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Fuel processing in a ceramic microchannel reactor: Expanding operating windows

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ABSTRACT

Ceramic microchannel reactors offer significant advantages to current microchannel reactor technology. Ceramic micro-reactors are able to operate at high temperatures and harsh chemical environments through the use of relatively inexpensive materials and manufacturing processes. Coupled with self-sustained operation through autothermal reforming (ATR) or catalytic partial oxidation (CPOX) of methane, ceramic microchannel reactors can increase reforming efficiency and expand the capabilities of hydrogen and syngas production. This work aims to assess the performance of a novel ceramic microchannel reactor for a wide variety of methane reforming conditions and reactive flow rates. Additionally, a computational fluid dynamics (CFD) model implemented in ANSYS FLUENT simulates fluid flow, heat transfer, and catalyzed heterogeneous chemistry in a three-dimensional model. Experimental testing demonstrates stable operation for both ATR and CPOX; no evidence of structural or catalyst degradation is observed in the presence of exothermic chemistry. Autothermal reforming in the novel ceramic microchannel reactor shows promising results, achieving ~90% methane conversion at a gas hourly space velocity (GHSV) of 75,000 h^{-1}. CFD model results accurately predict reactor outputs from experimental data and provide further insight into the internal reactor chemistry and reaction kinetics.

Introduction

This paper presents the operation and performance of a novel ceramic microchannel reactor for autothermal reforming (ATR) and catalytic partial oxidation (CPOX) of methane for H_2 and CO production. Microchannel reactors offer many improvements over current reforming technology by intensifying heat and mass transfer processes. Miniaturization of flow passages to the sub-millimeter scale has been found to significantly increase heat and mass transfer rates compared to traditional reactors [1–5]. This enhanced thermal regulation can improve product yield and selectivity, as well as increase catalyst lifetime by inhibiting hot-spot formation [1,2,6]. Several studies have demonstrated that microchannel reactors improve performance while simultaneously reducing footprint and initial cost compared to traditional reactor designs [7–10].

The majority of microchannel reactors described in the literature are fabricated from metals. While potentially costly, metallic microreactors benefit from favorable material properties and well-established fabrication techniques.

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Alternatively, the use of ceramic materials enables extremely high-temperature operation in harsh chemical environments not suitable for metallic equivalents [11].

Ceramic materials present unique engineering challenges not found in metallic microreactors. Temperature gradients across the reactor can induce mechanical strain and thermal shock that can fracture the ceramic. Metallic manifolds commonly supply working fluids to the ceramic reactor body. A robust, high-temperature ceramic–metallic seal is required at this interface. However, the low materials and fabrication costs of ceramic microchannel reactors, and the wider range of operation, motivate development of the technology [12,13].

Microchannel reactors have found application in hydrogen production [8]. Bulk H_2 is produced at large chemical plants through steam reforming of natural gas. The process intensification and small footprint enabled with microchannel reactors can increase reactor efficiency, reduce cost, and move H_2 generation towards distributed, point-of-use applications [14].

Natural gas is commonly converted into syngas through three primary methods: steam-methane reforming (SMR), catalytic partial oxidation (CPOX), and autothermal reforming (ATR) (Eqs. (1)–(3)):

$$CH_4 + H_2O \rightarrow 3H_2 + CO \tag{1}$$

$$CH_4 + \frac{1}{2}O_2 \rightarrow 2H_2 + CO \tag{2}$$

$$CH_4 + \frac{x}{2}O_2 + (1-x)H_2O \rightarrow (3-x)H_2 + CO. \tag{3}$$

Steam-methane reforming (Eq. (1)) yields the most hydrogen, and is the preferred process for large-scale hydrogen production. The exothermicity and quick response found with CPOX can be advantageous in smaller applications. Autothermal reforming (Eq. (3)) combines the properties of SMR and CPOX, where the heat released from oxidation is used to drive the steam-reforming reaction. ATR benefits from reduced thermal requirements compared to SMR and increased hydrogen yield compared to CPOX. Additionally, tuning of the steam-to-carbon and oxygen-to-carbon (O/C) ratios can optimize reformate composition for a wider range of applications.

