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Study on consolidated activated carbon: Choice of optimal adsorbent for refrigeration application

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ABSTRACT

For refrigeration condition in order to choose an optimal consolidated composite activated carbon (AC), the thermal conductivity, permeability, equilibrium adsorption performance, and non-equilibrium adsorption performance were studied. Six samples of three types of adsorbents (consolidated AC with expanded natural graphite treated with sulfuric acid (ENG-TSA), consolidated AC with expanded natural graphite (ENG) and granular AC) with different density and different grain size were produced and compared. Experiments showed that the thermal conductivity and permeability of consolidated AC with ENG-TSA are between 6.75 to 7.45 W/m K and 2.00×10^{-12} to 1.00×10^{-11} m^2, respectively, when the density is ranged between 350 kg/m^3 and 450 kg/m^3. The study on the equilibrium and non-equilibrium adsorption performance indicated that the addition of ENG-TSA helps to improve the concentration swing. The cycle time of consolidated AC with ENG-TSA decreases significantly compared with that of consolidated AC with ENG and granular AC. For the optimal results from the matrix of ENG-TSA although the bulk density of AC decreases, the volume cooling power is 30% improved in comparison with the results of granular AC because the cycle time decreases significantly.

1. Introduction

Adsorption refrigeration has been proved to be an energy saving and environmental benign refrigeration technology for the reason of being powered by low grade thermal energy and the application of green refrigerants [1]. Activated carbon (AC) has been widely utilized as adsorbent for its high mass transfer performance, stable adsorption characteristics, and no problems of corrosion with metal containers. The disadvantage of the AC is the low adsorption quantity. The highest adsorption quantity of AC is only up to 0.4–0.5 kg refrigerant/kg AC and normally the concentration swing is only less than 0.2 with ammonia or methanol as refrigerants [2]. Under such a condition a short cycle time is required for obtaining the high specific cooling power (cooling power per unit mass of adsorbent) because of the low cycle adsorption quantity. But for granular AC the heat transfer performance generally is critical and consequently the cycle time is always quite long. Thus the major technical challenge for the performance improvement is to achieve the high thermal conductivity within the bed while the mass transfer performance should not be deteriorated.

Thermal conductivity enhancement of the adsorbent is one effective way of improving the heat transfer in adsorption systems and thereby speeding up the process of adsorption/desorption [3]. Consolidated and composite adsorbents, which have higher thermal conductivity, have been studied by various researchers. For example Tamainot-Telto and Critoph [4] investigated AC mixed with a polymeric binder to produce monolith of a desired shape with the thermal conductivity up to 0.44 W/m K. Cacciola et al. [5] used polytetrauoroethylene (PTFE) to prepare carbon bricks leading to thermal conductivity ranged between 0.13 and 0.20 W/m K. Wang et al. [6] used solidified AC with mass flow channels to reduce mass transfer resistance in adsorbent beds. The composite consolidated adsorbent of AC and expanded natural graphite (ENG) could improve heat and mass transfer performance obviously [7–11]. Bonnissel et al. [7] manufactured a kind of compacted natural exfoliated graphite and the perpendicular thermal conductivity reached the highest value of 350 W/m K at 1350 kg/m^3. Biloe et al. [8] utilized consolidated AC and expanded graphite composite for natural gas storage leading to a relative high thermal conductivity. The addition of graphite reduced the charge time of methane by a factor of 10. Wang et al. [11] measured the thermal conductivity, permeability of consolidated AC and expanded natural graphite composites and the highest thermal conductivity of 2.47 W/m K was obtained.

Recently, we developed a new type composite consolidated AC with a host matrix of expanded natural graphite treated with sulfuric acid (ENG-TSA). The maximum thermal conductivity of consolidated ENG-TSA is 337 W/m K and the highest thermal conductivity of composite consolidated AC is 34.2 W/m K [12–14].

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Based on our previous studies on the consolidated composite AC with the matrix of ENG-TSA, the AC that was manufactured by the coconut shell is studied. The granular AC, consolidated AC with ENG as the matrix, and the consolidated AC with ENG-TSA as the matrix were compared, and the optimal adsorbent is chosen for the refrigeration by the comparison of thermal conductivity, permeability, equilibrium adsorption performance, and non-equilibrium adsorption performance.

2. Preparation for the experiments

2.1. Development of the adsorbent samples

The ENG-TSA is manufactured by Mersen in France which is detailed described in our previous work [12]. The composition of this material is shown in Table 1. The carbon content in the material is greater than 99.8%, and the ash content is less than 0.2%. The ENG is expanded by heating untreated natural graphite in an oven at the temperature of 600 °C for 10 min. AC used in the samples is YKAC with a large grain size (20–50 mesh) and with a small grain size (80–200 mesh) manufactured in Shanghai, China. The composition of YKAC is shown in Table 2. The carbon content in the material is greater than 97.5%.

