Application of central composite design for methyl red dispersive solid phase extraction based on silver nanocomposite hydrogel: Microwave assisted synthesis

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In this study microwave irradiation was used to synthesis the silver nanocomposite hydrogel and the usefulness as an adsorbent for the preconcentration, extraction and determination of methyl red (MR) from water by dispersive solid phase extraction method was evaluated. For this purpose, silver nanocomposite hydrogel (SNH) was synthesized and characterized by thermo-gravimetric (TG) analysis, FT-IR, TEM and SEM images. The effects of some important variables such as centrifuge time (min), the pH of the methyl red solution, amount of adsorbent (g), acetone volume (mL) and chloroform volume (mL) on methyl red removal were studied using central composite design (CCD) and the optimum experimental conditions were evaluated by the desirability function (DF) combined response surface methodology (RSM). Under the optimal experimental conditions, the detection limit was found to be 1.4 μg mL⁻¹ for MR adsorption. The limit of quantification and the linear range of calibration curve were 4.6 μg mL⁻¹ and 0.1–25 μg mL⁻¹, respectively.

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1. Introduction

Azo dyes are the largest group of dyes used in industry and their durability cause more pollution to the environment. MR is a commonly mono-azo dye applied extensively in laboratory assays, textiles and other commercial products. Because of the existence of the benzene ring in MR structure, it has a stable structure which is not biodegradable compound [1]. Though, it may cause eye and skin irritation and pharyngeal or digestive tract irritation if inhaled or swallowed [2].

In recent years, several clean-up methods have been tested to remove dyes from water samples such as solid phase extraction (SPE) [3,4], solid phase micro-extraction (SPME) [5] and liquid phase micro-extraction (LPME) [6]. Dispersive solid-phase extraction (DSPE) is a relatively new technique for clean-up operations. This pre-concentration method is based on solid phase extraction methodology that was introduced for the first time by Anastassiades and co-workers [7], and it is a very efficient procedure used to increase selectivity in analytical processes. More recently, dispersive solid-phase micro-extraction has been reported as a miniature model of DSPE based on the use of micro amounts of the adsorbed phase [8,9]. Influence of important variables can be examined and optimized by experimental design. This technique provides a systematic way of working that allows conclusions to be drawn about the variables or its combination, which is most influential in the response factors while carrying out the minimum possible number of experiments. Among experimental designs, central composite design (CCD), Doehlert matrices, Box-Behnken designs and three-level full-factorial designs [10,11], CCD was selected for investigating the importance of variables among the other experimental design method.

The microwave irradiation is one technique, which is used for preparation of the hydrogel in several researches. Because of the microwave heating process has high temperatures for outbreak the solution with relatively short times and consequently makes reactions faster than under conventional thermal conditions [12]. The hydrogel reaction time reduces by applying microwave irradiation time [13] and creates hydrogel with higher the equilibrium swelling ratios. Application of microwave irradiation has good advantages over the conventional thermal methods in improving properties of the polymer synthesis [14]. In the current work we used the main benefit of microwave-assisted polymer synthesis is reduced reaction time, absence of organic solvent and limitation of side reaction [15] and, the dispersive solid phase extraction methodology was successfully applied for MR removal from the waters. In order to apply the proposed method for MR extraction and determination, a good adsorbent should be applied, which, hydrogel nanocomposite, with unique properties and biological applications were used as an adsorbent. They are hydrophilic polymer networks, which could absorb up to thousands of times of their dry weight in water without dissolving [16].

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Salez is a powder, which has been used a lot, because of its nutritive and demulcent properties and could be used as a thickening and stabilizing agent [17].

The present study reports two methods for preparation of SNH, (i) under sunlight UV-irradiation mediated synthesis (UV); (ii) under microwave assisted synthesis (MW) and characterization of SNH; (iii) investigation of swelling properties of the prepared hydrogels in various conditions; (iv) applying SNH (MW) as an inexpensive and efficient adsorbent for MR removal from aqueous solutions; (v) spread over central composite design (CCD) as experimental designs for optimization of several effective parameters on MR removal efficiency. The optimized procedure was applied for dispersive extraction and determination of MR in different water samples. The main objective developed method was removing MR from industrial effluents and then MR determination in liquid samples was performed as the second goal.

2. Experimental section

2.1. Materials

Salez was obtained from a supplier in Kordestan, Iran (Mn = 1.17 \times 10^8 \text{ g mol}^{-1}, Mw = 1.64 \times 10^8 \text{ g mol}^{-1} (high Mw), PDI = 1.39, eluent = water, flow rate = 1 \text{ mL min}^{-1}, acquisition interval = 0.43 s from GPC results). Silver nitrate (AgNO3, from Fluka, St. Louis, MO), methylene bis acrylamide (MBA, from Merck, Darmstadt, Germany) as a crosslinker, ammonium persulfate (APS, from Fluka, St. Louis, MO) as a water soluble initiator, and acrylic acid (AA, from Merck) as a monomer, were analytical grade and used without further purification. All other chemicals were also analytical grade. In through experiments, double distilled water (DDW) was used for preparing solutions.

