Evaluation of the Integration of Direct Air Capture and Methanation Processes.

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Highlights

- Three DAC and Methanation processes with different integration strategies are evaluated: i) only heat integration; ii) sorbent regeneration with high-pressure H_2; iii) complete integration in a single unit.
- Auto-thermal operation can be achieved, since the heat generated in the methanation reactors exceeds the heat required for sorbent regeneration by the DAC units.

1. Introduction

Recently, ambient air has attracted interest as a potential source of virtually unlimited CO_2. Direct air capture (DAC) and CO_2 methanation could work particularly well together. With commercially available catalysts, the methanation reaction can be carried out at moderate temperatures (i.e. lower than 300 °C) and atmospheric pressure. These conditions are similar to the conditions required for the regeneration of typical solid sorbents developed for DAC. The aim of this work is to assess the challenges and the advantages related to the integration of adsorption-based DAC with CO_2 methanation for the production of synthetic natural gas (SNG) for energy storage and transport. Three processes with different levels of integration have been modelled and their performances compared, particularly: i) Only heat integration (HI-DACM); ii) Sorbent regeneration with high-pressure H_2 (HS-DACM); iii) Complete integration of DAC and methanation in a single unit (FI-DACM), schematically depicted in Fig. 1.

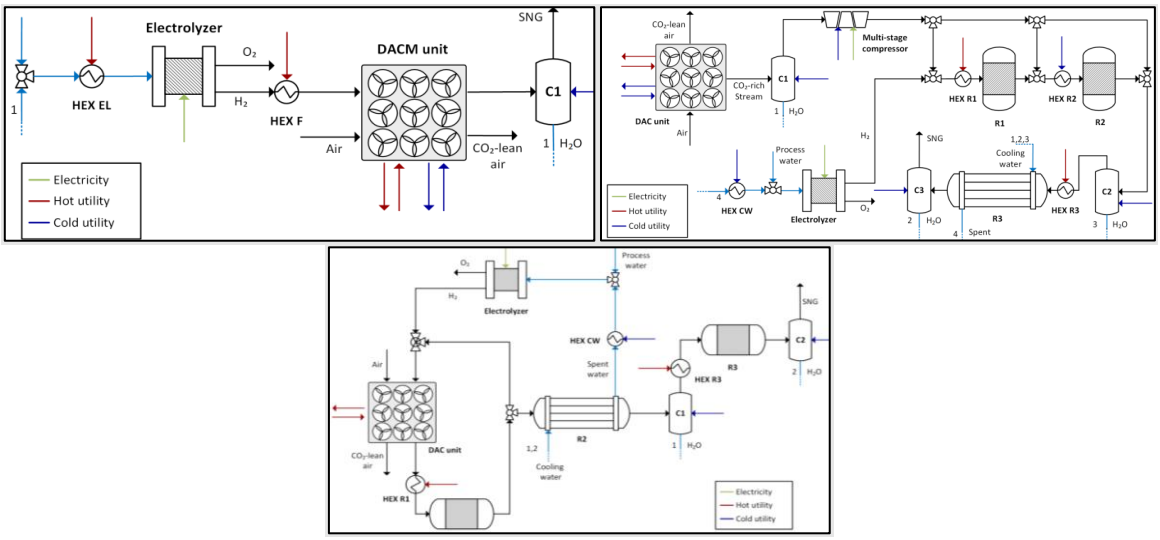

Figure 1:*(top-left) HI-DACM, (top-right) HS-DACM, (bottom) FI-DACM*

2. Methods

The HI-DACM process has been entirely modelled in Aspen Plus using the Peng-Robinson model to evaluate the thermodynamic properties of the system. The DAC unit is modelled as a black box, as its performance has already been assessed and optimized in our previous work [1]. The catalyst chosen is Ru/Al_2O_3 as it is the most active catalyst at low temperatures. The methanation reactors are modelled with the rate-based block RPlug, and the kinetics for the Sabatier reaction on Ru-based catalyst have been adapted from the work of Falbo et al. [2]. The HS-DACM has been modelled with the same methodology adopted for the HI-DACM. The cycle adopted for HS-DACM consists of four steps i.e. adsorption, pre-heating, regeneration and cooling. The different DAC cycle design required the

