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Experimental and modelling studies of CO₂ capture via Pressure Swing adsorption for zeolites and amine modified mesoporous silicas

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The capture of carbon dioxide via Pressure Swing Adsorption (PSA) has been examined experimentally and mathematically. The adopted method was a two bed/four step process, known as the Skarstrom Cycle.

Experimentally, the performance of zeolites and mesoporous silicas has been investigated, for different experimental conditions (cycle time, pressure ratio, feed/purge ratio). Recently, amine modified mesoporous silicas have received attention as an alternative option to zeolites for carbon dioxide capture. Several studies have been conducted in this area over the last few years. In the majority of these studies, the adsorption capacity of modified SBA-15 was measured by the injection of CO₂ into modified SBA-15 powder, under pressure, which is a much simpler approach than that normally encountered in more complex PSA processes. The relationship between the diffusion rates within and uptake characteristics of powder beds and the stationary columns of pelletised materials employed in PSA is not trivial and it was felt that a more direct investigation of PSA (and its variants) employing pellets of amine modified SBA-15 was warranted.

In this study, pelletised and calcined SBA-15 powder, modified with APTES ((3-Aminopropyl)triethoxysilane) has been tested in a PSA configuration. In order to investigate the diffusion mechanism (macropore/micropore) of carbon dioxide into the calcined SBA-15 pellets (unmodified), three diameters were selected for the production of the cylindrical pellets (cylinder diameters 1.2 mm, 1.7mm and 2.5mm each of length XX mm). A comparison of the breakthrough curves (Figure 1) indicated that macropore diffusion could be neglected and that the micropore diffusion mechanism plays a more important role in the control of the overall adsorption kinetics. The adsorption performance (Figure 2) of the APTES modified material was compared to the unmodified material, only to demonstrate that the poor performance of the calcined SBA-15 can be more than doubled with the presence of one amino group (APTES modified SBA-15).

For comparative purposes, PSA tests with APTES modified SBA-15 and zeolite 13X, which is a commercial adsorbent, were employed. In the case of zeolite 13X, capture of up to 99.9% of the CO₂ in a gas feed containing 10% carbon dioxide was achieved with a three-fold increase in the average composition of the CO₂, for a cycle time of 800 seconds, 6.25 bars pressure and 20°C temperature. APTES SBA-15 was less efficient than zeolite 13X in PSA, while RPSA (Rapid Pressure Swing Adsorption) was found to be more appropriate.

Capture of up to 45% of the CO₂ in a gas feed containing 10% carbon dioxide was achieved with a three-fold increase in the average composition of the CO₂, for a cycle time of 30

seconds, 3 bar pressure and 20°C temperature. This difference could be easily explained due to the different adsorption characteristics. Physisorption is the mechanism for CO₂ adsorption onto zeolites, while chemisorption is considered to be the primary mechanism for adsorption onto amine modified mesoporous silicas. Additional studies will be conducted in order to investigate the improvements in the limits of APTES SBA-15. A small amount of moisture will be introduced in the feed stream (to encourage the formation of carbonates in contrast to carbamates during adsorption) and higher operating temperatures will be tested. Moreover, the modification of SBA-15 with polyamines (more amino groups) is expected to extend the adsorption capacity of the material, so polyamine modified materials will be produced and tested.

In order to examine the temperature profile of PSA, three thermocouples were placed along the two columns of the PSA apparatus. In the case of zeolite 13X the profile demonstrated the existence of significant non-isothermal behaviour (Figure 3). The temperature profile for the APTES modified SBA-15 exhibited no clear non-isothermal trend, in view of its significantly different adsorption behaviour. The thermal characteristics of latter are under investigation both experimentally and theoretically.

For comparative purposes, two mathematical models have been employed in the numerical simulations of the two bed/four step process: the first model was originally suggested by Doong and Yang (1987)* and is based on uncoupled linear Fickian diffusion within bidisperse porous media subject to the linear driving force (LDF) approximation; the second approach is a more advanced model of linear coupled diffusion with a non-linear driving force approximation.

The experimental and simulated PSA results for both the zeolite and for the modified SBA-15 systems will be compared and discussed.

*Doong, S.J., Yang, R.T., *AIChE J.*, **33**, 1045-1049 (1987).

