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Effects of co-substrate and biomass acclimation concentration on the bioregeneration of azo dye-loaded mono-amine modified silica

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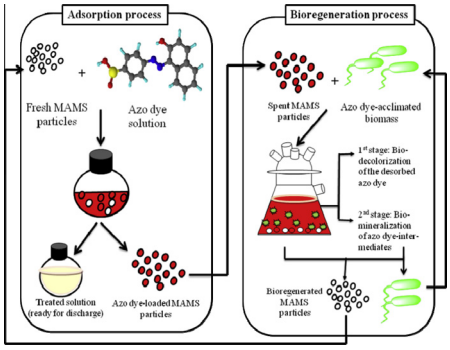
HIGHLIGHTS

- Bioregeneration of AO7-loaded MAMS in the absence of co-substrate.
- Bioregeneration of AY9- and AR14-loaded MAMS in the presence of co-substrate.
- Shortening of bioregeneration duration using higher acclimation concentration.
- No effect on bioregeneration efficiency under different acclimation concentrations.

GRAPHICAL ABSTRACT



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ABSTRACT

Bioregeneration of mono-amine modified silica gel (MAMS) adsorbent loaded with Acid Orange 7 (AO7), Acid Yellow 9 (AY9) and Acid Red 14 (AR14), respectively, was investigated under two different operational conditions, namely absence/presence of sucrose/bacto-peptone as the co-substrate and different biomass acclimation concentrations. The results revealed that the AY9- and AR14-loaded MAMS adsorbents could almost be completely bioregenerated but only in the presence of co-substrate whereas the bioregeneration of AO7-loaded MAMS could achieve up to 71% in the absence of the co-substrate. These differences could be related to the structural properties of the investigated azo dyes. In addition, the results showed that the bioregeneration duration of AO7-loaded MAMS could be progressively shortened by using biomass acclimated to increasingly higher AO7 concentration. However, the bioregeneration efficiencies were found to be relatively unchanged under different biomass acclimation concentrations.
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1. Introduction

One of the challenging tasks for chemists is to pursue green(er), safe(r) and clean(er) technique to remove the huge amount of dyes and byproducts generated from the textile industries. Unfortunately, cleaner dye wastewater treatment methods imply higher energy/operation costs. Nonetheless, significant reduction of costs and/or enhancement of dye removal can be achieved through the combination of different methods in hybrid treatments (Anjaneyulu et al., 2005). Among these hybrid treatments for azo dye removal is the combination of adsorption/bioregeneration which is considered as one of the greener protocols in the treatment of dye containing-wastewater due to the environmentally friendly nature and cost effectiveness of the processes (Aktas and Çeçen, 2007; Al-Amrani et al., 2012b).

Adsorption has been found to be superior to other treatment methods in terms of flexibility and simplicity of design, ease of operation and insensitivity to toxic pollutants. When the active sites available on the adsorbent are exhausted, they can be regenerated through microbial action to increase the service period of the adsorbent. The literature shows that the bioregeneration process effectively increased the life time of the spent activated carbon (AC) loaded with different pollutants such as phenolic compounds (Ng et al., 2009; Oh et al., 2011), surfactants (Klimenko

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et al., 2004), sulfur compounds (Li et al., 2006) and herbicides molinate (Coelho et al., 2006). However, the AC loaded with dyes such as azo dyes could not be regenerated by microbial activity due to poor desorption of the dye from the surface of AC (Walker and Weatherley, 1998; Al-Amrani et al., 2012b). Thus, the search for an alternative cost effective and bio-regenerable adsorbent for azo dyes is warranted.

It has been reported that organo–inorganic adsorbents have shown good adsorption capacity and fast kinetics in the removal of azo dyes from their aqueous solutions (Vrancken et al., 1995; Atia et al., 2009; de Menezes et al., 2012). Nonetheless, reports on the bioregeneration of organo-inorganic adsorbents loaded with azo dyes are extremely limited whereas biodegradation of many environmentally important azo dyes in their aqueous solution have been extensively studied (van der Zee and Villaverde, 2005; Saratale et al., 2011; Solís et al., 2012). More recently, the adsorption/bioregeneration of mono-amine modified silica (MAMS) as one of the organo–inorganic adsorbents loaded with Acid Orange 7 (AO7) has been investigated (Al-Amrani et al., 2012a; Al-Amrani et al., 2012b) and it was found that MAMS was a highly bio-regenerable adsorbent due to the higher reversibility of MAMS adsorption for AO7 and favorable pH_{pzc} value of the MAMS adsorbent.

Therefore, the objectives of the present study are: (i) to evaluate the bioregenerability of MAMS adsorbent loaded with other azo dyes, namely Acid Yellow 9 (AY9) and Acid Red 14 (AR14) which have different molecular structures and (ii) to investigate the effects of the presence/absence of the co-substrate and biomass acclimation concentration on the bioregeneration of azo dye-loaded MAMS.

