

13X-APG granular zeolite in a fixed bed adsorber of CO₂ adsorption characteristicsXiaojie Wu¹, Jingwei Xu², Jianling Tao²

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Abstract: 13X-APG granular zeolite as adsorbent, by measuring its fixed bed adsorber in the adsorption breakthrough curve, the adsorbent studied the dynamics of CO₂ adsorption characteristics were investigated gas temperature, gas flow rate, adsorbent particles the size of the adsorbent properties of CO₂ obtained adsorbent bed adsorption process temperature changes. The results show that with the increase of gas temperature and increase of the adsorbent particle size, adsorbent adsorption of CO₂ decreases; adsorption with little change in flow rate of gas; in the adsorption process, the adsorption of small diameter agent bed temperature changes significantly.

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1. Introduction

Fossil energy extraction and use of a large number of further exacerbate global warming, so CO₂ emissions are increasingly attracted worldwide attention [1]. China's energy mainly relies on coal, and coal CO₂ emissions from large to control CO₂ emissions is the implementation of the sustainable development strategy of urgent problems, and CO₂ capture and storage technology (CCS) is considered to be one of the effective solutions [2]. Of CO₂ capture and separation methods include chemical absorption, adsorption, membrane separation and cryogenic separation method, etc., of which the first two are the most important method for trapping. Compared with the traditional chemical absorption, adsorption CO₂ separation technology with less investment, easy operation, low energy consumption [3], in the field of CO₂ gas separation has broad application prospects. In order to improve sorbent CO₂ capture and separation performance of the scholars of these adsorbents decarbonization conducted extensive research. Li et al [4] conducted a zeolite 13X vacuum adsorption of CO₂ in the flue gas moisture experimental study showed that the presence of water reduces the CO₂ recovery and purity. Cavenati etc. [5] studied the high pressure zeolite 13X for CO₂, N₂ and CH₄ mixed gas adsorption properties of zeolite experimental results show that the largest amount of CO₂ adsorption, while the N₂ adsorption capacity is very small. Siriwardane etc. [6 at 120 °C, experimental study of four zeolite (4A, 5A, 13X, APG-II) Adsorption of CO₂, and through the calorimeter measuring the heat of adsorption. However, with the conventional use of zeolite 13X adsorbent compared to zeolite 13X-APG as a molecular sieve for air separation field has better adsorption and separation performance. Liang Hui and other port in the zeolite 13X-APG adsorbent, developed a vacuum coupling

temperature swing adsorption process flue gas adsorption of CO₂, effectively reducing the trapping heat energy; Konduru etc. [8 used as adsorbent zeolite 13X-APG the CO₂ content of 1.5 N₂: CO₂ gas mixture and temperature swing adsorption experiments carried out, after five cycles of CO₂ gas adsorption average recovery of 84%. ASU special screening of the CO₂ adsorption in terms of performance remains to be further studied.

In this paper, zeolite 13X-APG adsorbent, using a fixed bed adsorption experimental apparatus systems measure CO₂: adsorption breakthrough curves of CO₂ dynamic adsorption properties, and examine the adsorption gas temperature and flow, the particle size of the adsorbent, and the adsorption process adsorption bed temperature changes.

2. Materials and equipment

Adsorbent zeolite 13X-APG (Shanghai zeolite Ltd.), which is an alkali metal aluminosilicate, sodium X-type crystal structure, the formula Na [(AlO)(SiO₂)]-xH₂O, spherical particles. Before adsorption experiments, the first zeolite in a vacuum oven drying conditions in 200 °C activation 12h, then use the BET specific surface area analyzer (MicrometricASAP2020) to characterize the zeolite 13X-APG, the results shown in Table 1.

Table 1 Zeolite 13X-APG parameters

Sample	Specific surface area	Hole volume	Hole Diameter
Zeolite 13X-APG	466.88	0.27	2.29

CO₂ fixed bed adsorption experimental apparatus shown in Figure 1, the experimental system includes a fixed bed adsorber, a gas supply system, temperature control and measurement system. Fixed adsorption bed

to an inner diameter 430mm, long stainless steel cylinder 280mlTI bed filled with spherical particles of zeolite 13X-APG, filling height of 50mm; stable and uniform distribution of air into the adsorption bed in the adsorption bed filled with 6mm diameter at each end inert particulate glass beads 105ITIm, and a thickness 2mitt sieve adsorbents and glass beads are separated, in addition, in order to investigate the adsorption heat effect, the adsorption bed covered with a heat insulating material. Gas supply system by a CO₂ cylinder and N cylinders, valves, mass flow controllers (LF400, Chengdu Lai Feng Technology Co., Ltd.), mixing tank, back pressure valve and the mass flowmeter components. Temperature control and measurement system consists of wound electric heating mixing tank outside the tropics, temperature controller, and installed in the bed of the K-type sheathed thermocouples.

