Thermally self-sustaining tubular SOFC power generator with no moving parts

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Abstract. Hydrocarbon fuels is roughly 50 times higher energy density compared to commercial batteries. However, scaling down electrical power generators for portable devices have been far from successful due to the difficulties of minimizing heat and friction losses. Hence, an alternate solution without moving parts incorporating a micro-tubular solid oxide fuel cell (mT-SOFC), thermal transpiration (gas pumping in a nanoporous medium from temperature gradient) and catalytic combustion on platinum surface is proposed. The concept involves having thermal transpiration membrane utilizes the temperature gradient created by mT-SOFC and catalytic combustion to supply reactants at high temperature to mT-SOFC for its operation. With this concept, catalytic combustion modeling and thermal transpiration membrane modeling were studied individually and integrated together for an airbreathing cylindrical chamber. The chamber design involves an aluminum cylinder wrapped with glass microfiber filter as transpiration membrane. Aluminum structure provides practically uniform temperature on inside surface of the membranes. Platinum mesh was used as a catalyst for rich butane-air combustion, the products of which supply the mT-SOFC. It was found that the temperature, fuel flow rate and equivalence ratio can be varied over mT-SOFC operation range. The integrated system demonstrates capability in self-sustaining power generation using hydrocarbon fuel without moving parts.

1. Introduction

Portable electronic devices have ever-growing needs for electrical power source and/or energy storage unit. Batteries hold tremendously less specific energy compared to hydrocarbon fuels, whereas commercial batteries such as lithium ion has specific energy only 1.6% of the specific energy of hydrocarbon fuels. Even though energy density of batteries has significantly increased for more than twice in the last decade, the numbers remain distant from being able to support various mobile applications such as wheel chairs, mobile robots and portable military applications. In addition, such batteries have to be occasionally recharged and/or replaced unlike hydrocarbon fuels that can be stored in a container that can be replaced easily.

Scaling down of internal combustion engines and electrical generators to Micro-Electro-Mechanical Systems (MEMS) scales has been unsuccessful due to issues with heat losses, friction losses and the difficulty of precise manufacturing parts at such small scales. A small aircraft engine is an example of
small-scale power generator using hydrocarbon fuel. Menon et al. [1], Walther et al. [2] and Dessornes et al. [3] studied small-scale engine e.g. a reciprocating engine, a Wankel rotary engine and a microscale gas turbine; however, all of them were confronted with various issues including, unacceptable noise, manufacturing tolerance concerns, friction losses and heat losses, resulting in the efficiency of their engines to be too low for power generators.

Small-scale power generators with no moving parts have been studied throughout a lot of research groups due to friction loss concerns. Hsu et al. [4] proposed an idea of using thermoelectric to generate electricity from burning hydrocarbon fuel without any moving parts, but the efficiency of thermoelectric itself was proved too low. Another solution is a proton exchange membrane fuel cell (PEMFC), a portable power generator with no moving parts, has difficulty involving hydrogen storage and fuel impurity as described in Shan et al.’s work [5]. Solid Oxide Fuel Cell (SOFC) is a robust fuel cell that can operate with various hydrocarbon fuels, but it has to operate under high temperature at least 600°C. SOFC could be a feasible solution for a small portable electrical power generator.

To operate a fuel cell, oxygen has to be continuously supplied to maintain the reaction. Using small fan requires energy input and friction loss would be dominant at small scale. However, thermal transpiration pump from nanoporous membranes can be used to eliminate loss due to friction from moving parts. The pumping power is generated from temperature gradient across the membrane such that ambient air will be driven from the cold side to the hot side of the membrane. The temperature gradient can be provided from catalytic combustion on platinum foil placed inside a combustion chamber. One solution for small-scale power generator is a thermal transpiration membrane and catalytic combustion from butane combining with a solid oxide fuel cell (SOFC) for electrical power generation. This self-sustaining power generation device without moving parts was previously introduced by Wang et.al. [6] as in figure 1. It was designed without optimization of heat transfer and energy consumption; therefore, the power generator provided only 0.092% of efficiency due to heat losses and unconsumed fuel. To achieve the appropriate flow and power management, parameters for thermal transpiration membrane, catalytic combustion and SCFC have to be investigated separately. The system will be designed and optimized based on maximum efficiency.

