Micro Catalytic Combustor with Pd/Nano-porous Alumina for High-Temperature Application

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Abstract: A micro-scale catalytic combustor using high-precision ceramic tape-casting technology has been developed for high temperature applications such as thermophotovoltaic power generation. Pd catalyst on nano-porous alumina support fabricated through anodic oxidation was employed for the catalyst layer having good adhesion to the substrate. In order to minimize thermal stress at high operation temperatures, catalyst arrangement for minimum temperature gradient has been proposed based on a series of CFD analysis including surface reaction of butane. In combustion experiments with a prototype combustor, the wall temperature as high as 850 $^{\circ}$C and extremely high heat generation density of $2\text{-}5\times10^8$ W/m^3 has been achieved. It is also found that radial temperature gradient is significantly reduced with the localized catalyst arrangement proposed in the present study.

Due to recent advances of mobile electronic devices, worldwide demands for high-performance mobile power generation systems are growing rapidly. Small-scale combustor is a critical component for various mobile power generation systems such as thermophotovoltaic (TPV) generator [1]. The objective of the present study is to develop a high-temperature micro-scale catalytic combustor, which can be applied to micro TPV systems.

Okamasa *et al.* [2] employed Pd/nano-porous alumina fabricated through anodic oxidation of aluminum film for the catalyst layer having good adhesion to the substrate. They found its activity is as high as the catalyst prepared with the conventional sol-gel methods. High-precision ceramic tape-casting technology for LSI packaging is utilized for cost-effective fabrication of micro combustor integrated with an embedded electrical heater.

However, their ceramic combustors experience mechanical failure only at 500 $^{\circ}$C due to thermal stress caused by temperature gradient inside their combustors [2]. In order to propose combustor geometry and catalyst arrangement with minimum wall temperature gradient, a series of CFD analysis including surface reaction in micro channels has been made for radial-flow-type combustors using Fluent 6 (Fluent Inc.).

Figure 1 shows a radial-flow-type combustor considered in the present study. The height and radius of the combustion channel are respectively set to 0.3 mm and 10 mm. Since the channel height is much smaller than the quenching distance, homogenous reaction is neglected. A single-step irreversible reaction model, in which the reaction rate is proportional to the butane concentration, is employed to characterize the activity of the catalyst layer for *n*-butane oxidation. The reaction constants are estimated in a preliminary combustion experiment as follows [2]: pre-exponential factor $A = 6.181\times10^8$ (m/s) and activation energy $E = 1.163\times10^8$ (J/kmol). The overall reaction rate is diffusion-limited for the catalyst temperature higher than 380 $^{\circ}$C. It is noted that the reaction constants in the present are in accordance with those given by Wang *et al.* [3].

In the present CFD, a uniform velocity profile of butane-air mixture is assumed at the inlet. The volumetric flow rate Q_B is kept at 10 sccm with the equivalence ratio of unity, which corresponds to the heat generation of 20 W at complete fuel conversion. A heat exchanger with a temperature efficiency of 0.78 is assumed for heat recirculation. Thus, the mixture temperature at the combustor inlet is set to 630 $^{\circ}$C. Thermal conductivity of the ceramic wall material is set to 14 W/(m K). For the thermal boundary condition, radiative heat transfer is assumed at the bottom surface of the combustor. The emissivity is set to 0.3 based on the emission spectra of tungsten selective emitter for TPV [4]. At the top surface, adiabatic condition is assumed for simplicity.
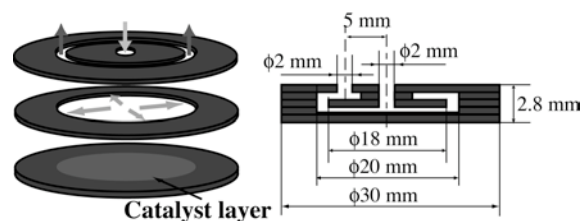

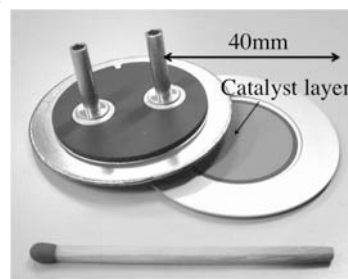Figure 1. Exploded view of a radial-flow-type combustor.

Figure 2. Micro combustor fabricated with ceramic tape-casting.

For the combustor with homogenous catalyst distribution, the wall temperature difference is as large as 260 °C, because the heat generation density at the center is about 1000 times larger than that at the trailing edge; most fuel has been converted upstream and the bulk mean concentration becomes very small near the trailing edge. Therefore, we try to minimize the heat generation density distribution by the localization of heat generation. By removing the catalyst layer from the center, the wall temperature difference can be reduced to 65 °C with a maximum wall temperature of about 900 °C (not shown).

