A New Thin-Layer Chromatographic System for the Identification and Selective Separation of Brilliant Blue Food Dye: Application of a Green Solvent

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Key Words:

Polyaniline modified silica gel
Food dyes
Separation
Densitogram
Thin-layer chromatography

Summary

Silica gel and polyaniline-modified silica gel were used as stationary phases with aqueous methanol as the mobile phase for the comparative study of the migration behavior of dyes in order to select the most suitable thin-layer chromatographic system for the resolution of coexisting dyes. A better separation efficiency was observed by modifying silica gel with polyaniline as compared to pure silica static phase. Densitographic presentation of separations achieved on polyaniline-modified silica gel was also presented. The separation patterns of the dyes were also examined by replacing methanol with other alcohols. The thin-layer chromatographic system comprising of polyaniline-modified silica gel [(Pani–SG (EB_1)] as the static phase and aqueous methanol (methanol–DDW, 1:9) as the mobile phase was the most favorable for the selective separation of brilliant blue from tartrazine and carmoisine. Silica gel and the modified silica gel nanocomposite of the static phase have been characterized by Fourier transform infrared (FTIR) spectroscopy, X-ray diffraction (XRD), and scanning electron microscopy (SEM), and transmission electron microscopy (TEM) studies were undertaken to characterize silica gel and modified silica gel (nanocomposite). Food dyes such as brilliant blue and tartrazine (in Ambrodil* S syrup) as well as carmoisine (in Flucold™ syrup) were successfully identified using the proposed thin-layer chromatographic method.

1 Introduction

Food dyes classified as natural, synthetic, and artificial dyes belong to an important group of food additives and are used to enhance attractiveness of food items, including sweets, to consumers [1]. Food dyes are added in order to counterbalance the partial or complete color loss of food products during processing and storing into some pharmaceutical products [2]. Natural dyes are unstable and easily degraded during the processing of foods as compared to synthetic dyes [3]. In addition, the cost of the production of synthetic dyes is low. Azo dyes containing azo groups –N=N– are the most commonly used synthetic dyes in food, cosmetics, and pharmaceutical products. Due to hazardous health effects, the analysis of these dyes has become very important. Azo dyes are easily degraded into aromatic amines in the intestine, and this offers serious health problems such as headache, neurological, genetic, and carcinogenic problems [4–7].

Thin-layer chromatography (TLC) has been one of the widely used analytical methods for the identification and separation of food dyes [8–14]. Generally, food dyes have been identified and separated by TLC using silica gel as the stationary phase and aqueous-organic or mixed-organic solvent systems as the mobile phases. Most of the studies reported so far involve the use of volatile organic solvents that have a negative impact on the environment.

In the present study, polyaniline-modified silica gel layer has been used as the stationary phase in combination of aqueous methanol as the green mobile phase. Polymer-modified silica gel enhances the selectivity and chemical stability of the stationary phases [15–17]. It is a well known fact that double distilled water (DDW) has occupied the first position in the list of green solvents due to its easy availability, non-toxicity, amazing solubilizing properties, and poor thermal conductivity [18]. On the other hand, methanol has been extensively used in thin-layer chromatography as a green solvent due to its low cost, easy availability, and environmental benefits [19].

Polyaniline-modified silica gel has been synthesized and used as the static phase with aqueous methanol as the mobile phase for the thin-layer chromatographic analysis of three food dyes, brilliant blue, tartrazine, and carmoisine, during the present study. The synthesized nanocomposite of the static phase has been characterized by Fourier transform infrared (FTIR) spectroscopy, X-ray diffraction (XRD), scanning electron microscopy (SEM), and transmission electron microscopy (TEM) studies.

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2 Experimental

All the experiments were performed at 25 ± 2°C.

2.1 Apparatus

20 cm × 3.5 cm glass plates coated with silica gel and polyaniline-modified silica gel using a TLC applicator (Toshniwal, India) were used as the stationary phase. A micropipette (Tripette, Germany) was used for spotting of analytes, and 24 cm × 6 cm glass jars were used to perform TLC.