2. Methods

2.1. Chemicals

The azo dyes of AO7, AY9 and AR14 with ~85%, 95% and 50%, respectively, and sulfanilic acid (SA) with 99% purity were purchased from Aldrich, Germany. The molecular structures of the investigated azo dye are shown in Fig. 1. Geometry optimization of the investigated azo dyes of AO7, AY9 and AR14 molecules was carried out using the semi-empirical Molecular Mechanics Minimizer (MM2) as implemented in software package Chem office 3D ultra 12.0. Silica gel of particle size 40–60 mesh (0.25–0.42 mm) and 3-aminopropyltriethoxysilane (3-APTS) of synthesis grade with 99% purity were purchased from Merck, Germany. All other chemicals were of analytical grade and were used as received.

2.2. Preparation of adsorbents

The commercial silica gel was functionalized using 3-APTS to give mono-amine modified silica (MAMS) particles according to a protocol which has been described by (Donia et al., 2009). The synthesized MAMS particles were dried at 120 °C for 2 h and kept in the desiccator until use. The physical and chemical characteristics of synthesized MAMS adsorbent are presented in Table 1.

Dried azo dye-acclimated biomass was prepared by collecting the biomass from the mixed liquor of a sequencing batch reactor (SBR) at the end of the REACT period as described in Section 2.4. The biomass was washed with distilled water, dried in the oven at 104 °C until constant weight, ground and kept in the desiccators until use.

2.3. Adsorption and desorption studies

For the adsorption isotherm study, samples of MAMS adsorbent, each of 0.1 g, were shaken with the initial investigated dye concentration in the range of 50–500 mg/L at 200 rpm using an orbital shaker (Stuart model SSL1) at pH 2 and 26 ± 1 °C for 3 h. The residual azo dye concentrations of AO7, AY9, and AR14 were measured using an UV–Vis spectrophotometer (JASCO model V-530) at their respective λ_{max}. In addition, the adsorption isotherm of each investigated azo dye on dried biomass was investigated by mixing 0.1 g of the dried biomass with nutrients at different initial dye concentrations in the range 5–100 mg/L at pH 6.62 and the contents were shaken at 200 rpm for 6 days. The equilibrium adsorption data of all azo dyes were fitted to the non-linear Langmuir isotherm model using the Matlab software (version 7.5.0).

In the desorption studies, 0.5 g of fresh MAMS adsorbent was first shaken at 200 rpm with 400 mg/L of AO7 and 300 mg/L of AY9 and AR14, respectively, at pH 2 until the equilibrium was attained. The selected azo dye concentrations were sufficiently high

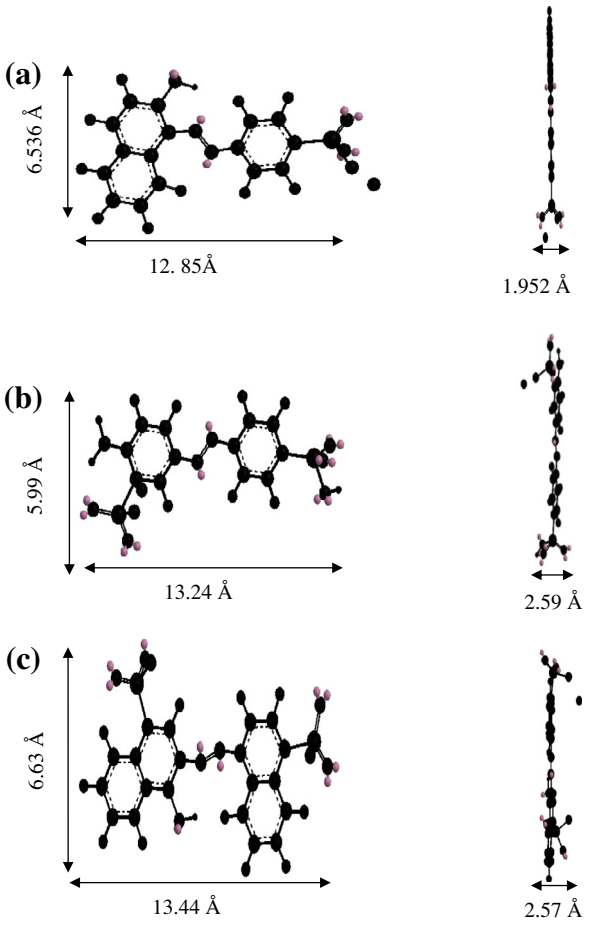

Fig. 1. The molecular structures of (a) Acid Orange 7, (b) Acid Yellow 9 and (c) Acid Red 14.

Table 1
Textural properties and pH_{pzc} of MAMS.

Property	MAMS
BET surface area (m^2/g)	244
Langmuir surface area (m^2/g)	363
t-Plot micropore area (m^2/g)	0
t-Plot external surface area (m^2/g)	251
Pore volume (cm^3/g)	0.57
Pore width (Å)	94
Pore diameter (Å)	70
Particle size (mm)	0.25–0.42
pH_{pzc}	6.81

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