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Research Paper

Simultaneous Determination of Binary Mixtures of Tartrazine and Quinoline Yellow Food Colorants in Various Food Samples and Cosmetic Products in Micellar Media by H-Point Standard Addition Method (HPSAM)

Kaur Amandeep and Gupta Usha*
Department of Chemistry, Punjabi University, Patiala-147002, Punjab, INDIA

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Abstract- *H-point standard addition method (HPSAM) was applied for simultaneous determination of Tartrazine and Quinoline yellow food colorants. The method is based on the complexation of food dyes Tartrazine and Quinoline yellow with potassium chromate or Cr (III) as complexing agent at pH 6.0 and solubilizing the complexes in Tween-80 micellar media. Tartrazine and Quinoline yellow can be determined simultaneously in the concentration range 2.67-12.0 and 0.954-2.39 µg /mL respectively. This method has satisfactorily been applied for the determination of Tartrazine and Quinoline yellow in synthetic binary mixture and commercial products. The accuracy and reproducibility of the method has been checked in their synthetic mixture.*

Keywords: Tartrazine, Quinoline yellow, Chromium chloride Cr (III), Micellar media, HPSAM.

Introduction

The term color additive can be applied to any dye, pigment or other substance artificially made or obtained from a vegetable, animal, mineral. Color [1], is a vital constituent of food and probably the first characteristic perceived by the human senses. Food coloring [2] is any substance that is added to food or drink to change its color probably, for economic reasons. These dyes are used to supplement and enhance natural colors destroyed during processing or storage and substantially increase the appeal and acceptability of food stuffs [3].

Tartrazine [4,6], known as FD and C Yellow 5 is a synthetic lemon yellow azo dye used as a food coloring . It is water soluble Many foods contain tartrazine in varying proportions, depending on the manufacturer commonly include confectionery, cotton candy, soft drinks (Mountain Dew), energy drinks, instant puddings .Tartrazine [7] appears to cause the most allergic and intolerance reactions of all the azo dyes, particularly among asthmatics and those with an aspirin intolerance.[8] A variety of immunologic responses have been attributed to tartrazine ingestion, including anxiety, migraines, clinical depression, blurred vision, itching, general weakness, heat waves, feeling of suffocation, purple skin patches, and sleep disturbance [9] .

Quinoline yellow [10,12] is a yellow food dye. The color, which lacks the sulfonate groups, is a related form that is soluble in water. Food colors and preservatives increased levels of hyperactivity and decreased levels of intelligence in children [13,14], but it is unclear which component of the mixture may be responsible for the effects. Quinoline yellow is one of the colors that the Hyperactive Children's Support Group recommends be eliminated from the diet of children. It is used in coloring food, and cosmetics. But it is banned in some countries as US, Australia, Japan and Norway, it may cause dermatitis.

A modified equilibrium standard addition method called the "H-point standard addition method" (HPSAM) [15,18] is developed for determination of unbiased [19], analyte concentration in the event that the presence of direct interferent is known [20,25]. The HPSAM has been applied with analytical spectroscopy to resolve mixtures of two components with extensively or fully overlapped spectra [26]. By plotting the analytical signals versus added analytical concentration, two straight lines are obtained that have a common point H with coordinates (C_H, A_H), C_H is the unknown analyte concentration and the A_H is the analytical signal due to the interferent species.

Micelles posses a high potential for multicomponent analysis, which can be used as special reaction media because they alter the rate, equilibrium position [27]. In the metal-dye complex, at a concentration above critical micelle concentration micelles form a ternary complex with advantageous properties, such as hyper chromic and bath chromic shifts that can modify the sensitivity of the method by affecting the interferences and matrix effects [28]. The ability of Micellar system to solublized slightly soluble or even insoluble complexes has been used to enhance the analytical merit of developed method [29].

In this work, H-point standard addition method (HPSAM) was optimized for the simultaneous determination of Tartrazine and Quinoline yellow as Cr (III) complexes in a Tween-80 micellar media at buffered pH 6.0.

