MULTISCALE SIMULATIONS FOR COMBUSTION PYROLYSIS OF NATURAL GAS

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Abstract

This work reports multiscale modelling of combustion pyrolysis of natural gas. The approach incorporates wide range of simulation techniques including microkinetic to computational fluid dynamics (CFD) models to investigate critical design parameters. Microkinetic models estimated theoretical limit as high as 30% of olefin yield under the adiabatic conditions. Experiments at the midscale reactor, however, achieved around 21% of olefin yield. Hence, CFD models were used to investigate transport limitations and heat losses on reactor performance. The detailed analysis suggests that the effectiveness of mixing between cracking gas and hot combusted gas at the mixer along with heat losses play the most important roles in determining reactor performance. To extend our investigation to more detailed chemistries, that includes formation of aromatics, a reactor network model was also developed to account for mixing effects on product distribution. This multiscale computational approach enabled us to optimize and scale-up the next generation of natural gas cracker.

Keywords

Combustion pyrolysis, Acetylene, CFD, Kinetic model, Cantera, Mixing

Introduction

The growing abundance of natural gas resources and their competitive advantage with respect to other light paraffinic hydrocarbons make the production of chemical intermediates and other high value chemicals from natural gas an attractive option. One method to convert the natural gas resource to high value products is through pyrolysis. Pyrolysis involves partial oxidation of hydrocarbons to produce smaller unsaturated molecules that can be used as building blocks for commodity chemicals. In addition, pyrolysis can also be used to upgrade low value off gases/ feed streams to produce valuable products instead of using these streams as fuel or flared. Although pyrolysis technology is seven decades old, it is not widely practiced commercially due to several reasons including operational and scalability challenges. The homogeneous nature of the reaction without catalysts, simple reactor design and favorable economics could make this process attractive in the coming decades. For example, non-oxidative thermal pyrolysis of methane achieved high acetylene yield and another study demonstrated 80% methane conversion and 80% acetylene selectivity on an electrical heated tungsten wire (Porsin et al.).

BASF commercialized single-stage 50 kTA acetylene plants in 1950 (BASF; Bartholomé). Reactor accommodates both preheated natural gas and oxygen stream in a single-stage reactor and utilized heat generated by partial oxidation for thermal pyrolysis of natural gas. HOECHST has developed the concept of two-stage

combustion pyrolysis (Holmen et al.; HOECHST). It used an oxy-fuel combustor to generate heat in the first section, followed by a mixer and pyrolysis reactor for thermal cracking of hydrocarbons. Two-stage combustion pyrolysis can mitigate back mixing and give more flexibility in operation. However, the main disadvantage is the high severity of operation in terms of temperatures and consequent material issues and heat loss.

SABIC carried out several experiments in the two-stage combustion pyrolysis reactor. This reactor intended to demonstrate the effectiveness of the combustion pyrolysis concept at relatively larger scale.

The scope of this work is to explore critical design factors affecting overall reactor performance of two-stage combustion pyrolysis reactor. For that, a multiscale computational approach is adopted. Focus was to investigate the effect of fluid dynamics, heat transfer, and kinetics on the performance of pyrolysis of natural gas.

Several experiments were carried out to measure heat flux, pressure drop, temperature, composition, etc. and these data was used to validate microkinetic and Computational Fluid Dynamics (CFD) models.

Microkinetic models with Metcalfe et al.'s detailed reaction mechanism estimated theoretical reactor performance at different secondary-to-primary ratios (S/P) and O_2 stoichiometry (OS). In addition, CFD simulations were used to further investigate the effect of heat transfer and transport phenomena on overall reactor performance, and identified that lower degree of mixing is the major hindrance to reach theoretical performance.

Figure1 shows a comparison between experimental data and simulations. Cantera kinetic models overestimate carbon yield because of the assumption of perfect mixing (ideal reactor model). Considering transport limitations in CFD models shows better agreement with experimental data.

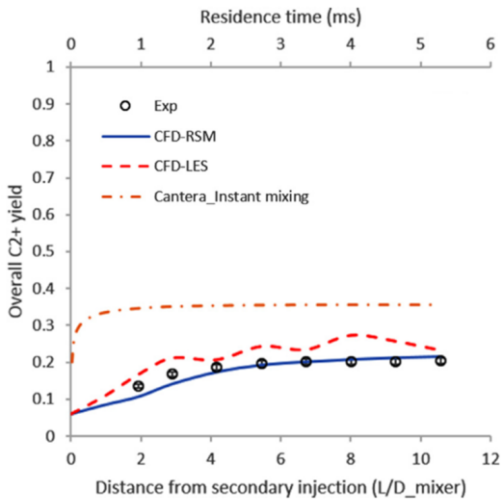

Figure1. A comparison between experiments and simulations (Lei et al.).

The computational expenses of CFD simulations, however, are the major hurdle to implement this technique over all interested reaction conditions. Therefore, a reactor-network model was further developed to accurately account for the effect of mixing on product distribution. This enabled us to explore broader ranges of design parameters with reasonable computational expenses while including very detailed chemistry models, such as Westbrook (Westbrook and Dryer), to estimate formation of aromatics with a variety of reactant feeds.

Conclusions

A multiscale computational approach for the combustion pyrolysis of natural gas enabled us to find the critical design parameters in this process. Comparing ideal reactor theoretical limit to CFD model concluded that mixing between hot combustor gas and feedstock gas plays an important role in determining reactor performance. Therefore, poor mixing presented in the experiments attributed to relatively lower reactor performance when compared with the ideal reactor models. To investigate all interested reaction conditions, the reactor network model was also implemented in this study. In addition, analyzing the problem at different simulation fidelity was successful in identifying the most critical design factors affecting reactor performance with optimal computational expenses.

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