Process design for SNG production through CO_2 methanation

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In recent years, the interest in catalytic methanation of CO_2 has grown exponentially as it has proved to be a valid solution for both the reutilization of CO_2 and for the storage of renewable energy, responding well to the need energy transition. Once-through methanation processes that provide, as an output, a synthetic natural gas (SNG) stream ready to be injected in the gas grid, are highly desirable. Since the CO_2 methanation reaction is strongly exothermic and occurs with a decrease of the number of moles, the CH_4 yield at the equilibrium is favoured at low temperatures and high pressures. Moreover, at high temperatures, the undesired production of CO via reverse-water-gas-shift reaction may occur, leading to a decrease in CH_4 selectivity. Nevertheless, adequate temperatures, strongly depending on the activity of the adopted catalyst, are required to sustain the methanation kinetics and reach interesting SNG yields. For these reasons, the choice of the right process configuration and conditions are of pivotal importance for controlling the purity of CH_4.

In this work, the "single bed" process configuration (one isothermal reactor) has been experimentally and theoretically compared to the "double bed" one (two isothermal reactor at different temperatures), with or without intermediate removal of water (Figure 1). Experiments were carried out on a Ru/Al_2O_3 home-made catalyst between 170–330 °C and 0–20 bar_g. The "CH_4 purity", defined as the molar fraction of methane at the reactor outlet on a dry-basis, has been considered as key-performance-index (KPI).

The highest CH_4 purity has been obtained with the double bed configuration (Figure 2), in which the two catalytic beds in series were operated at different temperatures: the first at high temperature to favour the kinetics, while the second at milder conditions to exploit the high CO_2 conversion and CH_4 selectivity at thermodynamic equilibrium. The performances were improved even further by the removal of water between the two reactors. Notably, when working at 10 bar_g or higher, keeping the first reactor at 270 °C and the second at 210 °C, we succeeded in obtaining and outlet stream with a dry-composition (97.5 % CH_4, 0.1 % H_2 and 2.4 % CO_2) compatible with the direct injection in the gas network without any additional treatment.

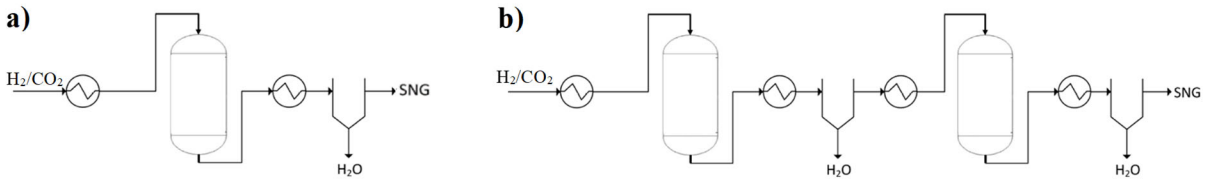

Figure 1: (a) Single bed configuration; **(b)** Double bed configuration with intermediate water removal.

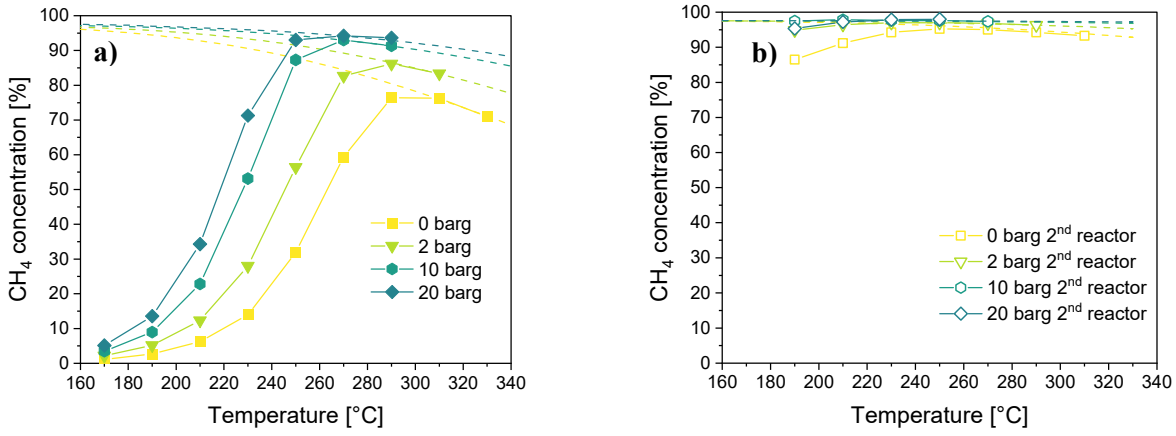

Figure 2. Methane purity at the outlet of **(a)** the first reactor and **(b)** of the second reactor in presence of intermediate water removal, at different pressures and temperatures.