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# Methane steam reforming in a novel ceramic microchannel reactor

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## ABSTRACT

Microchannel heat exchangers and reactors can deliver very high performance in small packages. Such heat exchangers are typically fabricated from aluminum, copper, stainless steel, and silicon materials. Ceramic microchannel reactors offer some significant advantages over their metallic counterparts, including very-high-temperature operation, corrosion resistance in harsh chemical environments, low cost of materials and manufacturing, and compatibility with ceramic-supported catalysts. This work describes a ceramic microchannel reactor that achieves process intensification by combining heat-exchanger and catalytic-reactor functions to produce syngas. A complete computational fluid dynamics (CFD) model as well as a geometrically simplified hybrid CFD/chemical kinetics model is used in conjunction with experimentation to examine heat transfer, fluid flow, and chemical kinetics within the ceramic microchannel structure. Heat-exchanger effectiveness of up to 88% is experimentally demonstrated. Reactive heat-exchanger performance for methane-steam reforming reaches 100% methane conversion and high selectivity to syngas at a gas hourly space velocities (GHSV) of 15,000 h<sup>-1</sup>. Model results agree well with experimental data and provide insight into physical processes underway during reactor operation.

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## 1. Introduction

Microchannel reactors offer exceptional thermal regulation and control of chemical processes. The high heat- and mass-transfer rates enabled by the microchannel design can increase reactor efficiency, reduce size and weight, and improve conversion, yield, selectivity, and catalyst life [1–3]. Izquierdo et al. [4] compared metal microreactors to conventional fixed-bed reactors for methane steam reforming on different catalysts. For all catalysts studied, microreactors reached higher hydrogen yield and methane conversion than fixed-bed

reactors operating at the same weight hourly space velocity (WHSV).

Microchannel reactors are typically fabricated using metal materials [5]. Ceramics can offer performance advantages over the metallic equivalent [6]. Ceramics are corrosion resistant under high-temperature oxidizing environments. Using inexpensive materials and high-throughput manufacturing processes, ceramic reactors can be fabricated at lower cost. The high-surface-area ceramic materials that commonly support catalysts adhere well within the microchannels of the ceramic reactor without additional material preparation.

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Ceramic reactors also present some unique design challenges. The low toughness of ceramics can lead to thermal-stress-induced fracture due to localized hotspots that can arise in chemical-processing applications. Long start-up times are necessary to mitigate the risk of thermal shock within a ceramic system; therefore, applications are limited to more steady-state operation rather than highly transient systems. Additionally, metal–ceramic interfaces required for gas manifolding can be a source of gas leakage due to thermal expansion mismatch.

In this paper, we present results on development of a ceramic microchannel reactor for use in steam-methane reforming. Hydrocarbon steam reforming in metal microreactors has been demonstrated previously [4], with some researchers using exothermic reactions in one set of layers to drive endothermic reactions on the other side [2,7–10]. Few have integrated stand-alone microreactor capabilities with ceramic materials. The use of ceramic materials for microreactor devices is uncommon; however, several examples of ceramic microreactors reported in the literature are described. Moreno et al. developed a square cordierite microchannel monolith for autothermal operation between methanol reforming and combustion [11]. Knitter et al. produced a ceramic microreactor with exchangeable components for partial oxidation of isoprene (300–500 °C) and oxidative coupling of methane (800–1000 °C) [12]. In an alternative approach, Lewinsohn et al. designed a ceramic microchannel membrane reactor which transports oxygen ions through the ceramic membrane to react with the adjacent methane stream to produce syngas [13].

Fig. 1a shows a photograph of the reactor used in this work, while Fig. 1b displays an exploded view highlighting the internal structure. The reactor consists of four identical layers: two for reactive flow and two for hot-gas flow to drive the chemical process underway on the reactive layers. Each layer of microchannels is fed by a single inlet port, with two exhaust ports in a counterflow arrangement.

Counterflow is optimal for heat-exchanger applications [8,14], and in this work, is used for both inert and reactive

operation. Frauhammer et al. [15], Petrachi et al. [16], and Kolb et al. [17] have shown that co-current flow configurations enable higher activity for reforming applications. However, such configurations also result in higher thermal stresses within the solid body, increasing the risk of mechanical failure. A counterflow configuration is used in this work to limit thermal stress within the ceramic microchannel reactor.

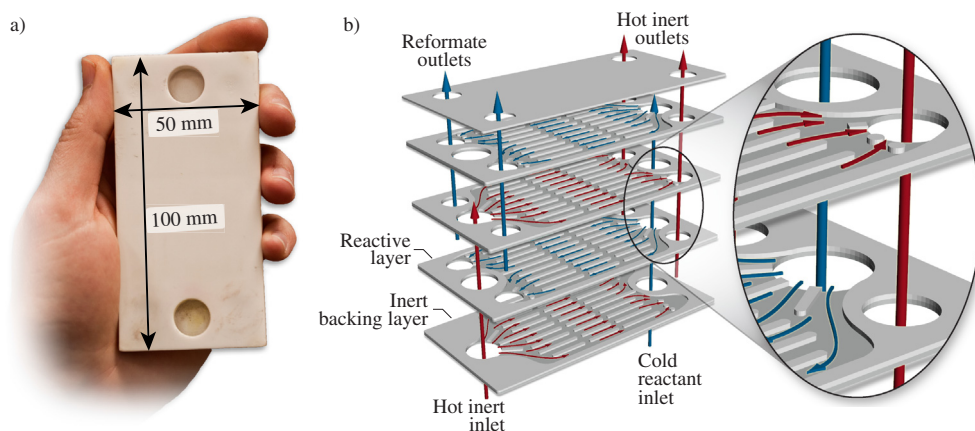
Reactor design and fabrication is presented in Kee et al. [18], including performance analysis under inert heat-exchanger conditions. Although this paper also presents some heat-exchanger results, the focus is on reactor operation and performance for steam-methane reforming.

## 2. Experimental methods

### 2.1. Fabrication

Reactors are fabricated by CoorsTek, Inc. (Golden, Colorado, USA) as four-layer Pressure Laminated Integrated Structures (PLIS) using CoorsTek AD-94 alumina material (93% Al<sub>2</sub>O<sub>3</sub>, 4% SiO<sub>2</sub>, 0.12% Fe<sub>2</sub>O<sub>3</sub>, 0.25% CaO, 0.7% MgO, 0.8% BaO, 0.12% Na<sub>2</sub>O, and 0.7% ZrO<sub>2</sub>). Fabrication begins by mixing ceramic powders with appropriate binders and spray drying. Current prototype designs are machined from dry-pressed un-sintered (green-state) rectangular plates to incorporate manifold and channel geometry. Once final designs are established, inclusion of such geometries into dry-pressing dies can improve manufacturing throughput and decrease product cost.

Green layers are assembled as shown in Fig. 1b and laminated together in a hydraulic press. After lamination, the assembled part is sintered at 1685 °C in an ambient environment. During sintering, the fired part becomes a single polycrystalline ceramic piece with no evidence of the bond lines between initially laminated layers. In fact, the layers are not “bonded” together; rather, the resulting part is a single polycrystalline ceramic with internal microchannels. The appropriate design, binder formulation, lamination pressure and temperature, and sintering environment were determined



**Fig. 1 – (a) Photograph of microchannel heat exchanger/reactor. (b) Exploded illustration showing hot inert (red) and cold reactive (blue) gas-flow paths in a counterflow configuration. Magnified section highlights lamination points required for hermetic sealing during fabrication. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)**

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