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Pre-concentration and determination of tartrazine dye from aqueous solutions using modified cellulose nanosponges

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ABSTRACT

In this study, a new absorbent based on cellulose nanosponges modified with methyltrioctylammonium chloride (aliquat 336) was prepared and used for pre-concentration, removal and determination of tartrazine dye, using UV–vis spectrophotometry. This adsorbent was fully characterized using various instrumental techniques such as SEM, FTIR and XRD spectra. The pre-concentration and removal procedures were studied in column and batch modes, respectively. The effects of parameters such as pH of the aqueous medium, methyltrioctylammounium chloride dose, adsorbent amount, desorbing conditions and interfering ions on the adsorption of tartrazine were investigated and optimized. The fitting experimental data with conventional isotherm models revealed that the adsorption followed the Brunauer-Emmett-Teller (BET) model and the maximum adsorption capacity for tartrazine was 180 mg/g with modified nanosponges. Under the optimized conditions, the calibration curve was linear over the range of 2–300 ng/mL and the limit of detection was 0.15 ng/mL. The relative standard deviation (RSD) for 20 and 100 ng/mL of tartrazine were 3.1% and 1.5%, respectively. The proposed method was applied for pre-concentration and determination of tartrazine dye in different water samples.

1. Introduction

Synthetic dyes have been used instead of natural colors because of their high stability to light, oxygen, heat and pH, color uniformity and relatively lower costs. Tartrazine (E102) is a synthetic lemon yellow azo dye which has been widely used as an additive in food, drinks, medicine and cosmetics. Tartrazine appears to cause the allergic and intolerance reactions, chiefly affecting individual's allergy to aspirin (Gupta et al., 2011). Recently, studies show that this dye poses potential risks to human health, especially when consumed in excess, due to the significant adverse effects on neurobehavioral parameters. However, presence and content of this dye must be controlled due to its potential harmfulness to human beings (Abu Shawish et al., 2013).

Several analytical techniques have been used for the determination of tartrazine dye, including spectrophotometry (Sahraei et al., 2013; Dinc et al., 2002), chromatography (Wu et al., 2013; Alves et al., 2008), electroanalytical methods (Gan et al., 2012; Ye et al., 2013) and capillary electrophoresis (Perez-Urquiza et al., 2000). However, most of these methods are expensive, long analysis time and sometimes it is necessary to make sample pretreatments. On the other hand, the combination of simple methodologies, such as spectrophotometric methods with pre-concentration techniques, represents a rapid, simple and cheap strategy for the determination of dyes (Pourreza et al., 2008). According to the above-mentioned reasons, determination of tartrazine requires a fast, simple, low-cost and reliable method, which can be used routinely.

Among the several techniques, solid phase extraction (SPE) has been generally used as a pre-concentration procedure for various target analytes. This widespread use of SPE is due to its well-known advantages such as simplicity, high enrichment factor, good recovery, low consumption of organic solvents, suitability for combination with different detection techniques and relatively low cost (Herrero Latorre et al., 2013). The basic principle of SPE is the transfer of analytes from the sample solution phase to bind to the active site of the solid phase (Pourreza et al., 2014).

Nanotechnology and nanoparticles are increasingly recognized for their potential applications in different branches of science. In the recent years, nanoparticles-based SPE offers great possibilities for development of new analytical methodology, because of their unique properties, such as their large surface areas and high adsorption capacity (Parham et al., 2012). Nanosponges (NSs) are a new class of tiny sponges which are made of microscopic particles with cavities a few nanometers wide, characterized by the capacity

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to encapsulate a large variety of substances that can be transported through aqueous media. NSs are highly efficient at entrapping different types of molecules (both organic and inorganic), and they can accomplish this by inclusion or non-inclusion complex formation (Lembo et al., 2013). These sorbents have been prepared by various chemical and physical techniques (Rojas et al., 2009; Watthanaphanit et al., 2010; Berry et al., 2006; Ma et al., 2005; Jiang et al., 2013; Wang et al., 2012; Mishra et al., 2011), and applied in drug therapy (Farooq et al., 2013; Bolmal et al., 2013; Naga et al., 2013; Rapolu et al., 2012), environmental studies (Mhlanga et al., 2007; Kozlowska et al., 2012–2013; Allabashi et al., 2007) and metal analysis (Yavuz et al., 2014).

In the present work, we focus on the potential application of new cellulose acetate-based nanosponges (CNSs) as novel solid phase. The prepared CNSs were modified with methyltrioctylammonium chloride (aliquat 336) and used for the removal, pre-concentration and determination of tartrazine dye, with UV–vis spectrometer detection. The parameters which affect the adsorption and elution efficiency of tartrazine were studied in batch and column modes.

