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A hybrid numerical approach for multi-responses optimization of process parameters and catalyst compositions in CO_2 OCM process over CaO-MnO/CeO_2 catalyst

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Abstract

A new hybrid numerical approach, using Weighted Sum of Squared Objective Functions (WSSOF) algorithm, was developed for multi-responses optimization of carbon dioxide oxidative coupling of methane (CO_2 OCM). The optimization was aimed to obtain optimal process parameters and catalyst compositions with high catalytic performances. The hybrid numerical approach combined the single-response modeling and optimization using Response Surface Methodology (RSM) and WSSOF technique of multi-responses optimization. The hybrid algorithm resulted in Pareto-optimal solutions and an additional criterion was proposed over the solutions to obtain a final unique optimal solution. The simultaneous maximum responses of C_2 selectivity and yield were obtained at the corresponding optimal independent variables. The results of the multi-response optimization could be used to facilitate in recommending the suitable operating conditions and catalyst compositions for the CO_2 OCM process.

Keywords: Weighted Sum of Squared Objective Functions; CO_2 OCM process; Multi-responses optimization; Pareto-optimal solutions

1. Introduction

The high CO_2/CH_4 ratio in Natuna's natural gas compositions, comprising of up to 71% carbon dioxide and 28% methane [1], should be strategically utilized for the production of higher hydrocarbons, liquid fuels and other important chemicals. Recently, the conversion of methane to C_2 hydrocarbons (ethane and ethylene) using carbon dioxide as an oxidant (carbon dioxide oxidative coupling of methane (CO_2 OCM)) has received considerable attention [2–9]. Eqs. (1) and (2) are the two main CO_2 OCM reaction schemes to produce C_2 hydrocarbons, while carbon monoxide and water are the by-products.

$$2CH_4 + CO_2 \rightleftharpoons C_2H_6 + CO + H_2O \quad \Delta H^\circ_{298} = +106\,\text{kJ/mol} \tag{1}$$

$$2CH_4 + 2CO_2 \rightleftharpoons C_2H_4 + 2CO + 2H_2O \quad \Delta H^\circ_{298} = +284\,\text{kJ/mol} \tag{2}$$

Catalyst screening of CeO_2-based catalysts for CO_2 OCM process over binary and ternary metal oxides [9] determined that the 15 wt.% CaO-5 wt.% MnO/CeO_2 catalyst as the most potential. Interestingly, the stability test showed that the 15 wt.% CaO-5 wt.% MnO/CeO_2 catalyst was stable with no obvious coking during 20 h of reaction time on stream. However, the process parameters and the catalyst compositions of the CO_2 OCM process have not been optimized.

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