2.2 Chemicals and Procedure

Silica gel (Fischer Scientific, India); methanol, ethanol, propanol-1, potassium persulfate, brilliant blue, copper sulfate, zinc sulfate, manganese sulfate, nickel nitrate, HCl (AR grade), and sodium salts of bromide, chloride, carbonate, acetate, and nitrate (Central Drug House; CDH, India); tartrazine, carmoisine (Roha Dyechem Pvt. Ltd., Mumbai, India); and aniline (E-Merck India Ltd.) were used as received. All chemicals were of analytical reagent grade. The water used in these experiments was double distilled.

The Fourier transform infrared (FTIR) spectra were recorded using Perkin-Elmer 1725 spectrometer operating in the 400–4000 cm^{-1} range. X-ray diffraction (XRD) data were recorded by using Bruker D8 diffractometer with Cu Kα radiation at 1.540 Å in the range of $5° \leq 2\theta \leq 70°$ at 40 kV. The morphology was observed by a JSM-6510LV system with a JEOL scanning electron microscope (SEM). Transmission electron micrograph (TEM) was done with the help of a JEM 2100, JEOL instrument to study the morphology and the particle size.

2.3 Test Solutions

Solutions of 5% (*w/v*) of dyes were prepared in double distilled water (DDW).

2.4 Stationary Phases

SG-S_1, Pani–SG (ES)-S_2, Pani–SG (EB1)-S_3, and Pani–SG (EB2)-S_4 were used as the stationary phases.

2.5 Mobile Phase

The solvent systems used as the mobile phases are listed in **Table 1.**

2.6 Composition of Flucold™ and Ambrodil* S Syrup

The Flucold™ (decongestant and antipyretic) syrup by Wallace Pharmaceutical Pvt. Ltd. (Bhatian, Nalagarh, Himachal Pradesh, India) and Ambrodil* S syrup by Aristo Pharmaceutical Pvt. Ltd. (Raisen, Madhya Pradesh, India) used in the present study to identify carmoisine, brilliant blue, and tartrazine. food dyes have the following compositions:

Flucold™ syrup:

– paracetamol Indian Pharmocopeia (I.P.) (125 mg);

– phenylpherine HCl I.P. (5 mg);

– chlorpheniramine maleate I.P. (1 mg);

– sodium citrate I.P. (60 mg) and

– coloring agents carmoisine and caramel I.P.

Ambrodil* S syrup:

– ambroxol hydrochloride (I.P. 15 mg)

– salbutamol sulfate (I.P. 1 mg)

– flavoured syrup base

– colorants brilliant blue FCF and tartrazine.

Table 1

List of the mobile phases used during the whole study.

Abbreviation	Mobile phase
M_1	Double distilled water (DDW)
M_2	Methanol
M_3	Methanol–DDW (9:1)
M_4	Methanol–DDW (7:3)
M_5	Methanol–DDW (5:5)
M_6	Methanol–DDW (3:7)
M_7	Methanol–DDW (1:9)
M_8	Ethanol–DDW (1:9)
M_9	Propanol–DDW (1:9)

2.7 Preparation of Polyaniline-Modified Silica Gel Nanocomposite

Pani–SG nanocomposites were prepared by simple *in-situ* oxidative polymerization of aniline in the presence of different amounts of SG using potassium persulfate as the oxidizing agent. One milliliter of aniline monomer was dispersed in 200 mL of 1 M aqueous solution of HCl. Different amounts of silica gel were added into the aniline solution. Later, the solution (100 mL) of potassium persulfate (made in 1 M HCl) was added drop-wise in the above solution for polymerization. The reaction mixture was put under continuous stirring for 22 h. The resultant mixture

Table 2

Preparation details of Pani–SG nanocomposites.

