

Utilization of raw and dried Victorian brown coal in the adsorption of model dyes from solution



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ABSTRACT

Being plentiful and readily available in many parts of the world, brown coal is probably one of the most potential substitutes for activated carbon. Many reports include the brown coal drying stage prior to adsorption despite the fact that it may lead to coal shrinkage and reduction of its pore volume, thus diminishing its adsorption capacity. In this study, various types of raw (wet) brown coal were investigated as adsorbents for anionic and cationic dyes in aqueous solution. The adsorption capacities of these raw brown coals were compared with those of oven dried brown coals and also with activated carbon. It was found that raw brown coals exhibit high adsorption capacities for cationic dye, but very low capacities for anionic dyes, suggesting the involvement of a cation-exchange mechanism. Additionally, the cation exchange capacities correlate positively with its cation adsorption capacity highlighting the major role of strongly acidic groups in the adsorption process. Drying the brown coal significantly reduces its adsorption capacity for cationic dye, supporting the hypothesis of coal shrinkage and the consequent reduced pore volume upon drying. These findings suggest that raw (wet) brown coal may perform as well as activated carbon for the removal of positively charged species in aqueous solution. Regeneration of the used adsorbent may also not be necessary, since the used brown coal can be simply burnt as fuel or composted.

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1. Introduction

Activated carbons are generally regarded as the most effective adsorbents used in water treatment processes due to their high surface area, microporous structure and high degree of surface reactivity. Activated carbons have been widely used in purification, decolorisation, deodorisation, dechlorination and separation processes [1]. However, the problems associated with high activation and regeneration costs are disadvantageous. As a result, relatively cheap alternative materials derived from industrial waste, agricultural waste, minerals and coal have been intensively investigated as alternatives for activated carbon [2,3].

Brown coal deposit in many parts of the world are vast and this provides the basis for power generation industries in many places [4]. Unfortunately, the high moisture content of the brown coal (up

to 60%) leads to inefficiencies for this purpose. Hence, alternative applications of brown coal are sought in order to make economic use of a cheap and locally available material [5].

Brown coal has some similarities to activated carbons, namely high porosity and high surface area. Brown coal has been used to prepare commercial activated carbons via energy intensive pyrolysis and activation procedures [6]. The use of raw brown coal itself, as well as sorbents derived from it, for colour and metal removal from aqueous solutions have also been reported [7–12]. It is commonly accepted that high adsorption capacities of brown coal for metal ions and dyes are mainly due to the cation-exchange mechanism of some oxygen containing groups on the coal surface [3,9,12]. However, the effects of moisture content and brown coal pre-treatment prior to adsorption, such as drying, are still poorly understood. When considered as an abundant and cheap material, brown coal optimisation for wastewater treatment processes is an attractive proposition.

The primary objective of this study is to characterise and optimize brown coal as a potential low-cost high-performance industrial adsorbent for aqueous process effluents. A series of Victorian brown coals, adsorbing either an anionic or a cationic dye

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Table 1
The moisture contents, ash yields and ultimate analyses of brown coals.

Adsorbent	Moisture content (%)	Ash Yield	Ultimate Analysis (% db)		
			Carbon	Hydrogen	Nitrogen
LYNA	34	5.1	62.5	4.0	0.5
MV	30	2.0	66.3	4.4	0.5
LY	12	6.2	62.7	4.3	0.67
YL	55	2.2	64	4.7	0.51
AC	6	12.6	83.3	0.5	0.4

were selected as model systems. The performance of brown coal was compared to coal-derived activated carbon. A secondary objective is to understand the mechanism of adsorption and to study the effect of pre-treatment (drying) on the adsorption efficiency.

2. Material and methods

2.1. Materials

Brown coals used in this experiment were Loy Yang high sodium (LYNA), Loy Yang (LY), Yallourn (YL) and Maryvale (MV) brown coals. All brown coal samples were mined from the Latrobe Valley region of Victoria, Australia. The bulk samples were coned and quartered to obtain representative samples, sieved and particles sizes between 150 μm to 212 μm were chosen. Dried brown coals were obtained by drying under nitrogen at 105 °C for 3 h.

For comparison, an activated carbon was also studied. The activated carbon was coal-derived and purchased from Activated Carbon Technology Australia with particle sizes between 150 μm to 212 μm .

