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Design, scale-out, and operation of a microchannel reactor with a Cu/CeO_{2-x} catalytic coating for preferential CO oxidation

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ABSTRACT

A preferential CO oxidation device consisting of an array of 26 parallel microchannel reactors with each a 5 wt% Cu/CeO_{2-x} catalytic coating was designed for application with a 100 W_e fuel cell system. The reactor device is able to reduce the carbon monoxide concentration in a realistic reformate gas from 1.5 vol.% to 10 ppm at an inlet oxygen to carbon monoxide ratio of 1.5, a WHSV of 240 $L\,g^{-1}\,h^{-1}$, and within the temperature range of 230–240 °C. The device outperformes similar reactor designs as presented in the literature.

1. Introduction

The low-temperature proton-exchanged membrane fuel cell (PEM FC) is an attractive power source for transport and portable applications. One of the promising methods of hydrogen production for application in a PEM FC is the on-board multi-stage process including reforming of fuels such as hydrocarbons, alcohols or ethers, followed by the CO water gas shift (WGS) reaction. Typically, the obtained hydrogen-rich gas contains 0.5–1.0 vol.% CO, in some applications (for example, after methanol or DME steam reforming) the low level of the outlet CO concentration (up to 2–3 vol.%) can be directly attained without WGS stage. CO poisons the FC anode catalyst and must be removed to a level below 10 ppm. Preferential oxidation (PrOx) of CO is an efficient method for CO removal from a hydrogen-rich gas mixture.

Several review papers [1–5] report on different types of PrOx catalysts developed and investigated over the last decade by various research teams. These catalysts can be classified into three groups: (i) supported platinum metal (Pt, Ru, Rh) based catalysts, (ii) supported Au-containing catalysts, (iii) and copper–cerium oxide based catalysts. Although considerable progress has been made towards the development of active and selective catalysts, the removal of CO to a level below 10 ppm and the simultaneous minimization of hydrogen losses are achievable only in a narrow temperature interval. For example, the suitable temperature window on platinum, gold or copper–cerium oxide based catalysts is usually less than 40 °C. Thus, thermal management is a key issue for efficient reactor operation in terms of highly selective CO reduction to levels acceptable for PEM FC application.

Microchannel reactors are characterized by high heat and mass transfer rates and therefore show large promise in the development of miniature fuel processors for small scale electricity production [6]. For portable power generation, microreactors may outperform conventional reactor designs, leading to lower reactor volumes and weights [7]. Several advantages of applying microreactors in comparison with conventional fixed-bed technology for the highly exothermic CO PrOx reaction were demonstrated in different studies [7–11]. A high rate of heat removal from the thin catalytic film deposited onto the microchannel walls allows for near-isothermal operation and in this way prevents the onset of side reactions (viz., hydrogen oxidation or reverse WGS) and extends the operational window [8–10].

To date, several PrOx microchannel reactor designs were proposed [8–16]. These microreactors were fabricated and tested using different catalyst washcoat compositions, such as Pt/Al_2O_3 [8,9,12,13], $Pt–Co/Al_2O_3$ [12,14], $Pt–Ru/Al_2O_3$ [12,15], $Pt–Rh/Al_2O_3$ [12], Ru/Al_2O_3 [12], Rh/Al_2O_3 [11,12], Au/CeO_2, $Au/\alpha\text{-}Fe_2O_3$ [16], and CuO/CeO_2 [10,16].

According to our previous work [10], the copper–cerium oxide catalytic system is a very promising one for efficient CO removal from a hydrogen-rich gas using microreactor technology. The selec-

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tivity of the copper–cerium oxide catalyst is considerably higher as compared to that of Pt metal supported catalysts at a similar level of CO conversion. The stability is also considerably higher [17,18] as compared to Au-containing catalysts. The latter may rapidly decrease their activity under reaction conditions. Furthermore, the price of the copper–cerium oxide catalyst may favorably compete with platinum containing catalysts.

The optimum temperature range for the CO PrOx reaction over the Cu/CeO_{2-x} catalyst is close to that for the low-temperature water gas shift reaction [10]. This facilitates the easy integration of CO PrOx and WGS microreactors without the need of an intermediate heat-exchanger to achieve higher efficiencies in the overall fuel processing.