Material and Methods

Reagents

All reagents used were of anal R grade unless otherwise stated. Stock solution of Tartrazine and Quinoline yellow (Loba chem.) were prepared in double distilled water respectively. A 1.0 % (w/v) solution of Cr (III) was prepared by dissolving Chromium Oxide (Loba Chem.) in double distilled water. A buffer solution of pH 6.0 was prepared by mixing appropriate amounts of 0.2M sodium acetate and 0.2M acetic acid solution. A 2.0 % (v/v) Tween-80 stock solution was prepared in hot double distilled water.

Apparatus

Digitized UV-Vis absorption spectra were recorded using a Shimadzu 1700 spectrophotometer, with 10mm Quartz cell and measurements of pH were made with digital century (C_p-901) pH-meter using a combined glass electrode.

Procedure

Individual Calibration

Appropriate volumes of tartrazine or quinoline yellow standard solutions (Beer`s law was obeyed in the concentration range of 2.67-12.0 µg /mL of tartrazine and 0.954-2.39 µg /mL of quinoline yellow), 2.0 mL of 2.0 % Tween-80, 2.0 mL of buffer solution (pH 6.0), 1.0 mL of 1.0 % Cr (III) solution were added into a 10.0 mL standard flask and volume was made up to the mark with double distilled water. A portion of the solution was transferred into a quartz cell and variations of absorbance were recorded for each sample.

Binary- HPSAM

In a 10.0 mL standard flask, 2.0 mL of 2.0 % Tween-80 , 2.0 mL of buffer solution (pH 6.0), 2.0 mL of 1.0 % Cr (III) (to ensure complete complexation when both tartrazine and quinoline yellow were present), appropriate amounts of tartrazine and quinoline yellow standard solutions in double distilled water were added. Absorbance of solution was measured at 422 and 410 nm (when standard solution of tartrazine was added) or 436 and 420 nm (when standard solution of quinoline yellow was added) against a reagent blank and graphs of absorbance vs. concentration were plotted. C_H and A_H were obtained from the point of intersection of the two derived straight lines.

Results and Discussion

HPSAM enables simultaneous determination of species X and Y in a mixture even if their analytical signals overlap extensively and their maxima are coinciding. Absorption spectra of tartrazine and quinoline yellow complexes with Cr (III) overlap as shown in (Figure 1). Since the spectral bands of the two complexes overlaps. So, simultaneous determination of tartrazine and quinoline yellow food colorants is possible only using Binary-HPSAM.

Optimization of variables

Effect of pH: The effect of pH in the range of 3.5–9.5 on absorbance of Tartrazine - Cr (III) and Quinoline yellow–Cr (III) is shown in (Figure 2). Tartrazine-Cr (III) and Quinoline yellow –Cr (III) complexes showed maximum absorbance at pH 6.0. Therefore, pH 6.0 was selected for simultaneous determination of tartrazine and quinoline yellow.

Effect of nature of the surfactant: Various surfactants such as Triton X-100, Tween-20, Tween-80, cetylpyridinium bromide (CPB), Cetyltrimethylammonium bromide (CTAB), and sodium Lauryal sulphate (SLS) were tried as solubilizing agents. Both for Tartrazine-Cr (III) and Quinoline yellow - Cr (III) complex absorbance was maximum with Tween-80 as shown in (Figure 3). So, for simultaneous determination of tartrazine and quinoline yellow, 2.0 mL of 2.0 % Tween-80 was selected as the working micellizing agent for further studies.

Effect of reagent concentration: Effect of different amounts of Cr (III) on the absorbance of Tartrazine-Cr (III) and Quinoline yellow -Cr (III) complexes was studied. The maximum absorbance in both the cases was observed when 1.0 mL of 1.0% Cr (III) was used for individual calibration as shown in (Figure 4). To ensure the complete complexation for simultaneous determination of tartrazine and quinoline yellow, 2.0 mL of 1.0% Cr (III) was used.

