Dynamic and equilibrium-based investigations of CO_2 removal from CH_4-rich gas mixtures on zeolites

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Introduction: CO_2 Removal from Methane

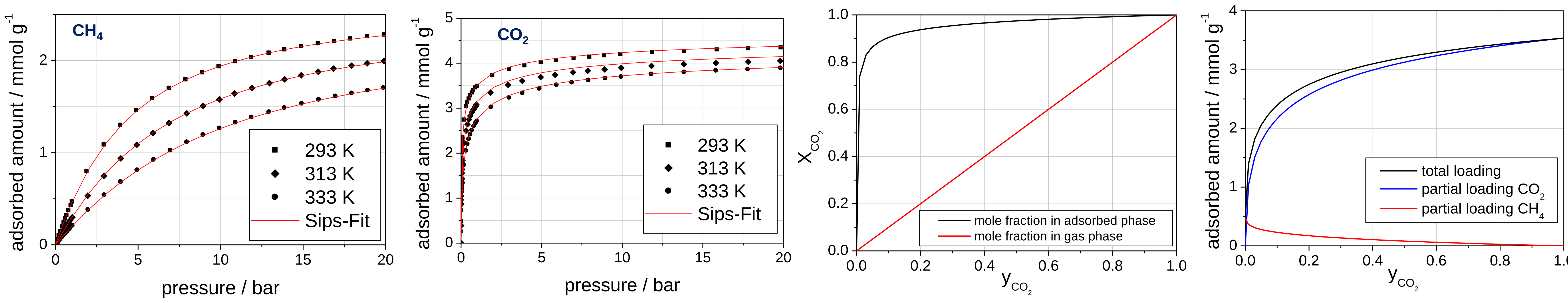
The adsorptive separation of gas mixtures along a fixed bed adsorber is one of the most effective and thus economical separation techniques. In general, the suitability of an adsorbent for the separation of a certain gas mixture cannot be deduced from texture data only, but is determined by a multitude of complex adsorption phenomena taking place within the adsorber column. Multicomponent adsorption equilibria, sorption kinetics, heat release and the flow conditions within the adsorber are just a few parameter that define the overall separation efficiency. The pure component sorption isotherms of CO_2 and CH_4 and the breakthrough curves of CO_2, CH_4 and CO_2/CH_4 mixtures have been measured at different temperatures on a binder containing zeolite 13X molecular sieve (Köstrolith® NaMSXK) in order to investigate the CO_2 removal from methane-rich gas mixtures (relevant in natural gas purification).

Single and multi-component sorption equilibria

- High pressure isotherms have been measured for CO_2 and CH_4 with an iSorb HP volumetric sorption analyzer
- The total adsorbed amount is much higher for CO_2 than for CH_4 over the whole pressure range
- The single component isotherms suggest a high separation performance of the zeolite for CO_2-CH_4 gas mixtures