2.2. Apparatus

SNH was synthesized under microwave irradiation by applying household microwave (Samsung Electronics company, temperature: 80 °C, power: 500 W). Fourier transforms infrared (FT-IR) spectra of samples in the form of KBr pellets were recorded using a Jasco 4200 FT-IR spectrophotometer. A Shimadzu UV-visible 1650 PC spectrophotometer was used for recording absorption spectra in solution. All samples were placed in a 1.00 cm quartz cuvette for UV measurements. The dynamic weight loss tests were conducted on a TA instrument 2050 thermo-gravimetric (TG) analyzer. All tests were conducted under N2 atmosphere (25 mL min\(^{-1}\)) using sample weights of 5–10 mg over a temperature range of 25–700 °C at a scan rate of 20 °C min\(^{-1}\). The morphology of the dried samples was examined using a scanning electron microscope (SEM) (Philips, XL30) operated at 10 kV after coating the dried samples with gold. Transmission electron microscopy (TEM) was taken on a Zeiss TEM at an acceleration voltage of 80 kV. Samples for TEM were prepared by putting a drop of solution on a carbon-coated copper grid. Duo to prove the stability of the prepared hydrogel as an adsorbent, silver content was measured before and after MR removal using a continuum source atomic absorption spectrometer model ContrAA (Analytikjena, Germany), using a graphite furnace atomizer (GFAAS). Transversely heated graphite furnaces made in pyrolitic graphite and equipped with L’Vov platform were used for atomization (Analytikjena, Germany) and microwave oven system UltraWAVE Milestone (USA) was also applied for digesting SNH in order to silver content measurement.

2.3. Hardware and software

All computations were carried out on a computer with 4 GB DDR3 memory and an Intel Core i5-3210M 2.5 GHZ with Turbo Boost up to 3.1 GHZ. The experimental design and statistical treatment of results we performed using Minitab 16 software (Minitab Inc., USA).

2.4. Preparation of SNH as an adsorbent

(i) Sunlight UV-irradiation mediated synthesis (UV): In general, a homogeneous solution of salep (1.0 g) in 80 mL DDW was prepared by dissolving appropriate amount of salep in DDW with mechanical stirrer (200 rpm). Silver nitrate solution (0.01 mol L\(^{-1}\)) was added to the mixture which was stirred for further 30 min. The reactor was placed in a thermostatic water bath preset at desired temperature (80 °C) and 5 mL of AA monomer (0.12 M), 5 mL of the MBA crosslinker (6.5 × 10\(^{-3}\) mol L\(^{-1}\)), and 5 mL of APS initiator (1.65 × 10\(^{-2}\) mol L\(^{-1}\)) were added and the reaction mixture was stirred until a gel-like product was observed after around 20 min. Finally, the reaction mixture was cooled to room temperature. The obtained hydrogel was dewatered by ethanol and dried in the vacuum oven at 50 °C, till constant weight was reached.

(ii) Microwave assisted synthesis (MW): MW synthesis contained the same weight ratios as UV synthesis, the mixed solution of salep, silver nitrate, AA, MBA and APS were dissolved in DDW. After that, the mixture was irradiated in a domestic microwave oven at 500 W for 5 min. At the end of the reaction, the resulting hydrogel was cooled to room temperature and then dewatered by immersing into 200 mL ethanol for overnight and at that time transferred into Petri dishes and dried for 6 h at 50 °C.

2.5. Swelling measurements

The accurately powdered SNH (0.1 ± 0.001 g) was transferred into the tea bag (i.e. a 100 mesh nylon screen) and they were immersed entirely in 200 mL distilled water and let them soak for 8 h at room temperature. Duo to remove the extra water of the tea bag, it was hung up for 10 min. The equilibrium swelling (ES) was considered according to the following equation:

\[
\text{ES(g/g)} = \frac{(W_c - W_d)}{W_d}
\]

where Wc and Wd are the weights of the swollen gel and the dry sample, respectively. The ES is reported as grams of water per grams of resin (g/g).

2.6. Evaluating swelling capability of SNH

In order to achieve the most stability hydrogel with high swelling capabilities, SNH components were studied and optimized. The swelling capability of the prepared hydrogel was investigated as a function of MBA concentration. Different concentration in the range of (0.001–0.1 mol L\(^{-1}\) was applied and the maximum water absorbance was achieved at 6.5 × 10\(^{-3}\) mol L\(^{-1}\). By changing the MBA concentration in the range of 0.001–0.006 mol L\(^{-1}\), no gel was formed in the MW, but SNH with a lower swelling capability was obtained in the UV. Flory proved the correlation between the swelling ratio and network structure parameters, presented as Eq. (2) [18].

\[
q_{\text{sw}} = \frac{1}{2} \frac{(1 - x)^{1/2}}{1 - x} \frac{1}{V_c/V_0} + \frac{(1 - x)^{1/2}(1 - x)}{V_c/V_0}\frac{1}{V_c/V_0}
\]

where, q_{sw} S-1, i, V_c, V_0, x, V_t are swelling ratio, ionic strength in the external solution, the valence of ionic groups, the ionic hydrogel concentration, the final volume of the swelled hydrogel and reference state volume dry hydrogel, the solvent interaction parameter and molar volume of swelling liquid, respectively. The terms of i/2V_n/V_c and (1/2 – %1)/V_t are the concentration of the fixed charges of the unswollen networks, the crosslink density, which refers to the number of effectively crosslinked points between chains in unit volume, the interaction parameter, i.e., affinity of the hydrogel to water, respectively.
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