

Adsorption of acetone and isopropanol on organic acid modified activated carbons



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ABSTRACT

Commercial activated carbon (AC) was modified using formic acid, oxalic acid, and sulfamic acid separately. The effects of the modification on the physicochemical properties of ACs were evaluated using specific surface area and pore distribution, scanning electron microscope, and Fourier transform infrared spectroscopy. After the modification, (i) the BET specific surface area and total pore volume of ACs decreased, (ii) uneven etching trace and white crystal particles were observed on the surface of AC, and (iii) more oxygen-containing functional groups such as O—H, C=O, C—O, and S=O were formed on the surface. The effects of the modifications on the adsorption behavior of acetone and isopropanol on ACs at different inlet concentrations were studied from adsorption equilibrium, kinetics, and energy point of view. The results show that the equilibrium adsorption capacity has been greatly improved by the modification of organic acids. The adsorption isotherms of acetone and isopropanol on ACs are well fitted by both Langmuir and Freundlich equations. Characteristic adsorption energy values of acetone and isopropanol slightly increase with increasing of oxygen functional groups on the ACs surface. The adsorption kinetics of acetone and isopropanol on ACs are best described by Bangham kinetics model.

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1. Introduction

Environmental regulations have been legislated to reduce the emission of volatile organic compounds (VOCs) both in gas and liquid streams. Among the different VOCs, acetone and isopropanol are the most common air pollutants emitted from the chemical, petrochemical, and similar industries; they are highly volatile at room temperature and cause acute or chronic poisoning to humans by prolonged contact.

Several techniques for VOCs have been investigated such as adsorption, catalytic combustion, membrane-assisted stripping and bio-filtration, etc. [1–3]. Among the different technologies, bio-filtration and membrane-assisted stripping are widely applied in polluted air and water treatment. Bio-filtration is based on the capture and biodegradation of VOCs into non-toxic compound using micro-organisms immobilized on the surface of a packing bed. Membrane-assisted stripping is a sophisticated technique that VOCs are removed from the polluted solutions by diffusion across the membrane [4,5]. However, the bio-systems exhibit important drawbacks such as poor adaptability to changes in gas composition and lack of accurate control of the PH, etc. The Membrane system

has the disadvantages of membrane pollution, short life and concentration polarization, etc.

Adsorption has been proposed as an excellent alternative for the removal of VOCs avoiding the aforementioned drawbacks. The advantages of the adsorption process are simplicity in operation, inexpensive compared to other separation methods and insensitivity to toxic substances. Due to the developed high surface area, porous structure, and tuneable chemical properties [6–10], activated carbon (AC) has been widely used in the purification of organic gas treatments. Previous research has already investigated adsorption of benzene, toluene, phenol, 1,2-dichloroethane, trichloroethylene, chloroform, acetone, ethanol, et al.

Surface chemistry and pore structure significantly affect the adsorption properties of AC [11–13]. Both of them can be controlled by activation condition and precursor in prepared process, and modified by using various methods including chemical oxidation, impregnation with metal particle, microwave heating, plasma and so on [14,15]. Among these modified techniques, chemical oxidation is the most common method, and it presents the advantages of simple step, low cost, energy saving and introducing more acidic oxygen groups on the AC surface. Most studies have generally applied inorganic acid (HNO₃, H₂SO₄, H₃PO₄ and HCl) and hydrogen peroxide to modify AC surface. However, few studies have been focused on the modification of AC with organic acids,

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and very little information exists about the use of adsorption energy to analyze the gas adsorption performance on AC.

For these reasons, this work investigated the feasibility of acetone and isopropanol adsorption by organic acids modification of commercial AC. Surface physicochemical properties were characterized using low-temperature N_2 adsorption, acid–base titration, SEM, and FTIR spectroscopy. Adsorption measurements of acetone and isopropanol on the modified ACs were examined and the effects of the porous properties and surface functional groups of the modified ACs on the adsorption characteristics were discussed by adsorption isotherm, adsorption kinetics and adsorption energy. This study developed a modification method for effectively removing hazardous acetone and isopropanol, and provided a good research idea.

2. Experimental

2.1. Materials and methods

Granular AC (250 g) was boiled in water for 30 min to remove the dirt and dust. Next, it was further washed with deionized water several times till the final pH was 6.8–7.2 and then dried in a vacuum oven at 383 K for 12 h. This sample was designated as AC-A.