**Figure 1.** Schematic of the thermal transpiration device.  
(a) assembled view and (b) exploded view

2. **Thermal transpiration membrane**

Thermal transpiration is a rarefied gas dynamics phenomenon describing the gas flow through a narrow channel with an imposed temperature gradient. Thermal transpiration as discussed in this study is refers to a gas interacting with a solid, stationary surface without vaporization or condensation. As gas flow from cold end to the hot end, a pressure gradient is created across the channel. The concept of thermal transpiration has been studied since Osborne Reynolds since 1879 [7] with high thermal conductivity porous media causing high thermal energy input for pumping power. For more recently study, Sone et al. [8], Vargo et al. [9] and Muntz et al. [10] have studied about the mass flow rate ($\dot{M}$) induced by thermal transpiration membrane. The relation of all related parameters is shown in equation (1)
\[ \dot{M} = P_{avg} \sqrt{\frac{m}{2kT_{avg}}} A \frac{L_r}{L_x} \left[ \frac{\Delta T}{T_{avg}} Q_T - \frac{\Delta P}{P_{avg}} Q_P \right] \]

(1)

where \( \Delta P \) is the pressure difference between both side of the membrane, \( P_{avg} \) is the average pressure across the membrane, \( \Delta T \) is the temperature difference across the membrane, \( T_{avg} \) is the membrane average temperature, \( Q_T \) is the thermally driven flow coefficient, \( Q_P \) is the pressure driven return flow coefficient, \( A \) is the channel cross-section area, \( L_r \) is the pore size radius, \( L_x \) is the capillary channel length, \( m \) is the molecular mass of the gas and \( k \) is Boltzmann’s constant. The relation of \( Q_T \) and \( Q_P \) as a function of Knudsen number, \( K_n \equiv \lambda/L_r \) (Knudsen number is the ratio of gas mean free path to channel radius) is shown in figure 2 and the thermal transpiration diagram and parameters are shown in figure 3.

In our study, fiberglass membranes have been used as transpiration membranes, while equation (1) is only applied for a cylindrical tube. The modeling of transpiration membrane from fiberglass has to include the backflow term which can be described by Darcy’s law for the flow through a porous media. Equation (1) has to be modified as

\[ \dot{M} = P_{avg} \sqrt{\frac{m}{2kT_{avg}}} A_{tot}(AF_{TT}) \frac{L_r}{L_{x,tot}} \left[ \frac{\Delta T}{T_{avg}} Q_T - \frac{\Delta P}{P_{avg}} Q_P \right] - \rho_{avg} \frac{\kappa(A_{tot}(1 - AF_{TT}))\Delta P}{\mu L_x} \]

(2)

where \( A_{tot} \) is the total area of the membrane, \( AF_{TT} \) is the area factor of the thermal transpiration membrane, \( \rho_{avg} \) is the average air density, \( \kappa \) is the permeability of the membrane which can be defined from the experiment and \( \Delta P \) is pressure drop across the membrane and \( \mu \) is the dynamics viscosity of the gas which is air for this study. Two K-type thermocouples were used to measure the temperature across the membrane. A bubble flow meter and a piezoelectric differential pressure sensor, 223BD-00001, were used to measure flow rate and pressure difference across the membrane, respectively. After the leak term was included, the performance curve from experiment fits with equation (2) is shown in figure 4. The fitted properties of this membrane are 1.25-μm effective pore radius, 0.28 thermal transpiration area factor and 1.932×10^{-13} m² permeability. The permeability was measured separately by applying pressure on one side of the membrane to push the air through the membrane without applying any heat on the membrane.
3. Catalytic combustion

Catalytic combustion is chemical reactions between oxidizer and fuel occurring only on catalytic surface. The catalyst helps decrease activation energy of the reaction so that the surface combustion can sustain at temperature lower than that of gas phase combustion. Well-known catalyst is a noble metal i.e. platinum which has been used in this study because platinum is one of the most reactive catalysts. Langmuir-Hinshelwood (LH) proposed adsorption rate model that is commonly used to express reaction rate for catalytic combustion as

\[ r_{ads} = k_{ads} C_s \theta(s) - k_{des} \theta \]  

(3)

where \( k_{ads} \) is rate constant for adsorption, \( k_{des} \) is rate constant for desorption, \( \theta(s) \) is empty space on the surface, \( \theta \) is fraction molecule adsorbed on the catalyst surface and \( C_s \) is gas phase concentration adjacent to the catalyst surface. The rate constant of adsorption and the rate constant of desorption for each molecule can be distinctly described by modified Arrhenius expression as

\[ k_{ads} = s \frac{RT}{\Gamma \sqrt{2\pi M}} \exp \left( - \frac{E_{a_{ads}}}{RT} \right) \left( \frac{T}{T_{ref}} \right)^{\beta_{ads}} \]  

(4)

and

\[ k_{des} = A \exp \left( - \frac{E_{a_{des}}}{RT} \right) \left( \frac{T}{T_{ref}} \right)^{\beta_{des}} \]  

(5)

where \( s \) is sticking coefficient, \( \Gamma \) is site density (2.5×10^9 mol cm\(^{-2}\) for platinum), \( R \) is universal gas constant, \( T \) is catalyst surface temperature, \( T_{ref} \) is reference temperature (300 K), \( E_a \) is activation energy of adsorption and desorption, \( \beta \) is temperature exponent and \( A \) is desorption coefficient. Generally, high temperature increases the rate constant of adsorption and desorption which tend to increase the global reaction rate of catalytic combustion.

