

## Catalytic Combustion in Microchannel for MEMS Power Generation

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### ABSTRACT

Catalytic combustion is a promising technique for producing thermal energy in MEMS- (Micro Electro-Mechanical Systems) scale electrical power generators. To examine the feasibility and possible benefits of catalytic combustion in MEMS-scale channels, catalytic reactions in small diameter (1 mm) flow reactor tubes at low Reynolds numbers are simulated by using the commercial fluid dynamics code FLUENT coupled to external subroutines that model the surface chemistry at the channel wall. Temperature and major chemical species distributions in the gas phase and on the Pt catalyst surface are obtained for fuel-lean methane/air mixtures as a function of the surface temperature. The catalyst surface area required for complete reaction is estimated by the characteristics of heat release caused by surface reaction. The transition from transport-limited to kinetically-limited reaction is quantified. Results suggest that Pt-catalyzed fuel-lean methane/air mixtures have potential for MEMS power generation applications, provided that the surface temperature can be maintained above about 1000K.

**Keywords:** catalytic combustion, MEMS power generator

### INTRODUCTION

MEMS is growing research area aimed at the development of micro-scale electrical and mechanical systems such as sensors, actuators, and communication tools [1]. Although functional components of MEMS are being miniaturized, power sources are often the largest and heaviest components of the system. The demand for smaller-scale, higher energy density power sources is expected to increase with the progress in this field.

Hydrocarbon fuels have numerous advantages over batteries for electrical power systems, most notably two or more orders of magnitude higher energy densities than those of existing batteries. However, devices that convert fuel energy into electricity at small scales have not yet been demonstrated. Several ongoing projects on MEMS power generation employ scaled-down versions of macroscopic combustion engines, however, the issues

of heat loss, friction losses, sealing, manufacturing, assembly, etc. are increasingly more difficult to solve at smaller and smaller scales.

Our approach for a MEMS electrical power generator is an integrated system called microFIRE. This system consists of a combustor, counterflow heat exchanger (Figure 1 (a)) and thermoelectric power generation devices. A Swiss-Roll [2,3] type heat exchanger is used to minimize heat loss from the device as well as recycle thermal energy produced by combustion back to the unburned mixture, enabling so-called "excess enthalpy" combustion. The Swiss Roll burner shown in Fig 1(b) that was first proposed by Weinberg [2,3] decreases heat losses, however, out-of-plane losses lead to quenching at small scales. To further reduce heat losses, we chose a toroidal 3-D geometry that minimizes heat loss from all surfaces as shown in Fig. 1(c). Electricity is generated by thermoelectric elements embedded in the walls between cold incoming reactants and hot outgoing products.

This complicated geometry can be constructed at micro-scale using EFAB (Electrochemical FABrication) solid freeform fabrication technology [4-6]. By this method, one can produce arbitrary 3-D structures by stacking hundreds of layers.

To obtain stable exothermic reaction for thermal energy at MEMS scales, catalytic combustion is a likely choice. Since chemical reactions only occur on the catalyst surface, the location of the heat source is fixed. This makes heat transfer design simpler than gas-phase combustion in which the location of reaction zone may change in undesirable ways. Furthermore, in MEMS combustors, the higher surface area to volume ratio makes catalytic combustion even more attractive. Finally, the lower temperature of catalytic combustion makes thermal stresses and heat losses less problematic.

In the present study, to examine the feasibility of catalytic combustion in MEMS-scale channels, catalytic reaction in small diameter flow reactor tubes at low Reynolds numbers is simulated by the commercial fluid dynamics code FLUENT [7] with an external surface chemistry subroutine [8]. In particular, the minimum temperature required to sustain combustion is

determined for a prescribed set of flow and reactant conditions. This initial study focuses on methane fuel on Pt catalyst because this is the only hydrocarbon fuel / catalyst combination for which a comprehensive chemical mechanism is available.

