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Improving temperature uniformity and performance of CO preferential oxidation for hydrogen-rich reformat with a heat pipe[☆]

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ABSTRACT

Preferential oxidation (PROX) is an effective, but highly temperature-sensitive, method of CO removal for hydrogen-rich reformates. In a packed-bed catalytic reactor, oxidation is strongest at the inlet side and the local catalyst pellets become over-heated with poor heat conduction. As a result, the enhanced parasitic H₂ oxidation consumes oxygen and suppresses CO conversion. This study applies a heat pipe to improve the temperature uniformity in a tubular one-stage packed-bed reactor by transporting heat downstream and thereby improve CO removal. In the experiments, the fuel mixture containing 2% of CO, 75% of H₂, and 23% of CO₂, further mixed with air at O₂/CO = 0.75, 1.0 or 1.25, is supplied with stepwise increase of feeding rate under a fixed environmental temperature of 99 ± 1 °C. The proposed simple method is found to significantly improve temperature uniformity and CO removal for the present test conditions with O₂/CO = 1.0 and 1.25.

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1. Introduction

The problems associated with hydrogen storage and safety limit practical hydrogen utilization. Reforming of liquid hydrocarbons provides safe and movable on-site hydrogen generation. Practically, the presence of poisonous carbon monoxide (CO) precludes direct charge of the reformat into PEM fuel cells [1]. A water–gas-shift reactor may be needed first to lower CO concentration to about 0.5–2%. To further remove CO to an acceptable level, less than 10 ppm for PEMFC, preferential oxidation (PROX) is a cheap and promising method [2–12]. However, the CO conversion and selectivity of PROX reactions are highly sensitive to reaction temperature. At

higher temperatures, the CO coverage on the catalyst surface gives way to H₂ and results in increased parasitic H₂ oxidation [13]. In packed-bed catalytic reactors, oxidation reactions of CO and H₂ are most intense at the reactor's inlet. The poor heat conduction of the catalyst support, e.g., alumina, or zeolite, etc., makes local catalyst pellets over-heated. At high local temperatures, the raised oxidation rates of CO and H₂ further increase the local temperature and deteriorate the performance. This phenomenon is especially serious at high feeding rates. Even though the reactors are equipped with temperature controllers, the local hot region exists due to the poor thermal conduction. Staged air-feeding has been used to reach significant improvements in CO removal [3,14,15]. When the amount of oxygen is split between stages in some proper ratio, the

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lower oxygen content in each stage reduces the possibility of H₂ oxidation. As a result, both CO conversion and CO selectivity can be improved. However, this method usually needs complex control systems of temperature and feeding [12]. The present study proposes an alternative convenient method to cool the over-heated inlet side of the PROX reactor by embedding a heat pipe. The heat pipe is a simple and cheap phase-change device which effectively delivers the heat generated at the inlet side to the downstream part. As the temperature is lowered at the inlet side, the parasitic H₂ oxidation can be suppressed over the reactor. In this work, we will compare the CO conversion performance in a tubular one-stage PROX reactor with and without a heat pipe. Under different feeding rates, the CO conversions will also be compared to find out the conditions when the heat pipe is favorable. In addition, longitudinal temperature distributions in the reactor are measured. To our knowledge, internal temperature distributions of packed-bed PROX reactors have not been examined in the literature. Such characteristics can be influential to the overall CO conversion performance since the internal temperature distribution of packed-bed reactors is highly non-uniform, even under temperature control of the integral reactor.