Applying Binary – HPSAM

When tartrazine is selected as the analyte, many pairs of wavelengths showing the same absorbance for the interferent i.e. Quinoline yellow-Cr (III) complex were possible. When selecting one pair of wavelengths for obtaining good accuracy, the absorbance difference at the two selected wavelengths for Tartrazine–Cr (III) complex must be as large as possible. On the basis of the absorption spectra of tartrazine or quinoline yellow (as analyte), one of the best pairs of wavelength is 422 and 410 or 436 and 420 nm, with these pairs of wavelength difference in absorbance and hence maximum sensitivity are attainable. C_H and A_H were obtained from the point of intersection of the two derived straight lines in the H-point graphs of absorbance versus analyte concentration where C_H was the unknown analyte concentration and A_H the analyte signal due to interferent species. HPSAM was also applied when tartrazine was selected as analyte and quinoline yellow as interferent. The selected pairs of wavelengths are 422 and 410 nm, where the absorbance difference for Tartrazine -Cr (III) complex is maximum and Quinoline yellow – Cr (III) complex showed the same absorbance.

Several experiments for evaluating HPSAM on the determination of tartrazine and quinoline yellow in a series of samples containing fixed amounts of quinoline yellow with different amounts of tartrazine (Figure 5) or fixed amounts of tartrazine with different amounts of quinoline yellow (Figure 6) were carried out by adding tartrazine standard solutions. The applicability of HPSAM was also tested for determination of quinoline yellow and tartrazine in a series of samples containing fixed amounts of quinoline yellow together with different amounts of tartrazine (Figure 7) or fixed amounts of tartrazine together with different amounts of quinoline yellow (Figure 8) were carried out by

adding quinoline yellow standard solutions. The results show that quinoline yellow and tartrazine contents in the samples could be determined accurately.

Reproducibility of the method: Under optimum conditions simultaneous determination of tartrazine and quinoline yellow were made by using H-Point Standard Addition Method (HPSAM). To check the reproducibility of the method five replicate experiments were performed and the results are given in (Table 1).

Accuracy and Precision: Several synthetic samples with different concentration ratio of tartrazine and quinoline yellow were analyzed using HPSAM. The concentration ranges of tartrazine and quinoline yellow for construction of calibration graphs were 2.67-12.0 and 0.954-2.39 µg /mL respectively. Several synthetic samples with different concentration ratios of tartrazine and quinoline yellow were analyzed using HPSAM. The results are given in (Table 2).

Application

Determination of tartrazine and quinoline yellow in Fruit Jelly (mango Flavor):5.0 gm of Fruit jelly was dissolved in hot double distilled water. After mixing, the residue was filtered, diluted with double distilled water up to mark in a standard volume. The sample solution was then analyzed by the developed procedure. The results are given in (Table 3).

Determination of tartrazine and quinoline yellow in Fruit Syrup (mango fruit syrup): 20.0 mL of double distilled water was mixed with 10.0 mL of fruit syrup. After mixing, the residue was filtered off and filtrate was diluted with double distilled water up to mark in 100.0 mL standard flask. The standard solution was analyzed by the developed procedure. The results are given in (Table 3).

Determination of tartrazine and quinoline: *yellow in Cream Biscuit (mango Flavor Cream):* 10.0 gm of cream sample was mixed with 20.0 mL of double distilled water. After mixing, the residue was filtered off and filtrate was diluted with double distilled water up to mark in 100.0 mL standard flask. The standard solution was analyzed by the developed procedure. The results are given in (Table 3).

Determination of tartrazine and quinoline yellow in cough syrup (Herbal soya product): 20.0 mL of double distilled water was mixed with 10.0 mL of cough syrup. After mixing, the residue was filtered off and filtrate was diluted with double distilled water up to mark in 100.0 mL standard flask. The standard solution was analyzed by the developed procedure. The results are given in (Table 3).

Determination of tartrazine and quinoline yellow in Candy (mango flavor): 5.0 gm of Candy sample was grounded with mortar in a pestle, dissolved in double distilled with. After mixing, the residue was filtered, diluted with double distilled water up to standard volume. The sample solution was then analyzed by the developed procedure. The results are given in (Table 3).

Conclusion

The important characteristic of this work are:

1. Simultaneous determination of tartrazine and quinoline yellow without the use of any expensive instrument is achieved. This reduces the cost of applied method.

1. 2. No extraction step is required as determination has been done in micellar media and hence the use of toxic and carcinogenic organic solvents is avoided. Most of the organic solvents that are being used for extraction are classified as toxic and environmental pollutants and some have been listed as carcinogenic by the US Environmental Protection Agency (EPA).

