ISCRE25 FLORENCE 2018 BRIDGING SCIENCE & TECHNOLOGY
The 25th International Symposium on Chemical Reaction Engineering
20-23 May 2018 Florence, Italy www.aidic.it/iscre25

Design of a Thermally Integrated Microreactor for Sabatier Reaction

Aswathy K. Raghu and Niket S. Kaisare*

Department of Chemical Engineering, Indian Institute of Technology Madras, Chennai 600036, India

**Corresponding author: nkaisare@iitm.ac.in*

Highlights

- Temperature profile in U-bend reactor is favorable for reversible exothermic reaction.
- CH_4 yield in U-bend for this case is 3.8 times higher than that in straight channel.
- Reasonable conversion even with inlet temperature as low as 300 K.

1. Introduction

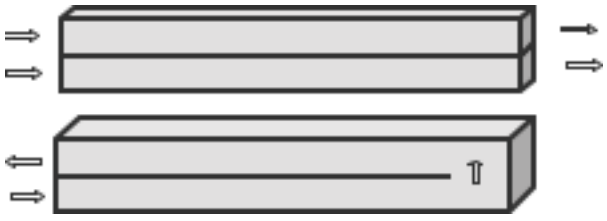
In recent years, global warming and climate change have closed in as prominent environmental issues that call for immediate resolution. Conversion of CO_2 to methanol or methane are being investigated as attractive options for recycling CO_2 to a fuel. The latter can be achieved by methanation or Sabatier reaction, given by the stoichiometric equation, $CO_2 + 4H_2 \Leftrightarrow CH_4 + 2H_2O$ $\Delta H = -165.0$ kJ/mol. Nowadays, methanation is gaining popularity as a promising strategy for CO_2 utilization for chemical storage of excess renewable energy by Power to Gas (PtG) concept[1]. PtG bases on the use of intermittent supply of electricity, generated from renewable sources of power, to electrolyze seawater and produce hydrogen which can be used for the methanation reaction.

Many of the reactor level studies on this reaction[2–4] have been conducted on externally cooled fixed bed reactors with emphasis on effective thermal management. Although higher temperatures are required for higher reaction rate, the reversible exothermic Sabatier reaction is thermodynamically favorable at lower temperatures. Such reactions are conducted with interstage cooling to approach equilibrium conversion.

This study is intended to explore the effect of heat recirculation in a microreactor for CO_2 methanation. Microreactors have the advantage of high surface area to volume ratio with enhanced heat and mass transfer rates. During heat recirculation, part of the reaction heat is utilized to heat up the reactants for accelerating the kinetics as in an autothermal operation[5]; heat transfer to the incoming cold fluid reduces the reactor exit temperature. In this study, we compare the performance of a microreactor when operated in two modes: As straight channel, and U-bend geometries to achieve favorable temperature profile through heat recirculation.

2. Methods

2D simulations are done in ANSYS Fluent 17.2 for a comparative study between a straight channel and a U-bend reactor (Figure 1). In the U-bend geometry, fluid takes a turn at the far end and the products exit through the outlet situated close to the inlet. In the straight channel reactor, there are two separate channels adjacent to each other, with reacting flow in a co-current heat exchange mode. The inlet and outlet of straight channel reactor are at the two extreme ends. Residence time for both the reactors are kept equivalent by introducing the same flowrate in each of them such that both the inlet velocity and reaction length in each straight channel is half of that in the U-bend.



Reactor dimensions	
Length	3 cm
Gap size	500 μm
Wall thickness	100 μm

Figure 1: Schematic of straight channels (top) and U-bend (bottom) for representational purpose. Arrows denote the flow direction.

Heat loss coefficient of 5 W/m^{2}K is used to account for heat loss by natural convection. Thermal conductivity of 1 W/mK (representative of ceramics) is used for all solid walls. Global reaction rate expression which also considers the reversibility of the reaction, fitted for Ru-TiO_2 catalyst, is adopted from Brooks et al.[6] and incorporated in Fluent using a user defined function (UDF). A parameter, F_{cat}, defined as the ratio of catalytic surface area to geometric surface area is employed to match the specific surface area in Brooks et al. Reactant mole ratio of 76:20 (H_2:CO_2) is given at the inlet with a volumetric flow rate 0.25 L/s. Argon is supplied as the inert. These conditions are similar to the ones in [6].

