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Electrochemical regeneration of a graphite adsorbent loaded with Acid Violet 17 in a spouted bed reactor

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HIGHLIGHTS

- A two-phase reactor is a promising alternative to traditional three-phase reactor.
- The spouted bed region defined an interesting operating domain.
- Study of current density and the liquid flow rate on the system performance.
- A model is developed for the adsorption and electrochemical regeneration process.

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ABSTRACT

A novel spouted bed reactor is evaluated for water treatment by an adsorption and electrochemical regeneration process. The adsorbent is a bisulphate graphite intercalation compound with low specific surface area but high electrical conductivity, suitable for adsorption of contaminants and simultaneous electrochemical regeneration within a single unit. The effects of current density and liquid flow rate on Acid Violet 17 removal were investigated. The hydrodynamic behaviour of the liquid spouted bed reactor was characterised by a flow regime map. A four-parameter model has been developed to describe the adsorption and electrochemical regeneration process in the liquid spouted bed reactor. It was found that the experimental data of dye removal agrees well with the modelled simulations.

1. Introduction

Adsorption technology is widely used for the removal of organic and inorganic contaminants from water and wastewater. Many absorbents are in use, among which activated carbon is the most widely used for removal organic pollutants. Once the activated carbon has been exhausted, it must either be regenerated, typically by an energy intensive thermal process, or disposed of, which is economically and environmentally unattractive [1].

There are two approaches suggested by many researchers to resolve the problems related to the exhausted activated carbon. The first is to develop low cost natural adsorbents that can be used once, such as using orange peel [2], plum kernels [3] and sunflower seed hull [4], etc. However, this approach only transfers the pollutant from the liquid to solid phase [5]. The second is by regenerating the adsorbent. Among regeneration methods, thermal regeneration is the choice for most industrial applications. This method, however, has high energy consumption (operating temperatures are 800–850 °C), and leads to 5–15% carbon loss due to oxidation and attrition [6]. Therefore, alternative regeneration methods have been investigated by researchers, including microwave [7], ultrasound [8], biological [9], Fenton oxidation [10], wet air oxidation [11] and electrochemical [12–15].

Electrochemical regeneration has been found to be effective for the regeneration of activated carbon, which can achieve regeneration efficiencies of 80–99% [12–15]. However, the adsorption and regeneration process was slow because of the limited rate of intra-particle diffusion. Thus long adsorption and regeneration times are required [5]. For example Zhang [12] reported that

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24 h was required to achieve adsorption equilibrium and 5 h was needed for 85.2% electrochemical regeneration for granular activated carbon.

An alternative approach was investigated by using a non-porous, highly-conducting graphite-based adsorbent material, a flake graphite intercalation compound (GIC) [5]. Because this adsorbent lacks internal surface area, it can significantly reduce the adsorption and regeneration time, but has a low adsorbent capacity. Anodic regeneration leads to oxidation of organic adsorbates on the surface of the GIC. The rapid adsorption and electrochemical regeneration have allowed the design of a treatment process that can adsorb contaminants and electrochemically regenerate adsorbents simultaneously within a single unit [16].

Most previous work exploited air-lift fluidized bed reactors for waste water treatment by adsorption and electrochemical regeneration [16,17]. This is because fluidized beds have certain unique characteristics such as enhanced mass transfer rates, high mixing rates and homogeneous reaction conditions [18]. However, their disadvantages are the possibility of forming a bubbling regime which would lead to non-uniform current distribution and high ohmic drop, i.e. increasing the energy consumption [17]. Mathur and Gishler [19] developed a spouted bed which can effectively deal with coarse particles with the same efficiency as a conventional fluidized bed. Since then, spouted beds have been used extensively in wheat drying, coating, granulation, coal gasification, combustion and wastewater treatment [20].

A novel spouted bed electrochemical reactor (SBER) for water treatment by an adsorption and electrochemical regeneration process is described in this work. Water to be treated is introduced at discrete locations to obtain a regular cyclic motion of particles in the spouted bed, to improve the mixing efficiency between fluid and particles [19,21–23]. The spouted bed has the advantage that parts of the bed are remain as a close packed moving bed, allowing the passage of current through the bed of adsorbent without the problems associated with intermittent contact that arise in a fluidised bed [17]. The main objective of the present study, therefore, is to evaluate the treatment of a model effluent by adsorption and electrochemical regeneration in an SBER under a range of operating conditions, to study the hydrodynamics of the spouted bed, and to develop a reactor model of the SBER.

