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Enhancing stability in parallel plate microreactor stacks for syngas production

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ABSTRACT

Recent work found that methane fueled small microreactor stacks for syngas production are unstable due to external heat losses primarily that cause the outmost combustion channels to fail. Methods for improving stability are investigated in this work using computational fluid dynamics (CFD) simulations of parallel plate microreactors consisting of alternating combustion and steam reforming channels with platinum and rhodium catalytic surfaces, respectively. To study the effect of combusting a more reactive fuel, 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 combusting hydrogen or increasing the platinum catalyst loading in the outmost combustion channels. These results should be applicable to other systems that may not be stable at small scales. Finally, several stability enhancement methods are contrasted in terms of throughput and efficiency.

1. Introduction

Parallel plate microreactor stacks are an attractive means to intensify exothermic and endothermic process elements by enhancing mass and heat transfer rates over conventional reforming processes, where reaction rates are limited by heat transfer to the catalyst bed (Rostrup-Nielsen, 1984). Parallel plate microreactors have been investigated, for this purpose, experimentally (Cremers et al., 2007; Tonkovich et al., 2007; Venkataraman et al., 2003) and numerically (Delsman et al., 2005; Kolios et al., 2005; Stefanidis and Vlachos, 2008; Stefanidis et al., 2009; Tonkovich et al., 2007; Zanfir and Gavriilidis, 2004). It is generally believed that results from a small number of channels can be used to linearly scale-out stacks to meet application-scale throughputs (Cremers et al., 2007; Glockler et al., 2004; Stefanidis and Vlachos, 2008; Venkataraman et al., 2003; Zanfir and Gavriilidis, 2004). This design principle tacitly ignores the finite size effects and heat losses.

In recent work, we have studied the effect of edge heat loss as a function of size for syngas production in small microreactor stacks (Mettler et al., 2010). Our stacks consist of alternating methane steam reforming (SR) and catalytic combustion (CC) microchannels. It was found that small stacks cannot function under typical laboratory heat loss conditions. Fig. 1 summarizes previous stability results and shows that small stacks with moderately conductive walls (corresponding to stainless steel) are unstable under laboratory heat loss conditions.

In order to improve stability, especially of small stacks for low throughput applications, several methods are explored that can lead to stability enhancement: (1) increase the net power input, (2) modify dimensions and wall materials, (3) increase catalyst loading, and (4) change combustion fuel in some of the channels. Stability enhancement methods are evaluated using computational fluid dynamics (CFD) simulations of a nine channel reactor (9CR).

2. Model

2.1. Nine channel reactor (9CR)

The parallel plate 9CR consists of alternating SR and CC channels, as shown in Fig. 2. The 9CR represents a middle ground in terms of stability, where high wall thermal conductivity stacks are stable under the laboratory heat loss conditions, while stacks with moderately conductive walls are not. The 9CR is preferred over smaller stacks, because inner and outmost combustion channels can be independently modified (in terms of flow rate, catalyst loading, etc.) to improve stability (in smaller stacks, this cannot be done as combustion channels are equidistant from edges, and thus respond identically to symmetric qualifier design changes). Simulations are conducted for a 9CR with highly conductive walls (100 Wm-K), which is characteristic of silicon carbide

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and low-alloy steels, and moderately conductive walls (23 W/m-K), which represents stainless steel (Green and Perry, 2007). In this work, SR channels are placed closest to edges in order to insulate combustion channels from heat losses. Different designs, such as having the outmost channels carrying out combustion rather than an SR, can also be considered. In the previous work, it was found that such design modifications do not significantly affect conclusions (Mettler et al., 2010), and thus the aforementioned design is considered here. Dimensions of the channels and walls are indicated in Fig. 2 and are the same as in the previous work (Mettler et al., 2010; Stefanidis and Vlachos, 2008). Inlet flow rates for combustion and steam reforming channels (shown in Table 1) are determined from infinite stack simulations and produce high SR throughput, while maintaining moderate wall temperatures. Throughput in outmost SR channels (closest to stack edges) is half that of interior SR channels, because outmost SR channels are adjacent to one combustion channel, whereas inner SR channels are sandwiched between two (thereby receiving twice as much heat). For inner SR channels (and all combustion channels), both walls are catalytic, while for outmost SR channels, only the lower wall (furthest from an edge heat loss) is catalytic. Placing catalyst in this manner keeps the contact time same for inner and outmost SR channels and also leads to an acceptable compromise in terms of (increasing) stability and (decreasing) hot spots (Mettler et al., 2010). Methane-fueled combustion channels operate with an inlet equivalence ratio of 0.92, while SR channels employ an inlet H_2O to CH_4 ratio of 2:1.