Sample ID	Volume of aniline (mL)	Weight of $K_2S_2O_8$ (g)	Weight of silica gel (SG) (g)
Pani–SG (ES)-S_2	1	5.0	30.0
Pani–SG (EB_1)-S_3	1	5.0	30.0
Pani–SG (EB_2)-S_4	1	5.0	60.0

turned slowly into greenish black slurry which was filtered, washed thoroughly with DDW, then undoped with 500 mL of 1 M ammonia solution for making Pani–SG (EB) and, in the case of Pani–SG (ES), not undoped, later washed with DDW and methanol to remove excess acid and potassium persulfate and other impurities until the filtrate became colorless. Thus, the prepared nanocomposites were dried in an air oven at 70–80°C for 12 h, converted into fine powders, and stored in a desiccator for further investigations. Different materials were assigned identity codes as silica gel (S_1) used as received, Pani–SG (ES)-S_2, Pani–SG (EB1)-S_3, and Pani–SG (EB2)-S_4 as given in **Table 2.**

2.8 Thin-Layer Chromatographic Separations

The preparation of TLC plates and the chromatographic procedure were carried out as reported earlier [20]. 0.1 µL of aliquot of dyes was spotted on the prepared TLC plates (S_1, S_2, S_3, and S_4), and developed with different mobile phases. After development, the spots were visualized, and the R_F values were calculated for the individual dyes. For the mutual separations of dyes, equal volumes (1 mL each) of dyes (tartrazine, carmoisine, and brilliant blue) were mixed and 0.1 µL of the resultant mixture was spotted on the TLC plates. The plates were developed with the selected mobile phase (M_7), the spots were detected, and the R_F values of the separated dyes were determined. The R_F values were calculated from the R_L (R_F of leading front) and R_T (R_F of trailing front) values of the spot as given below.

$$R_F = 0.5(R_L - R_T)$$

2.9 Effect of Alcohols on Separation

In order to examine the mobility pattern of dyes on S_3, methanol in M_7 was replaced with ethanol–DDW (M_8) and propanol–DDW (M_9) was also used as eluent.

2.10 Effect of Foreign Substances

For investigating the interference of metal cations and inorganic anions as impurities on the separation of the mixture, 0.1 µL of the standard test mixture of dyes solutions was spotted on the Pani–SG (EB1)-S_3 TLC plate followed by spotting of 0.1 µL of the metal cations and inorganic anions being considered as impurities. The plates were developed with M_7 and detected, and the R_F values of the separated dyes were calculated.

2.11 Limit of Detection

The detection limits of the dyes were determined by spotting 0.1 µL of tartrazine, carmoisine, and brilliant blue of different concentrations on the Pani–SG (EB1)-S_3 TLC plates which were developed with the selected mobile phase M_7, and the spots were visualized. This process was repeated by successive reduction of the concentration of dye until the detection of dye was not possible any more. The amount of dye just detectable was taken as the detection limit.

2.12 Application

The proposed method was applied for the identification of carmoisine in Flucold™ and brilliant blue and tartrazine in Ambrodil* S syrup with the chosen TLC system.

3 Results and Discussion

Thin-layer chromatography of three azo dyes (tartrazine, carmoisine, and brilliant blue) was performed on four different stationary phases: (1) as silica gel-S_1, (2) Pani–SG (ES)-S_2, (3) Pani–SG (EB1)-S_3, and (4) Pani–SG (EB2)-S_4 with nine different mobile phases for obtaining a novel TLC system for the selective separation of brilliant blue from the mixture of

Table 3

Mobility of food dyes in terms of R_F values on four different stationary phases with various mobile phases.

