



# Robust autothermal microchannel reactors<sup>☆</sup>



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## ARTICLE INFO

### Article history:

Received 3 November 2014

Received in revised form 10 March 2015

Accepted 19 March 2015

Available online 30 March 2015

### Keywords:

Catalytic plate reactors

Microchannel reactors

Process intensification

Process control

## ABSTRACT

Autothermal microchannel reactors are intensified process units that bring significant energy efficiency benefits over their conventional counterparts. Efficiency gains are obtained, however, at the cost of operational challenges. These stem from the loss of control handles that is inherent to combining several unit operations in a single physical device. In this paper, we investigate the impact of two recently proposed reactor design concepts (a segmented catalyst macromorphology and an embedded layer of phase change material) aimed at improving the steady state energy distribution and, respectively, preventing the advent of hotspots during transient operation, on reactor dynamics and control. Using an autothermal microchannel reactor coupling steam methane reforming with methane catalytic combustion as a prototype system, we demonstrate through rigorous simulations that these design innovations have a synergistic effect, resulting in superior steady-state performance and excellent disturbance rejection ability.

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## 1. Introduction

Microchannel reactors are one of the most successful applications of the process intensification (PI) concept (Stankiewicz and Moulijn, 2000; Lerou et al., 2010). With channel dimensions in the millimeter range, microchannel reactors achieve intensification by greatly increasing surface-area-to-volume ratios; studies have reported that transport properties in such reactors are up to two orders of magnitude higher than in corresponding conventional equipment (Lerou et al., 2010; Murphy et al., 2013). By eliminating transport limitations, processes are driven by intrinsic properties (i.e., the reaction kinetics), and the dimensions of microchannel reactors are often one order of magnitude lower than those of conventional processes of equivalent capacity (Zanfir and Gavriilidis, 2003).

Microchannel reactors have found several practical applications; most notable industrial successes include the generation of syngas via steam methane reforming, followed by Fischer Tropsch reactions for production of higher chain hydrocarbons (Thomas and Dawe, 2003; Wood et al., 2008; Khalilpour and Karimi, 2012; Butcher et al., 2014).

While microchannel reactor designs provide significant capital savings and size reduction, several operational challenges arise.

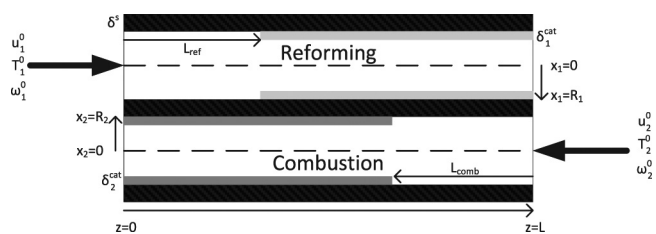
Most importantly, increasing the number of operations performed by a single physical device reduces the number of degrees of freedom available for control, compared to the equivalent conventional (non-intensified) process configuration. Further difficulties are related to geometric dimensions; the size reduction makes it infeasible to place measurement and actuation devices along the length of the reactors (Vaccaro et al., 2010; Vaccaro and Malangone, 2014), and smaller systems inherently have faster dynamics, making (feedback) control challenging (Nikačević et al., 2012).

The majority of literature studies have focused on the steady state designs of microchannel reactors, and on improving the temperature and conversion profiles of such devices. In steam methane reforming microchannel reactor applications, many designs feature autothermal coupling, i.e., the endothermic reforming reactions are supported by heat released in a separate, exothermic, reaction volume (Ramaswamy et al., 2006; Ramaswamy and Ramachandran, 2008), where the exothermic reaction is typically the catalytic combustion of methane. Autothermal reactor designs eliminate the need for an external heating medium, but make thermal management more challenging as the synchronization of heat flux along the reactor is difficult; the heat consumption in the reforming channels must match the heat released by the combustion channels. Improper alignment of heat fluxes can be detrimental to the performance of the system and can result in the formation of hot spots and steep temperature gradients that can damage the reactor structure (Zanfir and Gavriilidis, 2004; Stefanidis and Vlachos, 2009; Zanfir et al., 2011; Murphy et al., 2013; Granlund et al., 2014; Vaccaro and Malangone, 2014; Butcher et al., 2014). Several studies have attempted to address this problem by altering the design of the

<sup>☆</sup> A preliminary version of this work was presented at Foundations of Computer-Aided Process Design (FOCAPD) 2014, Cle Elum, WA, USA.

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**Fig. 1.** Two channels in the base case reactor structure. The catalytically active regions are offset optimally to synchronize heat generation and consumption. See Table 1 for the definitions and values of the variables.

reactor. Kolios et al. (2002) and Granlund et al. (2014) proposed distributed feed designs, whereby reactants are fed at multiple points along the reactor, and Kolios et al. (2005) suggested periodic switching of exothermic and endothermic reactions. Ramaswamy et al. (2006), Zanafir et al. (2011), and Jeon et al. (2014) showed that offsetting the catalytically active regions improves synchronization of endothermic and exothermic reactions by delaying the release and consumption of heat. Jeon et al. (2013) proposed a segmented catalyst concept for co-current flow designs in order to modulate heat generation axially, and Zanafir and Gavriilidis (2004) obtained similar results by varying the catalyst activity along the reactor, although the latter approach may be difficult to translate in practice. Stefanidis and Vlachos (2009) and Karakaya and Avci (2011) proposed improving the overlap of heat fluxes through proper selection of design and operating parameters (e.g., wall thickness, channel heights, catalyst selection, flowrates, etc.).

