



# Operating envelope of a short contact time fuel reformer for propane catalytic partial oxidation



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## HIGHLIGHTS

- Simulated and experimental propane cPOx results.
- Theoretical efficiency limit is 89%.
- Achieved a maximum experimental efficiency of 84%.
- Optimal operating point is  $O_2/C = 0.53$  at  $940\text{ }^\circ\text{C}$ .

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## ABSTRACT

Fuel cell technology has yet to realize widespread deployment, in part because of the hydrogen fuel infrastructure required for proton exchange membrane systems. One option to overcome this barrier is to produce hydrogen by reforming propane, which has existing widespread infrastructure, is widely used by the general public, easily transported, and has a high energy density. The present work combines thermodynamic modeling of propane catalytic partial oxidation (cPOx) and experimental performance of a Precision Combustion Inc. (PCI) Microlith<sup>®</sup> reactor with real-time soot measurement. Much of the reforming research using Microlith-based reactors has focused on fuels such as natural gas, JP-8, diesel, and gasoline, but little research on propane reforming with Microlith-based catalysts can be found in literature. The aim of this study was to determine the optimal operating parameters for the reformer that maximizes efficiency and minimizes solid carbon formation. The primary parameters evaluated were reformate composition, carbon concentration in the effluent, and reforming efficiency as a function of catalyst temperature and  $O_2/C$  ratio. Including the lower heating values for product hydrogen and carbon monoxide, efficiency of 84% was achieved at an  $O_2/C$  ratio of 0.53 and a catalyst temperature of  $940\text{ }^\circ\text{C}$ , resulting in near equilibrium performance. Significant solid carbon formation was observed at much lower catalyst temperatures, and carbon concentration in the effluent was determined to have a negative linear relationship at  $T < 750\text{ }^\circ\text{C}$ . The Microlith reactor displayed good stability during more than 80 experiments with temperature cycling from 360 to  $1050\text{ }^\circ\text{C}$ .

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## 1. Introduction

Over the past few decades, fuel cells have gained traction in the automotive and large-scale power generation industries, but have yet to gain a foothold in the residential sector. While there have been several fuel cell systems developed for residential use, there is a gap in systems that can deliver power on the order of 250 W–1 kW. Some examples of residential devices that require

these power levels include lawn mowers, leaf blowers, gas trimmers, and backup auxiliary power units. Presently, residential systems that require electrical power at this mid-level are dominated by small internal combustion engines (ICE) that suffer from low electrical conversion efficiency, high emissions, and loud operation. This provides a unique opportunity for fuel cell technology as it directly addresses these major drawbacks of ICE-based devices.

In spite of their myriad advantages, fuel cell systems capable of meeting medium-sized power requirements for the residential sector have yet to be commercialized at high volume, in part because of issues concerning the fuel quality requirements and attendant supply infrastructure. Hydrogen is predominately

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present in compound form and must first be processed, through a method such as hydrocarbon reforming, before it can be used as an energy carrier. Therefore, a key component of a viable fuel cell power generation system is that its fuel supply must be readily available, acceptable to regulatory/safety agencies, and conventional for the general public's acceptance. Propane, is an attractive fuel because it already has widespread infrastructure in place, the general public is familiar with its use, it is transportable, and has a high energy density. The Golisano Institute for Sustainability (GIS) at Rochester Institute of Technology (RIT) is developing a working fuel cell system that produces approximately 500 W, and runs on commercial propane. A general outline for the program involves three phases including the reformation system design, fuel cell stack configuration, and the final system integration. This work focuses on the first phase, fuel reformation.

Common catalytic reformation techniques include steam reforming (SR), catalytic partial oxidation, dry reforming (DR), and auto-thermal reforming (ATR). It is well known that SR is the most widely used process for hydrogen production at very large scales, however, SR may not be best suited for small scale portable applications due to additional system components such as an additional water tank and water management system [1]. Partial oxidation is a valid alternative since the process only requires fuel and air as oxidant, and is an exothermic reaction. However, when air is used as an oxidant, the products are diluted with nitrogen. The mechanism for cPOx has been investigated in many studies including [2–4] and is an exothermic reaction with CO and H<sub>2</sub> as the primary reaction products. Primary catalyst degradation modes include metal sintering from excessive thermal stress, coke deposition, and sulfur contamination [5–7].

While reformation processes have already been developed at very large scales with high efficiencies, scaling down the reformer for small mobile applications creates significant challenges due to the requirement for quick startup and transient response, a small operating volume, and high efficiencies. The Short Contact Time (SCT) reactor design is an approach that allows for high volumes of fluid flow such that the residence time of the gas mixture inside the catalyst bed is on the order of milliseconds [8]. Much of the research using SCT reactors has been focused on syngas/hydrogen production [9], however other applications include dehydrogenation [10–12], and Fischer-Tropsch synthesis [13]. Some of the more commonly studied SCT reactors include ceramic monoliths and foams [14–16].