Stationary phases	R_F value											
	S_1			S_2			S_3			S_4		
Dyes[a] / Mobile phases	CS	TZ	BB	CS	TZ	BB	CS	TZ	BB	CS	TZ	BB
M_1	0.75	0.90	0.22	0.89	0.93	0.25	0.89	0.91	0.12	0.70	0.98	0.10
M_2	0.69	0.90	0.86	0.70	0.84	0.71	0.83	0.85	0.78	0.80	0.85	0.79
M_3	0.92	0.93	0.90	0.88	0.84	0.73	0.72	0.875	0.56	0.705	0.87	ND
M_4	0.93	0.93	0.93	0.875	0.93	0.46T	0.91	0.93	0.70	0.78	0.94	ND
M_5	0.98	0.99	0.87	0.88	0.92	0.19T	0.90	0.94	0.25T	0.76	0.93	ND
M_6	0.96	0.95	0.60	0.81	0.91	0.10	0.91	0.93	0.09	0.70	0.97	ND
M_7	0.93	0.97	0.27T	0.82	0.88	0.08	0.90	0.94	0.05	0.75	0.95	0.07
M_8	0.85	0.99	0.39T	0.85	0.98	0.08	0.65	0.97	0.11	0.90	0.98	0.18
M_9	0.94	0.95	0.35T	0.93	0.89	0.18	0.97	0.93	0.28	0.95	0.91	0.39

T = tailed spot, ($R_L - R_T \geq 0.3$) and ND = not detected

[a]Here, CS, TZ, and BB represent carmoisine, tartrazine, and brilliant blue, respectively

other azo dyes. The obtained results have been tabulated in **Tables 3 and 4** and presented in **Figures 1–10** and are discussed below.

Table 4

Identification of carmoisine (in Flucold™) and brilliant blue as well as tartrazine (in Ambrodil* S syrup) according to their R_F values.

Food dyes	R_F value	
	Standard sample	Drug sample
Carmoisine	0.90	0.82
Brilliant blue	0.05	0.04
Tartrazine	0.96	0.92

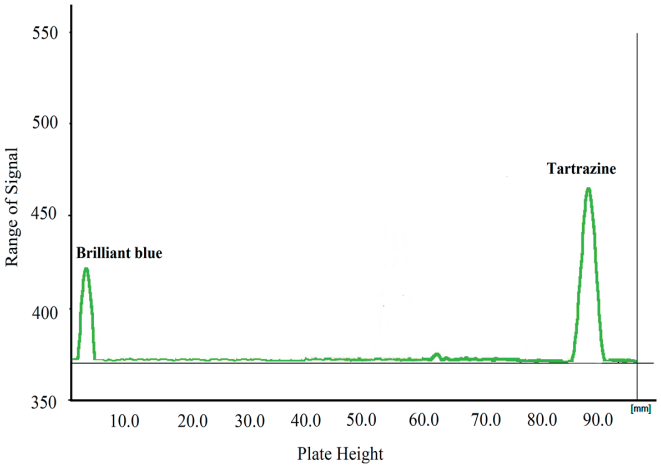

Figure 1

Densitographic representation of the separation of brilliant blue from tartrazine on S_3 with M_7.

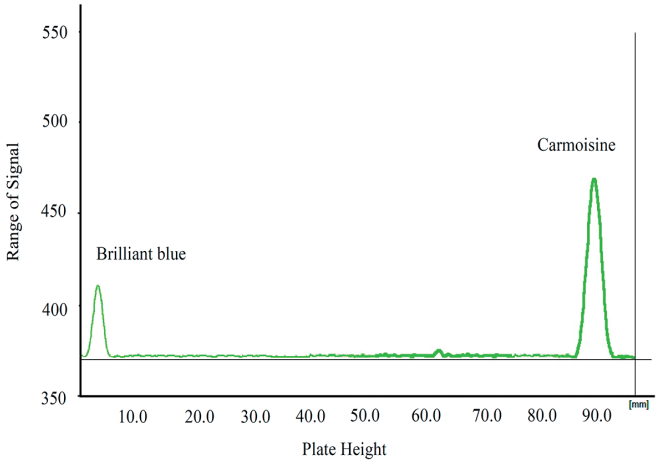

Figure 2

Densitographic representation of the binary separation of brilliant blue and carmoisine on S_3 with M_7.