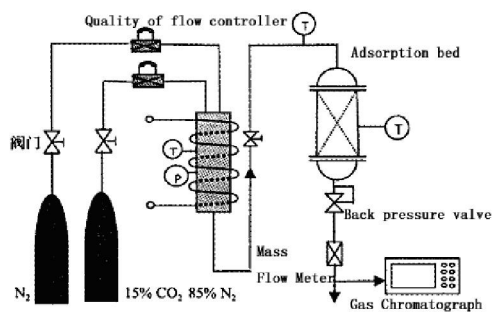


Fig 1 Adsorption experiment system

Walter experiment period prior to the N- tightness testing device, until the inspection is completed the experiment. During the experiment, the volume fraction of CO₂ and 85% N₂. The feed gas in the mixing tank is heated to the desired temperature, and then into the adsorption bed. Before adsorption bed is also preheated to the same temperature as the mixture, and the outsourcing of insulating material as in the adsorption / desorption process, the adsorption bed can be seen as insulation; adsorption bed temperature adsorption beds installed in the middle of the K- type sheathed thermocouple measurements. Adsorption bed adsorption pressure is controlled by a back pressure valve, the outlet gas flow from the LF-ID mass flowmeter to measure the concentration of CO₂ adsorption bed exports by gas chromatography (SC-2000 type, Chongqing Sichuan Instrument nine plants) were detected.

Operating conditions set benchmark experiments: gas inlet pressure 0.1MPa, the gas volume flow 1000mL/min, gas inlet temperature 21 °C, the zeolite particle size 3mm., Sorbent mass 26.0g. Adjusted separately mixed gas flow and temperature, and the use of different size adsorbents to study its effect on CO₂: adsorption performance. CO₂ adsorption using

adsorption breakthrough curves and adsorption capacity to characterize.

3. Experimental results and analysis

3.1 inlet gas temperature on the adsorption properties of

Figure 2 shows the temperature of the mixture of zeolite 13X-APG adsorption breakthrough curve of CO. From Figure 2, the initial adsorption, CO₂ is completely within the bed in the zeolite adsorption exit concentration is zero. Increase over time, gradually reaching saturation adsorption bed segment, continued down the mass transfer zone, outlet concentration increases gradually, gradually penetrating gentle curve, indicating that the mass transfer zone reach adsorption bed tail end of adsorption. With the gas temperature, CO₂ absorption through the point (adsorbate from the effluent occurs, the breakthrough curve corresponding points) ahead of the penetration time from the time of 21 °C 40 °C 4min reduced to the 1rain. Thus, when the adsorption time is the same, high inlet gas temperatures outlet CO₂ concentration, the faster the mass transfer zone reaches the end of the adsorption bed.

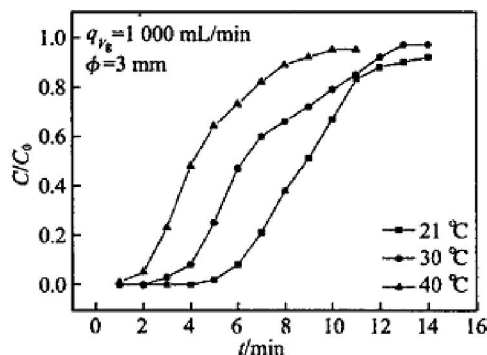


Fig 2 Influences of gas mixture on CO₂ adsorption

Figure 3 illustrates the inlet gas temperature of the adsorption curves it can be seen from Figure 3, the adsorption capacity increases gradually increases with time, but the adsorption rate (absorption rate of change with time) decreases, eventually reach equilibrium; with mixture temperature increases, the absorption rate is reduced, per unit mass of adsorbent in the packed bed adsorption equilibrium adsorption capacity was significantly reduced when from 21 °C when 51.24mg / g down to 40 °C when 25.24mg / g. This shows that the adsorption of zeolite 13X-APG CO₂ is the typical physical adsorption process, the temperature increases, the molecular thermal motion speed, sufficient to overcome the van der Waals force and leave the suction surface, thus reducing the system temperature is conducive for adsorption.