The manufacturing process of consolidated composite AC is shown in Fig. 1. Firstly the granular ENG-TSA or ENG and granular AC are dried in the oven at the temperature of 150 °C for 2 h to eliminate the mass error caused by moisture and impure gas. After that the AC is mixed with water by the mass ratio of 1:1 to make AC moist, which helps to mix with the granular ENG-TSA uniformly. The composite of AC, ENG-TSA or ENG, and water is then compressed into blocks or into the test units. Finally the block samples (1.4–2 g) for thermal conductivity test are dried in the oven at the temperature of 150 °C for 4–5 h; the samples for the permeability test (191–246 g) are dried in the oven at the temperature of 150 °C for 12 h; the samples in the adsorber (200–260 g) are dried in the oven at the temperature of 150 °C for 24 h to make sure that no moisture or non-condensable gas flows into the ammonia system.

2.2. Choice of the adsorbent samples

AC and ammonia is a reasonable working pair under the condition of refrigerating temperature lower than 0 °C. For such a situation, the permeability will be more significant because the saturated pressure and density of the refrigerant are low, which will influence the mass transfer performance. To maintain high volume cooling density and reduce the sensible heat of adsorbent as well as to consider the thermal conductivity and permeability, our previous experiments [14] showed that the sample with AC percentage of 67% is an optimal choice. For adsorption refrigeration the density cannot be too low because the volume refrigeration density will be influenced by it. The density also cannot be too high considering of the limitation of compressing process and mass transfer performance. Considered about the thermal conductivity, permeability, as well as the density, our previous experiments [14] showed that the optimal density should be around 400 kg/m^3. In this experiment the density ranged of 350–450 kg/m^3 was chosen.

Table 2
The composition of AC.

Item	Index
Carbon content (%)	>97.5
Ash content (%)	<2.5
Iodine sorption value (mg/g)	>1100
Methylene blue sorption value (mg/g)	>180
Resistance to temperature in oxidizing atmosphere (°C)	380
Density (kg/m^3)	450–600
BET surface area (m^2/g)	900–1100
BJH pore volume (cm^3/g)	0.8–1

Six samples with different addition, different AC ratio and different density were made and their parameters of density are shown in Table 3. The unit of mesh is standard US mesh size with 100 mesh equal to 0.152 mm sieve size. The bulk density of flake ENG-TSA and flake ENG are only about 6 kg/m^3 and 22 kg/m^3 respectively, while the bulk density of granular AC of 80–200 mesh and 20–50 mesh are about 518 kg/m^3 and 577 kg/m^3 respectively. The bulk density of AC, ENG-TSA or ENG is calculated by dividing mass of AC, ENG-TSA or ENG in composite adsorbents with total volume of composite adsorbents.

Samples are prepared according to the set apparent densities. The material is pressed into a container with fixed volume. The mass of material is calculated by multiplying the volume and the apparent density. During the calculation of the density of composite samples and ENG-TSA, the error was mainly caused by the balance for measuring the mass and the micrometer for measuring the size of samples. The absolute error of the balance is better than 0.01 g, the error of micrometer is better than 0.1 mm. Correspondingly the total relative error of the values of density is within 1%.

The surface area and pore parameters of different samples were tested by an Accelerated Surface Area and Porosimetry System (ASAP 2020) and the result is shown in Table 4. The system utilizes the principle of static volumetric technique to obtain adsorption and desorption isotherms and applies multiple gas sorption laws to get the information of surface area and pore parameters of a solid material. The surface area and pore volume of AC are shown in Table 2. AC has a much higher surface area compared to that of zeolite or silica gel [15]. But with the addition of ENG-TSA or ENG, the surface area and pore volume of composite materials decrease to some extent.

3. Experiments on the thermal conductivity and permeability

3.1. Experimental methods

The thermal conductivity is measured by the NETZSCH LFA 447 NanoFlash using the laser flash measuring method [16] as shown in Fig. 2. The unit includes infrared detector, sample changer, heater, optical filter, reflector and flash lamp et al.

In the experiments, under a certain set temperature T (controlled by a furnace heated condition), a beam of light pulses is

Table 1
The composition of ENG-TSA.

Carbon (%)	Content	Ash content (%)								Resistance to temperature in oxidizing atmosphere (°C)	Density (kg/m^3)
>99.8		<0.2								500	5–6
		Average ash content (ppm)									
		Fe	Si	Mg	Al	Ca	Cr	Mn	Cu		
		150	230	60	20	10	10	10	10		