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CONTENTS

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Electrochemical *in situ* regeneration of granular activated carbon using a three-dimensional reactor

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Abstract

Electrochemical *in situ* regeneration of granular activated carbon (GAC) saturated with phenol was experimentally investigated using a three-dimensional electrode reactor with titanium filter electrode arrays. The feasibility of the electrochemical regeneration has been assessed by monitoring the regeneration efficiency and chemical oxygen demand (COD). The influence of the applied current, the effluent flow rate, and the effluent path of the electrochemical cell have been systematically studied. Under the optimum conditions, the regeneration efficiency of GAC could reach 94% in 2 hr, and no significant declination was observed after five-time continuous adsorption-regeneration cycles. The adsorption of organic pollutants was almost completely mineralized due to electrochemical oxidation, indicating that this regeneration process is much more potentially cost-effective for application.

Key words: granular activated carbon; electrochemical regeneration; phenol; three-dimensional

Introduction

As an adsorbent with specific surface area, pore structure and surface functional groups, granular activated carbon (GAC) has been widely used for removing organic compounds in water and wastewater (Wang and Balasubramanian, 2009). During the use of GAC, the porosity becomes progressively saturated and inactive. It would not only be uneconomic but also bring environmental pollution if the exhausted GAC is subjected for landfill. Therefore, the regeneration of GAC has gained greater attention, especially carbon saturated with organic pollutants such as phenol (Zhang, 2002; Weng and Hsu, 2008). There are many well-established techniques used to regenerate GAC including extractive regeneration, thermal regeneration, wet air oxidation and chemical regeneration. However, these methods are either too expensive or have low efficiency (Bercic et al., 1998; Leng and Pinto, 1996).

Electrochemical regeneration methods are alternative strategies that have attracted less attention in the past decades. This technique presents some advantages compared to the conventional methods. Essentially, it can be conveniently operated at ambient temperature and pressure, with low energy consumption and with short time requirements. Moreover, it can allow the recovery, modification of organic pollutants into less hazardous compounds or even complete mineralization without additional chemicals (Berenguer et al., 2010; Narbaitz and Karimi-Jashni, 2009; Zhou and Lei, 2006).

In this article electrochemical *in situ* regeneration of GAC saturated with phenol was experimentally investigated using a three-dimensional electrode reactor with titanium filter electrode arrays. The feasibility of the electrochemical regeneration was assessed by monitoring the regeneration efficiency and chemical oxygen demand (COD). The influence of the applied current, the electrolysis time, the electrolyte concentration and the effluent path of the electrochemical cell were also evaluated.

1 Materials and methods

1.1 Materials

The GAC with a specific surface area of 900 m^2/g and an average pore diameter of 2.00 nm was purchased from Tianjin Kermel Chemical Reagent Co., Ltd. (Tianjin, China). GAC samples were washed several times with distilled water and dried in an oven at 105°C for 2 days to a constant weight prior to the experimentation. The phenol wastewater in the experiment was prepared dissolving phenol in distilled water with 1.0 g/L Na_2SO_4.

All other reagents used in the experiment are of analytical grade and used without further purification.

1.2 Apparatus and procedure

The regeneration experiments of GAC were carried out in a three-dimensional reactor that designed in our lab-

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oratory with a net working volume of 750 cm^3 (**Fig. 1**). The cylindrical reactor was constructed from polymethylmethacrylate and titanium filter electrodes with regular polygon staggered, which were cylinders of 0.6 cm internal diameter and 30–50 μm apertures. A known amount of exhausted GAC was fixed in the reactor. The electric power was supplied with regulated DC power supply (WSA-H 100V/30A, Shenzhen, China).

The fresh GAC was saturated with 2.00 g/L phenol solution by counter flow the GAC bed to a constant concentration of the outlet phenol at room temperature. The saturated GAC was regenerated by 1.0 g/L Na_2SO_4 solution counter flow the GAC bed in an electric field. The adsorption-regeneration cycle was repeated.

1.3 Analysis and calculation

The analytical determination of phenol was carried out using UV-spectrophotometer (Unico, model UV-2102PCS) by analyzing the color resulting from reaction of phenol with 4-aminoantipyrine at maximum wavelength 500 nm. The COD value of all solutions and samples were determined according to standard methods (Fochedey and Lierde, 2002). The saturated adsorption values of phenol before and after regeneration were analyzed to calculate the regeneration efficiency. The regeneration efficiency was computed comparatively to the saturated adsorption of fresh GAC under the same equilibrium solution concentration.