Figure 2 shows the present radial-flow-type ceramic combustor. The outer diameter and height of the combustor are respectively 40 mm and 3.5 mm. The fuel-air mixture from the inlet port is introduced into the center of the combustion chamber through a straight channel inside the combustor. The mixture flows in the radial direction in the combustion chamber, where the catalyst surface is formed at $7 < r < 10$ mm.

In order to examine the performance of the combustor, combustion experiment is conducted in a vacuum environment for thermal isolation. A coaxial pipe heat exchanger is used for exhaust heat recovery, and aluminum radiation shield is placed inside of the chamber. Temperature distribution of the bottom surface of the combustor is measured with a radiation thermometer (FTZ6, Japan Sensor Corp.) through a CaF_2 window. The exhaust gas is monitored with a CO_2 gas analyzer (Model 4300, Nippon Thermo) to calculate the fuel conversion efficiency. Pd catalyst is placed at $0 < r < 10$ mm and $7 < r < 10$ mm. The equivalence ratio is kept at 0.95, and the n-butane flow rate is changed with time. The combustor is preheated by joule heating up to 300 °C.

Figure 3 shows time traces of the wall temperature at the center, the chemical energy input, the electrical input, and the heat generation in Case 1. The heat generation is calculated with the butane flow rate and the conversion efficiency. When $Q_B > 5$-10 sccm, self-sustained combustion has been achieved. In Case 1, almost complete fuel conversion has been achieved up to $Q_B = 10$ sccm, corresponding to extremely large heat generation density of 2.1×10^8 W/m^3, which is based on the volume of the combustion chamber. The wall temperature is increased with Q_B, and reaches 850 °C at $Q_B = 30$ sccm with heat generation density as large as 5.2×10^8 W/m^3.

Figure 4 shows the radial temperature distribution in comparison with the CFD results. The emissivity of the bottom surface of ceramic substrate and the metal ring is assumed to be 0.94 and 0.3 respectively. By considering reflection from the radiation shield, the emissivity of the top surfaces are assumed to be half of that of the bottom surface.

For the combustor with the catalyst layer of $7 < r < 10$ mm, the temperature gradient for both $Q_B = 20$ and 30 sccm becomes much smaller as in the CFD results. Therefore, the temperature distribution of the combustor can be much improved with the modification of catalyst arrangement regardless of fuel flow rate.

However, the temperature level of the experimental data is much lower than the CFD results. This is because the heat loss is underestimated in the present CFD, in which only radiative heat transfer is assumed.

In summary, a micro-scale ceramic catalytic combustor has been developed using a high-precision tape-casting technology and Pd/nano-porous alumina catalyst fabricated through anodic oxidation. Combustor geometry and catalyst arrangement with minimum wall temperature gradient has been proposed based on a series of CFD analysis. In combustion experiments, radial temperature gradient is significantly reduced with the localized catalyst arrangement in the combustion chamber.

References
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[3] Wang, X., Zhu, J., Bau, H. & Gorte, R.J. 2001 Catal. Lett., 77, pp.173-177.
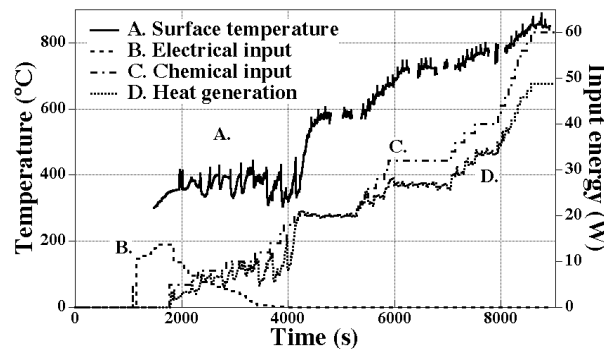[4] Sai, H. & Yugami, H. 2004 Appl. Phys. Lett. 85 pp.3399-3401.

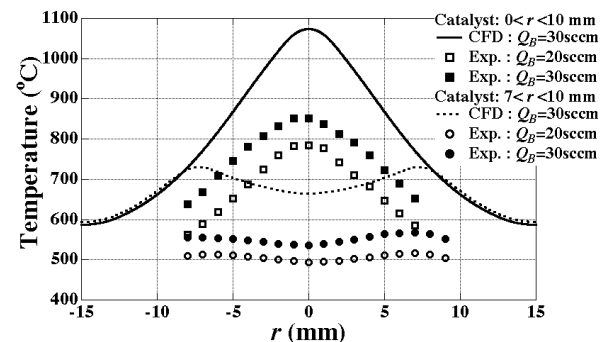Figure 3. Time trace of surface temperature and input energy (Catalyst $0 < r < 10$mm).

Figure 4. Wall temperature distribution.