3. Results and Discussion

Figure 2 shows the axial variation of temperature and conversion along the centerline for both the setups for an inlet temperature of 300 K. The straight channel reactor shows low conversion because the reaction is kinetically limited due to the prevalence of lower temperatures. In the U-bend reactor, conversion is rapid at temperatures in between 600–700 K. Progress of forward reaction tends to fall at higher temperatures due to the thermodynamic limitation. The temperature begins to level off near the U-bend and the reaction rate falls as it approaches equilibrium. The fall in temperature towards the end is due to heat exchange with the cold reactants and it brings about a low temperature methanation that improves conversion.

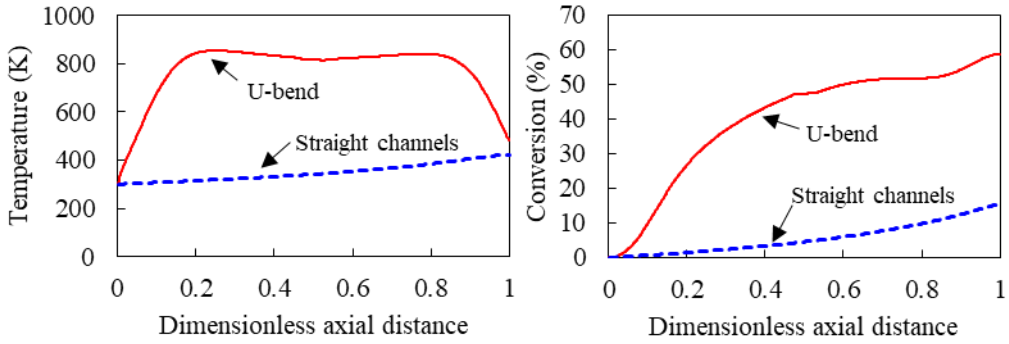


Figure 2: Temperature (left) and conversion (right) patterns for both reactors along the axis.

Thus, heat recirculation in U-bend enhances conversion by two simultaneous effects: (i) heating up of the cold reactants which helps cross the kinetic barrier, and (ii) cooling down of the hot products that helps overcome the thermodynamic limitation. Methane yield is 3.8 times higher in the U-bend (8.9E-5 kg/s of CO_2 at inlet is processed to 19.1E-6 kg/s of CH_4 in U-bend as against 5.0E-6 kg/s of CH_4 in straight channel). A thorough study of the role of various operating parameters, as well as the effect of additional reactions in the mechanism will be presented in the final paper.

4. Conclusions

The heat recirculation characteristics in U-bend reactor gives a temperature profile that results in a higher methane yield. Reasonable conversion was attained even with an inlet at room temperature. A more detailed parameter study will be conducted for further analysis.

References

(1) Gotz, M.; Lefebvre, J.; Mors, F.; McDaniel Koch, A.; Graf, F.; Bajohr, S.; Reimert, R.; Kolb, T. *Renew. Energy* **2016**, *85*, 1371–1390.
(2) Kiewidt, L.; Thöming, J. *Chem. Eng. Sci.* **2015**, *132*, 59–71.
(3) Ducamp, J.; Bengaouer, A.; Baurens, P. *Can. J. Chem. Eng.* **2016**, *9999* (November 2016), 1–12.
(4) Schlereth, D.; Hinrichsen, O. *Chem. Eng. Res. Des.* **2014**, *92* (4), 702–712.
(5) Kolios, G.; Frauhammer, J.; Eigenberger, G. *Chem. Eng. Sci.* **2000**, *55* (24), 5945–5967.
(6) Brooks, K. P.; Hu, J.; Zhu, H.; Kee, R. J. *Chem. Eng. Sci.* **2007**, *62* (4), 1161–1170.

Keywords
CO_2 Methanation; Sabatier Reaction; Heat Recirculating Reactor Design