2. Experimental

Fig. 1 shows the schematic layout of the experiments. The bottled sample gas has a composition of 2% CO, 75% H₂, and 23% CO₂, which simulates the reformat of methanol steam reformers. After mixing with air at a selected O₂/CO ratio, the mixture enters the PROX reactor. Three different O₂/CO ratios, 0.75, 1.0 and 1.25, are tested. The feeding rates before mixed with air range from 30 mL s⁻¹ to 60 mL s⁻¹, measured by an electric flow meter (Omega Inc., FMA1814). The space velocities for different test conditions are listed in Table 1. The product gas is sampled and analyzed for CO concentration by a gas chromatograph (GC, China Chromatography Co.) having a CO resolution of 2.5 ppm. The structure of the tubular PROX reactor embedded with a heat pipe is shown in Fig. 2. The reactor is made of a copper tube with 31.75 mm OD and 25.75 mm ID. The copper/water heat pipe is ϕ 6 mm and 120 mm long. The heat pipe is slanted at 15° with the evaporator downward to assist the return of the condensed water. Thermocouples (K-type, Omega, Inc.) T1 to T3 are placed adjacent to the inner surface of the copper tube. Thermocouples T4 to T6, embedded midway between the heat pipe and the copper tube wall, are used to monitor the longitudinal temperature distribution within the reactor. When the heat pipe is absent, thermocouples T4 to T6 are placed along the tube center. The resolution of the temperature measurement

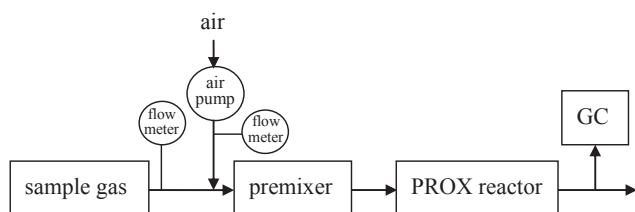


Fig. 1 – Schematic test layout.

Table 1 – Space velocities of different test conditions.

Flow rate [mL s ⁻¹]	Space velocity [h ⁻¹]	
	O ₂ /CO	
	1.0	1.25
30	3564	3645
40	4752	4860
50	5940	6075
60	7128	7290

is 0.1 °C. All data are taken at steady state when the temperature distribution and the CO concentration remain unchanged. The PROX reactor is immersed in a thermostat water bath at 99 ± 1 °C.

The catalyst pellets, 40 g in mass and 0.033 L in volume, are the commercial product TSSA-5 of Tanaka, Co. The TSSA-5 Ru–Al₂O₃ pellets, 1–3 mm in diameter, have 0.4 wt% (4.8 g L⁻¹) of Ru. Fig. 3 shows the CO and CH₄ yields measured by the producer [16]. Within 70–120 °C, CO can be fully removed for a sample gas of 0.61% CO, 59.00% H₂, 1.22% O₂, 16.00% CO₂, and balance N₂ at a space velocity 6600 h⁻¹. As commonly known, Ru-based catalyst yields considerable methanation [10] especially at high temperatures. But since CH₄ is non-poisonous to PEMFC [17], the disadvantage is only a slight reduction of H₂.

The CO conversion is determined by

$$\text{CO conversion} = \frac{[\text{CO}]_{\text{in}} - [\text{CO}]_{\text{out}}}{[\text{CO}]_{\text{in}}} \times 100\%$$

The CO selectivity is not determined in this work. One reason is that the main objective is to study the improvement of CO removal by applying heat pipes in packed-bed PROX reactors. Also, the Ru–Al₂O₃ catalyst has been known to have a high CO selectivity [2]. This will be exhibited in our results that high CO conversions can be obtained for a CO concentration of 2%, with the help of heat pipes, at O₂/CO ratios of 1.0 and 1.25.

3. Results and discussion

Fig. 4 compares the steady-state temperature distributions with and without a heat pipe at O₂/CO = 1.0 for two representative feeding rates, 30 mL s⁻¹ and 60 mL s⁻¹. These results are measured by thermocouples T4 to T6 embedded within the catalyst bed. The results of T1 to T3 near the inner surface

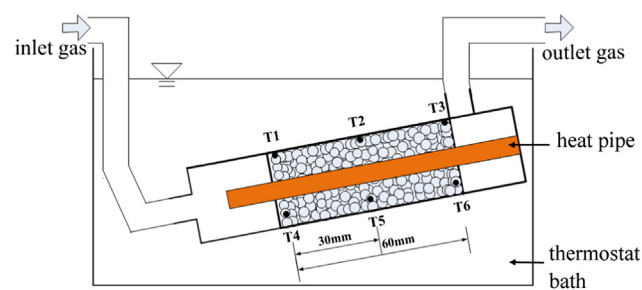


Fig. 2 – Structure of the heat-pipe-embedded PROX reactor in a thermostat water bath.

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