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Highly effective removal of basic fuchsin from aqueous solutions by anionic polyacrylamide/graphene oxide aerogels

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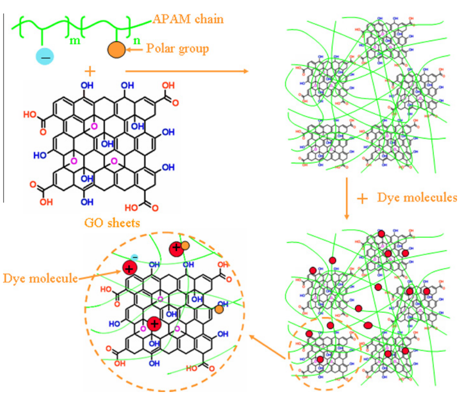
GRAPHICAL ABSTRACT



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ABSTRACT

Novel anionic polyacrylamide/graphene oxide aerogels were prepared by a freeze drying method and used to remove basic fuchsin from aqueous solutions. These aerogels were sponge-like solid with light-weight, fluffy and porous structure. The batch adsorption experiments were carried out to study the effect of various parameters, such as the solution pH, adsorbent dose, contact time and temperature on adsorption properties of basic fuchsin onto anionic polyacrylamide/graphene oxide aerogels. The kinetics of adsorption corresponded to the pseudo-second-order kinetic model. The Langmuir adsorption isotherm was suitable to describe the equilibrium adsorption process. The maximum adsorption capacity was up to 1034.3 mg/g, which indicated that anionic polyacrylamide/graphene oxide aerogels were promising adsorbents for removing dyes pollutants from aqueous solution.

1. Introduction

Dyeing wastewater has been a source of serious environmental pollution over the past decades. Basic fuchsin is the most commonly used organic dye in some industries, such as textile, printing, paper, leather and feather [1]. However, basic fuchsin has been listed as a suspected carcinogen in several countries due to

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its toxicity and carcinogenicity [2]. Different methods, including biological oxidation [3], membrane separation [4], photocatalytic degradation [5], ion exchange [6], chemical precipitation [7] and adsorption, have been explored to remove such organic dyes from wastewater. Adsorption is a simple and promising separation process among them [8,9]. The exploitation of novel and more effective adsorbents is always an important focus in related fields.

Graphene, a new member of carbon-based materials, has a unique two-dimensional structure [10]. Graphene oxide (GO) is an important precursor of graphene-based structure. On the surface of GO, large amounts of polar groups exist, such as carboxyl, hydroxyl and epoxy groups, which endow it extremely hydrophilicity [11]. Graphene and GO have been reported as effective adsorbents to adsorb hazardous substances in wastewater such as dyes [12,13] and antibiotics [14,15]. However, pure graphene and GO have tiny size, excellent chemistry stability and dispersibility in water, which make them difficult to be removed from water and can form secondary pollution. Three-dimensional structure endows graphene many new or improved properties, to overcome the deficiencies of two-dimensional graphene or develop new application, so the construction of three-dimensional graphene macrostructures has attracted more and more attentions [16–18].

As a kind of low-cost and effective flocculants, anionic polyacrylamide (APAM) is widely used to treat industrial and municipal wastewater. A number of polar groups exist in APAM molecular chain, which can adsorb the solid particles suspended in wastewaters [19–21]. Combining charge neutralization and excellent bridging cohesion, APAM molecular chain is immobilized on the surface of different particles, and polymer bridges are formed among the particles [22,23], which lead to aggregation of the particles to form large flocs and then sink. However, conventional APAM flocculants are mainly prepared dilute solution to use directly in water treatment, which lead to limited flocculation. Developing composite flocculants is more desired than using traditional flocculants.