Butane (C\(_4\)H\(_{10}\)) is used as fuel but its surface reaction contains a lot of detail reaction mechanism that requires a lot of computational time; therefore, Deshmukh et al. [11] and Kaisare et.al. [12] suggested simpler simulation model involving one-step mechanism to estimate catalytic combustion of butane on platinum surface with the following global reaction;

\[ C_4H_{10} + 6.5O_2 \rightarrow 4CO_2 + 5H_2O \]  

(6)

The reaction rate for fuel-lean mixture when C\(_4\)H\(_{10}\) is deficient molecule can be expressed as

\[ r = \left( \Gamma k_{C_4H_{10}}^{ads} C_s C_{4H_{10}} \right) \left( 1 + \frac{k_{O_2}^{des}}{k_{C_4H_{10}}^{ads} C_s O_2} \right)^2 \]  

(7)
However, under fuel-rich mixture condition when O\textsubscript{2} is deficient molecule, reaction rate was modified and expressed as

\[ r = \Gamma k_{O_2}^\text{ads} C_{O_2,0} \left[ \left( \frac{k_{O_2}^\text{ads} C_{O_2,0}}{k_{O_2}^\text{des}} \right) + 1 \right] + \left( \frac{k_{H_2O}^\text{ads} C_{H_2O,0}}{k_{H_2O}^\text{des}} \right) + 1 \right]^2 + 4 \left( \frac{k_{CH_4}^\text{ads} C_{CH_4,0}}{k_{CH_4}^\text{des}} \right) \]

For modeling purposes, an experiment of catalytic combustion on platinum was made in a 7-mm diameter quartz tube with suppling C\textsubscript{4}H\textsubscript{10}-air mixture at various equivalence ratios. The exhaust compositions were measure by gas chromatograph. These compositions will be reactants for mT-SOFC. A one-step catalytic reaction model for fuel-lean C\textsubscript{4}H\textsubscript{10}-air lean mixtures was modified for use with fuel-rich mixtures and used in conjunction with a simulation employing ANSYS-Fluent. The measured and predicted product compositions at various fuel-air equivalence ratios are shown in figure 5.

**Figure 5.** Measured and predicted exhaust compositions for combustion on platinum foil for 12 cm/s flow velocity of C\textsubscript{4}H\textsubscript{10}-air mixtures at varying equivalence ratios.

### 4. Self-sustaining and self-pressurizing combustion chamber

A transpiration membrane and platinum foils were integrated as a self-sustaining, self-pressurising combustion chamber to provide the appropriate temperature and reactant compositions for mT-SOFC. The combustion chamber is made from a 10-mm diameter aluminum tube. The wall along the tube was cut into 4 slots for the external air to be pumped in by transpiration membrane. Two platinum foils are placed inside the chamber for C\textsubscript{4}H\textsubscript{10} catalytic combustion which provides heat and temperature gradient across the membrane and for an mT-SOFC. The simulation from the models of transpiration membrane and catalytic combustion in previous sections were combined in ANSYS-fluent. The simulation and the actual chamber were compared as expressed in figure 6. The results show that the reaction occurs and self-sustaining combustion chamber can provide an appropriate temperature for mT-SOFC operation. Furthermore, the product compositions at the outlet of the combustion chamber were measured by a gas chromatograph at varying C\textsubscript{4}H\textsubscript{10} flow speed. The comparison between experimental results and computational results is shown in figure 7.
5. mT-SOFC for power generation

To generate electrical power, mT-SOFC with effective area of 4.0 cm$^2$ was used. The anode supported mT-SOFC was prepared by extrusion of the anode, dip-coating of the electrolyte, wet-powder spraying of the buffer layer, and dip-coating of the cathode. Yttria-stabilized zirconia (YSZ, Tosoh), Samaria Doped Ceria (SDC, Fuel Cell Materials), nickel oxide (NiO)-YSZ (60:40 w/w, J.T.Baker) and Lanthanum Strontium Cobalt Ferrite (LSCF) + SDC (Fuel Cell Materials) were used as the electrolyte, buffer layer, anode, and cathode materials respectively. For a portable power generator purpose, a mT-SOFC was integrated with the thermal transpiration membrane for supplying air from the outside to the combustion chamber. Boundary conditions and simulation results from ANSYS-Fluent are shown in Figure 6.

The comparison of product compositions at the outlet between simulation and experiment is shown in Figure 7.
cathode side of the fuel cell as shown in figure 8. Hydrogen, used as fuel for this setup, was supplied to the center of the fuel cell to the anode side. The transpiration membrane is able to supply air while the fuel cell generating power, which maximum power was found at 25 mW with 600°C and 45 ml/min H₂ supply.

![Figure 8. Integrated transpiration pump and mT-SOFC.](image)

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