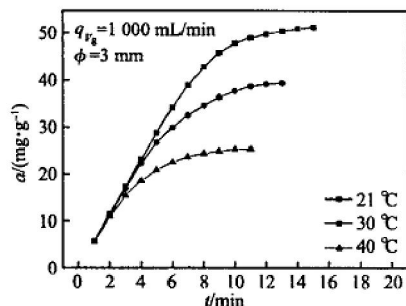


Fig 3 Influences of gas mixture temperature on adsorption capacity

Figure 4 is a different inlet gas temperature conditions, the center bed temperature versus time curve. Figure 4 shows that in the adsorption process, with the time increases, the first adsorption bed center temperature gradually increased and decreased slowly. This is mainly due to the initial adsorption, zeolite CO₂: adsorption is stronger, more heat generated by the adsorption, the adsorption bed temperature rise; but as for adsorption, the adsorption rate is reduced, the adsorption heat released per unit time decreases, and this the gas temperature is lower than the temperature of the adsorbent bed, the bed temperature is gradually decreased so. Furthermore, with the increase of gas temperature, the bed temperature rise decreases the center, the temperature rise at 21,30,40 °C volume reached 12.6,8.2,4.7 °C. This is due to the temperature rise of the zeolite adsorption capacity of CO₂ decreased, resulting in the release of adsorption heat is reduced, so the bed temperature amplitude decreases. From the above results, the adsorption wave of advancing the process, the gas temperature increases to a certain extent, the temperature is not conducive for adsorption. Therefore, strengthening the adsorption process heat can effectively improve CO₂: adsorption capacity.

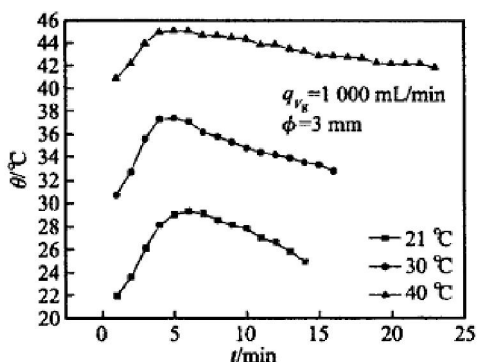


Fig 4 Influences of gas mixture temperature on center temperature of adsorption bed

3.2 Gas flow rate on the adsorption properties of

Figure 5 is a mixed gas flow changes on the

13X-APG zeolite CO₂: influence of adsorption breakthrough curve. Figure 5 shows that, with the increase of gas flow, at the same time, C₀. Gas outlet concentration, but in the early adsorption, gas flow rate of 1200mL/min the CO₂ concentration is less than 1000mL/min when export outlet concentration, mainly due to the gas flow rate was 1200mL/min, the particle external convection-diffusion enhancement, enhanced CO₂: molecules to the surface of the particles to pass, but with the adsorption of conduct, CO₂ concentration gradually increased exports and greater than the flow rate of 1000ml/min the outlet concentration.

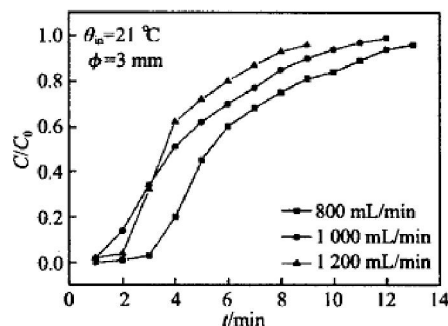


Fig 5 influences of gas mixture flow rate on CO₂ adsorption breakthrough curves

Figure 6 is different from the adsorption amount of the gas flow curve can be seen from Figure 6, when the gas flow rate is from 800mL/min to 1000mL/min, CO₂ adsorption rate and the packed bed adsorption equilibrium adsorption amount changes little when, but when the gas flow rate further increased 1200mL/min, CO₂ adsorption rate increases, the adsorption amount per unit time is large, this is mainly due to particle diffusion of the gas outside convection enhancement. However, with time, the adsorption rate decreases, equilibrium, the type of gas flow rate changes very little adsorption of CO. This is mainly because the gas in the adsorption process, through two external diffusion processes and internal diffusion, although the increase in the gas flow to some extent to enhance the external diffusion, but the zeolite L with an average pore diameter of 2.2nm, is filled with micro-rich holes in the internal diffusion control, limiting the transmission of gas. Thus, changes in gas flow adsorption affected.