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Table 1: Results of five replicate experiments for the analysis of quinoline yellow and tartrazine mixture

A-C-Equation	r^2	Present (µg /mL)		Found (µg /mL)	
		Quinoline-Y	Tartrazine	Quinoline-Y	Tartrazine
$A_{436}=0.086C_i+0.236$ $A_{420}=0.076C_i+0.225$	0.998 0.997	1.14	6.68	1.10	6.61
$A_{436}=0.062C_i+0.193$ $A_{420}=0.070C_i+0.202$	0.997 0.997	1.14	6.68	1.12	6.64
$A_{436}=0.074C_i+0.174$ $A_{420}=0.081C_i+0.183$	0.998 0.998	1.14	6.68	1.12	6.66
$A_{436}=0.061C_i+0.242$ $A_{420}=0.068C_i+0.253$	0.991 0.994	1.14	6.68	1.13	6.67
$A_{436}=0.069C_i+0.133$ $A_{420}=0.074C_i+0.141$	0.999 0.997	1.14	6.68	1.13	6.67
Mean				1.12	6.65
Standard deviation				0.007	0.014
R.S.D (%)				0.625	0.210
LOD				0.021	0.042
LOQ				0.07	0.14

Table 2: Results of several experiments for the analysis of tartrazine and quinoline yellow in synthetic samples by H-point standard addition

A-C Equation	r^2	Present (µg /mL) Tartrazine	Quinoline-Y	Found (µg /mL) Tartrazine	Quinoline-Y	% Recovery Tartrazine	Quinoline-Y
$A_{422}=0.040C_i+0.108$ $A_{410}=0.037C_i+0.099$	0.999 0.998	3.00	1.90	3.00	1.89	100	99.47
$A_{422}=0.016C_i+0.235$ $A_{410}=0.014C_i+0.227$	0.997 0.995	4.07	1.90	4.00	1.89	98.28	99.47
$A_{422}=0.008C_i+0.305$ $A_{410}=0.081C_i+0.183$	0.994 0.998	6.08	2.29	6.00	2.28	98.68	98.68
$A_{422}=0.018C_i+0.270$ $A_{410}=0.017C_i+0.263$	0.997 0.997	7.04	2.29	7.00	2.29	99.43	100
$A_{422}=0.014C_i+0.170$ $A_{410}=0.012C_i+0.161$	0.996 0.995	4.54	1.43	4.50	1.43	99.11	100
$A_{422}=0.018C_i+0.168$ $A_{410}=0.020C_i+0.177$	0.999 0.995	4.54	1.72	4.50	1.71	99.11	99.41
$A_{422}=0.014C_i+0.171$ $A_{410}=0.013C_i+0.166$	0.999 0.999	5.00	2.10	5.00	2.08	100	99.04
$A_{422}=0.011C_i+0.235$ $A_{410}=0.010C_i+0.230$	0.992 0.999	5.00	2.19	5.00	2.19	100	100

Table 3: Determination of tartrazine and quinoline yellow in different samples by HPSAM

Sample	Concentration (µg /mL)			
	Present (µg /mL)		Found (µg /mL)	
	Tartrazine	Quinoline-Y	Tartrazine	Quinoline -Y
Fruit jelly(mango Flavor)	3.32	1.52	3.35	1.56
Cream Biscuit	3.39	1.43	3.34	1.46
Fruit syrup (pineapple)	4.27	2.57	4.30	2.55
Cough syrup	4.43	2.31	4.46	2.33
Candy (Mango bit)	4.26	2.46	4.24	2.47

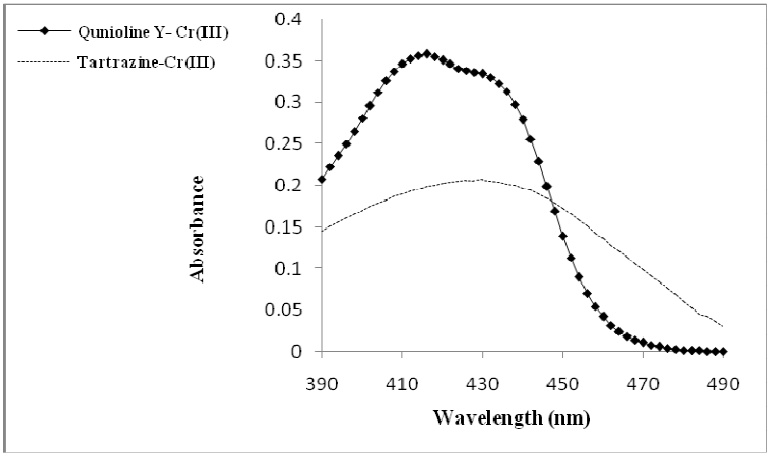

Figure 1: Absorption spectra of Quinoline yellow- Cr (III) and Tartrazine- Cr (III) complexes, in 2.0 % Tween-80 at pH 6.0.