2. Experimental

2.1. Apparatus

A Hach DR5000 UV–Visible Spectrophotometer (USA) was used for recording the spectra and the absorbance measurements of tartrazine, with 1.0 cm cell. The scanning electron microscope (SEM, Leo, 1455 VP, Germany) was performed to study the morphology of synthesized modified cellulose acetate nanosponges. The infrared spectra were obtained using a FTIR (BRAIC WQF-510, China) to identify the functional groups and chemical bonding of the modified materials. An IKA Works KS 130 Orbital Shaker (IKA Works Basic Model), a Jenway stirrer model 1000 (UK) and a Metrohm pH meter model 827 (Switzerland) were used during the experiments.

2.2. Reagents

All chemicals were of analytical grade were purchased from Merck (Merck, Darmstadt, Germany) and double distilled water was used throughout.

A stock solution of 500 mg/L of tartrazine was prepared by dissolving 0.050 g of the tartrazine dye in water and diluting to 100 mL in a volumetric flask. More diluted solutions were prepared daily using this stock solution. Alkyl dimethyl benzyl ammonium chloride (ADBAC), methyltrioctylammonium chloride (aliquat 336), Acetic acid (glacial) 100% and potassium hydroxide were used. The cellulose acetate was obtained from waste photographic film tapes. In all materials, reporting of genetic description was not observed.

2.3. Preparation of cellulose acetate solution

The cellulose acetate was treated by sodium hypochlorite solution (5%) in order to remove colored gelatinous layers. The transparent films were fragmented and washed by detergent solution and water several times. The cleaned discolored cellulose acetate films were dried in oven at 50 °C for 1 h. Finally, 0.5 g of fragmented cellulose acetate films was dissolved in 500 mL glacial acetic acid and then kept in an appropriate container prior to use.

2.4. Preparation of modified cellulose nanosponges

The cellulose acetate nanosponges (CNSs) were prepared according to immersion-precipitation method (Reuvers et al., 1987). In order to achieve 20 mg CNSs, 20 mL of the prepared cellulose acetate solution (in previous section) was rapidly injected into 300 mL stirring water (at 500 X), resulting in the formation of the CNSs. The prepared CNSs were filtered through a filter paper (Whatman No. 41) and rinsed with water for several times to remove the excess of the acetic acid. The washed CNSs were poured in the 200 mL of 5 M of KOH solution, and then the mixture was stirred for 24 h, filtered and afterward washed with excess water. In the next step, the obtained alkalized CNSs were added to the methyltrioctylammounium chloride solution (containing 0.5 g methyltrioctylammounium chloride in 200 mL water) and the mixture was stirred for 15 h. After this period, filtering-washing procedure was repeated on the modified-cellulose nanosponges (*m*-CNSs) to remove the extra of the methyltrioctylammounium chloride. The resulting M-CNSs were dispersed in 20 mL of water, so one milliliter of this solution contained almost 1 mg of M-CNSs.

2.5. Adsorption procedure

The adsorption of tartrazine by M-CNSs was performed by a batch procedure at room temperature. A beaker containing 50 mL solution of tartrazine 5.0 mg/L (pH 1.5), was shaken (400 X) with 5.0 mg of M-CNSs for 100 min. After this time the initial yellow colored solution became colorless; the nanosponges were collected using passed through a filter paper. Then, the residual concentration of the tartrazine in the supernatant solution was determined by UUV–Visspectrophotometer at 427 nm. The removal efficiency of tartrazine by the M-CNSs adsorbent was calculated according to the following equation:

$$R\% = \frac{C_0 - C_t}{C_0} \times 100$$

Where R is the removal efficiency of the tartrazine, C_0 is the initial concentration of tartrazine (mg/L), and C_t is the concentration of tartrazine (mg/L) remaining in the solution.

2.6. Pre-concentration and recovery procedure

This procedure was carried out using a polyethylene tube (5 cm length and 1 cm inner diameter) with a very fine bore. The outlet of the column was fitted with the glass wool, and then filled with the 5.0 mg of the M-CNSs solution (equivalent to 5.0 mL). To perform pre-concentration, 200 mL of tartrazine solutions (containing 2–300 ng/mL) which has been equilibrated at pH 1.5 were passed through the column, at a flow rate 3.0 mL/min. Then, the column was washed with 4.0 mL of ADBAC solution (8% w/w) to release the dye retained in the column and its absorbance determined by UUV–Visspectrophotometer. A blank solution was also prepared under the same analytical conditions without adding any tartrazine dye. The recovery of tartrazine adsorbed on the M-CNSs was calculated from its amount in the starting sample and eluted from the column.

2.7. Sampling

The proposed method was applied to different water samples including, Karoon River water (Khuzestan Province, Ahvaz, Iran), Caspian Sea (northern Iran) and waste water of beverage factory (Khuzestan Province, Ahvaz, Iran). All water samples were filtered through a 0.45 μm cellulose acetate membrane filter and the pre-concentration procedure was performed on the samples as described above.

2.8. PZC experiment

To obtain more information about the surface of the M-CNSs, an experiment to obtain its PZC was performed. In such experiment, 5.0 mg of this material were shaken up with 10.0 mL of the solutions whose pH varied from 1 to 10. The pH of the solutions was adjusted by the addition of HCl and NaOH solutions, and the