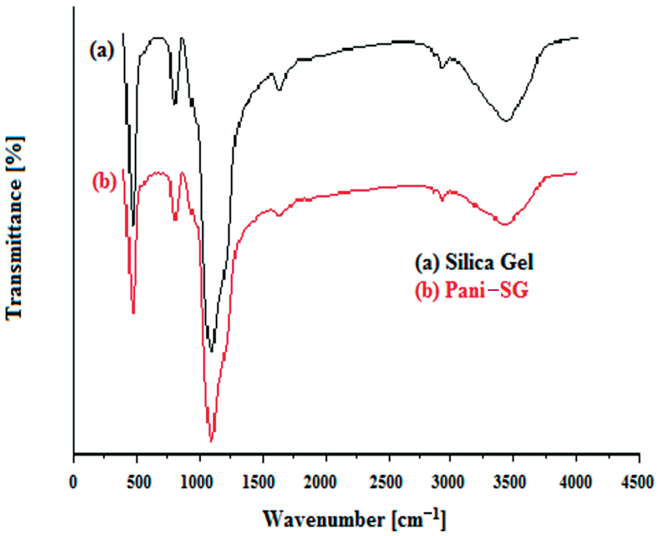

Figure 3

FTIR spectra of (a) silica gel (S_1) and (b) Pani–SG (EB1)-S_3 nanocomposites.

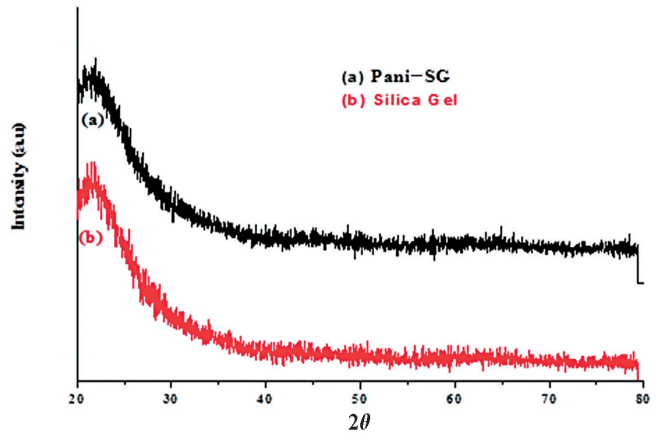

Figure 4

XRD spectra of (a) silica gel (S_1) and (b) Pani–SG (EB1)-S_3 nanocomposites.

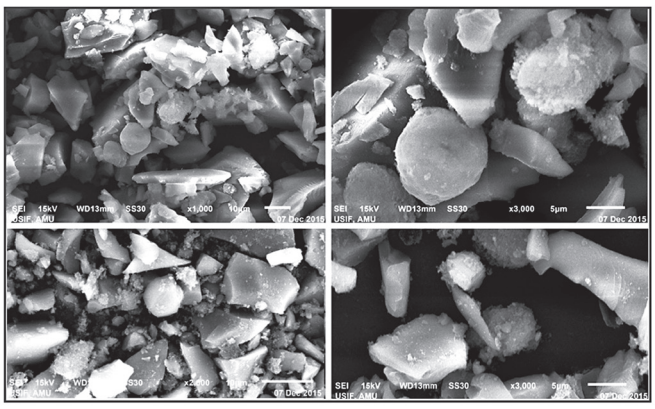

Figure 5

SEM micrographs of (a and b) silica gel (S_1) and (c and d) Pani–SG (EB1)-S_3 nanocomposite at different magnifications.

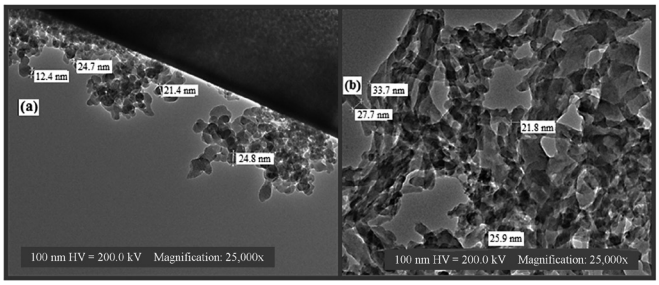

Figure 6

TEM micrographs of (a) silica gel (S_1) and (b) Pani–SG (EB1)-S_3 nanocomposite.

