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MILLIMETER-SIZED PITCH-BASED SPHERICAL ACTIVATED CARBONS WITH SMALL MESOPORES FOR CO_2 CAPTURE

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Introduction

The rapidly blossom of industry and the immoderate exploitation of fossil fuel has underscored the urgency of reducing CO_2 emission and exploring the related CO_2 capture, conservation, and utilization technologies [1].As a greenhouse gas, CO_2 emission has created global warming issues, rose of sea levels, and endangered the existence and security of humans and animals. Meanwhile, CO_2 is an important C1 industrial raw material for the synthesis of organic fine chemical intermediates [2]. Therefore, the development of a high-efficiency CO_2 capture and purification technology will be one of the principal themes for energy conservation and emission reduction.Active carbon materials adsorbent is a promising choice due to its high specific surface area, adjustable porous structure, and excellent thermal and chemical stability. This unique structure endows the carbon materials with high adsorption capacity, good selectivity and efficient regeneration [3]. In this work, PSAC was synthesized via suspension polymerization coupling with oxidative stabilization, carbonization and H2O steam activation, and deliberately designed microporous PSAC-2, micro-mesoporous PSAC-4 and PSAC-6 by simply regulating activation time. The result of CO_2 adsorption measurement indicate that PSAC-4 has better CO_2 uptakes than PSAC-2 and PSAC-6 under 1.0 bar.

Materials and Methods

In a typical preparation, coal tar pitch and naphthalene were smashed to particles about 0.1~2 mm. Finally, the particles were added into polyvinyl alcohol aqueous solution in autoclave at 150 ℃ under stirring about 3h. The obtained pitch-based spheres was stabilization, then it was further carbonized by heating at 920 ℃ for 1 h, and H_2O steam activated process was introduced about 2h, 4h, 6h, the resultant PSAC were named as PSAC-2, PSAC-4, PSAC-6.

Results and Discussion

Figure 1a show the CO_2 adsorption isotherms of PSAC-2, PSAC-4, and PSAC-6 at 25 ℃. All the samples show similar increasing trend of CO_2 uptake capacity with pressure boosting and no distinct saturation within the measurement pressure range. It is clearly that all samples display the similar CO_2 uptakes low pressure of 0.2 bar, implying that CO_2 uptakes are dominated by the ultramicropore volumes [11, 20]. With increasing the CO_2 pressure to 1.0 bar, PSAC-4 exhibits a higher adsorption quality of 2.59 mmol g^{-1} than PSAC-2 of 2.09 mmol g^{-1}. Such higher CO_2 uptake for PSAC-4 is mainly ascribed to the well-developed micropore (1 ~ 2 nm) volumes. In addition, the CO_2 incremental uptake is not proportional to the increment of specific surface area. The CO_2 uptake capacity of PSAC-6 with rich mesopores is 2.04 mmol g^{-1} and smaller than

PSAC-4 of 2.59 mmol g^{-1}, although they have approximate micropore (< 2 nm) volumes and microporous specific surface area, indicating that introducing more mesopores in the PSAC-6 could cause an explosion in decrease of CO_2 uptake. Interestingly, PSAC-2 and PSAC-6 deliver the comparable CO_2 uptake capacity, although their micropore volumes are in an entirely different state. In other word, physical absorption is mainly depends on the synergistic effect of diffusion rate and the amount of adsorption site. While at a moderate pressure of 5.0 bar, PSAC-4 exhibits larger CO_2 uptakes about 7.23 mmol g^{-1} than PSAC-2 of 4.99 mmol g^{-1}, and higher rate of CO_2 uptakes increment than 1.0 bar (7.23/4.99 = 1.45 > 2.59/2.09 =1.24). This result indicates that the introduction of mesopore can accelerate the diffusion of CO_2 into the interior of PSAC-4 and enhance the utilization percentage of microporous adsorption sites [13]. Compared with micropore dominated PSAC-2, micro-mesoporous PSAC-6 shows the higher CO_2 uptakes about 5.94 mmol g^{-1} than PSAC-2 of 4.99 mmol g^{-1} at 5.0 bar, which imply that CO_2 adsorption can be occurred in the developed small mesopores for capillary condensation. In addition, PSAC-4 and PSAC-6 share the higher incremental CO_2 uptakes than PSCA-2 within the same pressure fluctuation, indicating that appropriate introduction of small mesopore is beneficial to CO_2 uptakes. In order to probe the CO_2 adsorption rate of PSAC-4, CO_2 adsorption measurements were performed under different retention time at 25 ℃, where the retention time refer to the system pressure unvaried time. As shown in Figure 1b, the CO_2 capacity is constant under different pressure with the increasing retention time, indicating the rapid CO_2 adsorption rate. That is to say the length of the residence time has no influence on CO_2 adsorption, and PSAC-4 exhibits a typical physical absorption and quickly CO_2 adsorption kinetics.

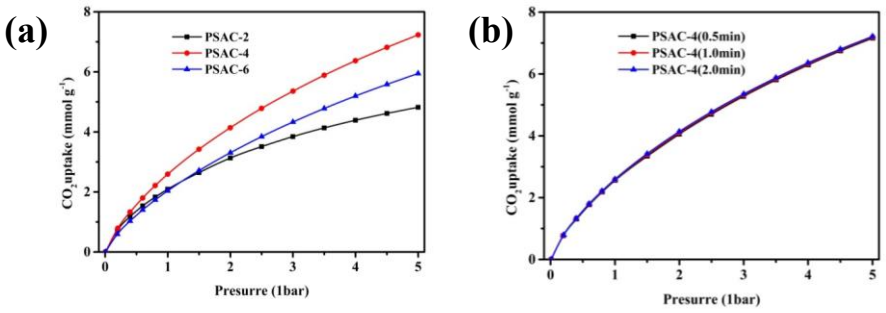

Figure 1 (a) CO_2 adsorption isotherms of PSAC-2, PSAC-4 and PSAC-6, (b) CO_2 adsorption isotherms of PSAC-4 under different retention time.

Conclusions

A series of pitch-based spherical activated carbons were synthesized via suspension polymerization coupling with oxidative stabilization, carbonization and H_2O steam activation method. The as-obtained PSAC-4 possess a high CO_2 adsorption capacity of 2.59 mmol g^{-1} at 1.0 bar and 7.23 mmol g^{-1} at 5.0 bar.

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