Sorption behavior of a hydrotalcite-based sorbent for CO₂-free hydrogen generation

Hydrogen is mainly produced via ‘steam reforming’ of natural gas. This is accompanied with large CO₂ emissions. The novel ‘sorption-enhanced water-gas-shift process’ allows hydrogen production with reduced CO₂ emissions, by capturing the CO₂ in the reactor on a solid sorbent. In the regeneration step the captured CO₂ is released and stored or reused. Researcher Kai Coenen investigated the complex adsorption behavior of different gases on a hydrotalcite-based sorbent and developed the first comprehensive kinetic model which adequately describes the complex interactions. He found that mainly the CO₂ release stage is slow but can be enhanced with steam.

The sorption-enhanced water-gas-shift (SEWGS) process combines the high-temperature water-gas-shift reaction with almost instant adsorption of CO₂. This means that hydrogen is produced, and CO₂ is captured in one single process step. Hydrotalcite based adsorbents impregnated with potassium have a high stability, work fast and have enhanced CO₂ capture and release capacities due to the potassium. To better understand and quantify the interactions between small gas molecules H₂O, CO₂ and H₂S and this sorbent, we investigated in detail the adsorption of these molecules on it, using different experimental techniques.

During my studies we found that especially the slow release kinetics determines the cyclic working capacity of the sorbent, while the adsorption is in general very fast. Therefore, the release stage (the regeneration of the sorbent) is the most important step to improve process efficiency.

Based on extensive experimental studies, combining the results of different experimental techniques such as thermogravimetric analysis (TGA), breakthrough experiments in a packed bed reactor (PBR), aluminum nuclear magnetic resonance (NMR), in-situ infrared spectroscopy (IR) and X-ray photoelectron spectroscopy (XPS), we developed a mechanistic model involving different adsorption sites, which explains and quantitatively describes the complex sorption and desorption behavior if steam is used for the sorbent regeneration. We found that the slow CO₂ desorption kinetics is caused by the heterogeneity of the sorbent surface which leads to different strengths with which the CO₂ is bound.

We developed the first comprehensive kinetic model which adequately describes the complex interactions of CO₂ and H₂O on the adsorption kinetics and cyclic working capacities of these sorbents. Additionally, we investigated adsorption of the highly toxic H₂S and its interaction with CO₂ and H₂O and present important findings which can help to use potassium-promoted sorbents also for other large-scale industrial sorption-processes, such as H₂S-removal from natural gas.

The results we presented in this thesis have not only led to several publications in the field of chemical engineering but can be used to improve the cycle design of adsorption columns, which can lead to a significant reduction of CO₂ emissions and H₂ production costs.