

A Multifunctional Reactor for CO₂ Capture and Conversion to Synthetic Natural Gas (SNG)

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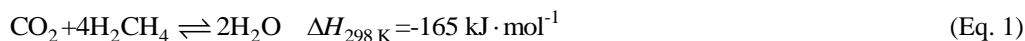
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Highlights

- A multifunctional reactor for capturing and converting CO₂ to SNG was successfully tested.
- Sorption capacity and desorption kinetics were improved under reactive regeneration mode.
- Complete conversion of captured CO₂ was obtained in dynamic operation.
- A max. CH₄ productivity of 2.32 mol_{CH₄} / (kg_{cat}·h) and 35 % purity was reached at 350 °C.

1. Introduction

New challenges have emerged following the growing contribution of renewables to the energy sector. For instance, the intermittent availability of resources like wind or the sun, frequently lead to a misalignment between production and demand, which requires the development of energy storage solutions to make these processes more efficient and versatile [1]. Among them, the Power-to-Gas (PtG) concept has stand out, particularly for synthetic natural gas (SNG) production, a pathway that enables balancing the power and gas grids [1]. In this approach, renewable power surpluses are supplied to a water electrolyzer to obtain H₂ that is subsequently used to produce SNG by reacting with CO₂ through the Sabatier reaction (Eq. 1).



CO₂ required to the reaction may be available from several processes (e.g. flue gas from power plants, refineries, cement industry, etc.). However, CO₂ purification requirements may differ substantially depending on the carbon source considered, which has a direct influence on PtG process configuration and flexibility. This work focuses on the study and development of a Carbon Capture and Utilization (CCU) technology able to, simultaneously, remove CO₂ from flue gas and further convert it to methane in a hybrid and cyclic sorption-reaction unit.

2. Methods

Sorption desorption/reaction tests were performed on a structured reactor containing alternating layers of a commercial K-promoted hydrotalcite (from SASOL) and a commercial Ni-based catalyst (from Clariant) with a sorbent/catalyst ratio of 3.15. Besides warm-up and materials activation, reactor operation included two steps: CO₂ from a synthetic flue gas stream (15 mol % of CO₂ in N₂) was firstly captured by sorption and afterwards converted in the same unit into synthetic natural gas. The later step begun by switching the feed to a pure H₂ stream for reactive regeneration of the sorbent bed fraction (or to N₂ for desorption only). The multifunctional reactor was tested at low pressure (<2.5bar) in a temperature range between 300-350 °C. The reactor performance was evaluated in terms of CO₂ sorption capacity, CO₂ conversion, CH₄ productivity and CH₄ purity at the reactor outlet.

3. Results and discussion

The sorbent working capacity at 350 °C was determined in a series of 5 sorption-desorption cycles under dry conditions and without reactive regeneration. In these experiments the sorbent was fully saturated before starting bed regeneration. The first sorption-reaction took place in cycle 6 where the bed undergone reactive regeneration for the first time. It was observed in the subsequent cycles that the sorption capacity increased and the initial bed capacity (of cycle 0) was recovered – cf. Figure 1.

The CO₂ that before was irreversibly sorbed could thus be removed due to the presence of steam that was produced *in situ* during the reactive regeneration of cycle 6 (see Eq. 1). This is a special feature of hydrotalcite-type sorbents (e.g. [2]), which makes them especially advantageous for this application and reaction. Fig. 1 also shows that around 92 % of previously captured CO₂ could be converted.

The reactor performance was also studied when the sorption step was stopped before CO₂ starts to break through the reactor, operating in a similar manner as a

cyclic pressure-swing unit. In this case, the conversion of captured CO₂ was almost complete (99 %) and the reactor performance was also assessed in terms of the CH₄ productivity and purity observed at the reactor outlet and along the reactive regeneration time (t_R).

Fig. 2 shows that both productivity and purity increase with the reactive regeneration time reaching a maximum before they start decreasing, providing information about the adequate time to stop this step. The maximum CH₄ productivity and purity attained was 2.32 mol_{CH₄} / (kg_{cat}·h) and 35 % at 350 °C, respectively.

4. Conclusions

A multifunctional reactor containing a structured bed of CO₂ sorbent and methanation catalyst layers was successfully tested for CO₂ capture and conversion into SNG. The sorbent performance was enhanced as a result of process intensification. Captured CO₂ at high temperature could be completely converted, but further reactor design is required to improve CH₄ purity at the outlet of the reactor in order to comply with natural gas grid specifications.

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References

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Keywords

“Power-to-Gas”, “Carbon Capture and Utilization”, “Multifunctional reactor”, “Reactive regeneration”.

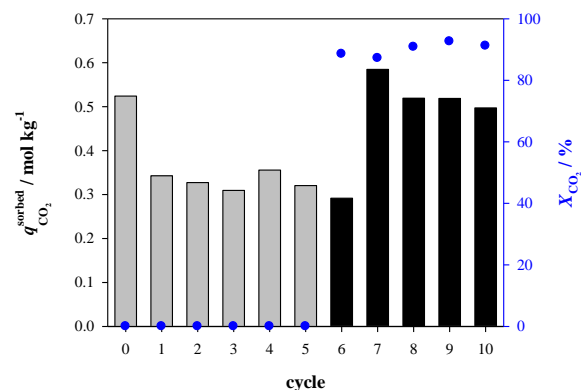


Figure 1. CO₂ sorption capacity and conversion during sorption-desorption/reaction cycles at 350 °C and $P=1.34$ bar. Grey and black bars stand for the sorption capacity obtained with normal (N₂ stream) or reactive regeneration (H₂ stream), respectively. Blue dots stand for the conversion of captured CO₂.

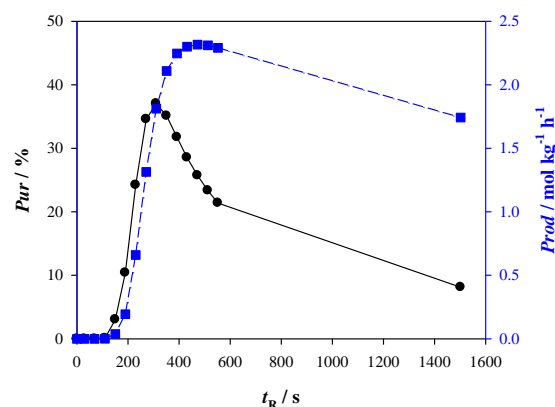


Figure 2. CH₄ productivity and purity obtained at 350 °C and for a total pressure of 1.34 bar.