ENHANCING STABILITY IN PARALLEL PLATE MICROREACTOR STACKS FOR SYNGAS PRODUCTION

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Summary

This paper develops the scale-out principles of microdevices for portable and distributed syngas production using computational fluid dynamics simulations with fundamental multiscale model-based rate expressions. It is found that small stacks fail under experimental conditions. Several methods for improving stability, such as combusting hydrogen in select channels, are evaluated. To simulate fuel-lean hydrogen combustion in a computationally efficient manner, a single step rate expression for hydrogen combustion on platinum is derived from a previously published microkinetic model. The most effective means for improving stability is found to be combusting hydrogen or increasing the platinum catalyst loading in the outermost combustion channels.

Keywords

Micro-reactors, Process Intensification, Hydrogen Production, Syngas, Scale-out, Methane, Platinum, Rhodium.

Abstract

Introduction

Parallel plate microreactor stacks are an attractive means to intensify exothermic and endothermic process elements as sub-millimeter gap sizes and wall thicknesses enhance heat transfer. Higher transport rates make microreactors a promising alternative to conventional syngas production processes, where reaction rates are limited by heat transfer to the catalyst bed[1]. Parallel plate microreactors have been investigated for this purpose experimentally[2, 3] and numerically[4-6]. It is generally believed that results from a small number of channels can be used to linearly scale-out stacks to meet application-scale production rates[2-6]. This design principle implicitly assumes that heat losses from stack edges affect all channels equally and their role is negligible. In our recent work, it was shown that edge heat losses cause nonlinearities during scale-out of methane steam reforming (SR) microreactor stacks[7]. It was found that methane fueled microreactor stacks for syngas production are highly unstable due to external heat losses. Based on this information, methods for improving stability are investigated in this work.

Methodology

Our stack design consists of alternating methane combustion and SR channels. Two dimensional microreactor stacks are simulated using the FLUENT® computational fluid dynamics (CFD) software. Reaction kinetics for methane combustion on Pt and methane SR and water-gas-shift on Rh are taken from previously published reduced expressions based on microkinetic modeling[8, 9].

Stability improvement techniques are based on two approaches: (1) increase stack temperatures by manipulating inlet streams and (2) improve combustion conditions by modifying stack design.

The first option sacrifices efficiency or throughput by increasing inlet combustion velocities or decreasing inlet SR flow rates, respectively. Additionally, inlet temperatures can be increased to boost energy input and approximate the effect of heat recirculation. The net effect of these options is that reactor temperatures, and thus reaction rates, are higher, thus improving stability to external heat loss.

The second approach modifies design to improve stability. Options such as different reactor sizes, reducing wall thermal conductivity, increasing the Pt catalyst loading and burning hydrogen rather than methane in the outermost channels are investigated. To model hydrogen combustion on Pt in a computationally efficient manner, a published microkinetic model[10] is reduced using reaction path analysis, sensitivity analysis and partial equilibrium analysis to produce a single step rate expression.

Results

Figure 1 shows that smaller stacks constructed of moderate wall conductivity materials (e.g., stainless steel) are not stable under experimental heat loss.

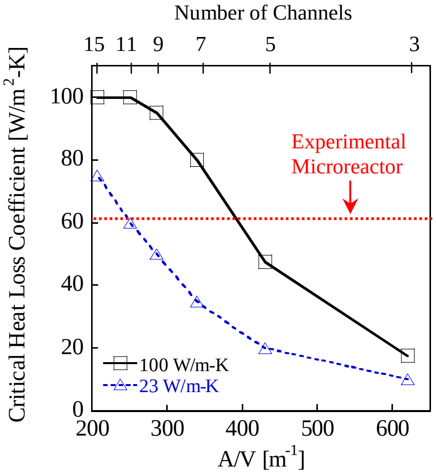

Figure 1. Stability of methane-to-syngas stacks as a function of stack size for high (100 W/m-K) and moderate (23 W/m-K) wall thermal conductivity. Experimental value is calculated from[11].

Failure is a result of low temperatures, and therefore reduced methane conversion, in the outer channels (**Figure 2**). Low conversion causes the outer combustion channel to fail and ultimately leads to total stack failure. Based on this information, methods for improving stability are investigated in this work.

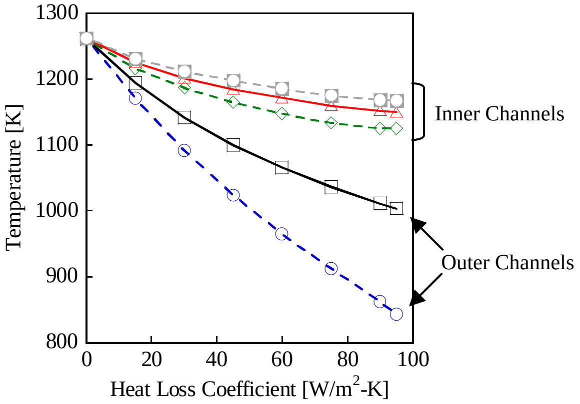

Figure 2. Exit temperatures as a function of heat loss coefficient for a nine channel reactor (9CR).

The mechanisms for enhancing stack stability for small sizes have been delineated. **Figure 3** indicates that the most effective means for improving stability is to increase the platinum catalyst loading or combust hydrogen in the outer channel.

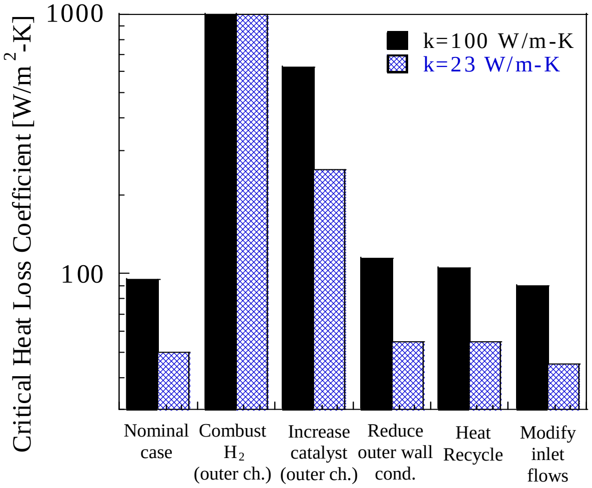

Figure 3. Methods for improving stability (in a 9CR).

Conclusions/Importance

The stability of microstacks is increased substantially for both high and moderate thermal conductivity wall materials by improving combustion in the outer channel of the stack using either an easy to activate fuel or a larger amount of catalyst. Our results provide guidelines for scale-out of microdevices at small sizes.

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