On the basis of above exemplary properties of GO and APAM, the combination of them is suitable for practically environmental protection applications. At the same time, construction of three-dimensional composite materials with them, can effectively solve the problem of secondary pollution of pure GO, and the ability of composite materials to treat wastewater is higher than that of pure APAM. Up to now, except for a few reports on the preparation of graphene-based PNIPAM hydrogels [24,25], there have been no investigations devoted to synthesizing APAM/GO composites for wastewater treatment. In this study, novel APAM/GO aerogels were prepared by freeze drying and used as adsorbent to remove basic fuchsin from aqueous solutions. Influencing parameters on the adsorption were detailedly studied, including the content of GO, initial solution concentration, solution pH, temperature, adsorption time and adsorbent dosage.

2. Materials and methods

2.1. Materials

APAM was purchased from Tianjin Guangfu fine chemical industry research institute. Basic fuchsin was purchased from Tianjin Hongyan Reagent Factory, China.

2.2. Preparation of GO

GO was prepared from expandable graphite by a modified Hummers method [26]. H_2SO_4 (115 mL, 98 wt.%) was added into a beaker in ice bath, and then $KMnO_4$ (15 g), $NaNO_3$ (2.5 g) and expandable graphite (2.5 g) were slowly put into the beaker in sequence and sufficiently blended. After the beaker was kept at 0 °C for 24 h, the mixture was stirred at 35 °C for 30 min. 230 mL deionized water was added into the beaker, while the temperature was gradually increased to 98 °C and kept for 15 min. H_2O_2 (30%) was added into the beaker to react with residual oxidant, and then the color of the mixture became golden brown. At last the mixture was washed and centrifuged with HCl (5%) and deionized water several times, and sonicated for 2 h to obtain GO.

2.3. Preparation of APAM/GO

APAM/GO aerogels were prepared by freeze drying. Firstly, APAM (2 g) was dissolved in deionized water to prepare 0.5% solution, then APAM solution was added into a beaker with GO (0.5 g). After slightly stirred and placed for 24 h, the gels of APAM/GO composites were formed. Secondly, the gels were divided into two samples with equal weight. One sample was heated by a chamber electric oven at 65 °C until dry completely (named oven heating sample). Another sample was put into a plastic bag and frozen in a freezer for 24 h, then it was placed in a vacuum freeze dryer to remove moisture for 72 h to form APAM/GO aerogels (named freeze drying sample). The samples with other different APAM and GO ratios and dried by two different methods were also prepared.

2.4. Characterization of the adsorbents

The surface morphologies of APAM and APAM/GO composites were characterized by scanning electron microscope (SEM, JSM 6700F, JEOL Ltd., USA). The surface functional groups of APAM/GO aerogels were tested by a Perkin–Elmer-283B FTIR spectrometer within the wave number from 400 to 4000 cm^{-1}.

2.5. Adsorption experiments

Basic fuchsin was dissolved in deionized water to make 1000 mg/L stock solution. To compare adsorption capacity of basic fuchsin onto APAM/GO composites (containing 20% GO and 50% GO) dried by freeze drying and oven heating methods, 25 mg of APAM/GO composites were added into basic fuchsin solution with initial concentration of 400 mg/L. Then the solution was vibrated in a water-bathing constant temperature vibrator for 72 h at room temperature (20 °C). Residual concentration of basic fuchsin in the solution was measured with a UV–visible spectrophotometer (TU-1810, Beijing Purkinje General Instrument Co., Ltd., Beijing). The amounts of basic fuchsin adsorbed by APAM/GO at equilibrium and time t were calculated from the following equations:

$$q_e = \left(\frac{C_0 - C_e}{m}\right)V \quad (1)$$

$$q_t = \left(\frac{C_0 - C_t}{m}\right)V \quad (2)$$

where C_0, C_e and C_t (mg/L) are the initial concentration, equilibrium concentration and certain concentration at time t (min), respectively, V is the volume (mL) of the basic fuchsin solution and m is the mass (mg) of the adsorbent.

The effect of initial concentration on adsorption was tested by adding 25 mg adsorbent into 50 mL solution with different basic fuchsin concentrations (100–400 mg/L). The effect of adsorbent dosage on adsorption was performed by adding different quantity of the adsorbent (5–65 mg) into 50 mL basic fuchsin solution with concentration of 400 mg/L. To determine the effect of pH, 25 mg adsorbent was added into 50 mL basic fuchsin solution with