The moisture contents, ash yields and ultimate analyses of the adsorbents investigated are supplied in Table 1. The ash yields and the C,H and N analyses were performed by the HRL Technology Pty. Ltd., Mulgrave, Victoria. The ash yields were determined according to the HRL Method No. 1.6 and the C,H and N analyses were determined on the Leco Truspec CHN analyser according to the HRL Method No.1.4. The nitrogen sorption (at 77.15 K) and carbon dioxide sorption (at 273.15 K) measurement was performed using a Micromeritics ASAP 2010 surface area analyser. Cationic and anionic dyes are safranin-O (SO) and alizarin red (AR), respectively. Their structures are given in Fig. 1.

2.2. FTIR analysis

The FTIR analysis was conducted to identify the lignite's functional groups. All lignite samples were dried at 105 °C under N_2 for 5 h to reduce the intensity of OH bands. The KBr pellets were obtained using 1 part of coal and 200 parts of KBr. The FTIR spectra were collected using a Perkin-Elmer Spectrum 100 FT-IR spectrometer. The analyses for all samples were performed at the same time

in order to reduce the possibilities of differences in the ratio against the background of the KBr pellets.

FTIR spectra of the dried brown coals are provided in Fig. 2. The important functional groups, phenols and carboxylic groups, give clear bands in the FTIR spectrum of the raw brown coal. The band at 1710 cm^{-1} is the C=O stretching due to the carbonyl and carboxyl groups. The absorption at 1630 cm^{-1} is dominated by (C...C) stretching of aromatic rings, unsaturated olefinic bonds and poly-aromatic structures, and C=O stretching of quinones bridged to acidic hydroxyl [13].

2.3. Batch adsorption

Known amounts of adsorbent were added to dye solutions in a batch reactor system and shaken for 20 h at 20 °C to attain equilibrium. The aqueous phase was filtered using a 0.45 μm syringe filter and then was analysed for residual concentration of dye using a UNICAM 8625 UV/vis spectrometer. The absorbance measured was then converted to concentration using calibration curves for each dye.

Due to the nature of brown coal, the addition of both raw and oven dried brown coal decreased the solution pH. For SO, the absorption at 516 nm did not change significantly as the pH of dye solutions changed. The absorption difference when the pH was changed from 6.5 to 2.5 is below 3% and is still acceptable. The solution pH after addition of raw and oven dried brown coal ranged from 3.8 to 4.8. For AR, the absorption at 425 nm changed dramatically as the solution pH changed from 3 to 6.5. However, the addition of brown coal did not considerably change the solution pH. The pH of various concentrations of AR solution in distilled water was in the range 3.9–4.5 and, after addition of brown coal, was still in the range 3.8–4.2. When activated carbon was used, the pH of the solution was adjusted to 4 using an aqueous solution of 0.1 M H_2SO_4 .

2.4. Cation exchange capacity analysis

All of brown coal samples used were analysed by the HRL Technology Pty Ltd., Australia for their effective cation exchange capacity (ECEC). The adsorbents were first dried, ground and sieved to less than 212 μm before submitted for the analysis of "Exchangeable bases" according to Method 15D3 on the Australian Handbook of Soil and Water Chemical Methods [14,15]

2.5. Theory and analysis

The adsorption capacity (q_e) represents the amount of dye molecules adsorbed per gram of adsorbent (mg/g) and can be calculated using equation 1.

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

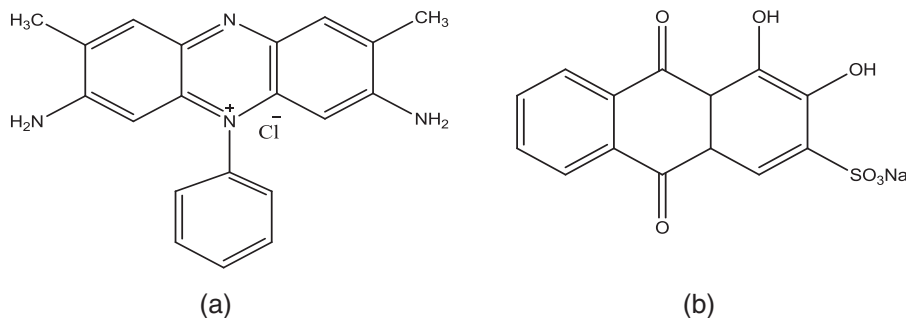


Fig. 1. Chemical structure of model dyes: (a) SO (cationic) (b) AR (anionic).

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