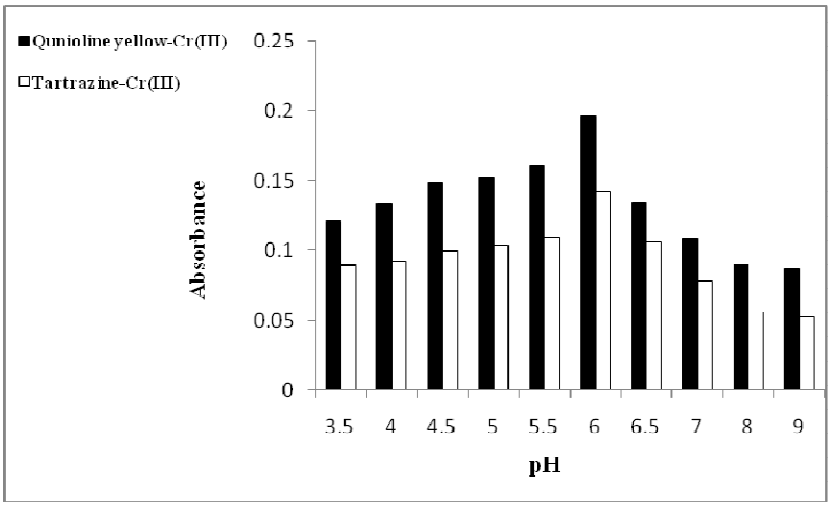

Figure 2: Effect of pH on the absorbance of Quinoline yellow- Cr (III) and Tartrazine- Cr (III) complexes.

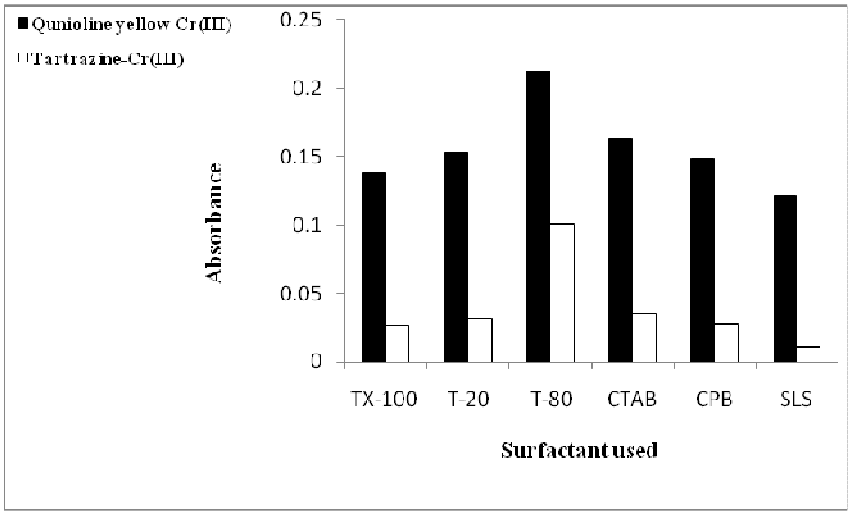

Figure 3: Effect of different surfactants on absorbance of Cr (III) complexes of quinoline yellow and tartrazine.