The reformer studied in this paper was provided by Precision Combustion Inc. (PCI), and is based on their patented Microlith® technology [17]. The Microlith substrate consists of a series of catalytically coated metal meshes with very small channel diameters, allowing for ultra-short-channel-length. As compared to other SCT substrates such as ceramic monoliths and foams, the Microlith reactor provides a higher rate of reaction, lower thermal mass, and significantly higher heat and mass transfer coefficients [18]. The Microlith substrate performs favorably because of its high geometric surface area (GSA) per unit volume of the reactor, and high specific surface area of the catalyst support/washcoat [19]. Additionally, the low thermal mass of the Microlith elements allows for rapid start-up and transition to steady operating temperatures. Using a lumped sum capacitance model, a 30-fold increase in thermal response time was determined when comparing a Microlith reactor to a ceramic monolith substrate [20]. Furthermore, near theoretical equilibrium reactions for partial oxidation experiments have been observed using these reactor designs which results in high selectivity of the desired product as well as high reformer efficiencies [19].

Fuel reforming research using Microlith based catalysts have focused on fuels including gasoline [8], methane [21], methanol

[22], JP-8 and diesel [23], but little research on propane reforming with a Microlith catalyst can be found in literature. The aim of this article is to assess the reforming performance of a 31.75 mm diameter Inconel single tube reactor for propane cPOx, and to examine various operating points. Previous collaborative work with PCI has yielded impressive results using their Microlith catalysts that were developed for syngas production [18]. The operating envelope for this reformer is described in terms of temperature range, and the diatomic oxygen (from air)-to-carbon ratio (O<sub>2</sub>/C). The catalytic performance is evaluated in terms of H<sub>2</sub> and CO yield, conversion efficiency, and solid carbon formation. Additionally, the optimal operating point that maximizes fuel conversion efficiency, and minimizes carbon formation is described. Thermodynamic modeling of propane cPOx was conducted using the Gibbs free energy minimization method with aspenOne® Engineering software, to complement the experimental data. All experimental testing was performed at an existing fuel cell and bio-fuel research facility at RIT's Golisano Institute for Sustainability (GIS).

## 2. Methods and materials

### 2.1. Thermodynamic modeling

In order to simulate the propane cPOx reaction, aspenOne® Engineering modeling software was employed. AspenOne Engineering is a comprehensive chemical process modeling software and is widely used in the petrochemical industry for refining simulation and optimization [24]. The propane cPOx model simulation was developed using Aspen HYSYS V8.4 of the aspenOne Engineering package, and primarily utilized the Gibbs reactor unit operation block. The Soave-Redlich-Kwong (SRK) equation of state was the global property model used. In order to simulate the effects of temperature and input O<sub>2</sub>/C ratio on propane cPOx, a case study was developed that varied the reformer temperature and O<sub>2</sub>/C ratio with a propane fuel flow of 1 kmol s<sup>-1</sup>. Previous investigations of propane reforming generally operate catalysts between 400 and 800 °C [4,25,26]. For the simulated case study, the reformer temperature was varied from 400 to 1000 °C with a step size of 10 °C.

For the propane cPOx simulation, the following assumptions were made:

- 1) Uniform temperature distribution within the reformer.
- 2) The reformate gas mixture behaves as an ideal gas and pressure gradients were ignored within the reactor.
- 3) Inlet propane and air were well mixed and the reaction reaches thermodynamic equilibrium. The considered reformate species in this analysis were H<sub>2</sub>, CO, O<sub>2</sub>, H<sub>2</sub>O, CH<sub>4</sub>, C<sub>2</sub>H<sub>4</sub>, C<sub>2</sub>H<sub>6</sub>, CO<sub>2</sub>, C<sub>3</sub>H<sub>8</sub>, C (solid graphite), and N<sub>2</sub>.
- 4) The Gibbs reactor unit operates adiabatically.
- 5) The major modeling independent parameters were reformer temperature and O<sub>2</sub>/C ratio. The effect of GHSV was not considered for this model.

The stoichiometric equilibrium for propane cPOx occurs at an O<sub>2</sub>/C ratio of 0.5 as can be seen in Equation (1) [27].



Thus the simulated reaction was designed to vary the O<sub>2</sub>/C ratio from 0.3 to 1.2 by changing the oxygen inlet flow from 1 to 3.5 kmol s<sup>-1</sup> with a step size of 0.1. Results are plotted herein from this analysis using Matlab®, including the effluent composition and reformer efficiency as a function of temperature and O<sub>2</sub>/C ratio. The reformer efficiency was calculated based on the following equation:

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