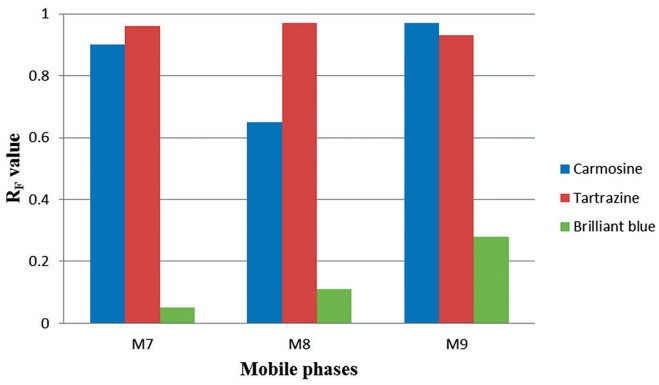

Figure 7

R_F value of azo dyes on S_3 with different mobile phases.

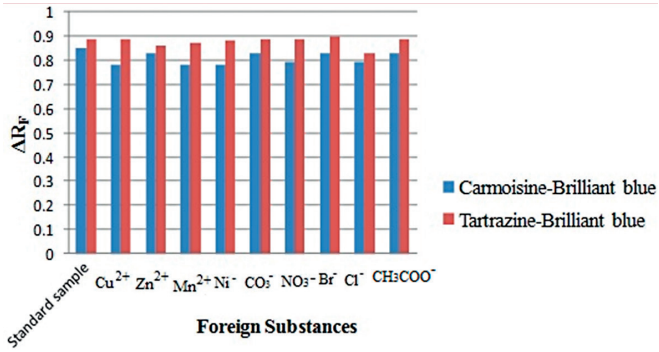

Figure 8

Effect of foreign substances on the ΔR_F values of the separated two-component mixtures of food dyes.

3.1 Silica Gel as Stationary Phase

– With double distilled water (M_1), on pure silica static phase (S_1), the separation of brilliant blue from the mixture of azo dyes has been observed with broad spot sizes.

– Using M_2 with S_1 static phase, poor resolution of carmoisine from tartrazine or brilliant blue has been obtained. As the volume of methanol in aq. methanol decreases (M_3 and M_4) and at equal volumes of methanol and DDW (M_5), separation was not obtained because all the three dyes show identical R_F values. Further decrease in the volume of methanol to the level of 1–3% (M_6 and M_7) and poor separation of brilliant blue from carmoisine or tartrazine have been observed due to the formation of tailed spots (Table 3).

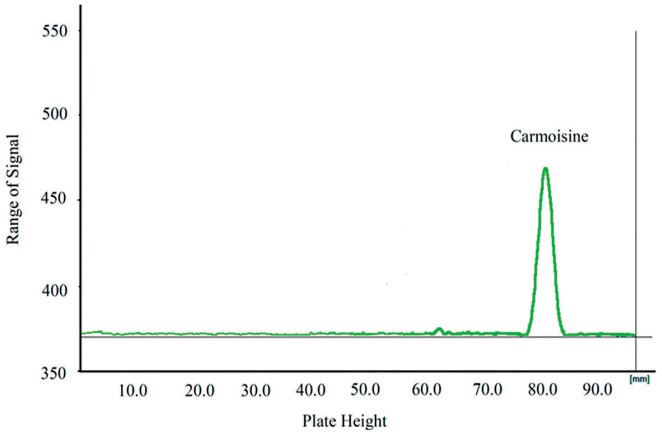

Figure 9

Densitographic illustration of the identification of carmoisine in Flucold™ syrup on S_3 with M_7.

