Investigating the Effect of Dilution and Pressure on Methane Synthesis Using Metamodels

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Keywords: Methane synthesis, Metamodels, Gaussian Processes, Neural Network, Singular Value Decomposition
Working group: WG3: Hybrid physics-based data-driven models

1. INTRODUCTION

Methane synthesis reactors use green hydrogen from renewable sources, and carbon dioxide from industrial combustion processes to produce synthetic methane. Synthetic methane boasts a high heating value (~50 MJ/kg) making it a viable carbon-neutral energy carrier and a key component for green chemical production. These reactors operate within a temperature range of 300 – 350°C and pressure levels of 1 to 10 bar to activate the Ni/Al_2O_3 catalysts effectively.

For computer-based optimization of methane synthesis reactors, one-dimensional simulation tools are employed. These models integrate detailed surface chemistry to simulate external and internal diffusion processes within the porous structure, as well as adsorption and desorption processes at the surface. Additionally, the heat balance in these models accounts for exothermic and endothermic chemical reactions, heat conduction through solids, and energy transport due to fluid flow. Despite their low fidelity, computational times can span from minutes to hours for a single operating point due to inclusion of intricate chemical details.

This research advances the development of a metamodel framework [1] designed to predict the effects of dilution and pressure on methane synthesis. The metamodel framework is intended to be integrated into Power-to-X optimization and digital twin development efforts.

2. DATASET PREPARATION AND METAMODEL TRAINING

The dataset for training and validation of the metamodels is generated by running the reactor model for 5,000 Uniform Latin Hypercube designs. The input parameters varied include inlet mole fraction of CO_2, H_2 and N_2, inlet temperature, inlet pressure, inlet flow rate, reactor length and reaction progress variable c. The reaction progress variable is defined based on the outlet species mass fraction: $c = Y_{CH4,out} + Y_{H2O,out} + Y_{CO,out}$.

The reactor outlet species mole fractions of CH_4, CO, CO_2, H_2, H_2O, and temperature are then extracted according to reaction progress variable (ranging from 0 – 1 with a step size of 0.01) and reactor length (ranging from 0 – 0.1 m with a step size of 0.05 m). Consequently, the total dataset expands from 5,000 up to 220,720 points. To decrease the computational effort for metamodel training and validation, the total dataset is reduced using a uniform greedy algorithm, resulting in a reduced dataset containing 5,000 points.

For the training, a 6th order polynomial [2], a feedforward neural network [3] and Gaussian processes [4] are selected. One metamodel is trained for each outlet species and temperature, and the change rate of reaction progress variable dc/dt.

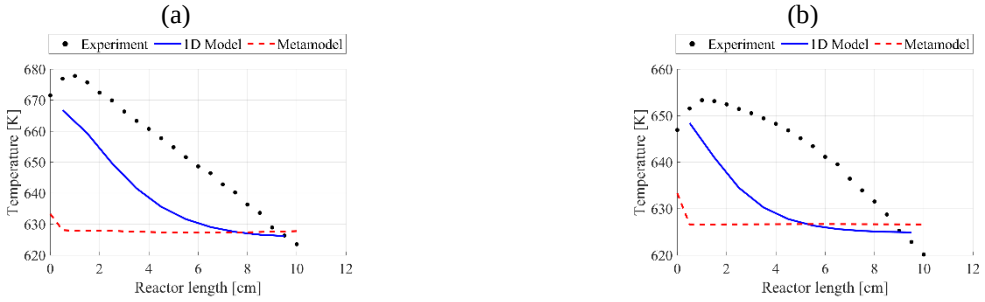

Figure 1: Temperature along reactor length for (a) No dilution and (b) 60% N_2 dilution at H_2:CO_2=4:1, inlet temperature of 623 K, and inlet pressure of 1bar.

