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Regular Article

Sodium dodecyl sulfate intercalated and acrylamide anchored layered double hydroxides: A multifunctional adsorbent for highly efficient removal of Congo red

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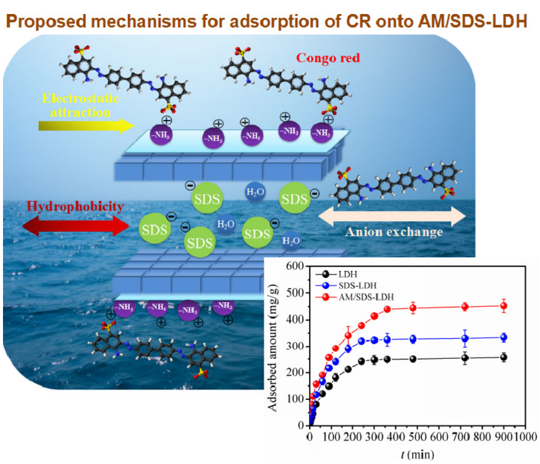
G R A P H I C A L   A B S T R A C T



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In this study, a novel adsorbent was designed and synthesized via intercalation of sodium dodecyl sulfate (SDS) in the interlayer, and subsequent grafting of acrylamide (AM) on the layer of layered double hydroxides (LDH) (named as AM/SDS-LDH), and applied for congo red (CR) removal. The morphological structure, wettability and physicochemical properties of the adsorbents were thoroughly characterized using SEM, EDS, N_2-adsorption/desorption isotherm, static contact angle, XRD and FTIR. Results elucidated that SDS and AM were successfully introduced into the interlayer and onto the layer of LDH, respectively. Adsorption experimental results suggested that the maximum adsorption capacities of CR on SDS-LDH and AM/SDS-LDH at pH 5.0 and 293 K were 714.29 and 1118.78 mg/g, respectively, which were much higher than that of CR on LDH (588.24 mg/g). Based on the BET, XRD and FTIR analysis, the higher adsorption capacity of AM/SDS-LDH was mainly attributed to high surface area, large basal spacing as well as the abundant $—NH_2$ groups. The experimental data can be well fitted by pseudo-second-order kinetic and Langmuir isotherm models. Thermodynamic parameters indicated the adsorption process was favorable under the higher temperature condition. The synergistic effect

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existed during the adsorption process of CR onto AM/SDS-LDH, including the electrostatic interactions, anion exchange and hydrophobic-hydrophobic interactions. Overall, this study provided a strategy for design and fabrication of highly efficient adsorbents.

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1. Introduction

Dyes are widely used in many industries including papermaking, electroplating, textile, rubber, plastics, and printing industries. More than 7×10^5 tons of different commercial dyes are produced by mentioned industries around the world annually [1]. And about 5–10% of untreated dyestuffs are discharged into natural waters as wastes without reasonable treatment [2,3]. The extensive use of dyes poses pollution problems in forms of colored wastewater bodies. This not only influences aesthetic merit but also reduces light penetration and photosynthesis. Moreover, most dyes have complex aromatic molecular structure, making them highly toxic, non-biodegradable, mutagenic and carcinogenic for aquatic life and human being [4,5]. Thus, it is extremely important and urgent to remove various dyes from industrial wastewaters before discharging into natural water bodies. Congo red (CR) is a popular benzidine-based anionic bisazo dye used in many industries. It is known to metabolize to benzidine, which is a potential danger of bioaccumulation and exposure to this dye can creates allergic problems.

Most of the dyes are stable to photo-degradation and biodegradation. Meanwhile, the conventional biological treatment process was ineffective in removing dyes from aqueous solution [6]. Therefore, various physic-chemical technologies have been employed for the treatment of dyes sewage, including flocculation [7], chemical coagulation [8], advanced oxidation [9], membrane separation [10], and ion-exchange [11]. However, the application of these techniques has been limited due to poor efficiency and high cost. Adsorption is regarded as a promising method because of its low cost, easy operation and simple design. Adsorption technique also easy scaling from laboratory scale to field scale, and effectiveness in removing dyes in low concentration [12].

