



Numerical study of methane steam reforming and methane combustion over the segmented and continuously coated layers of catalysts in a plate reactor



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ABSTRACT

Four separate 2D steady state numerical models are developed for a catalytic plate reactor (CPR), designed with the four different configurations between segmented and continuously coated layers of combustion and reforming catalysts for hydrogen production by combustion assisted methane steam reforming (MSR). MSR is simulated on one side of a plate by implementing experimentally validated surface microkinetic model for nickel/alumina catalyst. Required heat to an endothermic MSR is provided by simulating catalytic methane combustion (CMC) on an opposed-side of the plate by implementing reduced surface microkinetic model for platinum/alumina catalyst. Four different combinations of coating configurations between reforming and combustion catalysts are studied in terms of reaction heat flux and reactor plate temperature distributions as well as in terms of methane and hydrogen mole fraction distributions. These combinations are: (1) continuous combustion-catalyst and continuous reforming-catalyst (conventional CPR design), (2) continuous combustion-catalyst and segmented reforming-catalyst, (3) segmented combustion-catalyst and continuous reforming-catalyst, and (4) segmented combustion-catalyst and segmented reforming-catalyst. For the same reforming-side gas hourly space velocity, the study has shown that the CPR designed with the segmented catalysts requires 66% less combustion-catalyst to achieve similar methane conversion and hydrogen yield in MSR compared to the conventional CPR design. The study has also shown that maximum reactor plate temperature, thermal hot spots and axial thermal-gradients are reduced significantly in the CPR designed with the segmented catalysts than the CPR designed with the conventional continuous catalysts configuration.

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1. Introduction

Fuel cells are expected to have extensive applications as an alternative clean power source from stationary use to distributed to portable use. Due to an increase in global energy demand and as a result increase in environmental pollution, it is also expected that the demand of alternative clean power sources such as fuel cells will continue to rise. Consequently, the demand for hydrogen, the most important fuel for fuel cells and now emerging as universal energy carrier with energy security, will also continue to rise [1–3]. Though hydrogen is considered as the most abundant fuel in the universe, pure hydrogen exists in very limited quantity on our planet. Therefore, to fulfill the demand of hydrogen, various methods are developed to produce hydrogen from its primary sources. Two methods are most commonly in use for the production of hydrogen: reformation and electrolysis. Reformation involves hydrogen separation from the water and carbon compounds such as methane, whereas electrolysis separates hydrogen from oxygen in water [3]. Fuel cells combined with electrolysis is not a viable solution for electricity

generation, considering the fact that electrolysis requires electricity to produce hydrogen. Hence, to increase the wide spread use of fuel cells, onsite production of hydrogen via methane reformation is the most cost effective solution, especially after the recent discoveries of vast reserves of shale gas [4]. In reformation, steam reforming (SR) of hydrocarbons provides the maximum hydrogen concentration compared to other processes such as catalytic partial oxidation (CPOX) and autothermal reforming (ATR) [5]. However, SR is highly endothermic process and requires an effective way of supplying heat in a reactor. Mostly, SR is carried out in a fixed-bed catalytic reactor with high residence time [6]. But the fixed-bed design suffers from limitations such as heat-transfer and internal diffusion, which has led to the study of alternate reactor design such as catalytic plate reactor (CPR).

A CPR design provides excellent heat and mass transfer characteristics compared to the conventional fixed-bed design. It allows to carry out highly endothermic reactions like methane steam reforming (MSR) and exothermic reactions like catalytic methane combustion (CMC) side by side in the separate flow channels [7]. The close proximity of exothermic heat source with endothermic reaction sites, reduces significantly the overall heat transfer resistance [8]. Also, the use of catalytic combustion to supply heat offers advantages over gas-

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phase combustion. Catalytic combustion takes place at a lower temperature than the gas-phase combustion, which reduces NO_x formation and the lower operating temperature allows more material choices for designing a CPR [9]. However, the use of catalytic combustion in a CPR creates localized thermal-gradients (hot-spot) especially near the inlets due to the imbalance between generated and absorbed heat in combustion and reforming reaction zones, respectively [10]. Such localized thermal imbalance causes problems of material failure and also catalyst delamination due to different thermal expansion coefficients of the coated catalyst and the metal plate. Large thermal-gradient also reduces the catalytic active surface area, and as a result conversion rate of reactants [11]. To overcome these issues, recently Pattison et al. [12] and Jeon et al. [13] have proposed a segmented layers of combustion-catalyst (SLCC) for CMC coupled with MSR in a microchannel CPR. Pattison et al. [12] have explored numerically, a method for emulating distributed feed configuration in a microchannel CPR via SLCC macromorphology, consisting of alternating active and catalytically inactive sections. They have employed LHHW (Langmuir-Hinshelwood-Hougen-Watson) type Xu and Froment's [14] global kinetic model for MSR on nickel catalyst and power law type global rate expressions for both gas-phase methane combustion and CMC on platinum catalyst. Similarly, Jeon et al. [13] have proposed a stripe configuration for combustion-catalyst to minimize the formation of hot-spots in a microchannel CPR. They have considered nearly 50% combustion-side plate section coated with SLCC and the remaining 50% with the continuous layer of combustion-catalyst. They have also employed LHHW type Xu and Froment's global kinetic model for MSR on nickel catalyst and power law type global rate expression for CMC on palladium catalyst. Both studies have shown disappearance of hot-spots without any loss of methane conversion in MSR. Recently Settar et al. [15] have proposed a segmented reforming-catalyst layers' pattern for MSR with non-reactive metal foam to improve the mixing and hence methane conversion for a micro combined heat and power (CHP) system. With the segmented reforming-catalyst layers' pattern, they have predicted faster methane conversion compared to the conventional continuously coated reforming-catalyst. Use of non-reactive metal foam has improved mixing of reactants and hence better heat-transfer is achieved and resulted into increased hydrogen production. Settar et al. [16] have also carried out a computational comparative study between the segmented and conventional continuous reforming-catalyst layers. Their numerical analysis has shown performance enhancement factor up to two in terms of methane conversion with the segmented reforming-catalyst layers for MSR. However, no experimental evidence has been reported to support their finding. Mundhwa et al. [17] have investigated the internal diffusion limitation of continuously coated reforming-catalyst by evaluating the effectiveness factors for the steam reforming of diesel surrogate as a function of the CPR length. The study has found that independent of the type of catalysts, the initial 20% of the CPR length has shown high diffusion limitations. Therefore, the use of graded or segmented reforming-catalyst can be an advantageous in optimizing the amount of reforming-catalyst for its maximum utilization.