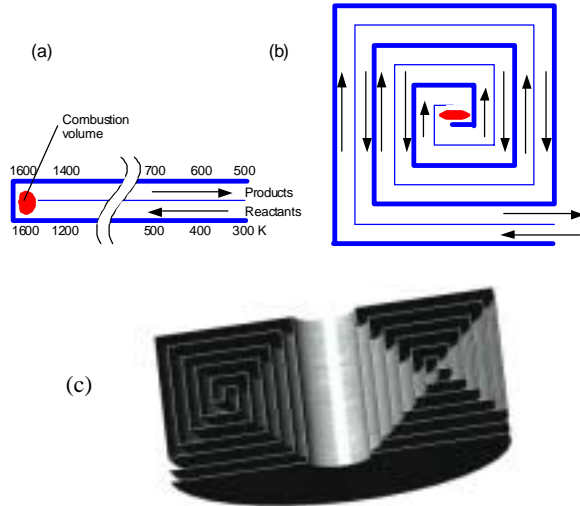


Figure 1. Heat exchanger and Swiss Roll type burner used in the microFIRE power generation system.

### Computations

The microscale channel configuration is assumed to be a cylindrical tube 1 mm in diameter and 10 mm in length as shown in Fig. 2. The usual two-dimensional, axisymmetric Navier-Stokes equations were solved by the computational fluid dynamics code FLUENT [7], which was coupled to external subroutines [8] that model the heterogeneous chemical reaction at the catalytic channel walls. The subroutines calculate the mass fluxes at the wall due to the surface reactions and the surface coverage with chemical species adsorbed by the walls. The governing equations and treatment of surface reactions used were the same as those employed by Deutschmann et al [9]. At the catalytic wall, prescribed temperature boundary conditions were applied to examine the catalytic activity. Although this boundary condition does not rigorously correspond to the operating condition of Swiss-Roll burners in which the heat release due to combustion and the heat loss environment will determine the actual temperatures, our approach determines the required catalytic temperature for the design of successful MEMS power generators. A fuel lean methane/air mixture is supplied from the inlet of flow reactor tube at a velocity of 0.16 m/s, which corresponds to a Reynolds number of 10. The concentration of the mixtures was chosen for equivalence ratio of 0.4, which is outside of conventional flammability limit. Davis *et al.* [10] concluded that for methane/air mixtures on Pt catalyst at equivalence ratios from 0.3 to 0.5, homogeneous (gas-phase) reaction occurs only for temperatures higher than 1275 K. This is beyond the temperatures of interest in this study, consequently, no gas phase reactions are modeled here. To avoid numerical complications associated with the catalyst leading edge coinciding with the inlet boundary conditions, the first 1 mm of the wall at the inlet is assumed to be an inert wall. The remaining 9 mm of the channel wall is assumed to be Pt catalyst. The wall temperature at the inlet is kept at 300 K for the first 0.5 mm of

channel and ramps linearly to a specified constant temperature ( $T_w$ ) over the remaining 0.5 mm of the inert wall. The surface reaction mechanism [11] employed in this work consists of 24 heterogeneous reactions involving eleven surface species (Pt(s),  $\text{CH}_3(\text{s})$ ,  $\text{CH}_2(\text{s})$ ,  $\text{CH}(\text{s})$ ,  $\text{C}(\text{s})$ ,  $\text{CO}(\text{s})$ ,  $\text{CO}_2(\text{s})$ ,  $\text{H}_2\text{O}(\text{s})$ ,  $\text{OH}(\text{s})$ ,  $\text{H}(\text{s})$  and  $\text{O}(\text{s})$ ) and seven gaseous species ( $\text{CH}_4$ ,  $\text{O}_2$ ,  $\text{N}_2$ ,  $\text{H}_2\text{O}$ ,  $\text{CO}_2$ ,  $\text{CO}$  and  $\text{H}_2$ ).

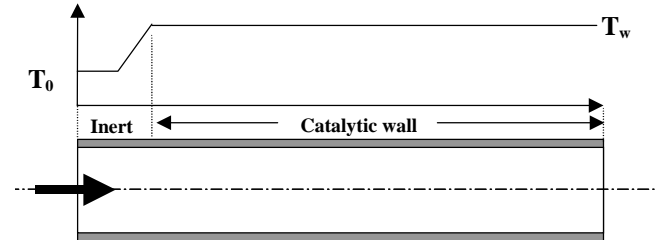


Figure 2. Simulated geometry of flow reactor tube of 1 mm in i.d. and 10 mm in length. Inner wall surface is assumed to be Pt catalyst except inert wall of 1 mm in length at the inlet.