Figure 7 shows that, when the gas flow rate increased from 800mL/min 1000mL/min, the increase rate of temperature rise of the adsorbent bed is mainly due to the adsorption does not reach saturation until the gas flow rate increases, C₀. The load increases, the adsorption amount per unit time increases, the adsorption heat released per unit time is greater, resulting in temperature rise rate increases; 1200mL/min further increased when the gas flow rate,

the temperature rise rate is not improved, primarily As traffic increases, so that the adsorption of gas per unit time away more calories, increased flow causes an increase in the adsorption heat and gases away the heat of adsorption equilibrium. Temperature increase from the point of view, 800mL/min temperature increase when the flow rate is less than the temperature increase 1000mL/min, mainly due to low gas flow, the adsorption equilibrium time is longer, so although the gas per unit time taken away Smaller heat 1000mL/min, but the overall balance of time away more heat. The gas flow rate was 1200mL/min, although the adsorption reached equilibrium time shortened, but the heat away the gas per unit time significantly increased, resulting in the condition of the adsorption bed temperature amplitude decreased. Shows that the adsorbent bed temperature rise jointly determined by two factors: First, the gas flow caused by different adsorption heat release changes; two different gas flow is led to changes in gas heat away.

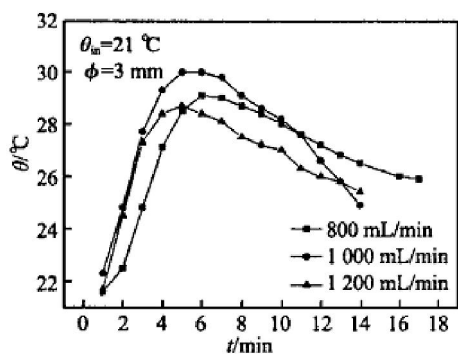


Fig 6 influences of gas mixture flow rate on center temperature of adsorption bed

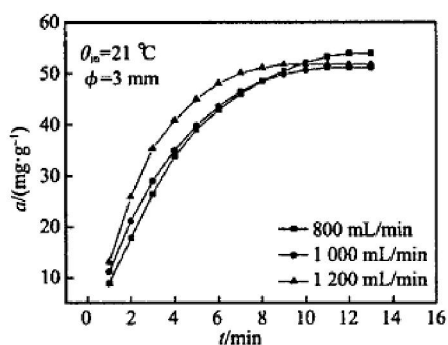


Figure 7 is a mixed gas flow rates of different bed temperature curve center.

3.3 sorbent particle size on the adsorption properties of

Figure 8 shows the particle size of the adsorbent for CO₂: the impact of the adsorption breakthrough curve. Figure 8 shows, 2mm diameter adsorption equilibrium time required is longer, the particle size is

much greater than the equilibrium q53mm time, and 3mm diameter and 5mm diameter required for the adsorption equilibrium time is almost the same, and the gas outlet concentration C₀ when the particle size is much larger than the rate of rise rate of rise of 2mm. This is mainly due to the adsorption equilibrium reached except by the particle diffusion mass transfer process control, the packed bed particles within the flow channels between the state, convection and mass transfer characteristics of the adsorption equilibrium has important implications. For the same quality of small particle size (2mm) packed bed of zeolite formed within the flowing gas in contact with the macroscopic particle surface area is greater than the case when a large particle size, and the accumulation of particles within the bed larvae L tract gas convection-diffusion mass transfer ability, and thus the same inlet gas flow conditions, small particle size adsorbents wave forward speed is relatively slow, reflecting the level of the adsorbent bed adsorption equilibrium time is longer.

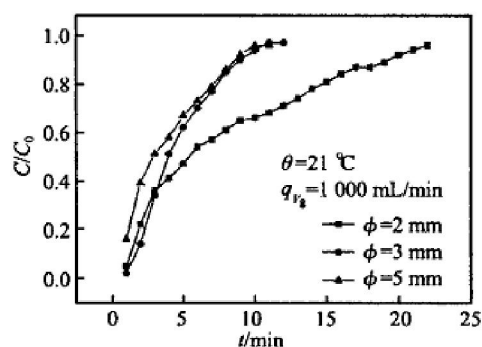


Fig 8 Influences of particle diameter on CO₂ adsorption breakthrough curves

Figure 9 is the type of adsorbent particle adsorption curve of Figure 9 shows that the particle size decreases as the adsorbent, the stacked-bed adsorption equilibrium adsorption capacity for CO₂ increases as the particle diameter of 2mm adsorption capacity up 91.21mg / g, and the adsorption capacity of the particle diameter 5mm only 42.06mg / g. This is mainly because as the particle size decreases, the stacked bed and macro flowing gas contact surface area of particles becomes large, and the same inlet gas flow conditions, the channels between the particles stacked bed convective mass transfer of the gas ability, more accessible for the adsorption sites. Thus for the packed bed adsorption reached equilibrium when a large amount.

