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A comparative parametric study of a catalytic plate methane reformer coated with segmented and continuous layers of combustion catalyst for hydrogen production

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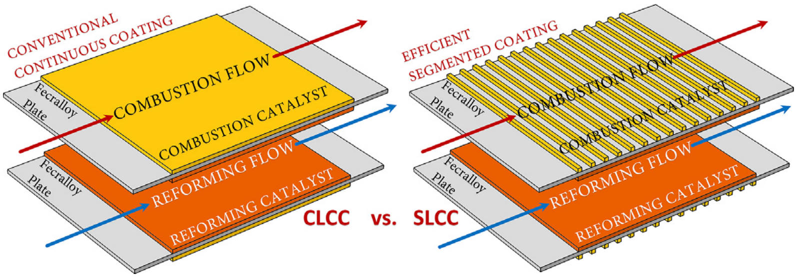
HIGHLIGHTS

- Proposed a segmented layer of combustion-catalyst (SLCC) design to produce H_2 via MSR.
- Parametric comparison between SLCC and continuous layer of combustion-catalyst (CLCC).
- H_2 production in MSR is increased when SLCC configuration is used compared to CLCC.
- SLCC requires 8% less combustion-fuel and 70% less combustion-catalyst than CLCC.
- Reactor plate temperature and axial thermal gradients are reduced with SLCC design.

GRAPHICAL ABSTRACT



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ABSTRACT

A parametric comparison study is carried out between segmented and conventional continuous layer configurations of the coated combustion-catalyst to investigate their influence on the performance of methane steam reforming (MSR) for hydrogen production in a catalytic plate reactor (CPR). MSR is simulated on one side of a thin plate over a continuous layer of nickel-alumina catalyst by implementing an experimentally validated surface microkinetic model. Required thermal energy for the MSR reaction is supplied by simulating catalytic methane combustion (CMC) on the opposite side of the plate over segmented and continuous layer of a platinum-alumina catalyst by implementing power law rate model. The simulation results of both coating configurations of the combustion-catalyst are compared using the following parameters: (1) co-flow and counter-flow modes between CMC and MSR, (2) gas hourly space velocity and (3) reforming-catalyst thickness. The study explains why CPR designed with the segmented combustion-catalyst and co-flow mode shows superior performance not only in terms of high hydrogen production but also in terms of minimizing the maximum reactor plate temperature and thermal hot-spots. The study shows that the segmented coating requires 7% to 8% less combustion-side feed flow and 70% less combustion-catalyst to produce the required flow of hydrogen (29.80 mol/h) on the reforming-side to feed a 1 kW fuel-cell compared to the conventional continuous coating of the combustion-catalyst.

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1. Introduction

The production of electricity using small scale internal combustion engine (1–10 kW) has low efficiency of about 15% to 25% [1]. Reducing the environmental impact by minimizing the emission of greenhouse gases from such low efficiency systems is of particular concern in many parts of the world, where electricity generators running on natural-gas/diesel/kerosene are used as power backup for small businesses and residences, especially in all developing countries [2]. Apart from greenhouse gases, conventional generators also emit NO_x and particulates that have significant impact on health [3]. By comparison, fuel-cell based electricity generators produce no particulates and no NO_x due to the absence of high temperature gas-phase combustion. Unlike electric batteries, a fuel-cell does not run down or require recharging; it produces continuous electricity as long as hydrogen and an oxidizer are supplied continuously to the cell. At present, there is no hydrogen infrastructure to employ wide spread use of fuel-cell generators. An alternative solution is to develop an efficient and compact fuel reformer to produce hydrogen, which can take advantage of the natural-gas (95% methane) supply infrastructure in many countries. Compact reformers can be integrated easily with fuel cell generators which are deployed for stationary and portable use.

A fuel reformer can be built in a variety of configurations depending on the desired system efficiency, dynamic response and ease of manufacture. Three possible reformer designs to produce hydrogen from methane are: (1) Steam Reforming (SR), (2) Autothermal Reforming (ATR) and (3) Partial Oxidation (POX). SR is a highly endothermic reaction, whereas POX is exothermic. ATR is a combination of SR and POX and can offer the advantage of thermoneutral operation. SR provides the maximum hydrogen concentration as compared to ATR and POX [4], however, due to its highly endothermic nature, SR requires an effective way of supplying heat to the reaction sites. One efficient solution is to supply the heat indirectly by means of a catalytic combustion of methane.