In this paper, we present experimental results on the design, scale-out, and operation of a preferential CO oxidation microdevice, consisting of a numbered-up array of 26 microchannel reactors running in parallel. In this design, the external surface area of the individual microchannel reactors is used for heat-exchange with the environment to maintain the desired operation temperature. Thus, the heat flux via the external surface areas of the parallel microchannel reactors and the fittings is equal to the heat produced by the reaction. While this design does not allow to recover the heat produced by the PrOx reaction, it is simpler and cheaper as compared with integrated microreactor/heat-exchanger configurations as proposed by Cominos et al. [12] and Delsman et al. [14,15]. This paper presents different aspects of the CO PrOx microreactor assembly design as well as experimental results from catalyst testing.

2. Experimental

2.1. Substrate and washcoat preparation

The 5 wt% Cu/CeO_{2-x} coatings were prepared as described elsewhere [10]. Stainless steel platelets were manufactured using a commercial photoetching technique based on wet chemical etching with an aqueous iron trichloride solution. The platelets each having 14 semicylindrical channels of 500 μm width, 250 μm depth, and 25 mm length were first cleaned with isopropanol for 10 min in an ultrasonic bath and after drying calcined at 800 °C for 2 h. This treatment presumably generates a thin oxide surface on the channel walls and good adhesion of oxide materials is obtained. A polymer film was used to cover the metal parts of the surface that were not intended to be coated.

The ceria support with a BET surface area of $80\,m^2\,g^{-1}$ was prepared by calcination of $Ce(NO_3)_3{\cdot}6H_2O$ at 400 °C for 2 h in air. In a first step, 1.7 g polyvinyl alcohol (binder) was dissolved in 25 g deionized water by smoothly stirring with a magnetic stirrer at 65 °C for 2 h and left without stirring overnight. Then 7 g CeO_2 powder and 0.3 g acetic acid were added successively without stirring. The resulting suspension was stirred at 65 °C for 2 h, cooled to room temperature and stirred for 3 days.

The microchannels were completely filled with the prepared suspension and excess suspension was wiped off. After drying at 110 °C in air, the platelets were calcined at 400 °C for 2 h. To produce the 5 wt% Cu/CeO_{2-x} catalyst, the washcoated platelets were impregnated with the required quantity of an aqueous solution of $Cu(NO_3)_2$, then dried at 110 °C in air, and calcined at 400 °C for 2 h. The obtained coatings demonstrated good impact resistance and were still intact after contact with water [10]. For the catalyst tests, 52 coated platelets were prepared. The average amount of the catalyst coating deposited on a single platelet was 14 ± 4 mg.

Sets of two platelets with the 5 wt% Cu/CeO_{2-x} coating were sealed by laser welding to create circular microchannels. Inlet and outlet tubes were attached to the microplatelets by laser welding to create a sandwich type of microchannel reactor (see Fig. 1) with dimensions of 4 mm × 14 mm × 50 mm.