Keywords: Carbon dioxide capture, Pressure/Vacuum Swing Adsorption, Skarstrom Cycle, Mesoporous Silica, Zeolites, Modelling-Simulations

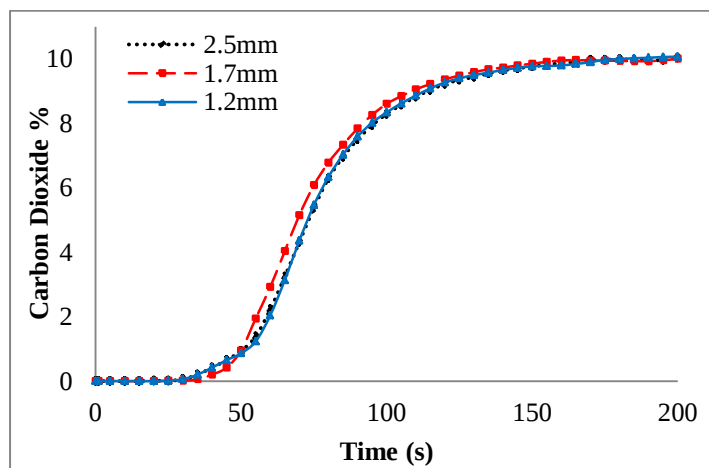


Figure 1: Breakthrough curves of SBA-15 for the experimental examination of micropore and macropore diffusion in PSA (feed flow: 1000cm³/min, feed stream composition: 10% CO₂)

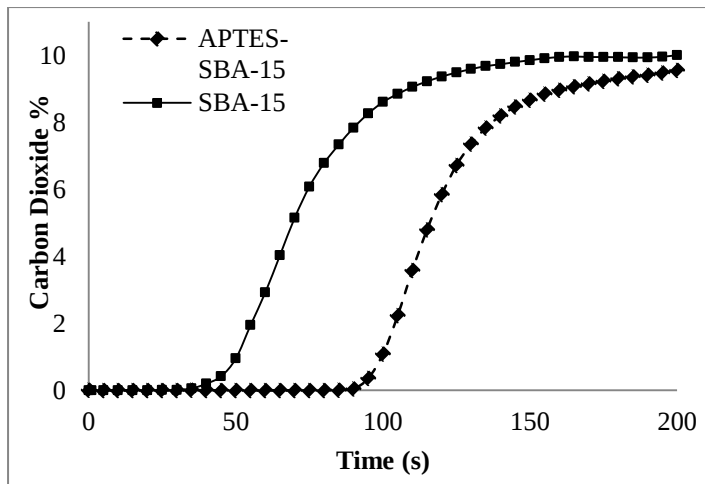


Figure 2: Breakthrough curves of unmodified and APTES modified SBA-15 pellets

(feed flow: $1000\text{cm}^3/\text{min}$, feed stream composition: 10% CO_2)

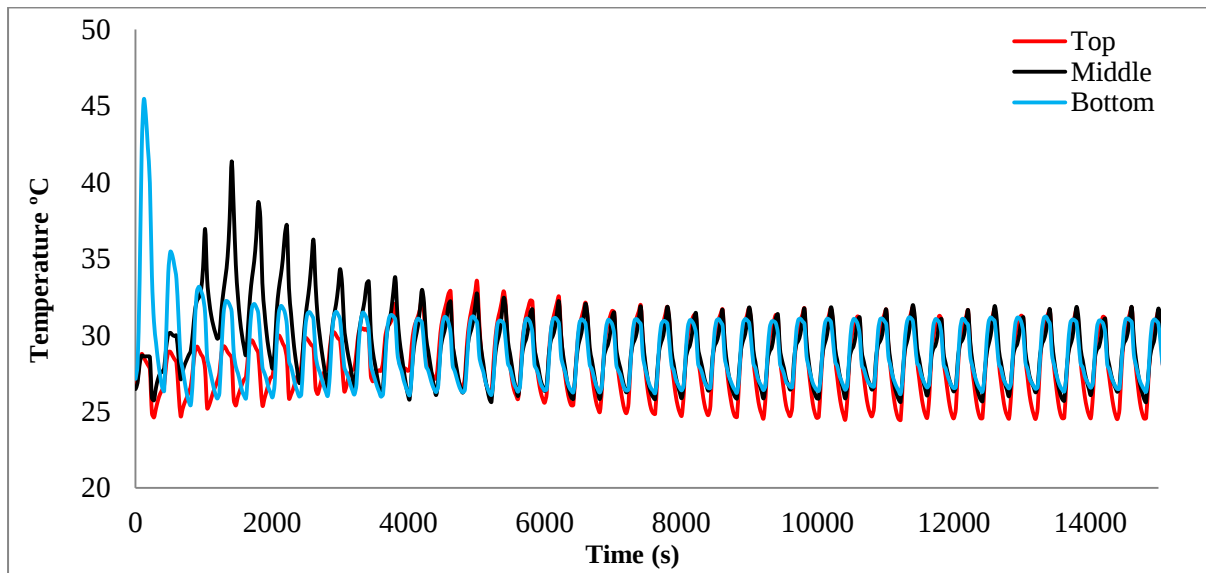


Figure 3: Temperature profile of carbon dioxide capture in PSA for zeolite 13X - Non isothermal behaviour. The feed enters at the bottom of the column and the purge at the top.