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Fast removal and recovery of congo red by modified iron oxide magnetic nanoparticles

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Abstract: The adsorption and removal of congo red from an aqueous solution by iron oxide nanoparticles (IONPs) coated with cetyltrimethylammonium bromide (CTAB) as adsorbent was reported. The novel magnetic separation was quite efficient for the adsorption and desorption of congo red. The effect of temperature, pH of aqueous medium, and interfering ions on the recovery process were also investigated. The results indicated that the CTAB-coated IONPs could be employed in the removal of the anionic dye from wastewater. The congo red was removed successfully in spiked samples of Karoon River water.
Keywords : Dye; Spectrophotometry; Azo; Adsorption isotherm; Karoon River.

1. Introduction

The development of nanotechnology has been developed fastly in recent years. Nanosized magnetic particles possess high performance in the separation process due to the high specific surface area and the absence of internal diffusion resistance [1]. Nanosized magnetic iron oxide particles have a wide range of applications in ferrofluids, high-density information storage, magnetic resonance imaging (MRI), biological cell labeling and sorting, separation of biochemicals, targeting, and drug delivery [2]. They can be easily recovered with an external magnetic field [3].

Surface modification of magnetic nanoparticles is a challenged key for different applications and can be accomplished by physical/ chemical adsorption of organic compounds by four major methods: organic vapor condensation, polymer coating, surfactant adsorption and direct silanation [2]. Removing of hazardous compounds from industrial effluents is one of the growing needs in the present time. Different techniques including coagulation, adsorption, chemical oxidation and froth floatation have been used to remove organics and/or inorganics from wastewaters.

Many dyes and pigments are toxic in nature with suspected carcinogenic and mutagenic effects [4] that affect aquatic biota and humans [5]. Congo red (CR) (1-naphthalenesulfonic acid, 3,3ó-(4,4ó-biphenylenebis (azo)) bis (4-amino-) disodium salt) is a benzidine-based anionic disazo dye. This dye is known to metabolize to benzidine, a known human carcinogen.

Dyes and pigments represent one of the problematic groups; they are emitted into wastewaters from various industrial branches, mainly from the dye manufacturing and textile finishing [6] and also from foodcolouring, cosmetics, paper and carpet industries. Synthetic dyes have complex aromatic structures which provide them physico-chemical, thermal and optical

stability [7,8]. CR containing effluents are generated from textiles, printing and dyeing, paper, rubber, plastics industries, etc. Due to its structural stability, CR is difficult to biodegrade. Physico-chemical or chemical treatment of such wastewaters is, however, possible[7,9]. Adsorption is considered an attractive option in treating such wastewaters [10,11].

The present investigation reports a new, simple and fast method for removal and recovery of congo red by iron oxide nanoparticles coated with cetyltrimethylammonium bromide (CTAB) as adsorbent. The main objective of the present work may be formation of bilayer surfactant-coated magnetic nanoparticles for the separation of dye.

2. Materials and methods

2.1. Materials

congo red, methanol (99.9% m/m), ammonia solution (25% m/m), hydrochloric acid (37% m/m), $FeCl_3$ (96% m/m) and $FeCl_2$. $4H_2O$ (99.9% m/m) were purchased from Merck (Darmstadt, Germany) and were used without further purification processes. CTAB was obtained from Sigma. All chemicals and reagents were of analytical grade purity.

The spectrophotometric measurements were carried out with a Perkin Elmer lambda25 spectrophotometer. A Metrohm pH meter model 632 (Herisau, Switzerland) for pH adjustments and a magnet (1.2 T, 10 cm × 5 cm ×2 cm) for settlement of magnetic nanoparticles were used.

2.2. Preparation of iron oxide magnetic nanoparticles

Iron oxide nanoparticles (IONPs) were prepared according to the method described by Berge et al. (1999) without using any surfactant. The reaction for the formation of nanoparticles was suggested according to the following equation:

$2FeCl_3 + FeCl_2 + 8NH_3 + 4H_2O \rightarrow Fe_3O_4\ 8NH_4Cl$ ..... (1)

The particles were ageing in the solution under mechanical stirring, decanted by magnetic settling, and washed three times with distillated water. The IONPs were imaged with TEM (906E, LEO, Germany) and their sizes were in the range of 20–80 nm.

2.3. Separation process

The adsorption of congo red by magnetic nanoparticles was carried out in a 0.02 M phosphate buffer (pH 6) solution at 25 ^oC. Phosphate buffer solution (pH 6) was prepared by adding appropriate amounts of sodium hydroxide into a 1 M phosphoric acid solution. In General, 0.3g of damped IONPs (equivalent to 0.030 g of dried particles) and 0.75 mL of 0.5% (m/v) of CTAB were added to a 50 mL solution of congo red (2 lg mL^{-1}) containing 1 mL of phosphate buffer (1 M). After mixing for 1 min, magnetic nano-adsorbents were separated magnetically from solution by the magnet. Removal percent and adsorbed amount of congo red was determined by photometric measurement of absorption of the sample solution before and after removing process at 521 nm.

