Process optimization studies of dual-bed catalytic reactor system for conversion of methane to liquid hydrocarbons using design of experiments (DOE)

N. A. Saidina Amin*, Sriraj Ammasi

Chemical Engineering Department, Faculty of Chemical and Natural Resources Engineering, Universiti Teknologi Malaysia, 81310 Skudai, Johor, Malaysia

Abstract

A reactor system with two catalyst beds is proposed for converting methane directly to liquid hydrocarbons. In this dual-bed system, methane is converted in the first stage to oxidative coupling of methane reaction (OCM) products by selective catalytic oxidation with oxygen over a La supported on MgO catalyst. The second bed comprises of a nickel loaded HZSM-5 zeolite catalyst that has been tested as an oligomerization function to convert the OCM products mainly ethylene to liquid hydrocarbons. Thus, the research objective is to establish the factors that exert a greater impact on the dual-bed system and to determine the optimal reaction conditions to achieve the maximum yield of C_{5+}. Three independent factors (temperature (X_1), wt% of Ni nickel loading (X_2) and methane to oxygen ratio, CH_4/O_2 (X_3)) were investigated on the response, yield of C_{5+} hydrocarbons products (Y_{C5+}). These independent variables were coded at three levels and their actual values were selected on the basis of preliminary experimental results. The central composite design (CCD) coupled with response surface methodology (RSM) based on the design of experiments was successfully applied to map the response and to obtain the optimal reaction design. The experimental data was analyzed using an analysis of variance (ANOVA) to determine the significance of the factors tested. Based on the significance of each coefficient, the variable with the largest effect was the quadratic term of temperature, followed by the quadratic term of CH_4/O_2, linear term of Ni wt% and quadratic term of Ni wt%. Numerical results indicated that the optimum C_{5+} yield of 8.901% was attained at reaction temperature = 742 °C, Ni loading = 0.67 wt% and CH_4/O_2 ratio = 9.68. The

* Corresponding author. Tel: 607-5535588; Fax: 607-5581463 & Email: noraishah@fkkksa.utm.my

model had a satisfactory coefficient of R^2 (=0.903) and was verified experimentally. The actual experiment results were in agreement with the prediction. This verification confirmed the validity of the models built, thus indicating the suitability of the model employed and the success of RSM in optimizing the reaction parameters. Furthermore, this optimization strategy led to an increase in the yield of C_{5+} in the dual- bed system. This exploration also suggests that the concept of this dual catalyst bed system is an interesting candidate for application in methane utilization to produce liquid hydrocarbons.

Keywords: Dual-bed; Optimization; Liquid Hydrocarbons; DOE