development of a robust and computationally light model, exploiting the Sharp-Front-Approach method (SFA) [3]. The FI-DACM has the DACM unit at its core, as here CO_2 is both captured and converted. The unit is described with a 1D pseudo-homogeneous reactor model. K_2CO_3 on Al_2O_3 sorbent was chosen thanks to its thermal stability [4]. The corresponding CO_2 isotherm was fitted from the experimental data [5]. The same Ru-based catalyst was considered, with the equilibrium and kinetic parameters obtained through the experimental data in the open literature. Two productivities are used as performance indicators: volume-based (Pr_V), defined as the mass rate of CH_4 produced divided the total volume of the air contactors and reactors, and the Ru-based (Pr_{Ru}), defined as the produced CH_4 divided by the mass of the used Ru catalyst.

3. Results and discussion

The HI-DACM process achieved high CO_2 conversion (98%) coupled with excellent CH_4 selectivity. Although the Pr_V is only 2.5 $kg_{CH4}\,m^{-3}\,h^{-1}$, Pr_{Ru} is much higher at 344 $kg_{CH4}\,kg_{Ru}^{-1}\,h^{-1}$. Heat integration allowed the reduction of the hot utility to zero, thus achieving overall auto-thermal operation. In the HS-DACM, a higher Pr_V of 7.2 $kg_{CH4}\,m^{-3}\,h^{-1}$ has been estimated. This outcome is in part a consequence of water adsorption not being considered, and in part resulting from the cycle design itself. On the other hand, a low CO_2 conversion of 32% is achieved, determining the lower Pr_{Ru} of 220 $kg_{CH4}\,kg_{Ru}^{-1}\,h^{-1}$. The overall duty demand amounts to 13.1 MJ kg_{CH4}^{-1}, which is considerably lower than what was estimated for the HI-DACM process. It was not possible to reach auto-thermal operation in this case. The FI-DACM provides the highest Pr_V, with an estimated value of 6.4 $kg_{CH4}\,m^{-3}\,h^{-1}$. This result is in part due to the lower total process volume, as the DACM units perform the functions of both air contactor and methanation reactors. However, this aspect also brings a considerable disadvantage, namely a poor use of the valuable Ru-based catalyst. As a result, a low Pr_{Ru} of 1.2 $kg_{CH4}\,kg_{Ru}^{-1}\,h^{-1}$ is obtained.

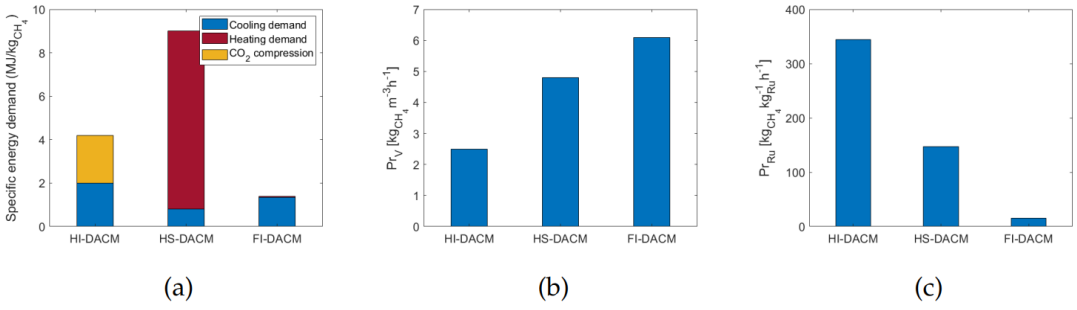

Figure 2: *a) Specific energy demand, b) Volume based productivity, c) Ru-based productivity*

4. Conclusions

This work provided a first indication of the potential synergies of integrating DAC with CO_2 conversion. It has been demonstrated that benefits can be achieved, especially in terms of energy demand. Future research should focus on the design of more realistic DACM cycles and heating strategies.

References

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Keywords

Direct Air Capture, CO_2 Utilization, Power-to-X, Heat integration