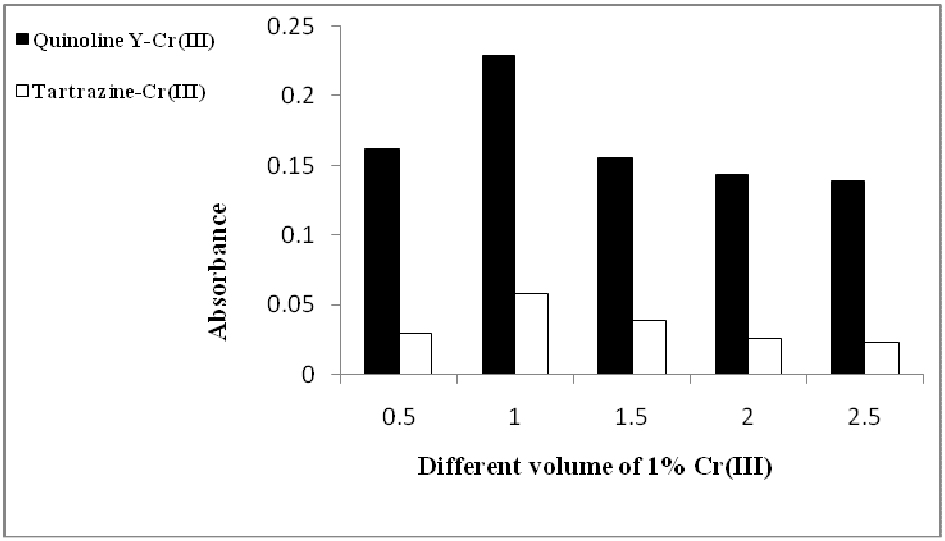

Figure 4: Effect of different volume of 1.0 % Cr (III) on absorbance of Quinoline yellow –Cr (III) and Tartrazine- Cr (III).

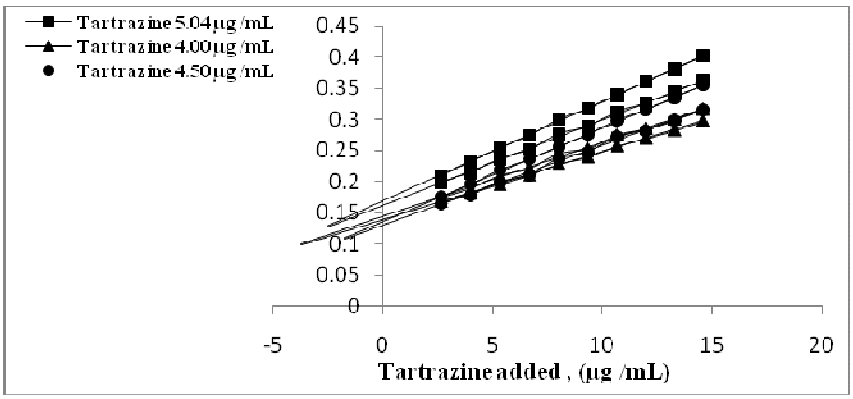

Figure 5: Plot of H-point standard addition method for simultaneous determination of fixed amounts of quinoline yellow and different amounts of tartrazine. Condition: pH 6.0, 2.0 mL 0f 2.0 % Tween-80, 2.0 mL of 1.0 % Cr (III) and 0.954 µg /mL quinoline yellow and different concentration of tartrazine, when different amounts of standard tartrazine solutions are added.

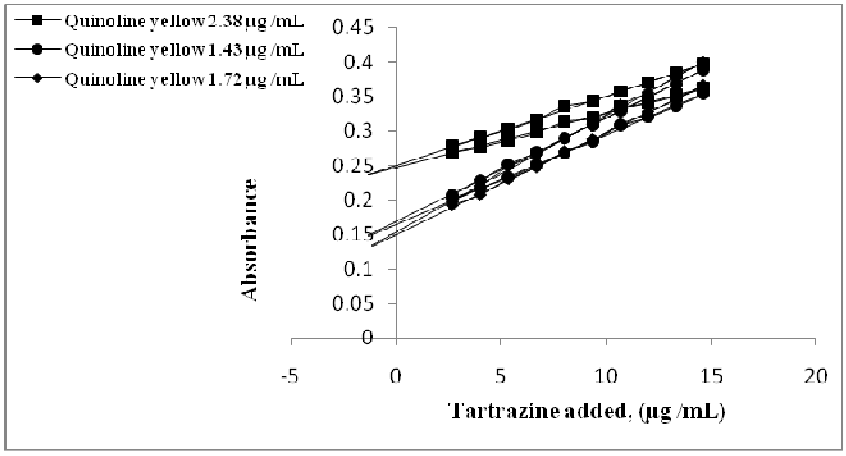

Figure 6: Plot of H-point standard addition method for simultaneous determination of fixed amounts of tartrazine and different amounts of quinoline yellow. Condition: pH 6.0, 2.0 mL 0f 2.0 % Tween-80, 2.0 mL of 1.0% Cr (III) and 2.50 µg /mL tartrazine and different concentration of quinoline yellow, when different amounts of standard tartrazine solutions are added.