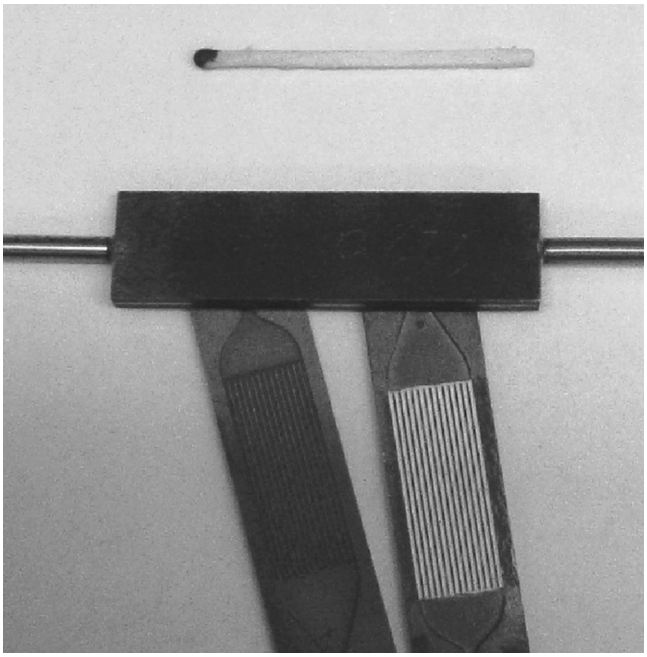

Fig. 1. A single laser welded microchannel reactor (top) and the corresponding platelets with 5 wt% Cu/CeO_{2-x} (bottom left) and CeO_2 (bottom right) washcoats. The platelets have 14 semicylindrical channels of 500 μm width, 250 μm depth, and 25 mm length.

Using the described procedure, 26 microchannel reactors were manufactured and tested in the CO PrOx reaction. The catalyst weight distribution in these microchannel reactors is shown in Table 1; the total catalyst loading was 732 mg. No deactivation of the copper–cerium oxide catalyst was observed and the catalyst retained stable activity for more than 300 h on stream.

2.2. Catalytic activity study

2.2.1. Catalytic activity tests with single microchannel reactors

For the catalytic activity screening, the prepared microchannel reactors were tested individually. Each microchannel reactor was placed in a holder heated by a heating cartridge. Three chromel–alumel thermocouples were placed at the outer surface of the reactor along the reactor centerline at distances of 1, 12, and 24 mm from the beginning of the washcoat layer. The corresponding temperatures are referred to as T_1, T_2, and T_3. The difference between the measured temperatures T_1, T_2, and T_3 was always below 1 °C in the whole temperature range studied. Temperature T_2 will be taken as the reactor temperature, hereafter.

Table 1
Catalyst weight distribution in the individual microchannel reactors.

	Catalyst weight (mg)	Number of microchannel reactors
	23	1
	25	9
	26	4
	27	2
	28	2
	29	1
	30	2
	31	3
	32	2
	33	3
Total	732	26

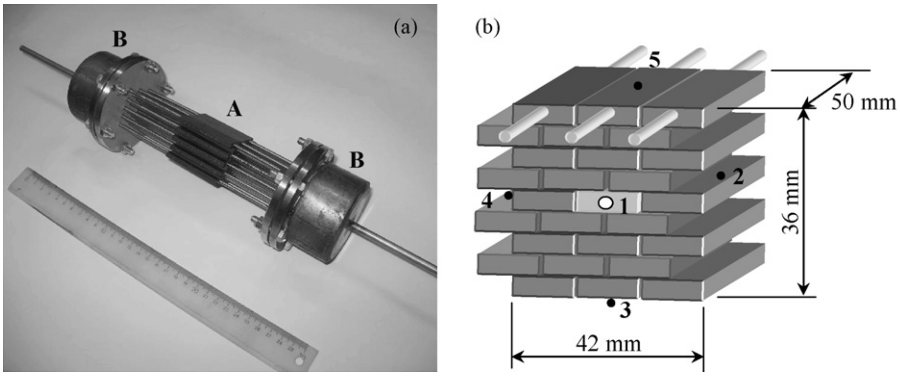

Fig. 2. Microreactor assembly (A) with gas distribution fittings (B) (a) and schematic view of the microreactor assembly (b). Temperature measurement points are indicated by black dots and are numbered for reference. The light gray rectangle with the white circle at the center of the microreactor assembly designates the stainless steel platelet with the channel for the T_{R1} thermocouple. For reasons of clearness the inlet and outlet tubes are only shown for the first microchannel reactor layer.

The experiments were performed with the following gas feed compositions: 0.5–3.0 vol.% CO, 0.25–4.5 vol.% O_2, 57 vol.% H_2, 10 vol.% H_2O, 20 vol.% CO_2, with He as balance. The total feed flow rate was varied in the range of 0.02–0.4 L min^{-1} (STP) to achieve a Weight Hour Space Velocity (WHSV) in the range of 50–1000 L g^{-1} h^{-1}.

2.2.2. Microreactor assembly tests

Fig. 2a presents a general view of the microreactor assembly (A) with the gas distribution fittings (B). For inlet reformate gas stream preheating to a temperature in the range of 150–250 °C (depending on the experimental conditions), the inlet gas distribution fitting was heated by a heating cable. The gas temperature (T_{gas}) was measured by a 0.2 mm thick chromel–alumel thermocouple located inside the inlet gas tube.