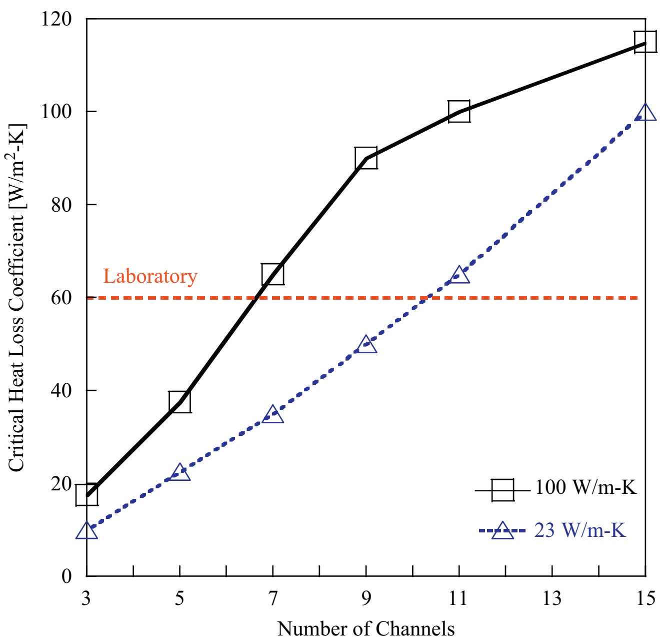

Fig. 1. Stability as a function of stack size for stacks with highly (100 W/m K) and moderately (23 W/m K) conductive walls. Redrawn from Mettler et al. (2010). The critical heat loss coefficient is the maximum h, where combustion within stacks is sustained.

2.2. Quantifying stability

The heat lost (Q) through stack edges is given by Newton's law of cooling

$$Q = h\int_A (T - T_a)dA \tag{1}$$

Here, T and T_a are wall and ambient temperatures, respectively, and h is the external heat loss coefficient. In our model, the latter parameter lumps convective and radiant heat loss from all exterior surfaces. A heat loss coefficient of zero corresponds to an adiabatic stack. The critical heat loss coefficient (h_c) is defined as the maximum possible heat loss coefficient, above which stacks are no longer autothermal. Our previous work has shown that high heat losses cause small stacks to fail, because of low temperatures and reduced reaction rates in the combustion channel nearest to the stack edge (Mettler et al., 2010). Single channel microburners (not stacks) fail as a result of either extinction or blowout. The demarcation between these mechanisms depends on operating conditions and device design (Federici and Vlachos, 2008; Kaisare and Vlachos, 2007; Norton and Vlachos, 2004). Our recent work with methane SR microreactor stacks revealed that the failure mechanism is more complicated and is size-dependent, where small stacks fail because of extinction and larger stacks, due to blowout under certain conditions (Mettler et al., 2010). The failure mechanism suggests combustion flow rates (for inner and outmost channels) can be modified to improve stability, as discussed below.

The relative stability improvement is defined as the ratio of critical heat loss coefficient (h_c) of improved over that of the nominal case.

$$\text{Relative stability improvement} = \frac{h_c(\text{improved})}{h_c(\text{nominal})} \tag{2}$$

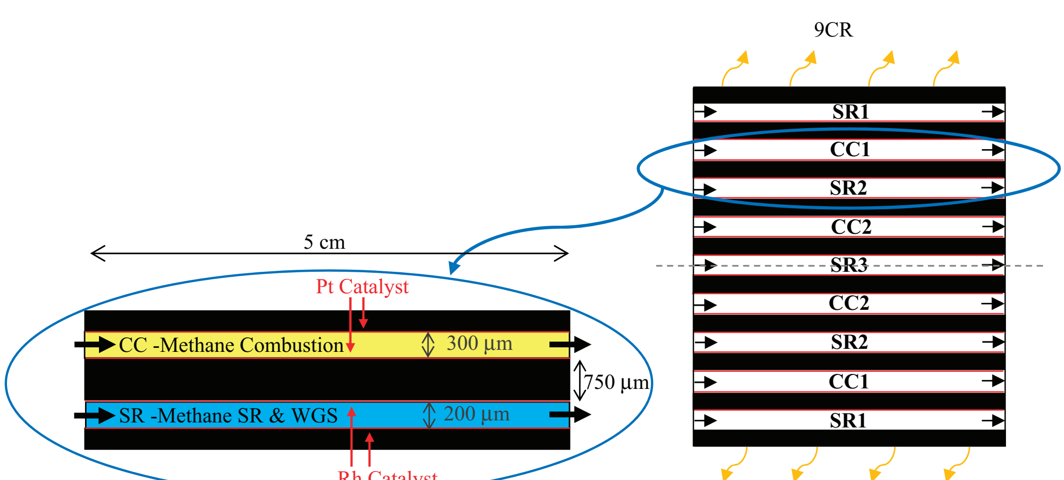

Fig. 2. Schematic representation of the 9CR. Red lines indicate catalytic surfaces in steam reforming (SR) and catalytic combustion (CC) channels. The dashed (gray) line represents a symmetry plane. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)