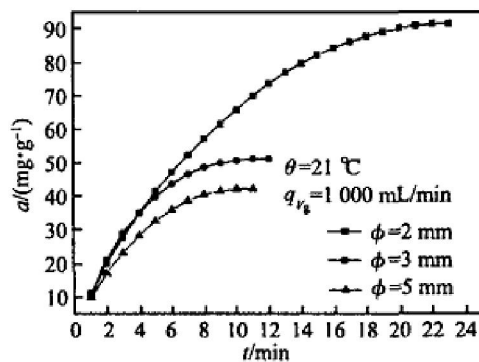


Fig 9 influences of gas mixture flow rate on CO₂ adsorption breakthrough curves

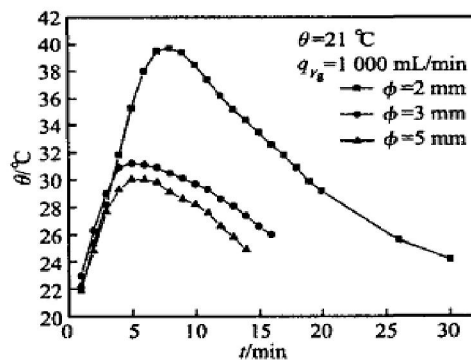


Fig 10 influences of particle diameter on center temperature of adsorption bed

Figure 10 is a size change on the adsorbent bed temperature of the center of. As can be seen from Figure 10, as the particle size decreases, the bed temperature significantly increased, when the diameter of the adsorption bed 2mm temperature reached 18.7 °C, far greater than the particle diameter 5mm 3mlTI and Adsorption bed temperature rise. Which on the one hand due to the adsorption of 2mm diameter large, the adsorption heat released during the adsorption more, and smaller particle size, the heat transfer to the particles from the interior of the particle heat transfer resistance outside the small, adsorption release faster transfer of heat energy to the particle exterior, thus packed bed adsorption temperature was significantly increased. Also seen in Figure 10, when the adsorbent particle size is from 2mm to 3mm adsorber temperature increase sharply, and the particle diameter to 5mm 3ram, the adsorption bed reduction temperature decrease, mainly due to two adsorption those little difference (see Figure 9). This indicates that the zeolite particles increases to a certain size, and then continue to increase sorbent particle size, reducing the influence on the adsorption properties. But it is worth noting that the temperature of the adsorption properties of known, the temperature rise is not conducive to the zeolite adsorption of CO₂. Therefore, the small particle adsorbent, on one hand the increase of the surface area in favor of CO₂ adsorption, on the other hand, the increase of the adsorption heat of adsorption on the latter in turn negative. However, if the packed bed and the external environment to enhance the heat transfer, the bed temperature will drop. It can be seen, the use of small particle size adsorbent, the adsorption process while strengthening cooling, reducing the operating temperature of the fixed bed adsorber, can effectively improve the performance of a fixed bed adsorption.

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4. Conclusions

In zeolite 13X-APG particulate adsorbent using ASAP2020 physical adsorption analyzer to characterize the adsorbents; fixed bed adsorber and studied for CO₂ adsorption characteristics discussed inlet gas temperature, gas flow rate, the adsorbent particle size. The results showed that: the gas temperature ahead of CO₂: adsorption breakthrough point, and significantly reduces the adsorption capacity of the adsorbent; adsorbed gas volume flow rate increases resulting in increased absorption, but little effect on the adsorption, the adsorption process is internal diffusion controlled process. The adsorption capacity of 2mm diameter up to 91.21mg / g, the adsorption capacity 5mm diameter 2.1 times, but the small size of the adsorbent in the adsorption process the temperature rise is larger, reaching 18.7 °C. Therefore, the use of small particle size adsorbent, the adsorption process to enhance heat dissipation and reduce the operating temperature of the fixed bed adsorber, in order to effectively improve the performance of a fixed bed adsorption.

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