Development of a pressure swing adsorption process for pre-combustion CO_2 capture

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One of the most promising technologies for CO_2 capture from power generation plants based on fossil fuels is Integrated Gasification Combined Cycle (IGCC), where the CO_2 is separated from H_2 before its combustion. In contrast to post-combustion capture, where the CO_2 partial pressure is below 1 bar, the CO_2/H_2 separation process profits of the specific feed characteristics, namely a pressure up to 40 bar and a CO_2 concentration of about 40 vol%. Therefore pressure swing adsorption (PSA) offers an attractive alternative to absorption- or membrane-based techniques.

The present study investigates both the experimental and theoretical aspects related to the PSA process. First a sound characterization of different promising adsorbents, namely activated carbon (Chemviron, Germany), MOF[1] and MCM-41[2] is done by measuring equilibrium adsorption isotherms of CO_2, H_2 and N_2 using a Magnetic Suspension Balance (Rubotherm, Germany). Additionally physical material properties such as material and bed densities as well as heat capacities were defined.

Then a two column PSA unit has been built, to allow for dynamic adsorption measurements and continuous H_2 and CO_2 production. The PSA unit is equipped with thermocouples at different locations inside the columns, pressure sensors and a mass spectrometer for online composition analysis. The behavior of the pressure and temperature inside the column as well as the outlet composition of the H_2 and CO_2 product during continuous production is shown in Figure 1.

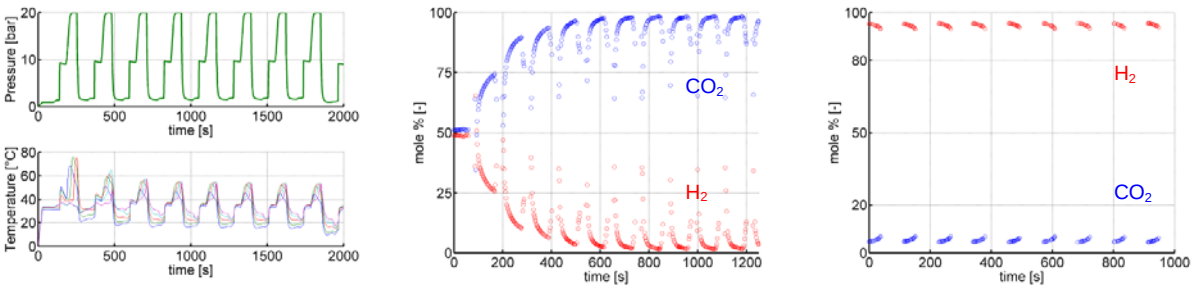

Figure 1: PSA experiment on activated carbon at 25°C and cycling the pressure between 1 and 20 bar.

[1] A. Bjørnar et al., *Adsorption* **14**, 755 (2008)

[2] Y. Belmabkhout et al., *Chem. Eng. Sci.* **64**, 3721 (2009).

The interpretation of the dynamic experiments is done by describing the process with a detailed one-dimension model consisting of mass and heat balances and several constitutive equations such as adsorption isotherms, equation of state and pressure drop correlation. The process parameters of the mathematical model, which cannot be separately measured, such as mass and heat transfer coefficients, are fitted to the dynamic adsorption column experiments.

For the design of the PSA process several aspects are important: the energy penalty has to be minimized whereas the capture rate is aimed to be higher than 90% and the CO_2 purity has to meet the specifications for the subsequent transportation and storage.

One characteristic of PSA processes is the large number of process layouts as the individual steps, i.e. pressurization, adsorption, pressure equalization, blowdown and purge, can be combined in many different ways. Then defining a sequence of the basic steps, the process parameters such as temperature, adsorption and desorption pressure, flow rates and cycle time have to be defined and optimized. For every new material as well as for different gas mixtures the optimal process can be rather different.

Therefore in this work, after the validation of the process parameters, the simulation tool is used to describe various complete PSA cycles with different combinations of the individual steps. To assess the performance of the newly developed PSA cycles a multi-objective optimization including the CO_2 capture rate and the CO_2 purity is conducted. This analysis leads to a Pareto set comparing all operating points that are optimal with respect to all objective functions, i.e. no better operating points can be found where all objective functions can be improved. Additionally the adsorbent productivity is calculated given that it has a big impact on the capital and operational costs. In this manner the process performance of the new adsorbents (MOF[1] and MCM-41[2]) is compared to the base case with commercial activated carbon.

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