3. RESULTS

3.1. PREDICTION OF TEMPERATURE ALONG REACTOR LENGTH

Figure 1 illustrates the reactor temperature along the reactor length as determined by experiments, 1D simulation and metamodel. For reactor temperature, the neural network is chosen due to its superior performance indicated by the lowest root mean squared error and the highest correlation coefficient (R^2). While the metamodel struggles to accurately predict the temperature profile along the reactor length, it successfully captures the effect of dilution on the temperature at the reactor outlet. The poor prediction accuracy for temperature profile may be attributed to the dataset reduction and the absence of a comprehensive quality check.

3.2. PREDICTION OF SPECIES MOLE FRACTION FOR DIFFERENT DILUTION RATE AND PRESSURE

Figure 2 compares the experimental data, 1D simulation results and metamodel predictions for the outlet mole fraction of CH_4 and H_2 under varying dilution rates and inlet pressures. For predicting CH_4 mole fraction the neural network is selected, whereas Gaussian processes are chosen to predict H_2 mole fraction. The neural network accurately predicts the decrease of CH_4 mole fraction with increasing N_2 dilution. This decrease exhibits a nonlinear trend, attributable to the reduction in reactor temperature and subsequent lower conversion rate. Gaussian processes capture the trend of H_2 mole fraction at high dilution rates, but their predictions for points without dilution are less accurate.

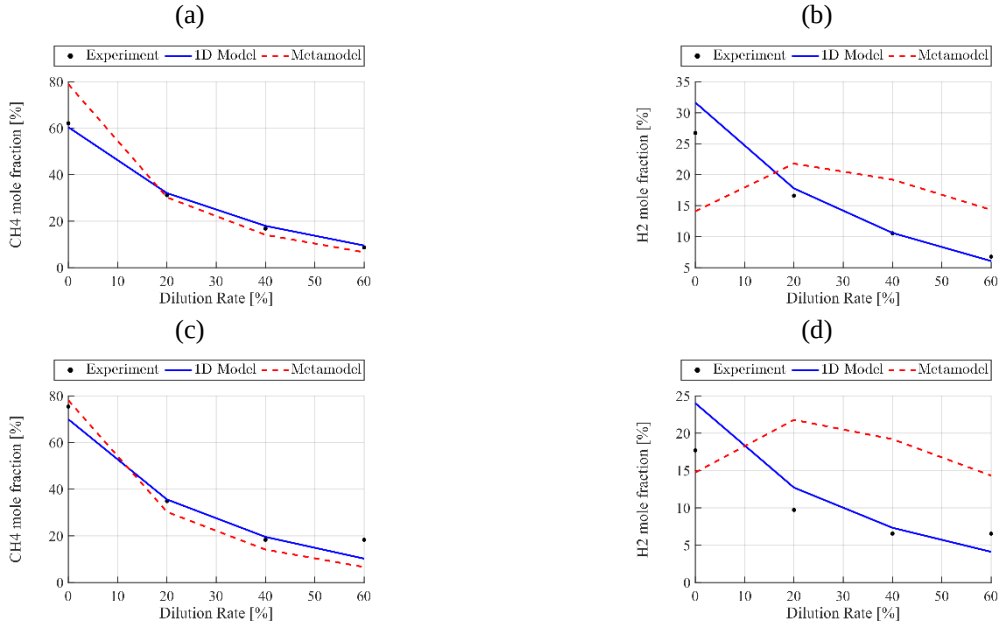

Figure 2: Species mole fraction at reactor outlet for different N_2 dilution rates. (a) CH_4 mole fraction at 1bar, (b) H_2 mole fraction at 1bar, (c) CH_4 mole fraction at 4bar, and (d) H_2 mole fraction at 4bar.

4. CONCLUSIONS AND RECOMMENDATIONS

The metamodel framework for methane synthesis reactors is extended by including the reactor length and N_2 dilution in the input parameters. Even though the metamodels well captured the reactor outlet temperature and species mole fraction, further enhancements are required for predicting the temperature profile along the reactor length. Enhancing the dataset's quality could potentially improve the metamodel's predictive performance.

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