Experimental Evaluation of Micro Heat Exchanger Using Catalytic Combustion

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Abstract
Catalytic micro heat exchangers were fabricated with stainless steel plates. Microchannels having 0.3-0.5 mm of width, 0.2 mm of depth and 20 mm of length were formed on 40 × 40 × 0.3 mm^3 of metal plates by photolithography and chemical etching. The metal plates with microchannels were stacked and then brazed to seal up the gaps between the metal plates. Alumina to support palladium catalyst was deposited on the microchannels of the brazed metal plates by sol-gel process. The coated alumina was present in 5-10 µm of thickness, which was not uniform in the alumina layers. The fabricated micro heat exchangers were evaluated in the efficiency of heat exchange with or without catalytic combustion of LPG. In the heat exchange without catalytic combustion, heat exchange efficiency increased with decreasing Reynolds number of heating air and increasing that of cooling air. This result suggested that the heat exchanger without catalytic combustion could be operated at high Reynolds numbers of heating and cooling air in order to obtain high heat transfer rate as well as high efficiency. On the other hand, in the heat exchange with catalytic combustion, there was a region to operate the micro heat exchanger, where the stable LPG combustion was obtained by adjusting the inlet concentration of LPG and the Reynolds number of the cooling air. The heat exchange efficiency of the heat exchange with catalytic combustion was lowered at high inlet concentration of LPG.

Keywords: heat exchanger, microchannels, palladium catalysts, LPG combustion

1 INTRODUCTION
As microstructured reactors’ benefits include their inherent safety, improved process control, and rapid implementation, much consideration has been paid to micro structured reactors. Especially, microstructured reactors give high surface-to-volume ratios which lead to efficient heat and mass transfer. Thus, microstructured reactors have been studied for many chemical reactions such as partial oxidation where kinetically fast reactions must be quenched to prevent full oxidation, reactions which are highly exothermic or involve explosive mixtures and reactions which involve toxic precursors or products. Janicke et al. [1] combined a microstructured reactor and heat exchanger in order to control hydrogen oxidation from an explosive gas mixture. A microstructured reactor/heat exchanger was applied for measuring reaction kinetics of ammonia oxidation in an explosive region [2]. Another application of microstructured reactor/heat exchanger would be micro fuel processors for micro fuel cell development. Most micro fuel processors require providing heat to produce hydrogen from fuel by steam reforming. Thus, it has been studied to generate heat by burning a part of fuel fed and provide a steam reformer with combustion heat.

The microchannel reactors and heat exchangers have been made of mechanically microstructured metal foils of stainless steel, hastelloy, aluminum, copper, palladium, silver, and others [3]. A reactor is assembled from individual metal plates with microchannels having cross sections in the range of 50–300 µm. These metal plates are assembled by bolting, gaskets such as graphite, or diffusion bonding. On the other hand, for a wider application with chemical reactions it is necessary to provide oxide coatings as carriers for metal catalysts to increase the overall inner surface area of the microchannels [3]. Several techniques have been utilized for coating microchannels with porous oxides: anodic oxidation which has been used to provide a porous layer on aluminum [3,4]; chemical vapor deposition [1]; deposition of nanoparticles [6]; and sol–gel process [3].

This study introduces vacuum brazing method for assembly of microchanneled stainless steel plates. Also micro heat exchanger having microchannels coated with Pd/γ-Al₂O₃ catalyst is evaluated in its heat exchange efficiency and heat transfer rate.

2 EXPERIMENTAL

2.1 Fabrication and Catalyst Coating
In the design of the micro heat exchanger, uniformity in fluid velocity distribution was considered, referring to Commenge et al. [7]. Main design factors were curvature...
ratio at the end of microchannels, ratio of depth to width in microchannels, number of microchannels, length of microchannels and area ratio of wall to microchannels. Top and bottom covers were prepared by machining in 4 × 4 cm² of size. Microchannels were created by photolithography and chemical etchings and the details for microchanneled plates are summarized in Table 1. For assembly of the microchanneled plates, they were stacked up for a cross-flow heat exchanger and nickel foils were inserted between the microchanneled plates for vacuum brazing. The assembled microchanneled plates were tested for leakage with a leak detector (ASM 142, Alcatel). For incorporation of catalysts, a microchanneled stainless steel plate was washcoated with a mixture of alumina sol and Pd/γ-Al₂O₃ powder catalyst into the microchannels, followed by drying in air at 120 °C overnight.