### Results and discussion

Figure 3(a) shows the temperature distribution in the flow reactor tube at the catalytic wall temperature  $T_w$  of 1100 K. The channel wall close to the inlet has a comparatively low temperature because of the specified wall temperature profile, then rises to 1100K farther downstream. Obviously temperature cannot be used as a measure of the degree of completeness of reaction because of the prescribed temperature boundary condition. Figure 3(b) shows the corresponding distribution of  $\text{CH}_4$  mole fraction. It can be seen that almost all of the methane is absorbed and reacted with oxygen very close to the leading edge of the catalytic wall. The distribution of product  $\text{CO}_2$  mole fraction is shown in Fig. 3(c). With the progress of the reaction of methane,  $\text{CO}_2$  is produced such that  $\text{CO}_2$  is distributed throughout almost the entire region downstream of where  $\text{CH}_4$  starts to react. The  $\text{H}_2\text{O}$  mole fraction distribution (not shown) is similar to that of  $\text{CO}_2$ . Figure 3(d) shows the distribution of  $\text{CO}$  mole fraction as a representative of intermediate species. It can be seen that  $\text{CO}$  is localized at the leading edge of the catalytic wall in the region where  $\text{CH}_4$  disappears but before  $\text{CO}_2$  is produced, in a manner similar to that typical of gas-phase combustion.

Figure 4 shows the total heat release due to surface reaction for several different values of  $T_w$ . It can be seen that for  $T_w = 900\text{K}$ , about half of the total possible heat release occurs within the 10 mm length of channel, whereas at  $T_w = 1000\text{K}$  and above, nearly complete reaction is obtained, and at  $T_w = 800\text{K}$ , almost no reaction occurs over this length of channel. Catalytic activity often shows hysteresis as temperature is increased or decreased [12], however, in the present computations, no significant difference was found between results obtained by progressively increasing or decreasing  $T_w$ .

Figure 5 shows the axial profile of  $\text{CH}_4$  mole fraction in the gas phase at the radial location adjacent to the catalytic wall. When the catalytic wall temperature is 1100K, the  $\text{CH}_4$  mole fraction decreases to zero close to the leading edge of the catalytic wall, implying that  $\text{CH}_4$  is completely consumed by surface reaction in this region. With the decreasing  $T_w$ , however,

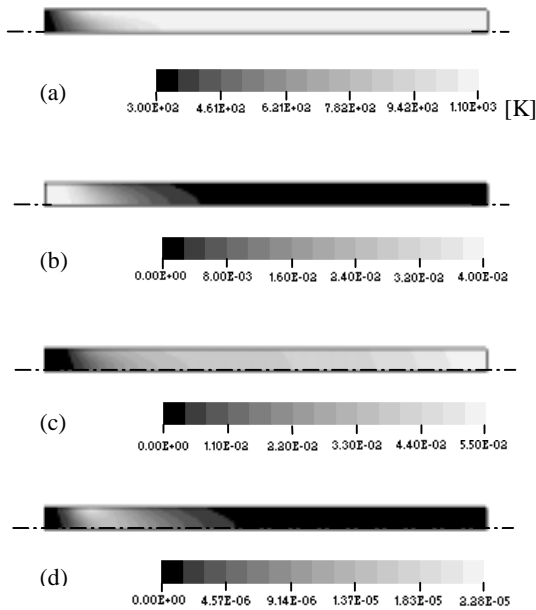


Fig. 3 (a) Temperature profile, (b) CH<sub>4</sub>, (c) CO<sub>2</sub> and (d) CO mole fraction distributions on the central cross section of the channel. (upper half)

catalytic reactivity decreases and the CH<sub>4</sub> concentration at the wall increases. Clearly the fuel mole fraction at the wall is an indication of whether the heat release rate is transport limited or kinetically limited. At  $T_w = 1100\text{K}$  there is no fuel at the catalyst surface (except very near the inert inlet section), indicating that the wall is acting as a sink of fuel whose strength is limited only by the rate at which fuel can be transported to the surface. In contrast, at  $1000\text{K}$  there is significant fuel that reaches the layer of gas adjacent to the surface but does not react, leading to a buildup of fuel. Still, over the 10 mm length of tube, nearly all of this fuel eventually reacts. At  $900\text{K}$  the surface reaction rate is sufficiently slow that there is substantial fuel accumulation at the wall over the entire length of the tube.