2 Results and discussion

2.1 Effect of electric current

Different electric current was applied to investigate the electrochemical regeneration of GAC (**Fig. 2**). It was found that the regeneration efficiency increased with increasing electric current. The maximum regeneration efficiency was from 90% to 98% for the electric current of 1.0 to 3.0 A. The results indicated that the effect of the apparent electric current on the regeneration efficiency was insignificant

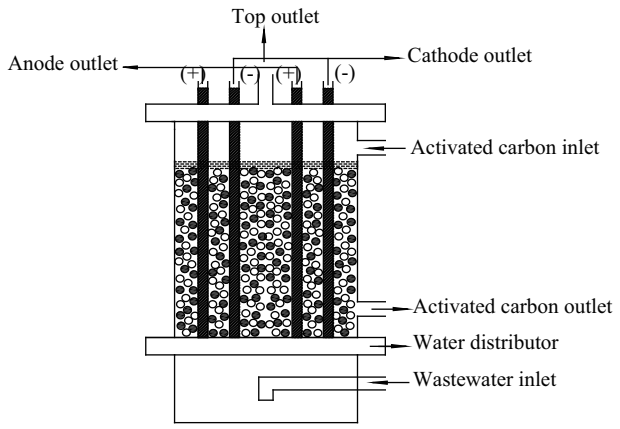

Fig. 1 Schematic diagram of the three-dimensional electrode reactor.

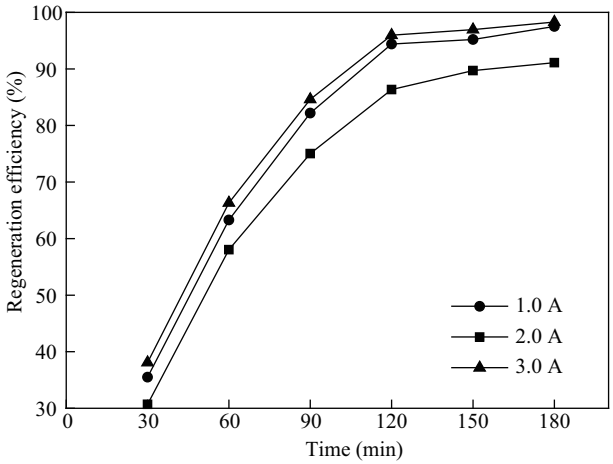

Fig. 2 Effects of electric current on regeneration performance. Experimental conditions: Na_2SO_4 concentration 1 g/L, effluent flow rate of 20 mL/min and top effluent.

when the apparent electric current exceeded 2.0 A. Since the consumption of electric energy increased with the electric current, the best applied current in this system was 2.0 A when the accepted regeneration efficiency and the energy consumption were considered simultaneously.

2.2 Effect of effluent flow rate

In general the Na_2SO_4 and NaCl solutions were chosen as a supporting electrolyte for electrochemical oxidation of organic compounds. But the highly toxic intermediates may be generated in using the latter. Therefore, 1.0 g/L Na_2SO_4 solution was select as the effluent in electrochemical regeneration of GAC. The effect of effluent flow rate on the regeneration process is shown in **Fig. 3**. At the electric current 2.0 A, the maximum values of electrochemical regeneration under the effluent flow rate of 5, 20 and 30 mL/min were about 94%, 97% and 99%, respectively. The higher flow rate caused higher regeneration efficiency, but higher flow rate increase the amount of phenol in the

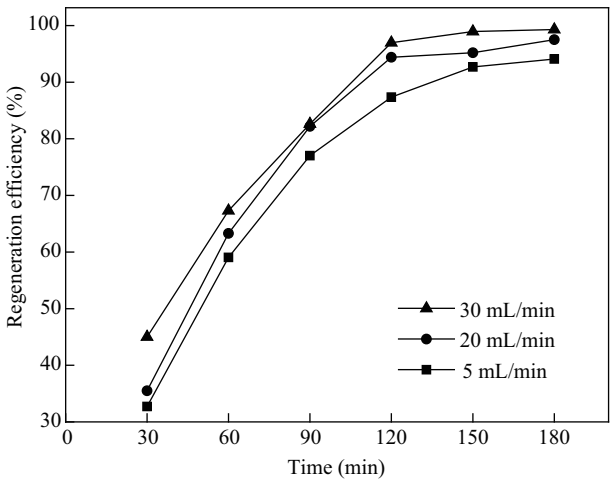

Fig. 3 Effects of flow rate on regeneration performance. Experimental conditions: Na_2SO_4 concentration 1g/L, electric current of 2.0 A and top effluent.

