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Transport of anionic azo dyes from aqueous solution to gemini surfactant-modified wheat bran: Synchrotron infrared, molecular interaction and adsorption studies

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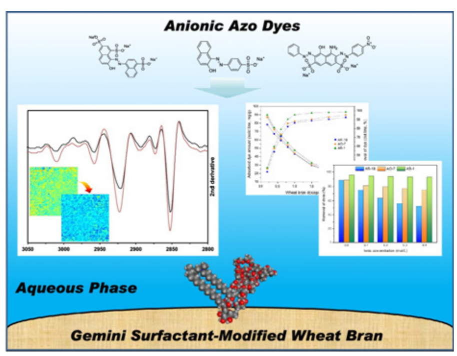
HIGHLIGHTS

- Investigated the adsorption of azo dyes on gemini surfactant-modified wheat bran
- The equilibrium and kinetic adsorption studies were conducted.
- The interaction between azo dyes and surfactant-modified wheat bran was explored.
- New insights from synchrotron IR analysis and molecular interaction simulation
- Modified wheat brans have the potential to be used as adsorbents in dye removal.

GRAPHICAL ABSTRACT



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ABSTRACT

From the view of economic efficiency and technology sustainability, biomass adsorbent has a high potential for pollution control. In the present study, the performance of gemini 12-2-12 surfactant-modified wheat bran (MWB) for the removal of anionic azo dyes from aqueous solution was investigated. A new insight was gained into the modification mechanism through synchrotron-assisted infrared analysis and molecular interaction simulation. The equilibrium and kinetic studies for the adsorption of Acid Red 18 (AR-18), Acid Orange 7 (AO-7) and Acid Black 1 (AB-1) on MWB were conducted. The Langmuir model well fit the adsorption isotherm data. The adsorption kinetics could be described by the pseudo-second-order and intra-particle diffusion models. The results of thermodynamic studies indicated the adsorption of AR-18 and AB-1 onto MWB was endothermic and spontaneous, while the adsorption of AO-7 was exothermic. The optimum pH for the adsorption of anionic azo dyes on MWB was 3. The adsorbed amount of anionic azo dyes onto MWB decreased when NaCl concentration increased from 0 to 0.4 mol L^{-1}. The potential of modified wheat bran as a suitable adsorbent for the removal of dyes from wastewater was presented in this study. The results can help understand the migration patterns of organic pollutants at wheat bran-water interface.

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1. Introduction

Azo dyes are a major class of synthetic organic compounds, which account for more than half of the annually produced amount of dyes (Ferreira et al., 2016; Vacchi et al., 2013). Various techniques such as chemical sedimentation, photocatalytic degradation, membrane filtration, advanced oxidation, and electrochemical decolourisation have been used to remove azo dyes from effluents (Ahmad et al., 2015). Adsorption is an alternative technology with the advantages of simple operation and good removal efficiency (Cui and Gan, 2013; Ehsan et al., 2017; Ersan et al., 2016; Wang et al., 2014; Zhao et al., 2016). A wide range of adsorbent materials has been applied in the removal of azo dyes (Shoukat et al., 2017). It was reported that activated carbon could be used to remove azo dyes (Vargas et al., 2012). However, its application is limited due to its high cost (Arami et al., 2006). Some inorganic materials like kaolin, zeolite, sepiolite, bentonite, and perlite have also been utilized in the removal of azo dyes as alternative low-cost adsorbents.

From the view of economic efficiency and resource sustainability, biomass adsorbent has a high potential to be used in pollution control. In particular, considerable attention was recently given to the use of low-cost agricultural residues and by-products as adsorbents (Rosales et al., 2015; Safa et al., 2016). Some researchers have examined the ability of soy meal hull (Arami et al., 2006), rice bran (Ogata et al., 2015), wheat straw (Han et al., 2010), and wheat shells (Bulut et al., 2007) as adsorbents to uptake dyes from wastewater. Wheat bran, the outer shell of wheat grain, is available as a by-product of the wheat-milling process. It is the outer shell of wheat grain, accounting for about 20% of wheat grain weight. In structure, wheat bran consists of outer pericarp, inner pericarp, testa, hyaline and aleurone layers. Dietary fiber which is composed of plant cell walls and components obtained from these walls is the major contributor to wheat bran structure (Harris et al., 1996; Esposito et al., 2005). The large surface area of these fibers is favorable for their binding with other compounds (Ryden and Robertson, 1995). The potential of using wheat bran as effective adsorbent to remove azo dyes has not been explored to present.