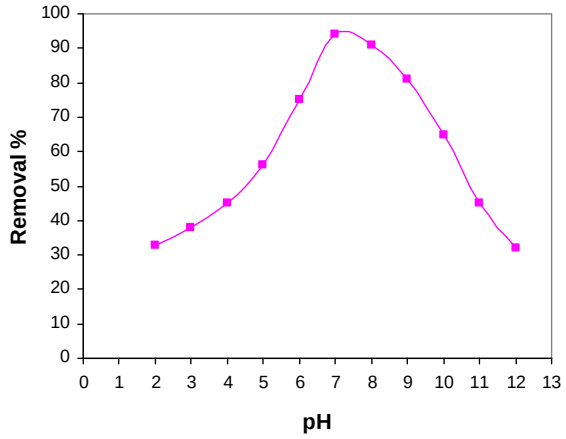

Fig. 1. Adsorption of congo red on S-IONPs as a function of pH.

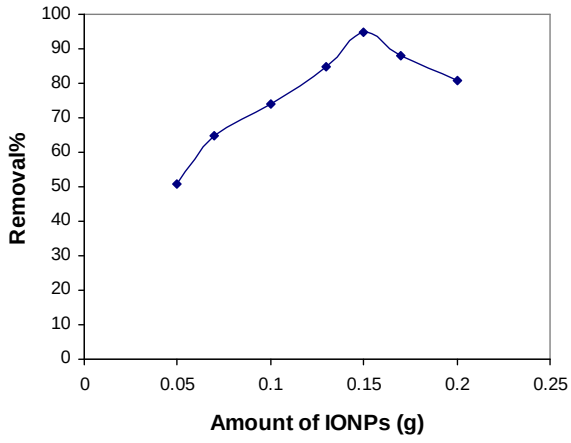

Fig. 2. Effect of different amounts of IONPs on the adsorption of CR.

3. Results and discussion

The removal and preconcentration of congo red by surfactant modified IONPs (S-IONPs) showed fast separation of this dye from the bulk of the water solutions. Moreover, the removal of congo red reached equilibrium within about 3 min. Such a fast adsorption rate could be referred to the absence of internal diffusion resistance [12]

3.1. Effect of pH variation

The effect of pH on the adsorption of congo red (2.0 µg mL^{-1}) by 0.4 g of damped IONPs at 25 ^{0}C showed (Fig. 2) that the adsorption and removal of congo red remains constant at pH range from 3 to 9. The mean pH value (6) was used as optimum pH for further works.

3.2. Effect of the amount of IONPs

The amount of IONPs for complete removal of CR (2 µg mL^{-1}) in a 50 mL solution at pH 6 was investigated. The results are shown in Fig. 2 and indicated that addition of 0.15 g of damped IONPs per 50 mL solution of CR (2 µg mL^{-1}) lead to maximum separation of dye

3.3. Effect of the CTAB amount

The results revealed that by increasing the amount of surfactant, the rate of removal of CR was nearly constant. Therefore, 0.75 mL of 0.5% (m/v) solution of CTAB per 50 mL solution was chosen as optimum amount of CTAB for further experiments.

3.4. Effect of temperature

The adsorption of CR (15 µg mL^{-1}) in a 50 mL solution using 0.15 g of damped S-IONPs at pH 6 under different temperatures showed that the adsorbed amount of CR remained almost constant at different temperatures and the removal was greater than 95%.

3.5. Desorption solvent (eluent)

The study revealed that the desorbed CR was increased with increasing the content of methanol in the solution and the dye could be completely desorbed in the presence of pure methanol. In this study more than 96% of CR could be desorbed and recovered by pure methanol.

4. Removal of congo red from spiked Karoon River water

To determine the ability of the proposed method for the removal of congo red from sample, Karoon River water was spiked. The determination of congo red in Karoon River water spiked with 15 µg mL^{-1} and 30 µg mL^{-1} indicated that the spiked solutions were successfully removed in different river water samples. More than 95% was removed and the complex matrix of river water samples does not interfere with the removal of congo red.

5. Conclusion

A fast, simple and new magnetic removal of congo red from aqueous solution has been successfully developed with CTABcoated IONPs as adsorbent. The adsorbent could be manipulated magnetically and exhibited high adsorption capacity and fast adsorption and desorption rates for the removal of congo red due to the high specific surface area and the absence of internal diffusion resistance. The adsorption behavior could be described by Langmuir isotherm. The adsorbent may also be useful for the removal of other anionic dyes from aqueous solutions. The adsorbed congo red could be desorbed using methanol solvent. The whole adsorption–desorption processes can be completed within 3 min☺☺☺. In addition the proposed procedure offered higher removal and recovery percents and also shorter adsorption–desorption times of the dye compared with most of the previously reported methods.

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