.03$	$1.13 \pm 0.03$	101.61	101.61
	1.00	1.00	$1.72 \pm 0.02$	$1.62 \pm 0.03$	100.06	99.43

<sup>a</sup>CL is confidence limits,  $(\frac{ts}{\sqrt{n}})$ ; n=5

$$^b\text{Recovery} = \frac{(C_2 - C_1)}{C_3} \times 100$$

$C_1$  = Measured sample concentration (blank)

$C_2$  = The measured "spiked sample" concentration (blank + spike)

$C_3$  = Spike concentration

### Comparison with reported methods

Table 3 shows the comparison of some remarkable properties of the new method with similar methods in the literature available for determination of TAR and IC in syrups and food samples. All these methods for determination of TAR are based on CPE. The limit of detection value obtained for TAR in the present study is excellent compared to similar methods (Nambiar *et al.* 2017; Ates *et al.* 2011; Sha *et al.* 2014) [22, 2,

29]. Moreover, the present method has been obtained better result than repeatability and recovery values of these methods. A similar literature comparison for IC is given in Table 4. However, this comparison is very limited because, there is no study in the literature for the spectrophotometric determination of IC after CPE.

### Conclusion

The work explains the use of CPE for the extraction of TAR and IC from commercially available pharmaceutical preparations and food samples. The extraction of TAR and IC by CPE using Brij 58 before spectrophotometric determination was reported for the first time. The developed CPE technique has some advantages such as environmental friendliness, sensitivity, selectivity, high extraction efficiency, and low cost. This novel CPE method, developed for precise, reproducible, and selective spectrophotometric determination of TAR and IC in a short time, can be used in ordinary analysis.

**Table 3:** Comparison of similar extraction method for determination of TAR reported in the literature

Surfactant	Salt	pH	Inc. <sup>a</sup> Temp. (°C)	Inc. <sup>a</sup> Time (dk)	Dilution Solvent	Linear Range $\mu\text{g mL}^{-1}$	LOD $\mu\text{g mL}^{-1}$	RSD %	Recovery %	Remarks	Ref.
Triton X-100 0.025 M, 5 mL, CTAB $0.4 \times 10^{-3}$ M, 2 mL	KCl 0.14 M, 7 mL	2.5	70	60	Water 2 mL	0.04-4.00	$13.3 \times 10^{-3}$	1.92	96.5- 103.9	1. Analysis time is very high, 2. A second surfactant was used.	(Nambiar <i>et al.</i> 2017) [22]
Triton X- 114% 10, 3 mL	NH <sub>4</sub> OAc 0.1 M, 1 mL	-	50	30	Acetone 10 mL	0.02-0.30	$4.0 \times 10^{-2}$	5.30	69.60- 116.00	1. LOD is high, 2. Repeatability is low.	(Ates <i>et al.</i> 2011) [2]
[C <sub>4</sub> MIM]Br 0.3 mL	K <sub>2</sub> HPO <sub>4</sub> 3.00 g	2.0-11.0	-	-	-	0.01-50.00	$5.2 \times 10^{-3}$	5.60	93.50- 98.00	1. Repeatability is low.	(Sha <i>et al.</i> 2014) [29]
Brij 58% 10, 0.75 mL	Na <sub>2</sub> SO <sub>4</sub> 2 g	~5.5 (5.0- 6.0)	25	-	Water 1 mL	0.04-2.50	$4.6 \times 10^{-3}$	1.18	98.26- 101.06	1. Analysis in less time, 2. Repeatability is high. 3. Highly sensitive and selective	This work

<sup>a</sup>Inc.: Incubation**Table 4:** Comparison of similar extraction method for determination of IC reported in the literature

Surfactant	Salt	pH	Inc. <sup>a</sup> Temp. (°C)	Inc. <sup>a</sup> Time (dk)	Dilution Solvent	Linear Range $\mu\text{g mL}^{-1}$	LOD $\mu\text{g mL}^{-1}$	RSD %	Recovery %	Remarks	Ref.
BMIM.PF <sub>6</sub> , 1 mL	KCl 0.1 M	3.0	45	-	-	-	-	-	>95.00		(Fat'hi <i>et al.</i> 2012) [11]
Brij 58% 10, 0.75 mL	Na <sub>2</sub> SO <sub>4</sub> 2 g	~5.5 (5.0- 6.0)	25	-	Water 1 mL	0.50-2.50	$21.3 \times 10^{-3}$	1.18	98.17- 104.92	1. Analysis in less time, 2. Repeatability is high. 3. Highly sensitive and selective	This work

<sup>a</sup>Inc.: Incubation

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