Table 1. Details of microchanneled stainless steel plates

<table>
<thead>
<tr>
<th>Material</th>
<th>Stainless Steel</th>
</tr>
</thead>
<tbody>
<tr>
<td>Geometry</td>
<td>40 × 40 × 0.3 mm³</td>
</tr>
<tr>
<td>Microchannel area</td>
<td>20 × 20 mm²</td>
</tr>
<tr>
<td>Microchannel width</td>
<td>300 µm</td>
</tr>
<tr>
<td>Microchannel depth</td>
<td>200 µm</td>
</tr>
<tr>
<td>Number of channels</td>
<td>34</td>
</tr>
</tbody>
</table>

2.2 Performance of Micro Heat Exchanger

The catalytic micro heat exchanger was characterized in an experimental set-up. First, pressure drop was measured with varying flowrate of air. Heat-exchanging performance without catalytic combustion was tested by measuring temperatures of inlets and outlets of heating and cooling air. The heater was controlled to obtain a desired temperature of the heating air. A quantity characterizing the performance of a heat exchanger is the efficiency, which is the ratio of the actual heat transfer rate for a heat exchanger to the maximum possible heat transfer rate. The efficiency of the heat exchanger without catalytic combustion (E) was calculated with equation (1).

\[
E(\%) = \frac{V_2(T_{out,2} - T_{in,2})}{V_1(T_{in,1} - T_{out,1})} \times 100
\]

where E is the efficiency of the heat exchanger (%), V is flowrate of air (l/min), T_{out,2} and T_{in,2} are temperatures of outlet and inlet of cooling air, and Q_{combustion} is heat of LPG combustion. C_p and \( \rho \) are heat capacity and density of air (1 kJ/kg·K and 0.93 kg/m³), respectively.

\[
E_c(\%) = \frac{C_p \rho V_2(T_{out,2} - T_{in,2})}{Q_{combustion}} \times 100
\]

where E_c is efficiency of the heat exchanger with catalytic combustion (%), V_2 is flowrate of cooling air (l/min), T_{out,2} and T_{in,2} are temperatures of outlet and inlet of cooling air, and Q_{combustion} is heat of LPG combustion. C_p and \( \rho \) are heat capacity and density of air (1 kJ/kg·K and 0.93 kg/m³), respectively.

3 RESULTS AND DISCUSSION

3.1 Heat Exchanging without Catalytic Combustion

Figure 1 exhibits pressure drop of air in microchannels as a function of Reynolds numbers calculated by equation (3). As flowrate of air is varied from 7 – 26 l/min, Reynolds numbers are less than 65, which reveals that the flow pattern of air in microchannels is present in a laminar regime.

\[
Re_{Dh} = \frac{uD_h}{\mu}
\]

where u is velocity in microchannels (m/s), D_h is hydraulic diameter (0.24 mm), \( \rho \) is density of air (1.1614 kg/m³), and \( \mu \) is viscosity of air (184.6×10⁻⁷ N·s/m²). Furthermore, for laminar flow (Re_{Dh} < 2300), the hydrodynamic entry length may be obtained from an expression of the following form.

\[
\frac{x_{fd,h}}{D_h} \approx 0.05 Re_{Dh}
\]

where \( x_{fd,h} \) is hydrodynamic entry length (mm), and D_h is hydraulic diameter (0.24 mm). In this study, as the