A wide variety of materials have been used as adsorbents [1–4,6,12]. Among them, layered double hydroxides (LDH) have received increasingly attraction in environmental pollution cleanup in recent years due to its extraordinary ion-exchange ability, large surface area and simple synthesis method [13]. More importantly, LDH can be intercalated with different compounds such as crown ether, lignin, EDTA, and sulfur [14,15]. It confirms that the organic modification not only improved the adsorption capacity by optimizing the lipophilicity and the spacing of layers of LDH, but also imparted some significant characteristics, such as high retention affinities of organic compounds, hydrophobic interactions and buffering property over a wide solution pH range [16,17].

Polymers have abundant specific functional groups [18–21]. Much attention have been paid to graft polymers onto materials like kaoline [22], attapulgite [23], bentonite [24], chitosan [25], and cellulose [26]. Shirsath et al. [21] reported a poly (acrylic acid) hydrogel composite (PAA-K hydrogel), and the PAA-K hydrogel exhibited a adsorption capacity of 26.31 mg/g for Brilliant Green dye at 308 K. Wang et al. [22] successfully synthesized chitosan-g-poly (acrylic acid) (CTS-g-PAA) and CTS-g-PAA/attapulgite composites (CTS-g-PAA/APT), and the maximum adsorption capacities of CTS-g-PAA and CTS-g-PAA/APT for methylene blue reached 1873 and 1848 mg/g, respectively. Chitosan-poly(vinyl alcohol)/bentonite nanocomposites prepared by Wang et al. [23] could remove Hg(II) ions with the capacity up to 460.18 mg/g. Yu et al. [24] found that chitosan derivative/Fe_3O_4 composite magnetic nanoparticles could effectively adsorb Hg(II) with a adsorption capacity of 1.18 mmol/g at 298 K. Wang et al. [25] prepared a polyacrylamide grafted quaternized cellulose to remove anionic dyes, the maximum adsorption capacity of this material was 380.084 mg/g for CR and 349.284 mg/g for Eriochrome blue SE. However, to the best of authors' knowledge, there was no study using both sodium dodecyl sulfate (SDS) and acrylamide (AM) for the modification of LDH for pollutants adsorption.

In this study, a novel LDH-based material was designed and synthesized via intercalation of SDS in the interlayer, and subsequent grafting of AM on the layer of LDH (named as AM/SDS-LDH). The structure and textural properties of the adsorbents were thoroughly characterized. The adsorption properties of CR onto AM/SDS-LDH were studied as a function of the influencing parameters, such as initial solution pH, contact time, initial concentration, and temperatures. The adsorption kinetics, isotherms and thermodynamics parameters for CR adsorption on the AM/SDS-LDH were systematically investigated, and the possible adsorption mechanisms were also proposed. This study provided an comprehensive strategy for design and fabrication of high effective adsorbent for removing CR from wastewater.

2. Materials and methods

2.1. Reagent and materials

Congo red(CR) was purchased from Tianjin GuangFu Fine Chemical Reagent Co., Ltd., China. Azodiisobutyronitrile (AIBN), Triethoxyvinylsilane (KH151), Dimethylformamide (DMF), sodium dodecyl sulfate (SDS), acrylamide (AM), NaOH, $Mg(NO_3)_2 \cdot 6H_2O$, and $Al(NO_3) \cdot 9H_2O$ were purchased from Sinopharm Chemical Reagent Co., Ltd., China. All of these reagents were analytical grade and used as received without further purification. Double distilled water was used in all the experiments.

2.2. Synthesis of adsorbents

The synthetic procedure of adsorbents is depicted in Text S1 (Fig. S1) in Supporting information.

2.3. Characterization of adsorbents

The morphologies of adsorbents were determined using scanning electron microscopy (SEM, Hitachi-4800, Japan). Powder X-ray diffraction (XRD) pattern were collected with a Rigaku D/Max-2500 powder diffractometer using Cu Kÿ radiation with a scan step of 0.02ÿ. The Brunauer Emmett Teller (BET) surface areas and the pore size distributions were determined by Nitrogen adsorption/desorption isotherms on an Autosorb-iQ automated gas-sorption analyzer (Quantachrome Instruments, USA). Fourier transform infrared spectroscopy (FT-IR) was obtained using the KBr pressed pellet method with a meter (Thermo Nicolet, Nexus-470,USA) in range of 4000–400 cm^{-1} with a resolution of 0.5 cm^{-1}.

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