Many numerical studies of MSR coupled with CMC in a CPR have been carried out for various applications. However, except Pattison et al. [12], Jeon et al. [13] and Settar et al. [15–16], all literature reviewed in this study, have investigated MSR coupled with catalytic combustion of hydrocarbons over conventional continuously coated layers of reforming and combustion catalysts.

Zanfir and Gavriilidis [18] have carried out a sensitivity analysis of several design and operating parameters including reaction kinetic parameters. They have demonstrated that different catalysts can show similar thermal behaviour and performance but exhibit different sensitivity behaviour. Major finding of their study is the strongest influence on the reactor sensitivity comes from the reaction activation energies. Zanfir and Gavriilidis [9] have also performed parametric investigation of reforming-catalyst thickness and flow-channels height of a CPR for operating conditions similar to conventional industrial methane

reformer. They have developed a simplified 2D model of a CPR by implementing Xu and Froment's global kinetic model for MSR and power law rate expression for CMC. Zanfir and Gavriilidis have observed that by increasing reforming-channel height at constant inlet velocity, methane conversion decreases. They have concluded that MSR is feasible, provided that flowrates, catalyst thickness and channel heights are properly designed. In a separate study, Zanfir and Gavriilidis [19] have conducted numerical study of co-flow and counter-flow modes between MSR and CMC. They have determined higher methane conversion on the reforming-side with counter-flow than co-flow design. However, they have observed the thermal hot-spots in counter-flow configuration and suggested to optimize catalyst distribution on the combustion-side to reduce them.

Arzamendi et al. [20] have developed a 3D computational fluid dynamic (CFD) model of a microchannel reactor integrating MSR and CMC. They have implemented simplified rate expressions [21] for MSR and CMC. They have investigated the influence of catalyst loading, flow-modes between the two channels, and gas hourly space velocities (GHSVs). Their results have shown the possibility of 96% of methane conversion with 2–4 mg/cm^2 of catalyst loading at 930–1000 °C in the range of 10,000–30,000 h^{-1} reforming-side GHSV_{ref} . In a separate work, Arzamendi et al. [22] have developed 3D CFD models of a plate reactor with square microchannel and microslits designs to investigate the effects of characteristic dimension with different aspect ratios. Their results have shown that methane conversion decreases with increase in characteristic dimension, however microchannel design has provided methane conversion slightly higher than that of the microslits design. Stefanidis and Vlachos [23] have studied MSR on a rhodium catalyst coupled with propane combustion over a platinum catalyst in a CPR and have reported that increasing catalyst loading and decreasing possible internal mass transfer limitations results in considerable process time reduction. Also by lowering steam to carbon (SC) ratio yielded higher power output at relatively low reactor temperatures. In an another study, Stefanidis et al. [24] have reported that the use of low thermal conductivity plate materials increases fuel conversion and power output in the incomplete conversion regime. However, the use of very low thermal conductivity materials has shown high thermal-gradients in a CPR and thus recommended to use intermediate thermal conductivity materials, such as stainless steel to trade-off between thermal-gradients and conversion. Zhai et al. [25] have developed 2D computational fluid dynamic (CFD) model of a CPR using surface microkinetic for MSR on rhodium and LHHW type global kinetic model for CMC on platinum. They have investigated the influence of wall thermal conductivity, ratio of combustion to reforming feed, channel size and space time. They have reported that the performance of the metallic wall is superior against ceramic wall and also recommended to use the wall material with intermediate thermal conductivity to avoid the thermal hot-spots formation.

This work presents the four separate 2D steady state numerical models of a CPR to study the influence of segmented layers of reforming and combustion catalysts on composition and temperature distributions in a CPR compared to the continuous catalysts layers. In the first model, abbreviated as CCCR, reacting flows of CMC and MSR are simulated over conventional continuously coated layers of combustion and reforming catalysts, respectively. In the second model, abbreviated as CCSR, CMC is simulated over continuously coated layer of the combustion-catalyst but MSR is simulated over segmented layer of the reforming-catalyst. In the third model, abbreviated as SCCR, CMC is simulated over the segmented combustion-catalyst, whereas MSR is simulated over the continuous reforming-catalyst. In the fourth model, abbreviated as SCSR, both CMC and MSR are simulated over the segmented layers of combustion and reforming catalysts, respectively. For this study, methane flow-rate of 9.94 mol/h at the inlet of the reforming-side is determined based on the required inlet hydrogen flow-rate of 29.80 mol/h in a 1 kW fuel-cell, consists of 158 cells with 0.7 cell voltage and 90% hydrogen utilization on the anode-side and

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