Cyclically-integrated CO_2 Capture and Conversion into Synthetic Natural Gas

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Highlights

- A sorptive reactor was conceived for CO_2 capture and conversion.
- The unit was operated cyclically, providing continuous CO_2 capture and CH_4 production.
- Proof-of-concept was carried out for both flue gas and biogas.

1. Introduction

In the perspective of decarbonizing several sectors, the "Power-to-Gas" approach has gained significant prominence and could play a significant role in the future energy system; it is based on converting electricity from a renewable source into a gas easily transported and stored [1]. Among the existing options, the production of methane benefits from the existence of a well-established natural gas infrastructure where it can be injected, enabling the integration of power and gas sectors; alternatively, it can be used in neighbor facilities.

Carbon dioxide may be available, for example, from flue gas streams of various industrial processes. However, in such streams CO_2 is often too diluted for subsequent use; thus, its previous capture is required, which typically involves different process units. The work carried out aimed the development and demonstration of a cyclic adsorption and reaction unit, with continuous operation, capable of, simultaneously, removing CO_2 from flue gas streams [2] (or even from biogas [3]) and converting it into CH_4 (synthetic natural gas).

Such unit can be operated as schematically illustrated in Fig. 1. In a first phase, one of the reactors (operating in the sorption mode) is fed with the CO_2-containing gas, so that the CO_2 is retained in the selective sorbent (e.g. hydrotalcite) contained in the mixed bed while the other species (N_2 or CH_4, for flue gas and biogas, respectively) leaves the column, until almost complete saturation of the bed. In the second stage – called reactive regeneration, H_2 (obtained from a renewable source, for example via water electrolysis) is fed to the same column and reacts with the CO_2 previously concentrated in the mixed bed (that contains both the CO_2-selective sorbent and the methanation catalyst), producing methane by the Sabatier or methanation reaction (Eq. 1):

$$CO_2 + 4H_2 \rightleftharpoons CH_4 + 2H_2O \qquad \Delta H^{298K} = -165\ kJ \cdot mol^{-1} \qquad (1)$$

Thus, to operate continuously, the process requires at least two columns operating in complementary steps: when a CO_2-saturated bed is being regenerated (with green H_2) and is producing CH_4, the other is in the CO_2 capture phase and vice-versa.

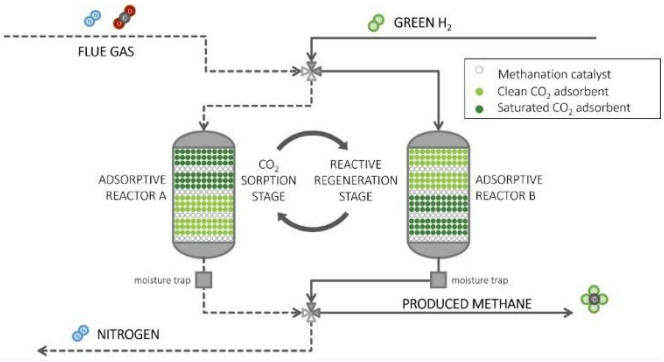

Figure 1. Scheme of the integrated process for capturing and converting CO_2 into CH_4 with two beds operating 180º out of phase; illustration is made using flue gas as the CO_2 source stream.

2. Results and discussion

The proof-of-concept of the integrated sorption-reaction process was done while also analyzing the effect of a wide range of experimental conditions: flue gas/biogas inlet flow rate, feed CO_2 content (to simulate either different typical flue gas – in various industrial processes – or biogas streams), stage duration, temperature, and H_2 inlet flow rate. Moreover, different process indicators were defined: i) CO_2 sorption capacity – which reflects the amount of CO_2 captured per mass of sorbent in the specified cycle conditions; ii) CO_2 conversion – which is the fraction of the previously sorbed CO_2 that is converted in the reactive regeneration stage; iii) methane productivity – amount of CH_4 produced per mass of catalyst and time unit; iv) ratio between H_2 fed and CH_4 produced (important for economic reasons), and v) methane purity – which is the average outlet content of methane during the reactive regeneration stage (for flue gas) or during both stages (for biogas). The effect of some process parameters on those indicators will be shown, for each CO_2-containing gas stream.

It was possible to reach CH_4 purities above 80% (for biogas feed) and up to 70-80% (for flue gas). Furthermore, the commercial tested materials showed absolute compatibility (a key factor for the success of the technology) and stability under operation for more than 150 h.

The parametric studies allowed to understand the effect of some operating conditions on the process performance, contributing to the future unit modeling and optimization, before the respective scale-up to reach a higher TRL level.

3. Conclusions

The multifunctional reactor approach presented envisions the capture of CO_2 from different streams and its conversion into synthetic CH_4 through the use of a sorptive reactor that combines, in the same unit, CO_2 separation/concentration and its subsequent catalytic methanation. In this way, it is possible to store renewable energy using CO_2 and H_2 coming from e.g. flue gas/biogas and water electrolysis, respectively, contributing to the decarbonization of the industrial ecosystems (net-zero scenario).

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Keywords

Multifunctional cyclic reactor, CO_2 capture and conversion, Flue gas and biogas, Power-to-Methane