Computational Fluid Dynamics based design of a novel reactor technology for the oxidative coupling of methane

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Highlights

- The gas-solid vortex reactor (GSVR) is proposed as a novel reactor technology for OCM.
- Computational fluid dynamic (CFD) simulations are needed to design/optimize the GSVR.
- It is possible to operate the GSVR quasi-isothermally.
- For the selected catalysts, C2 yields as high as 15-20% can be obtained in the GSVR.

1. Introduction

The decreasing crude oil reserves, the large amount of natural gas resources (shale gas) and the renewability of methane (biogas) have created a strong economic interest in developing processes that allow methane conversion into more valuable products [1]. One of the most promising direct routes to convert methane to ethylene and higher hydrocarbons is the oxidative coupling of methane (OCM). Ever since the pioneering work of Keller and Bhasin [2], OCM continues to attract both industrial as well as academic interest. Although the benefits of OCM have been known for over 30 years, a crucial issue among researchers still remains to find a viable catalyst with the performance needed for commercialization of the process [3]. Furthermore an appropriate reactor technology needs to be developed to address the low yields of ethylene and the strong exothermicity of the reaction. The main challenges to be overcome before OCM can be used as an alternative to steam cracking for the production of ethylene and higher olefins are hence twofold, with catalyst development on the one hand and novel reactor design on the other. In this work, the focus is on the latter. The gas-solid vortex reactor (GSVR), developed at the Laboratory for Chemical Technology (UGent) is proposed as an excellent reactor choice to demonstrate the OCM process.

In the GSVR, gas is injected tangentially via a number of inlet slots. The swirling gas then transfers its momentum to the particles, which start rotating as well. When the drag force exerted by the gas balances the apparent weight of the particles in the centrifugal force field, a fluidized state is obtained. In contrast to gravitational fluidized beds higher gas throughput, more uniform particle beds, higher slip velocities, and better heat and mass transfer can be achieved using centrifugal fluidization. The GSVR therefore emerges as an excellent candidate for process intensification [4, 5].

2. Methods

A schematic view of the GSVR setup is shown in Figure 1. It basically consists of a cylindrical unit positioned along a vertical axis with eight gas injection slots of 1 mm width, equally distributed over the circumferential wall and tangentially inclined at a 10° angle. A reactor diameter of 80 mm and length of 15 mm are defined. The design of this reactor is very flexible, meaning that the number of inlet slots, as well as their width and angle, can easily be adjusted.

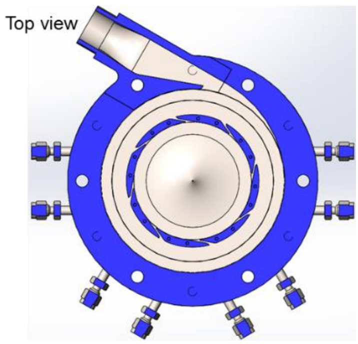

Figure 1: Schematic representation of the GSVR (top view).

The Eulerian-Eulerian approach is used to simulate the reactive two-phase flow, using the commercial CFD software package OpenFOAM. The most widely used kinetic model for OCM was used. This comprehensive 10 step kinetic model was originally developed for La_2O_3/CaO [6], but was later also fitted for other catalysts (e.g. Mn/Na_2WO_4/SiO_2 [7]). The GSVR is simulated as an adiabatic unit.

3. Results and discussion

Preliminary reactive simulations in the original eight slot geometry have shown that there is still a chance of hot spot formation when performing OCM in the GSVR. Furthermore, C2 yields were rather low for the selected catalyst and conditions. These observations initialized an optimization study, in which both the reactor geometry and operating conditions (flow rates, temperature, pressure, type of catalyst) were fine tuned in order to improve C2 yields and temperature uniformity in the bed. For example, it was found that increasing the number of inlet slots has a beneficial effect on the catalyst bed uniformity, which implies a better temperature uniformity and hence reduces the risk for hot spot formation.

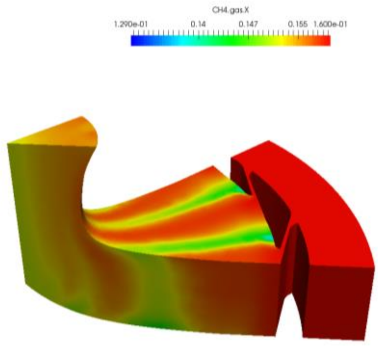

Figure 2: Methane mole fraction profile in a 16 slots GSVR geometry.

4. Conclusions

In this work a new reactor technology is proposed for the oxidative coupling of methane (OCM). Using the gas-solid vortex reactor (GSVR) it is possible to obtain higher gas throughput and better heat transfer, both of which are important to improve the industrial relevance of OCM. Because of the complex flow patterns inside the GSVR, simple 0D/1D reactor models are insufficient to estimate the reactor performance. Hence computational fluid dynamic simulations are necessary to optimize and design the GSVR specifically for OCM. Based on CFD simulations it was found that, for selected operating conditions and reactor design, OCM in the GSVR can be performed quasi-isothermally, and for the selected catalysts C2 yields as high as 15 – 20 % can be obtained.

Keywords
Computational Fluid Dynamics, oxidative coupling of methane, gas-solid vortex reactor

References

[1] E. V. Kondratenko, M. Schlüter, M. Baerns, D. Linke, and M. Holena, "Developing catalytic materials for the oxidative coupling of methane through statistical analysis of literature data," *Catal. Sci. Technol.,* vol. 5, pp. 1668-1677, 2015.
[2] G. E. Keller and M. M. Bhasin, "Synthesis of Ethylene via Oxidative Coupling of Methane," *Journal of Catalysis,* vol. 73, pp. 9-19, 1982.
[3] A. Galadima and O. Muraza, "Revisiting the oxidative coupling of methane to ethylene in the golden period of shale gas: A review," *Journal of Industrial and Engineering Chemistry,* vol. 37, pp. 1-13, 2016.
[4] J. De Wilde and A. de Broqueville, "Experimental investigation of a rotating fluidized bed in a static geometry," *Powder Technology,* vol. 183, pp. 426-435, 2008.
[5] K. Niyogi, M. M. Torregrosa, M. N. Pantzali, G. J. Heynderickx, and G. B. Marin, "Experimentally validated numerical study of gas-solid vortex unit hydrodynamics," *Powder Technology,* vol. 305, pp. 794-808, 2017.
[6] Z. Stansch, L. Mleczko, and M. Baerns, "Comprehensive Kinetics of Oxidative Coupling of Methane over the La2O3/CaO Catalyst," *Industrial & Engineering Chemistry Research,* vol. 36, pp. 2568-2579, 1997.
[7] M. Daneshpayeh, A. Khodadadi, N. Mostoufi, Y. Mortazavi, R. Sotudeh-Gharebagh, and A. Talebizadeh, "Kinetic modeling of oxidative coupling of methane over Mn/Na2WO4/SiO2 catalyst," *Fuel Processing Technology,* vol. 90, pp. 403-410, 2009.