![Figure 1. Pressure drop against Reynolds number of air in microchannels.](image-url)
hydrodynamic entry length exists in a range of 0.2 - 0.8 mm which is 1 - 4 % of the whole length of the microchannels, it is considered that the laminar flow of air in the microchannels is fully developed at the entrance of the microchannels. Heat exchange efficiency (E) without catalytic combustion increases with raising Reynolds number of cooling air and lowering that of heating air (see Figure 2). However, it is worth noting that heat transferred to the cooling air increases with large Reynolds numbers of heating and cooling air, where the heat exchange efficiency (E) is also high (see Figure 3). Halbritter et al. [8] observed similar behavior of heat exchange in a crossflow micro heat exchanger. It was reported that the efficiency decreases with increasing flowrate of cooling water and heat transfer rate increases with increasing flowrate of cooling water.

### 3.3 Heat Exchange with Catalytic Combustion

It is necessary that operation conditions should be found for heat exchange with catalytic combustion. First, it is found that LPG concentration should be adjusted to obtain a stable reaction temperature at least for 2 h of reaction time. The LPG concentration is affected by Reynolds number of heating air as shown in Figure 4 (line ABCD). Even though the theoretical equivalent concentration of LPG for its combustion is about 40000 ppm based on the reaction equation (5), the LPG concentration for a stable reaction temperature is less than 10400 ppm. The LPG concentration above the line ABCD causes likely overheating of the heat exchanger, and it is also difficult to maintain the reaction temperature at the LPG concentrations below the line ABCD. Thus, the LPG concentration for heat exchange with cooling air is kept to be the values on the line ABCD.

\[
C_3H_8 + 5(O_2 + 3.76 N_2) \rightarrow 3CO_2 + 4H_2O + 18.8N_2 \tag{5}
\]

It is observed that there is a limitation of Reynolds number of the cooling air under the conditions for a stable reaction temperature as shown in Figure 4 (line EFGH). At the Reynolds numbers of the cooling air above line EFGH, the reaction temperature decreases rapidly below an ignition temperature of LPG combustion, which may result from that heat removed by the cooling air is larger than that generated by LPG combustion. Thus, it is suggested that the micro heat exchanger with catalytic combustion should be operated at a condition in the surface AEGFHDCA.
On the other hand, heat transferred into the cooling air increases with Reynolds number of the cooling air in microchannels as shown in Figure 5, which is similar to the result from the heat exchange without catalytic combustion. Heat generated by LPG combustion is calculated with LPG conversion, and the generated heat increases with increasing inlet concentration of LPG (see Figure 6 (a)). Heat transfer rate of the cooling air is lowered at high inlet concentration of LPG as shown in Figure 6 (b), which is influenced by flowrate of the cooling air. Heat exchange efficiency ($E_c$) is 40 - 85 % in this study (see Figure 6 (c)). It is also found that the heat exchange efficiency ($E_c$) is low at high inlet concentration of LPG due to low heat transfer rate of the cooling air in spite of high heat generation by LPG combustion. Thus, it is considered that optimal operation conditions of inlet concentration of LPG and flowrate of cooling air would be present for the micro heat exchanger with catalytic combustion.

4 CONCLUSIONS

Microstructured stainless steel plates were successfully brazed with nickel filler under vacuum. It was also demonstrated that microchannels could be coated with Pd/γ-Al$_2$O$_3$ catalyst after brazing stainless steel plates. In the heat exchange without catalytic combustion, heat exchange efficiency increased with decreasing Reynolds number of heating air and increasing that of cooling air. However, it was suggested that the heat exchanger without catalytic combustion could be operated at high Reynolds numbers of heating and cooling air in order to obtain high heat transfer rate as well as high efficiency. On the other hand, in the heat exchange with catalytic combustion, there was a region to operate the micro heat exchanger, where the stable LPG combustion was obtained by adjusting the inlet concentration of LPG and the Reynolds number of the cooling air. The heat exchange efficiency of the heat exchange with catalytic combustion was lowered at high inlet concentration of LPG. Thus, it is considered that optimal operation conditions of inlet concentration of LPG and flowrate of cooling air would be present for the micro heat exchanger with catalytic combustion.

REFERENCES