dynaSim

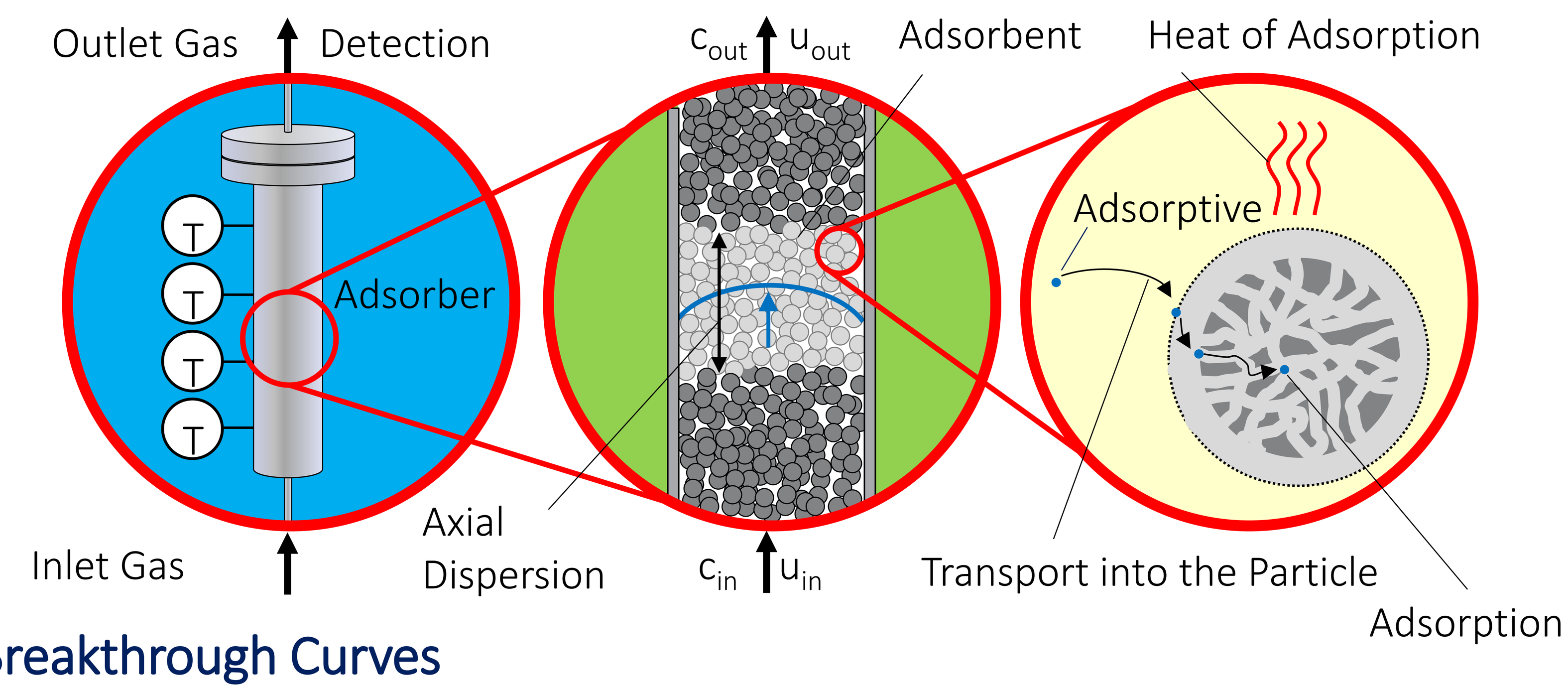
- The simulation tool dynaSim was used to calculate the mole fraction of CO_2 in the adsorbed phase with respect to the mole fraction of CO_2 in the gas phase at a total pressure of 1 bar and a temperature of 293 K
- In the same way the total loading and the partial loadings have been calculated based on multi-component SIPS



Breakthrough Curves: Measurements and Simulations

Analysis conditions

- Pressure: 5 bar (absolute), Gas flow: 2.5 L min^{-1} (STP), inner diameter of the adsorber: 30 mm, Temperature: 293 K
- Gas mixture 15 % CH_4 and 5 % CO_2 in He