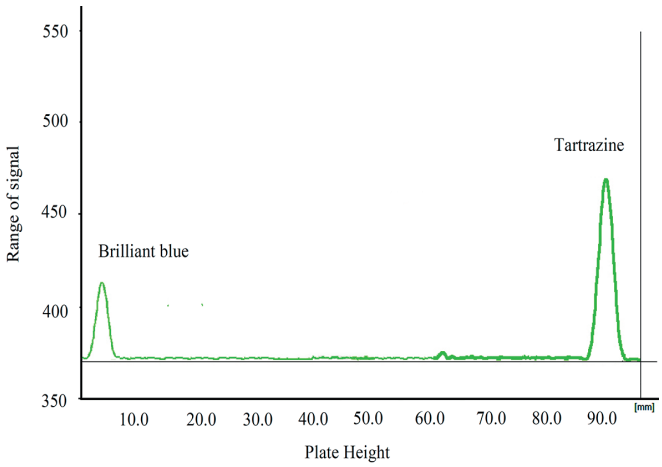

Figure 10

Densitographic illustration of the identification and separation of brilliant blue and tartrazine in Ambrodil* S syrup on S_3 with M_7.

3.2 Polyaniline-Modified Silica Gel in Different Ratios

– On S_2, S_3, and S_4 with DDW (M_1), brilliant blue has been selectively separated from the mixture of azo dyes with slightly broader spot sizes. With M_2, no separation of dyes was possible because of the identical migration trend (almost equal R_F values) of all three dyes.

– With decreasing volume of methanol (M_3–M_6) on S_2, S_3, and S_4, carmoisine and tartrazine show the same R_F values, while brilliant blue has not been detected on S_4, but it showed tailed spots on S_2 and S_3 (Table 3).

Though brilliant blue has been selectively separated on S_2, S_3, and S_4 with M_7, very compact spots of all dyes were observed on S_3 with M_7. Hence, the TLC system comprising of Pani–SG (EB1)-S_3 as the stationary phase and methanol–DDW (1:9)

as the mobile phase has been found the most favorable for the selective separation of brilliant blue from the mixture of other azo dyes (carmoisine and tartrazine) (Figures 1 and 2).

3.3 FTIR Spectroscopic Studies

The FTIR spectra of silica gel (SG) and Pani–SG (EB1)-S_3 nanocomposite are shown in Figure 3. The main characteristic peaks of the silica gel in curve are the broad absorption band around 3436 cm^{-1} which corresponds to O–H stretching vibrations from Si–OH; the strong absorption band at 1098 cm^{-1} is due to the asymmetric stretching vibration of Si–O–Si bond because of the formation of a SiO_2 network, and the absorption peak at 800 cm^{-1} which represents Si–O–Si bending vibrations and the band at 468 cm^{-1} is due to the deformation vibration of Si–O–Si [21–23]. The absorption peak at 1634 cm^{-1} indicates H–O–H bending vibrations of a water molecule [24].

In the case of Pani–SG (EB1)-S_3 nanocomposite apart from the peaks of SIG, the characteristic peak of 3434 cm^{-1} due to the free (non-hydrogen bonded) N–H stretching vibration is present, confirming the presence of Pani in Pani–SG [25–28]. The absorption peak around 2926 cm^{-1} is attributed to C–H stretching vibrations. The FTIR spectrum of Pani–SG (EB1)-S_3 nanocomposite is similar to that of the silica gel, and no additional peaks have been found. Thus, it can be inferred that the incorporation of SG does not damage the backbone of Pani.

3.4 X-Ray Diffraction (XRD) Studies

The XRD patterns of silica gel and Pani–SG (EB1)-S_3 nanocomposites (Figure 4) are indicative of an amorphous nature with low intensity centered at an angle of $2\theta = 23$ [29–31] in the case of silica gel. However, Pani exhibits the characteristic peak around $2\theta = 23.44$ [32]. The XRD pattern of Pani–SG nanocomposite is similar to that of the silica gel.