Fig. 2b illustrates the arrangement of all 26 microchannel reactors in the assembly and the thermocouple locations. The assembly consists of 9 layers. There were 3 microreactors per layer in layers 1–4 and 6–9 and 2 microreactors in layer 5. The even layers were shifted laterally against the odd layers by 7 mm for easy assembling. A stainless steel platelet with a thermocouple channel was placed in the middle of the 5th reactor layer (see a light gray rectangle with a white circle at the center in Fig. 2b). The outside dimensions of this thermocouple platelet were the same as those of each of the microchannel reactors. A 0.2 mm thick chromel–alumel thermocouple (T_{R1}) was placed into the central platelet at a distance of 25 mm from the beginning of the channel. The metal temperature at the outer walls of the microreactor assembly was measured by four external thermocouples (T_{R2}, T_{R3}, T_{R4} and T_{R5}). The difference between the measured temperatures was always below 5 °C in the whole range of temperatures studied under steady-state conditions. Temperature T_{R1} will be taken as the microreactor assembly temperature, hereafter.

The microreactor assembly was operated in an integral mode during the catalytic activity measurements to examine the effects of temperature and space velocity on catalyst activity and stability. Tests were carried out under atmospheric pressure and at total flow rates of 1, 2 and 3 L min^{-1} (STP) which corresponded to a WHSV of 80, 160, and 240 L g^{-1} h^{-1}, respectively. In these experimental runs the microreactor assembly was heated by an electrical heater. The experiments were performed with the feed simulating a realistic reformate stream composition: 1.5 vol.% CO, 2.25 vol.% O_2, 57 vol.% H_2, 10 vol.% H_2O, 20 vol.% CO_2, with He as balance.

The external dimensions of the microreactor assembly excluding fittings and insulation are 49 mm × 50 mm × 36 mm. The volume and the weight of the assembly are 88 cm^3 and 520 g, respectively.

For experiments without electrical heating, the microreactor assembly was covered with a double-layer insulation to reduce heat losses to the environment. The inner layer was a 0.5 mm thick glass fiber and the outer layer was a 10 mm thick glass wool material. The preferential oxidation reaction produced 26 W of heat at a total flow rate of 3 L min^{-1} (STP) corresponding to the amount of hydrogen needed for a 100 $\mathrm{W_e}$ PEM FC. The heat losses to the environment from the non-insulated assembly amounted to 57 W in the temperature range of 200–230 °C as it was determined in a separate experiment. The 0.5 mm thick glass fiber insulating layer reduced the heat losses by 50%. The remaining heat losses were exactly matched by the heat released in the PrOx reaction. For the reactor start-up, the following procedure was applied. An electrical heater was used to preheat the reactor to 160 °C in order to prevent water condensation at the beginning of the start-up procedure. Then the electrical heater was switched off and it was replaced with the double-layer insulation mentioned above. The reformate stream was preheated to 215 °C before entering the microreactor assembly. The outer insulation layer was removed when the O_2 conversion reached 100%.

2.3. Gas analysis

The inlet and outlet concentrations were determined using a Kristall 2000 chromatograph equipped with molecular sieves NaX and Porapak Q columns and with thermal-conductivity (TCD) and flame-ionization (FID) detectors. The combination of a methanator (containing a reduced NKM-4 nickel catalyst) and the FID allowed highly sensitive analysis of CO, CO_2, and the hydrocarbons in the gas mixture. The detection limit of the CO, CH_4, and CO_2 concentrations was 1 ppm, and that of the O_2 concentration was 10 ppm. The conversions of CO (X_{CO}) and O_2 (X_{O_2}), and the selectivity towards oxygen consumption (S) were calculated from the following equations:

$$X_{CO} = \frac{[CO]_{inlet} - [CO\ ]_{outlet}}{[CO]_{inlet}} 100\% \tag{1}$$

$$X_{O_2} = \frac{[O_2]_{inlet} - [O_2]_{outlet}}{[O_2]_{inlet}} 100\% \tag{2}$$

$$S = \frac{1}{2} \frac{[CO]_{inlet} - [CO]_{outlet}}{[O_2]_{inlet} - [O_2]_{outlet}} 100\% \tag{3}$$

where $[CO]_{inlet}$ and $[O_2]_{inlet}$ are the inlet concentrations of CO and O_2, respectively; $[CO]_{outlet}$ and $[O_2]_{outlet}$ are the outlet concentrations of CO and O_2, respectively.