To date, very few literature studies have considered the transient operation of such microchannel reactors (Vaccaro and Malangone, 2014); the majority of these focused on the start-up behavior (Robbins et al., 2003; Jeon et al., 2014; Vaccaro et al., 2010). There is also very little information on the behavior of microchannel reactors subject to operational disturbances (Baldea and Daoutidis, 2007; Baldea et al., 2009; Pattison and Baldea, 2013). Motivated by the above, in this paper, we focus on achieving resilient (in a similar sense as defined, e.g., by Lenhoff and Morari (1982)) reactor performance. We investigate the impact of two recently proposed reactor design concepts (a segmented catalyst macromorphology (Pattison et al., 2013) and an embedded layer of phase change material (Pattison and Baldea, 2013)) aimed at improving the steady state energy distribution and, respectively, preventing the advent of hotspots during transient operation, on reactor dynamics and control (Pattison and Baldea (2014)). Using an autothermal microchannel reactor coupling steam methane reforming and methane combustion as a prototype system, we demonstrate through rigorous simulations that these design innovations have a synergistic effect, resulting in superior steady-state performance and excellent disturbance rejection ability.

## 2. Prototype system

We consider the case of a catalytic plate microchannel reactor with alternating channels of reforming reactions and catalytic combustion that is shown in Fig. 1 with parameters given in Table 1. The catalytically active regions in the channels are offset in order to optimally synchronize the rates of heat generation and consumption and maximize conversion (Zanafir et al., 2011).

The following reactions occur in the reforming channels:

- Methane steam reforming (1)
 
$$\text{CH}_4 + \text{H}_2\text{O} \rightleftharpoons \text{CO} + 3\text{H}_2$$

$$\Delta H = +206 \text{ kJ mol}^{-1}$$

**Table 1**  
Nominal reactor system parameters (Zanafir et al., 2011; Pattison and Baldea, 2013; Pattison et al., 2013).

Parameter	Variable	Value
Reactor Length	$L$	63.4 cm
Reforming Channel Half-Height	$R_1$	1.0 mm
Combustion Channel Half-Height	$R_2$	1.0 mm
Plate Thickness	$\delta^s$	0.5 mm
Reforming Catalyst Height	$\delta_1^{\text{cat}}$	20 $\mu\text{m}$
Combustion Catalyst Height	$\delta_2^{\text{cat}}$	20 $\mu\text{m}$
Reforming Inlet Temperature	$T_1^0$	793.15 K
Combustion Inlet Temperature	$T_2^0$	793.15 K
Reforming Inlet Velocity	$u_1^0$	4.0 m/s
Combustion Inlet Velocity	$u_2^0$	3.2 m/s
Reforming Catalyst Offset	$L_{\text{ref}}$	26.8 cm
Combustion Catalyst Offset	$L_{\text{comb}}$	27.3 cm
Reforming Inlet Composition (Mass Fraction)	$\omega_1^0$	19.11% CH <sub>4</sub> 72.18% H <sub>2</sub> O 2.94% CO <sub>2</sub> 0.29% H <sub>2</sub> 5.48% N <sub>2</sub>
Combustion Inlet Composition (Mass Fraction)	$\omega_2^0$	5.26% CH <sub>4</sub> 22.09% O <sub>2</sub> 72.65% N <sub>2</sub>

- Water-gas-shift (2)
 
$$\text{CO} + \text{H}_2\text{O} \rightleftharpoons \text{CO}_2 + \text{H}_2$$

$$\Delta H = -41 \text{ kJ mol}^{-1}$$
- Reverse methanation (3)
 
$$\text{CH}_4 + 2\text{H}_2\text{O} \rightleftharpoons \text{CO}_2 + 4\text{H}_2$$

$$\Delta H = +165 \text{ kJ mol}^{-1}$$

The reaction kinetics for a 15.2% Ni/MgAl<sub>2</sub>O<sub>3</sub> catalyst are described using the Langmuir–Hinshelwood–Hougen–Watson kinetics developed by Xu and Froment (1989). The catalytic combustion of methane occurs in half the channels operating in countercurrent to the reforming channels.

- Methane combustion (4)
 
$$\text{CH}_4 + 2\text{O}_2 \rightarrow \text{CO}_2 + 2\text{H}_2\text{O}$$

$$\Delta H = -803 \text{ kJ mol}^{-1}$$

The reaction kinetics for catalytic combustion on a noble metal catalyst (e.g., Pd or Pt) are first order with respect to methane concentration and zeroth order with respect to oxygen. There is a small but measurable impact of homogeneous combustion that occurs in the channels and is given by a rate law of order  $-0.3$  and  $1.3$  with respect to methane and oxygen, respectively.

The nominal reactor model considers multiple interacting domains around each wall plate: combustion channel, combustion catalyst, plate, reforming catalyst, and reforming channel. The flow channels are modeled by 2-dimensional laminar convection-diffusion-reacting flow with a parabolic flow profile between infinite parallel plates. We assume symmetry at the channel centers which is enforced through a no-flux boundary condition. The catalyst layers are modeled in 1-dimension assuming a negligible thickness, and the plate is modeled by a 2-dimensional heat equation (Zanafir and Gavriilidis, 2003).

The system model was developed and solved in gPROMS (gPROMS, 2014). The partial derivatives in the axial domains of the reforming and combustion channels were discretized using backward and forward finite differences, respectively, and the partial derivatives in the axial domain of the plate were discretized with central finite differences. The partial derivatives in the longitudinal domains were discretized using orthogonal collocation of finite elements. The model equations are presented in the Appendix for

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