The binding of a particular pollutant to adsorbent depends on its ability to interact with both adsorbent and other components in the system (Gan et al., 2003a, 2003b). To achieve the desirable adsorption efficiency, some efforts have also been made to modify the biomass adsorbents through appropriate treatments. The phosphorylation and thermochemical esterification were used to modify the crops straw and improve its adsorption capacity (Gong et al., 2007; Gong et al., 2008). There is also a particular interest in surfactant-assisted surface modification. It was reported that hexadecylpyridinium chloride monohydrate-modified barley straw was used to remove emulsified food and mineral oils from wastewater (Ibrahim et al., 2009). The adsorption of Congo red by hexadecyltrimethyl ammoniumbromide-modified wheat straw was also investigated (Zhao et al., 2014). Although these findings are encouraging, knowledge about the adsorption of pollutants onto surfactant-modified biomass is still limited. A number of issues about the interface-transport characteristics are poorly understood.

Gemini surfactants are composed of two monomeric surfactant molecules which are chemically bonded together by a spacer (Wei et al., 2011; Wei et al., 2012). Compared with conventional surfactants, gemini surfactants have the advantages of higher surface activity, lower critical micelle concentration (CMC), lower Krafft temperature and better viscoelastic features (Zhao et al., 2015). They are ten to hundred times more efficient at reducing the surface tension of water than conventional surfactants (Kumar and Tyagi, 2014). It is a cost-effective option for the use of gemini surfactants in environmental applications.

The present study therefore investigated the adsorption of anionic azo dyes onto gemini surfactant-modified wheat bran in the solution. The adsorption performance in both equilibrium and kinetic terms were studied. The effects of aqueous chemistry characteristics such as temperature, pH and ionic strength were also evaluated. This is the first study to our knowledge to explore the interaction between anionic azo dyes and gemini surfactant-modified wheat bran. Innovative attempts were presented to obtain new insights into the nature of surfactant-assisted biomass modification through synchrotron infrared spectroscopy analysis and molecular interaction simulation.

2. Materials and methods

2.1. Chemicals

Acid Red 18 (AR-18, C.I. 16255, MW: 604.47), Acid Orange 7 (AO-7, C.I. 15510, MW: 350.32) and Acid Black 1 (AB-1, C.I. 20470, MW: 616.49) obtained from the Aladdin Industrial Corporation (Shanghai, China) were used as the representative anionic azo dyes. Cationic gemini surfactant (N1-dodecyl-N1, N1, N2, N2-tetramethyl-N2-octylethane-1,2-diaminium bromide, 12-2-12) was obtained from Chengdu Institute of Organic Chemistry (Chinese Academy of Sciences, Chengdu, China). The chemical structures of selected anionic azo dyes and gemini surfactant are shown in Fig. S1 and S2, respectively. All other chemicals used were of reagent grade quality or higher.

2.2. Wheat bran adsorbents

The wheat bran used in this study was a by-product obtained from a flour mill (Shandong, China). The wheat bran was washed with distilled water for several times to remove dust and soluble material. Then the sample was dried in an oven at 338 K for 24 h and was ground, screened and stored in a glass container. It was labeled as raw wheat bran (RWB). The surfactant-modified wheat bran was prepared by mixing RWB in 1 mmol L^{-1} gemini surfactant solutions at a ratio (wheat bran weight (g):liquid volume (mL)) of 3:50. The mixture was stirred on a reciprocal shaker at 200 rpm at 298 K for 24 h. The treated wheat bran was separated from the solution and washed with distilled water several times to eliminate surface retained surfactant. Then the treated wheat bran was dried and designated as modified wheat bran (MWB).

2.3. Adsorption studies

All experiments were conducted in 250 mL conical flasks with Parafilm sealing film. For each experiment, appropriate MWB and 100 mL dye solution at certain concentration were added to conical flask. Different amounts of MWB ranging from 0 to 2.5 g were added to conical flasks to investigate the effect of MWB dosage on dye adsorption. Adsorption isotherms were measured at different temperatures of 293, 303 and 313 K, with initial dye concentrations varied from 0 to 100 mg L^{-1}. The effect of pH on dye adsorption was studied using adsorbent dosage of 1 g L^{-1}, initial dye solution of 50 mg L^{-1}, and temperature of 293 K. The initial pH of each solution was adjusted by 0.1 mol L^{-1} HCl and 0.1 mol L^{-1} NaOH solution. To study the effect of ionic strength, appropriate NaCl were added into each conical flask with 100 mL dye solution at concentration of 50 mg L^{-1} and 0.1 g MWB. All flasks in these experiments were placed on a reciprocal shaker at 200 rpm for 12 h to reach the adsorption equilibrium. Preliminary experiments showed that 12 h were enough for reaching equilibrium of dye adsorption. Then the aqueous samples were taken from flasks and the concentrations of anionic azo dyes were measured after centrifugation. The adsorbed amount of dyes can also vary with time. In experiments of adsorption kinetics, 100 mL dye solution at the concentration of 50 mg L^{-1} and 0.1 g MWB were added to flasks. Then, the adsorption experiments were conducted at 293, 303 and 313 K. Dye concentrations were monitored at different time points. The dye concentration in aqueous solution was determined using a UV–Vis spectrophotometer (Unico Com., China). The absorbance values for AR-18, AO-7 and AB-1 were