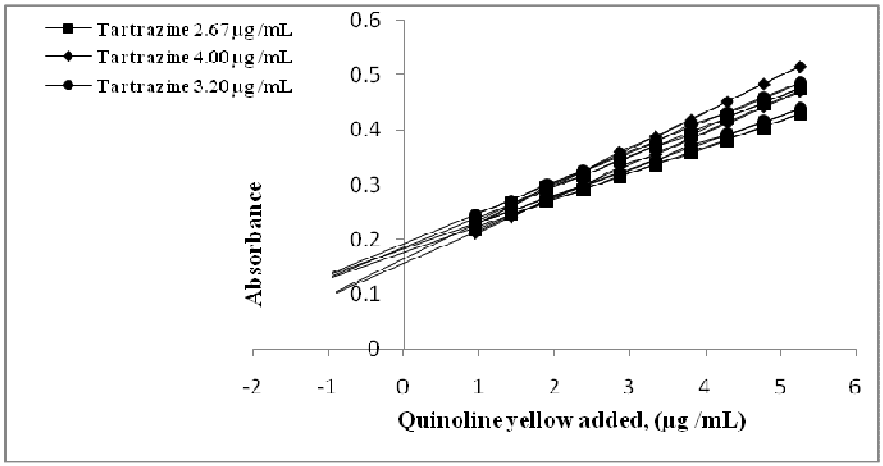

Figure 7: Plot of H-point standard addition method for simultaneous determination of fixed amounts of quinoline yellow and different amounts of tartrazine. Condition: pH 6.0, 2.0 mL 0f 2.0 % Tween-80, 2.0 mL of 1.0 % Cr (III) and 1.43 µg /mL quinoline yellow and different concentration of tartrazine, when different amounts of standard quinoline yellow solutions are added.

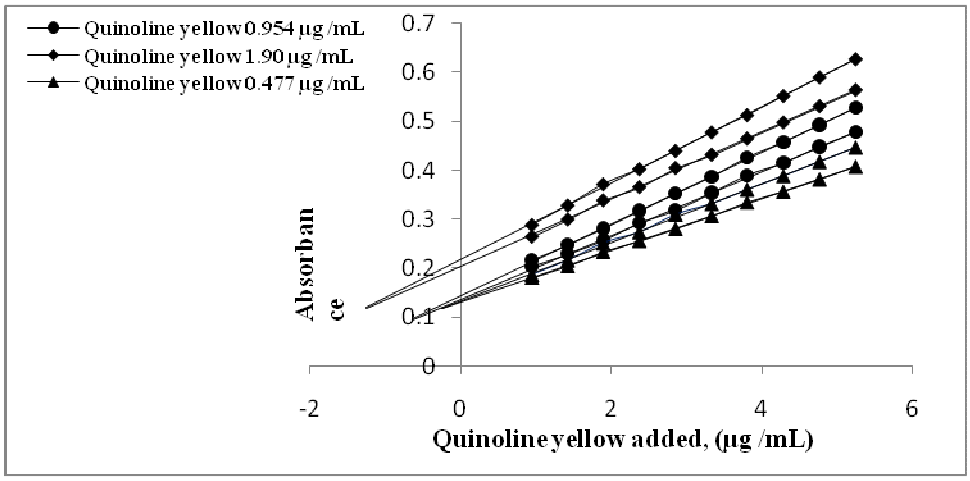

Figure 8: Plot of H-point standard addition method for simultaneous determination of fixed amounts of tartrazine and different amounts of quinoline yellow. Condition: pH 6.0, 2.0 mL 0f 2.0 % SLS, 2.0 mL of 1.0 % Cr (III) and 5.34 µg /mL tartrazine and different concentration of quinoline yellow, when different amounts of standard quinoline yellow solutions are added.