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Behaviors and kinetics of toluene adsorption — Desorption on activated carbons with varying pore structure

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ABSTRACT

This work was undertaken to investigate the behaviors and kinetics of toluene adsorption and desorption on activated carbons with varying pore structure. Five kinds of activated carbon from different raw materials were selected. Adsorption isotherms and breakthrough curves for toluene were measured. Langmuir and Freundlich equations were fitted to the equilibrium data, and the Freundlich equation was more suitable for simulating toluene adsorption. The process consisted of monolayer, multilayer and partial active site adsorption types. The effect of the pore structure of the activated carbons on toluene adsorption capacity was investigated. The quasi-first-order model was more suitable for describing the process than the quasi-second-order model. The adsorption data was also modeled by the internal particle diffusion model and it was found that the adsorption process could be divided into three stages. In the external surface adsorption process, the rate depended on the specific surface area. During the particle diffusion stage, pore structure and volume were the main factors affecting adsorption rate. In the final equilibrium stage, the rate was determined by the ratio of meso- and macro-pores to total pore volume. The rate over the whole adsorption process was dominated by the toluene concentration. The desorption behavior of toluene on activated carbons was investigated, and the process was divided into heat and mass transfer parts corresponding to emission and diffusion mechanisms, respectively. Physical adsorption played the main role during the adsorption process.

Introduction

Air pollution has increased significantly with the acceleration of industrialization. As one of the main causes of air pollution, volatile organic compounds (VOCs) have attracted wide attention recently (Jayakodi et al., 2013; Elham et al., 2016). The toxicity and irritation caused by VOCs can harm animals, plants and even human life (Karuppiaha et al., 2012; Rino and Vandenbroucke, 2011; Kim et al., 2008). VOC treatments have included recycling and destruction technologies (Zaitan et al., 2016; Peia and Zhang, 2012; Qin et al., 2016). Destruction technologies generally consume a great deal of energy and generate intermediates, resulting in secondary pollution. Recycling technologies, including absorption, adsorption, membrane separation and condensation, have emerged as established technologies (Li et al., 2011; Mo et al., 2009; He

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et al., 2004; Guillaume et al., 2015a). Among them, owing to its stable operation and low energy consumption, adsorption is favored over other technologies (Saeid et al., 2016). Due to its large specific surface area and pore volume, well-developed pore structure and easy regeneration, activated carbon has drawn more and more attention relative to other commercial adsorbents (Wang et al., 2016; Lillo-Rodenas et al., 2005). Many scholars have conducted research on VOC adsorption and desorption on activated carbons in recent years (Lillo-Rodenas et al., 2011; Mangun et al., 2001; Guillaume et al., 2015b). In a study on mixed VOC adsorption on activated carbon, it was found that the boiling point of the adsorbate was the main factor affecting adsorption kinetics (Wang et al., 2015). The desorption behavior of mixed VOCs on activated carbon was studied by Naoto et al. (2016), who found that the desorption rate varied greatly for each VOC, based on its volatility and affinity with activated carbon. A great deal of research has been carried on VOC adsorption on activated carbon with different physicochemical properties (Wang et al., 2012; Son et al., 2016; Francisco et al., 2009). However, the effect of variations in the physicochemical properties of activated carbons on the adsorption and desorption of VOCs has not been systematically investigated. The adsorption and desorption behaviors of activated carbon are related to its properties as well as the properties of adsorbate. For a specific adsorbate, the pore structure of activated carbon is one of the most important factors affecting adsorption. Maroto-Valer et al. (2005) investigated the effect of pore structure on the mercury capacity of a fly ash carbon and an activated sample, and the result showed that mesopores were the major adsorption sites for the adsorbate, and that the pore size also played an important role in the mercury capacity of the adsorbent. The effect of the pore structure of activated carbon on ethanol removal was studied, and it was found that the pores of the activated carbon could accommodate two adsorbed layers of ethanol (Albero et al., 2009). Therefore, the motivation for this work was to study the effect of activated carbons with varying pore structure on VOC adsorption and desorption, which can provide a theoretical basis for the design of industrial processes for their removal.

As a representative of VOCs, toluene was selected as the adsorbate. Activated carbons that were made from different raw materials, including wood, coal and coconut shells, were selected. The samples were characterized by nitrogen adsorption isotherms and Fourier transform infrared spectroscopy (FTIR). Adsorption isotherms and breakthrough curves of toluene were investigated, and the effect of pore structure on adsorption capacity was studied. Adsorption kinetics were analyzed, and toluene desorption from different activated carbons was compared.

1. Experimental

1.1. Samples

Activated carbons, which were made from different raw materials, were supplied by Beijing Sinopharm Group, Jiangsu Sense Charcoal Industry, Modern Coal Factory of Gongyi, Changzhou Charcoal Factory and Beijing Pu Linsen Environmental Protection Technology Companies, respectively (designated as AC-1, AC-2, AC-3, AC-4 and AC-5). The basic parameters, including raw materials, traits, water and ash contents, apparent density and pH, were provided by the suppliers of the activated carbons as shown in Table 1. AC-1 was made from wood chips, had a large specific surface area and was mesoporous. AC-2, AC-3 and AC-4 were coal activated carbons, with developed micropores and slightly advanced mesoporous. The coconut shell activated carbon, AC-5, was a typical microporous adsorbent. All activated carbons were crushed and sieved to 20–40 mesh, then washed several times with deionized water to remove adsorbed impurities and dried at 110°C for 5 hr.

1.2. Methods

1.2.1. Adsorption isotherms of toluene

Adsorption isotherms were measured by the volumetric method, and adsorption capacities were determined using a static adsorption device as shown in Fig. 1. After adding 1 g of activated carbon to the adsorption ball, the device was then placed in a water bath at 30°C. A certain volume of toluene was adsorbed and the pressure was reduced after equilibrium was reached. Toluene was considered to be an ideal gas under the experimental conditions (for which the pressure was less than 1 atm). The adsorption capacity (M) of the activated carbons was calculated by Eq. (1):

$$M = \frac{P_1 V_1}{RT} - \frac{P_2 V_2}{RT} \quad (1)$$

where P_1, V_1 (Pa and m^3) and P_2, V_2 (Pa and m^3) are pressure and volume of toluene before and after adsorption; R and T are 8.314 J/(mol·K) and 303 K, respectively.

1.2.2. Breakthrough and desorption curves of toluene

Fig. 2 shows a schematic diagram of the experimental device, including gas distribution, adsorption-desorption and

Table 1 – Basic parameters of activated carbons.

	AC-1	AC-2	AC-3	AC-4	AC-5
Raw materials	Wood	Coal	Coal	Coal	Coconut shell
Traits	Granular	Columnar	Columnar	Columnar	Atypical
Water (%)	≤10	≤8	≤10	≤5	≤3
Ash (%)	≤3	≤10	≤10	≤15	≤10
Apparent density (g/cm^3)	0.08–0.45	0.49–0.55	0.45–0.65	0.38–0.45	0.45–0.55
pH	5–7	8–10	5–7	6–9	8–10

AC-1, AC-2, AC-3, AC-4 and AC-5 refer to Section 1.1.

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