Figure 6 shows the profile of heat release rate due to surface reaction on the channel wall. This figure clearly shows that exothermic chemical reaction occurs mainly close to the leading edge of the catalytic wall when its temperature is high enough. In contrast, a comparatively flat profile of heat release is observed at the lower temperature region. This again shows that the required area of the catalyst surface for total conversion of fuel depend on the surface temperature. In the present flow conditions, the requirement varies roughly from  $30\text{ mm}^2$  to  $10\text{ mm}^2$  (corresponding to heat release rates of  $0.6$  to  $1.8\text{ W/cm}^2$ ) for temperatures of  $1100$  to  $1200\text{ K}$ .

Within the context of the current model, complete reaction can be obtained for any  $T_w$  if the length of the tube is extended sufficiently since  $T_w$  is a prescribed quantity. Figure 5 suggests a length of about  $20\text{ mm}$  (corresponding to a heat release rate of  $0.3\text{ W/cm}^2$ ) would be required for  $T_w = 900\text{K}$ . In a practical combustor,  $T_w$  is not prescribed but rather is determined by the balance between heat generation and heat losses. For example, for a typical natural convection heat loss coefficient of  $10$

$\text{W/m}^2\text{K} = 10^{-3}\text{ W/cm}^2\text{K}$  and a temperature of  $1100\text{K}$  (i.e.  $800\text{K}$  above ambient), the heat loss rate would be  $0.8\text{ W/cm}^2$ , which is close to the heat release rates obtained at  $1100\text{K}$ . Thus a wall temperature of  $1100\text{K}$  could be sustained with a reactor length of  $10\text{ mm}$ . However, using a longer tube at a lower  $T_w$  would lead to a heat loss rate greater than the heat generation rate and could not be sustained. For example, as mentioned above, at  $T_w = 800\text{K}$  a  $20\text{ mm}$  reactor is required for nearly complete combustion. For this case the heat release rate is  $0.3\text{ W/cm}^2$ , but with the same heat loss coefficient as before the heat loss rate would be  $0.5\text{ W/cm}^2$ . Since this level of heat loss could not be sustained,  $T_w$  would drop further, leading to flame extinction.

Figure 7 shows the surface coverage of O(s) for  $T_w = 900\text{ K}$  and  $1100\text{K}$ . At  $T_w = 900\text{K}$ , more than  $90\%$  of the catalyst surface is covered by O(s), which inhibits the adsorption of CH<sub>4</sub> that could lead to higher rates of surface reaction. At  $T_w = 1100\text{ K}$ , where surface reaction is mostly transport limited, more than  $20\%$  of surface sites are open for surface reaction. This implies that the adsorption and desorption reaction steps of O(s) play an