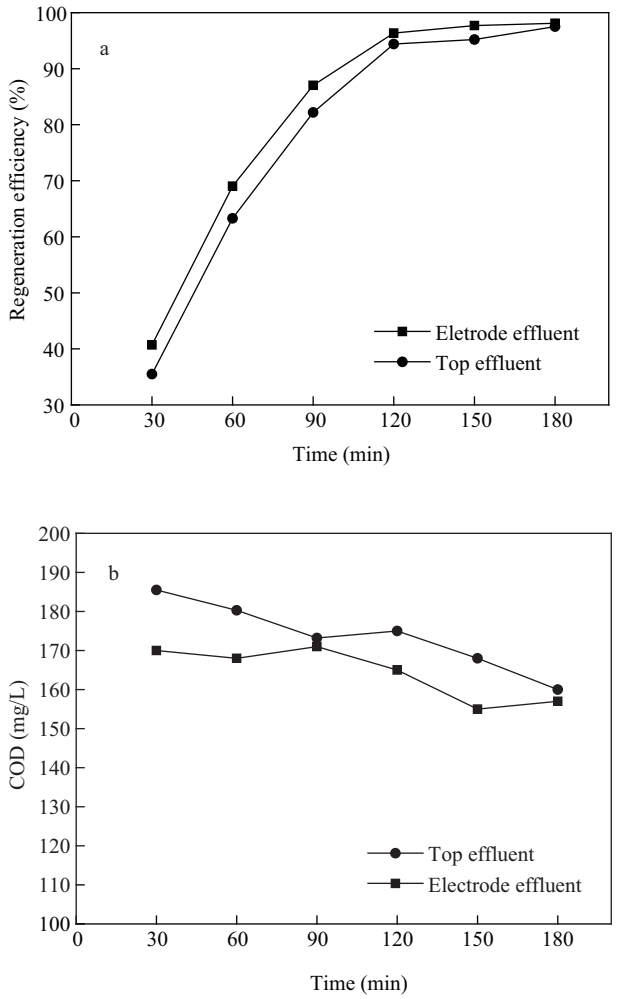

Fig. 4 Effects of effluent path on regeneration efficiency (a) and COD values (b). Experimental conditions: Na_2SO_4 concentration 1 g/L, electric current of 2.0 A and effluent flow rate of 20 mL/min.

effluent because of part of phenol being rushed out of the reactor instead of being oxidized. Thus, the optimal flow rate is 20 mL/ min.

2.3 Comparison of the top and anode effluent

The regeneration efficiency by the top effluent was compared with that by the anode as shown in **Fig. 4a**. The two tests were carried out under the same experimental conditions at Na_2SO_4 concentration 1 g/L, electric current 2.0 A and flow rate 20 mL/min. For the top effluent, the regeneration efficiency was lower than that of the electrode in initial stage of the experiment, while with the time increase, the efficiency was consistency.

But it was found that COD value of the top effluent was lower than that of electrode effluent (**Fig. 4b**), which illuminated the more phenol was mineralized by the electrode effluent.

2.4 Effect of adsorption-regeneration cycles

Under the optimum conditions, the GAC regeneration efficiencies after 1, 2, 3, 4, 5 times adsorption-regeneration cycles were 94.4%, 93.2%, 91.2%, 90.8%, and 91.0%, respectively. No significant declination was observed after five-time continuous adsorption-regeneration cycles.

3 Conclusions

Electrochemical *in situ* regeneration of GAC saturated with phenol was experimentally investigated using a three-dimensional electrode reactor with titanium filter electrode arrays. Under the optimum conditions of electric current 2.0 A, flow rate of the electrolyte 20 mL/min, the regeneration efficiency of GAC could reach 94% in 2 hr, and no significant declination was observed after five-times continuous adsorption-regeneration cycles. The adsorption of organic pollutants was almost completely mineralized due to electrochemical oxidation, indicating this regeneration process is much more potentially cost-effective for application.

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