3.5 Scanning Electron Micrograph (SEM) Studies

The shape and surface morphologies of silica gel and Pani–SG (EB1)-S_3 nanoparticles have been demonstrated in Figure 5 (a–d) at two different magnifications. SEM images of silica nanoparticles shown in Figure 5 (a–b) are indicative of irregular, porous, and spherical shape with relatively smooth surface. In Figure 5 (c–d), Pani–SG (EB1)-S_3 nanoparticles show a rougher surface, probably due to the polymerization of aniline on the surface of the silica gel, and porosity may increase due to the presence of polyaniline [33, 34]. However, both micrographs are relatively similar and SiO_2 nanoparticles are mechanically very strong and not very distinct in nanocomposite which confirms that the aniline has polymerized on the surface of SiO_2 as is evident from the FTIR analysis.

3.6 Transmission Electron Micrograph (TEM) Studies

Silica gel and Pani–SG (EB1)-S_3 nanocomposites were characterized to identify the size and shape of their particles by TEM. The results are shown in Figure 6. Most of the silica gel particles were spheroidal or ellipsoidal with particle sizes ranging between 8 and 25 nm. However, some irregular particles were also observed [35]. In the case of Pani–SG (EB1)-S_3 nanocomposite, it appears that the Pani undergoes a polymerization on the surface of the silica gel, leading to the formation of interconnected tubular and fibrous nanostructures containing silica gel in the core. However, the SG surfaces became rough and a large amount of amorphous films or particles were observed, indicating the formation of Pani over SG, and the thickness size range was 15–40 nm. Thus, the material particles belong to the nanosize. The Pani is tightly bound to SiO_2 which is beneficial for its use in electronic and separation techniques [36].

3.7 Effect of Alcohol on Separation

In order to get more separation possibilities, methanol in M_7 was replaced with ethanol (M_8) and propanol (M_9). The chromatographic performance of alcohols towards the separation efficiency of dyes was in this order: methanol > ethanol > *n*-propanol. Thus, as the chain length of alcohols increases, the separation efficiency of dyes decreases (Figure 7).

M_7 – methanol–DDW (1:9), M_8 – ethanol–DDW (1:9), and M_9 – propanol–DDW (1:9)

3.8 Effect of Foreign Substances

The effect of metal cations and anions on the magnitude of the ΔR_F value for the separation of a two-component mixture consisting of carmoisine or tartrazine and brilliant blue has been examined, and the results are presented in Figure 8. From the results, it is clear that the magnitude of these parameters is slightly changed in the presence of these foreign substances, but separation was always possible in each case. The change in the value of these parameters is due to the slight increase in the spot size of the analyte because of certain interactions of the dyes with these foreign substances.

3.9 Limit of Detection

The lowest possible detectable amounts (μg $spot^{-1}$) of carmoisine, tartrazine, and brilliant blue on Pani–SG (EB1)-S_3 developed with M_7 (methanol–DDW in 1:9 ratio) were 0.75, 1.25, and 1.0, respectively, showing a reasonably good sensitivity.

3.10 Application

The proposed thin-layer chromatographic method comprising of polyaniline-modified silica gel Pani–SG (EB1)-S_3 as the static phase and aqueous methanol (methanol–DDW, 1:9) as the mobile phase is applicable for the identification of carmoisine in Flucold™ syrup and brilliant blue and tartrazine in Ambrodil* S syrup (Table 4 and Figures 9 and 10).

4 Conclusion

It can be concluded that the proposed thin-layer chromatographic system consisting of polyaniline-modified silica gel as the static flat phase with aqueous methanol as the eco-friendly mobile phase is the most favorable for the identification and selective separation of brilliant blue from the mixture of carmoisine and tartrazine. The chosen TLC system is also successfully applicable for the identification of these food dyes in pharmaceutical products (Flucold™ and Ambrodil* S syrup).

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