AC-A (75 g) was impregnated with an oxalate dihydrate solution (0.5 M) and a sulfamic acid solution (0.5 M) separately. Then, the mixtures were stirred at room temperature for 1 h, washed with deionized water several times, and dried to a constant weight. After this treatment, the samples were referred to as AC-OA and AC-SA, respectively. Another sample of the initial AC was impregnated with pure formic acid. The preparation procedure was basically the same as for the oxalate dihydrate-modified AC. This sample was named as AC-FA. To avoid moisture, the ACs were stored in sealed plastic bottles inside desiccators till further use.

2.2. Methods

2.2.1. Characterization of adsorbents

The textural properties (specific surface area and pore volume) of the adsorbents were evaluated by N_2 adsorption isotherms (77 K) obtained using a BECKMAN COULTER SA3100 apparatus. The apparent BET surface area was calculated using the BET equation. The micropore volume was obtained by the t -Plot method. The pore-size distribution curves were calculated from the adsorption branch using the Barrett–Joyner–Halenda (BJH) method. The total pore volume, V_{total} , was assessed from the adsorbed amount at a

relative pressure of 0.9814 by converting it to the corresponding volume of liquid nitrogen. The texture and chemical elements present on the surface of ACs was observed using a scanning electron microscope (SEM, QUANTA 200, FEI Instrument Co., NED). The chemical structure of the ACs was qualitatively evaluated using an FTIR spectrometer (NEXUS670, Nicolet, USA). The IR spectrum was obtained in the frequency range $400\text{--}4000\text{ cm}^{-1}$. Three samples were taken for each adsorbent, and characteristic analysis of each sample was duplicated.

2.2.2. Experimental

Fig. 1 shows the experimental apparatus for measuring the amount of acetone and isopropanol adsorbed. The adsorption tests were carried out in a quartz glass column with 1.1 cm inner diameter. Both the ends of the column were packed with glass fiber filter. The ACs (~2 g, average particle size: 0.5 mm) were packed on the glass fiber filter inside the column. The adsorption temperature was maintained at 283 K using a constant-temperature vessel. The amount of flowing gas was regulated using mass-flow controllers: one for pure nitrogen and one for the mixture of acetone vapor and nitrogen. The gas concentration of the outlet stream was monitored continuously by gas chromatography (GC) to obtain the breakthrough curves. The breakthrough profile was measured by monitoring the outlet concentration as a function of time. When the outlet concentration reached 5% of the initial concentration, the breakthrough time was determined. The saturation adsorption time was measured once the outlet concentration was equal to 95% of the inlet concentration [16]. The adsorption isotherms of acetone and isopropanol were determined using a gravimetric apparatus. The adsorption runs were repeated three times for all experimental concentrations.

3. Results and discussion

3.1. Effects of modification on textural properties

Table 1 shows the microstructural properties including the surface areas, micropore volume, total pore volume, and macropore volume of the ACs. The surface areas and micropore volume of AC-A are higher than those of other modified ACs. The decrease in the surface area can be attributed to the blocking of some of the pores by oxide functional groups introduced by the chemical modification and the erosion of carbon skeletal structure [17,18]. The surface area and total pore volume decrease in the order AC-FA > AC-OA > AC-SA, indicating that the acidity of organic acids increase the surface area and total pore volume. Moreover, the

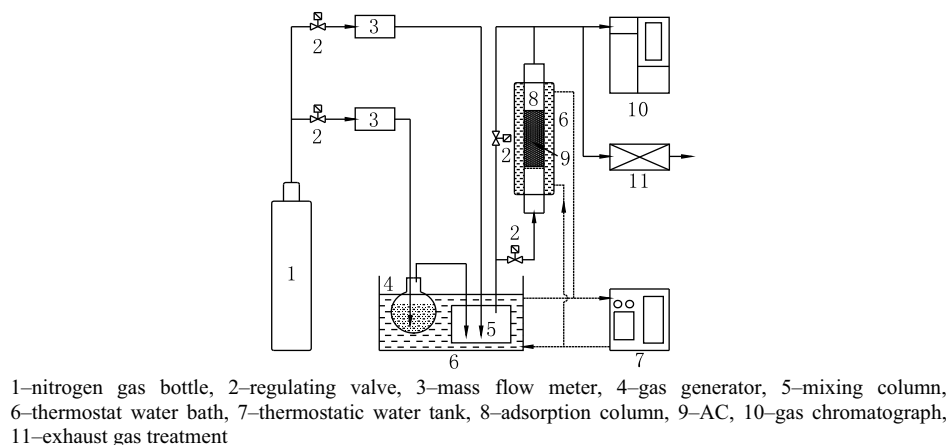


Fig. 1. Schematic diagram of experimental apparatus for adsorption of acetone/isopropanol.

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