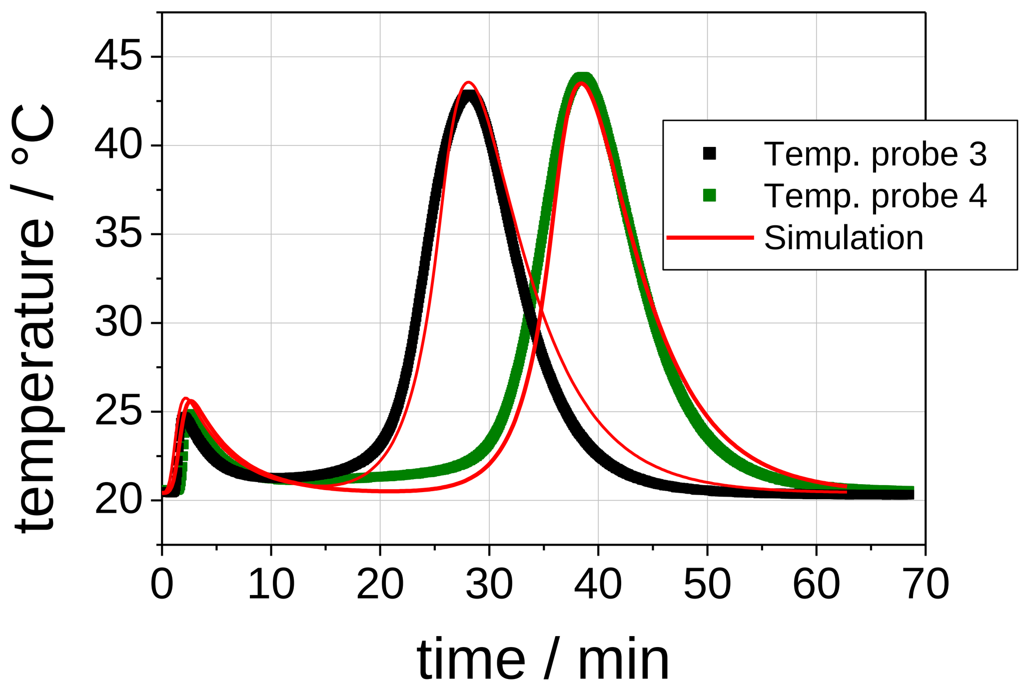
Breakthrough Curves

- The measured breakthrough curves (BTC's) prove the good separation performance of the zeolite under authentic process conditions
- The measured BTC's are in accordance with the predictions based on the pure component isotherms

Simulation

- The linear driving force constants k_{LDF} of CO_2 and CH_4 can be calculated by fitting a model curve to the measured BTC's
- That fitting is based on mass- and energy balances

Heat profiles measured within the fixed-bed during adsorption

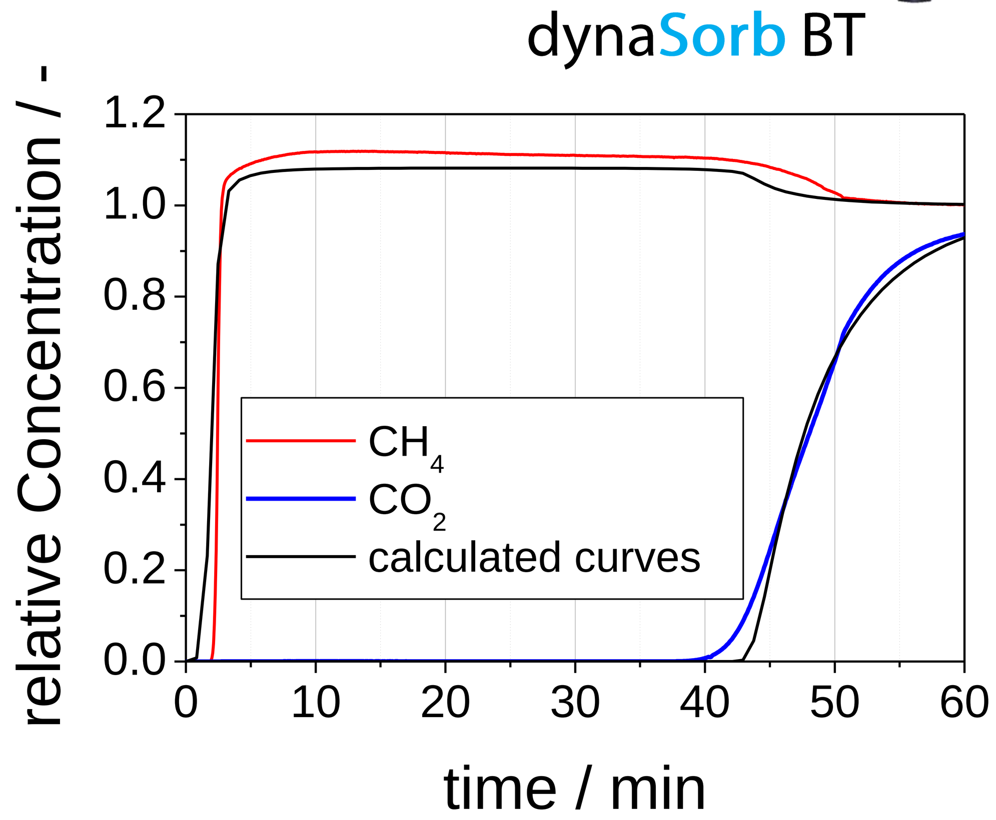


dynaSorb BT

Linear driving force constants

k_{LDF} (CO_2) = 0.48 min^{-1}

k_{LDF} (CH_4) = 5.30 min^{-1}



Conclusions

- The investigated zeolite reveals an outstanding separation performance in CO_2 removal from methane-rich gas mixtures
- The separation efficiency under process conditions is in perfect agreement with the predictions based on pure component isotherms
- The higher kinetic coefficient found for CH_4 has no negative impact on the separation performance
- The excellent separation is exclusively caused by the strongly preferential adsorption of CO_2 compared to CH_4
- The dynaSorb BT in combination with the simulation tool dynaSim provides a powerful set-up to investigate technical separation processes under authentic conditions

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