Indirect supply of heat from catalytic combustion of methane can be achieved effectively in a catalytic plate reactor (CPR). A CPR design consists of number of thin metal plates, each having both sides coated with appropriate catalysts for the endothermic and exothermic reactions and are arranged in a stacked configuration [5]. The close association of the exothermic and endothermic reaction zones reduces significantly the overall heat transfer resistance [6]. The use of catalytic combustion to supply heat offers advantages over gas-phase combustion. Catalytic combustion takes place at a lower temperature than gas-phase combustion which reduces NO_x formation, and the lower operating temperature allows more material choices in designing the plate reactors [7]. However, the use of catalytic combustion in a CPR creates localized thermal-gradients or hot-spots especially near the inlets due to the imbalance between the rate of heat generation in combustion and the rate of heat absorption in reforming [8]. Such localized thermal imbalance causes problems of material failure and catalyst delamination due to different thermal expansion coefficients of the coated catalyst and the metal plate. Large thermal-gradients also reduce catalytic active surface area as well as catalyst support area, and thus reactants conversion rates [9]. To overcome these issues, recently Pattison et al. [10] and Jeon et al. [11] have proposed a segmented layer of combustion-catalyst (SLCC) configuration for catalytic methane combustion (CMC) coupled with methane steam reforming (MSR) in a microchannel CPR. Pattison et al. [10] have explored numerically, a method for emulating distributed feed configuration in the 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 [12] kinetic model for MSR and power law rate models for gas-phase and catalytic methane combustion. Similarly, Jeon et al. [11] have proposed a stripe configuration for combustion-catalyst to minimize the formation of hot-spots in a microchannel CPR. They have considered nearly half the combustion-side plate section coated with SLCC and the remaining half coated with a continuous layer of combustion-catalyst (CLCC) configuration. They have also employed Xu and Froment's [12] kinetic model for MSR and power law rate model for CMC. Both studies have shown disappearance of hot-spots without any loss of methane conversion (X_{CH4}) in reforming. More recently, in one of our studies [13], we have proposed a segmented combustion-catalyst and segmented reforming-catalyst configurations over the entire length of the plate by employing one-step reduced microkinetic model for CMC and multi-step microkinetic model for MSR. The study has illustrated that apart from reducing hot-spots, segmented catalyst configuration has improved effective thermal conductivity of both reforming and combustion catalysts and decreased the back diffusion of hydrogen in the reforming-channel for the SLCC configuration compared to the CLCC configuration.

Many numerical studies of CMC coupled with MSR in a CPR have been carried out for various applications. However, except Pattison et al. [10], Jeon et al. [11] and Mundhwa and Thurgood [13], none of the literature reviewed in this study have investigated MSR coupled with CMC over SLCC. None of the studies in our knowledge have investigated the influence of reactor parameters on MSR coupled with CMC over SLCC in a CPR and its comparative study with the CPR coated with CLCC.

Zanfir and Gavriilidis [14] have performed a parametric investigation of reforming-catalyst thickness and flow-channel 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 [12] kinetic model for MSR and power law rate model for CMC. Zanfir and Gavriilidis [14] have observed that by increasing reforming-channel height at constant inlet velocity, X_{CH4} decreases. They have concluded that MSR coupled with CMC is feasible in a CPR if flow-rates, catalyst thickness and channel heights are properly designed. Zanfir and Gavriilidis [15] have also conducted numerical study of co-flow and counter-flow modes between MSR and CMC. They have determined higher X_{CH4} on the reforming-side with counter-flow than co-flow design. They have also observed thermal hot-spots in counter-flow mode and have suggested to optimize combustion-catalyst distribution to reduce thermal hot-spots. In a separate study, Zanfir and Gavriilidis [16] 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 behavior and performance but exhibit different sensitivity behavior. The major finding of their study is that the strongest influence on reactor sensitivity comes from the reaction activation energies. Stefanidis and Vlachos [17] 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 a different study, Stefanidis et al. [18] 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 the CPR and thus recommended to use intermediate thermal conductivity materials, such as stainless steel as a trade-off between thermal-gradients and conversion. Zhai et al. [19] have developed 2D computational fluid dynamic (CFD) model of a CPR using surface microkinetics for MSR on rhodium and