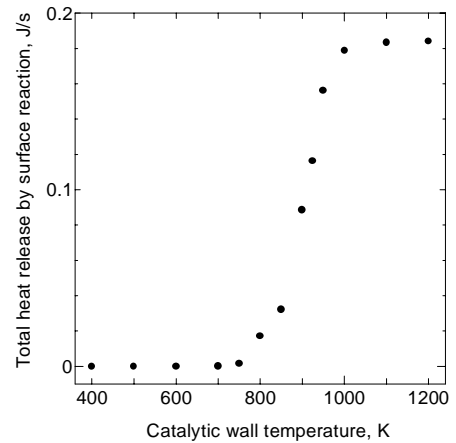


Fig. 4 Total heat release by the surface reaction

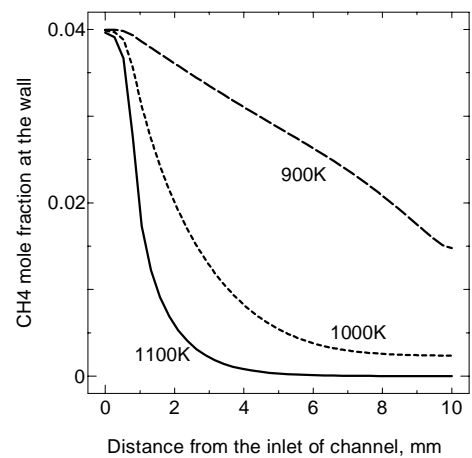


Fig. 5 CH<sub>4</sub> mole fraction in the gas phase along the catalytic wall.

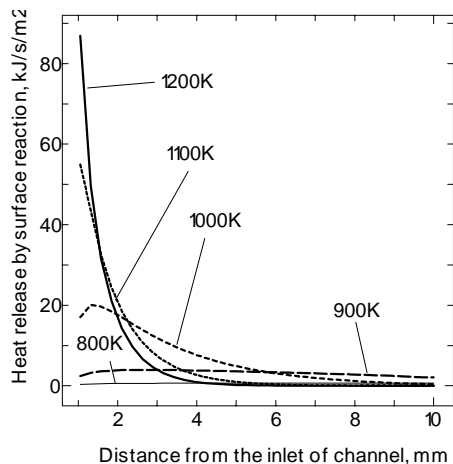


Fig. 6 Heat release rate distribution at the catalytic wall.

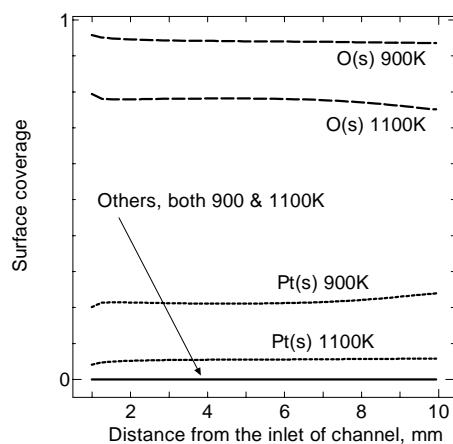


Fig. 7 Surface coverage at the catalytic wall temperature of 900 K and 1100K. Others are OH(s), H<sub>2</sub>O(s), H(s), CO(s), C(s) and CH(s).

important role in the ability of the catalyst surface to sustain chemical reaction. If the wall temperature further decreased to 800 K (not shown), almost all surface sites are covered by O(s).

In Ref. [8], a decrease in ignition temperature with increasing methane concentration was reported for rich CH<sub>4</sub>/O<sub>2</sub> mixtures reacting on platinum foil. This tendency is analogous to the present results. In contrast, H<sub>2</sub>/O<sub>2</sub> and CO/O<sub>2</sub> systems exhibited an increase in ignition temperature (i.e., the temperature at which the transition from kinetically-controlled to transport-controlled reaction occurs) with increasing fuel/oxygen ratio [11]. These results suggest that it may be possible to minimize ignition temperature by appropriate use of excess air vs. exhaust gas recirculation to control the amount of O<sub>2</sub> available for adsorption on the catalyst surface.

It is well known that for higher hydrocarbons, combustion can be sustained at lower temperatures than for methane; when mechanisms become available for these fuels, it is anticipated that the approach employed here can also be used to obtain

insight for these fuels as well. In future work, different Reynolds and Knudsen numbers as well as different compositions will be employed to determine scaling relationships for MEMS-scale catalytic combustion.

## Conclusion

Catalytic combustion in small diameter reactor tubes at low Reynolds numbers was studied numerically to examine its feasibility for MEMS power generator applications using a combination of commercial fluid dynamics code and external subroutines that calculate the surface chemical reactions. Results show that Pt catalyst for methane/air mixture has potential for the application when the catalyst wall temperature is above 1000 K.

Required surface areas of the catalyst for total conversion of supplied fuel in the present specified condition are from 10 mm<sup>2</sup> to 30 mm<sup>2</sup>. In future work, scaling relationships for MEMS-scale catalytic combustion will be determined.

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