2. Materials and methods

2.1. Materials

2.1.1. Adsorbent

The adsorbent used in this study was a bisulphate GIC and was supplied by Arvia Technology Ltd under the trade name Nyex™ 1000. This material has been used in several previous studies of adsorption/electrochemical regeneration process [16,24]. The particles of adsorbent have a characteristic flake like appearance (Fig. 1) associated with the graphite precursor. GIC is more hydrophilic than graphite flake, and has surface functional groups [25] that enhance the adsorption performance. The GIC used in this study had a carbon content of about 95% w/w, a density of 2.225 g cm^{-1} with particle diameters of 100–700 μm, and mean particle diameters of around 500 μm [16]. All particles of size less than 140 μm were sieved out to avoid leaving the reactor because of the small particle size. Based on nitrogen adsorption, the value of Brunauer Emmet Teller (BET) surface area was determined to be 1.0 m^2 g^{-1}. This is very low compared with typical activated carbons with surface area in the range 600–2000 m^2 g^{-1} [26]. By mercury porosimetry, it was revealed that essentially no internal pores existed in the material. GIC has a high concentration of free electron carriers at room temperature leading to a relatively high bed electrical conductivity of 0.16 $(\Omega \cdot cm)^{-1}$ compared to around 0.025 $(\Omega \cdot cm)^{-1}$ for GAC [5].

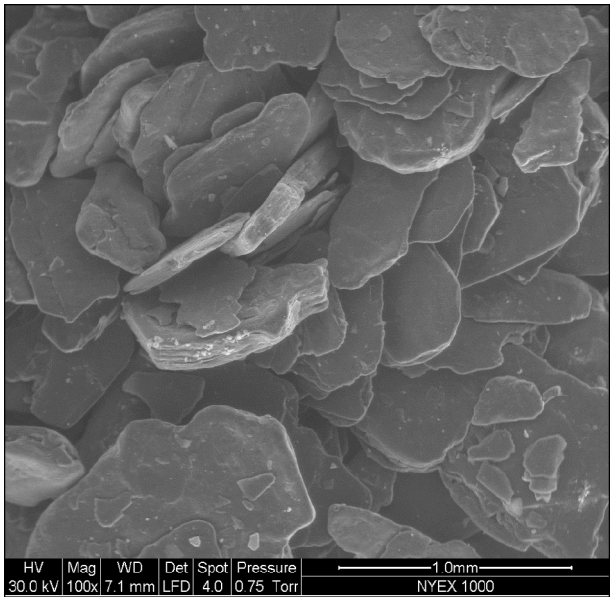

Fig. 1. SEM micrograph of the GIC adsorbent used in this study (Nyex™1000).

2.1.2. Adsorbate

Acid Violet 17 (AV 17) was used as the adsorbate in this study and was supplied by Sigma-Aldrich Company Ltd UK under the trade name Coomassie® Violet R200. It was chosen as the target compound because it has low toxicity and used in previous studies [16]. It is commercial grade and was used in the experiments without further purification. The supplier indicated that the AV 17 content of the Coomassie® Violet R200 was 50%, the remainder is an inorganic salt used in the dye manufacture process. The dye content was confirmed by total organic carbon (TOC) analysis. The AV 17 solutions were prepared using distilled water and mixing for 30 min. The chemical structure and the characteristics of AV 17 are shown in Table 1.

2.2. Experimental set up and procedures

The removal of colour from wastewater and the electrochemical regeneration of the GIC adsorbent were performed in a liquid-lift cell at ambient laboratory temperature of 20 °C and atmospheric pressure. A schematic diagram of the experimental set up is shown in Fig. 2. This reactor operated with simultaneous adsorption and electrochemical regeneration occurring within a single unit. The main body of the Liquid-Lift reactor consisted of two rectangular sheets of transparent polyvinyl chloride (PVC) of 6 mm thickness (see Fig. 3(a)) and the internal dimensions of the process unit were 40 cm tall, 20 cm wide and 2.5 cm deep. Two liquid inlets were provided on either side of the lower sidewalls of the unit. The adsorbent (solid phase) formed a bed at the bottom of the anodic chamber. The liquid to be treated was injected into an inlet chamber below the anodic chamber and distributed through a perforated plate (see Fig. 3(c)), which had four equidistant inlets, each of diameter 1 mm. During operation liquid and solid phases flowed concurrently to the top of the adsorbent bed where they were separated under gravity, with the solid phase circulating back to the base of the reactor and the liquid phase flowing over a weir at the top of the reactor to provide a uniform flow at the outlet. For the range of flowrates used, the 35 cm high chamber above the