While a number of metallic microchannel reactors have been developed for use in autothermal reforming [8,15,14,16], ceramic microreactors have witnessed less development. A number of different fabrication methods have been explored. Knitter et al. [17] describes additive manufacturing of a modular ceramic microchannel reactor for use in oxidative coupling of methane. Wang et al. utilized deep x-ray lithography to fabricate modular ceramic microreactors [18]. These reactors demonstrated good performance for steam-ethanol reforming, with 100% ethanol conversion at 600 °C. Moreno et al. [19] extruded a tubular, cordierite reactor that achieved methanol conversions >90% at a reactor temperature of 400 °C. Thermal load for the endothermic MeOH steam reforming was supported by methanol combustion in neighboring microchannels.

The ceramic microchannel reactor presented in this paper is an integrated heat exchanger/reactor, designed to transfer heat between two fluids in a counterflow configuration. Coflow designs, in which the cool and hot gases flow in the same direction down the microchannels, have been shown to be superior when coupling endothermic and exothermic reactions in opposing channels. This coflow configuration is also associated with increased thermal stresses that can lead to structural failure [20]. The alumina reactor shown in Fig. 1 is comprised of five layers. Two of the layers have been wash-coated with a rhodium catalyst, and are fed with methane-air-steam mixtures. Hot inert gas flows through the three opposing layers that provide heat to the two reactive layers. Reactor design, fabrication, and performance under steam-methane reforming have been published previously [21,22,23]. In this work, reactor operation is extended to exothermic chemistries. A detailed computational model is used to understand and interpret the observed performance.

Experimental methods

Reactor fabrication

The ceramic microchannel reactors are fabricated in collaboration with CoorsTek Inc., (Golden, Colorado, USA) using the Pressure Laminated Integrated Structure (PLIS) processing. PLIS is presented in detail in Kee et al. [21], and is briefly reviewed here. Carefully synthesized alumina powders (~94% Al_2O_3) are compressed into thin rectangular blank plates.

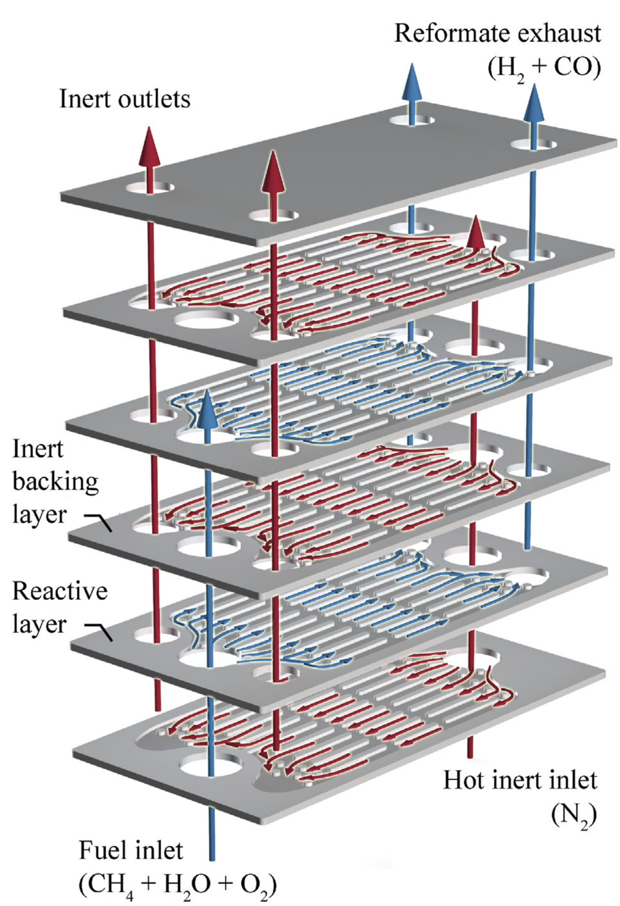

Fig. 1 – Exploded view of a five-layer microchannel reactor.