3. Results and discussion

3.1. Catalytic activity tests in each single microchannel reactor

To determine the optimum reaction conditions, the catalyst performance of each single microchannel reactor was examined as a function of the temperature, the inlet CO concentration, the O_2/CO ratio, and the space velocity. These tests were performed with all 26 microchannel reactors.

The outlet CO concentration, the O_2 conversion, and the selectivity are shown in Fig. 3 as a function of the reactor temperature at different WHSVs. It can be seen that the conversion curves shift to higher temperature with increasing the space velocity. As the reactor temperature increased, the CO concentration decreased, reached a minimum at an optimum temperature and then increased. At a WHSV of 250 $L\,g^{-1}\,h^{-1}$, this optimum temperature was 230 °C, corresponding to a CO concentration of 4 ppm. As the WHSV increased to 470 and then to 660 $L\,g^{-1}\,h^{-1}$, the optimum temperatures were 250 and 255 °C, corresponding to CO concentrations of 19 and 25 ppm, respectively. Full oxygen conversion at the reactor outlet was observed at these temperatures at a selectivity of 33.3%. A further increase of the temperature to 270 °C resulted in a dramatic increase in the outlet CO concentration up to 250 ppm, with a corresponding decrease of the selectivity to 32.8% at complete oxygen conversion at all flow rates. These results are in good agreement with our earlier data reported in [10]. For comparison, several microchannel reactors with different catalyst loadings were tested. The microchannel reactors loaded with 23 and 31 mg of catalyst showed similar performances at the same WHSV. The separate screening tests with each of the 26 microchannel reactors gave the same results. The deviations were small and the shifts of the conversion curves at the same WHSV were within 5 °C.

The activity and selectivity data at different O_2/CO ratios and inlet CO concentrations were in line with the earlier results [10] as well. Summarizing, as the flow rate increases, the highest reachable CO conversion decreases while the optimum reactor temperature, corresponding to the highest CO conversion, shifts to higher values. The CO conversion does not depend on the CO inlet concentration. It does depend on the O_2/CO ratio. Increasing the O_2 concentration increases the highest reachable CO conversion and increases the optimum temperature. For oxygen conversions below 100%, an increase of the inlet O_2 concentration does not change the amount of O_2 reacted, and therefore does not change the selectivity. This is because both the hydrogen and CO oxidation rates have zero order in the O_2 concentration. Thus, an increase of the inlet O_2 concentration allows to reach higher CO conversion and expands the PrOx operational window (where the CO outlet concentration is below 10 ppm, $\Delta T_{10\,\text{ppm}}$), with a simultaneous decrease of the selectivity. The area below a curve in Fig. 4 demonstrates the PrOx operational window ($\Delta T_{10\,\text{ppm}}$) as a function of the WHSV at the $[O_2]_{\text{inlet}}/[CO]_{\text{inlet}}$ ratios of 0.8, 0.9, 1.0, and 1.5 for the inlet CO concentrations in the range of 0.5–3.0 vol.%. It can be seen that the PrOx operational window becomes wider at higher oxygen concentration and lower WHSV. For example, at a carbon monoxide concentration of 1.5 vol.% and a $[O_2]_{\text{inlet}}/[CO]_{\text{inlet}}$ ratio of 1.5, this interval decreases from 47 to 5 °C as the WHSV increases from 50 to 275 $L\,g^{-1}\,h^{-1}$.

3.2. Catalytic activity tests in the microreactor assembly

Based on the activity results with the single microchannel reactors, a number of experimental runs with the total microreactor assembly were carried out to characterize the influence of the operating conditions on the CO conversion and the temperature field and to determine optimal process conditions.

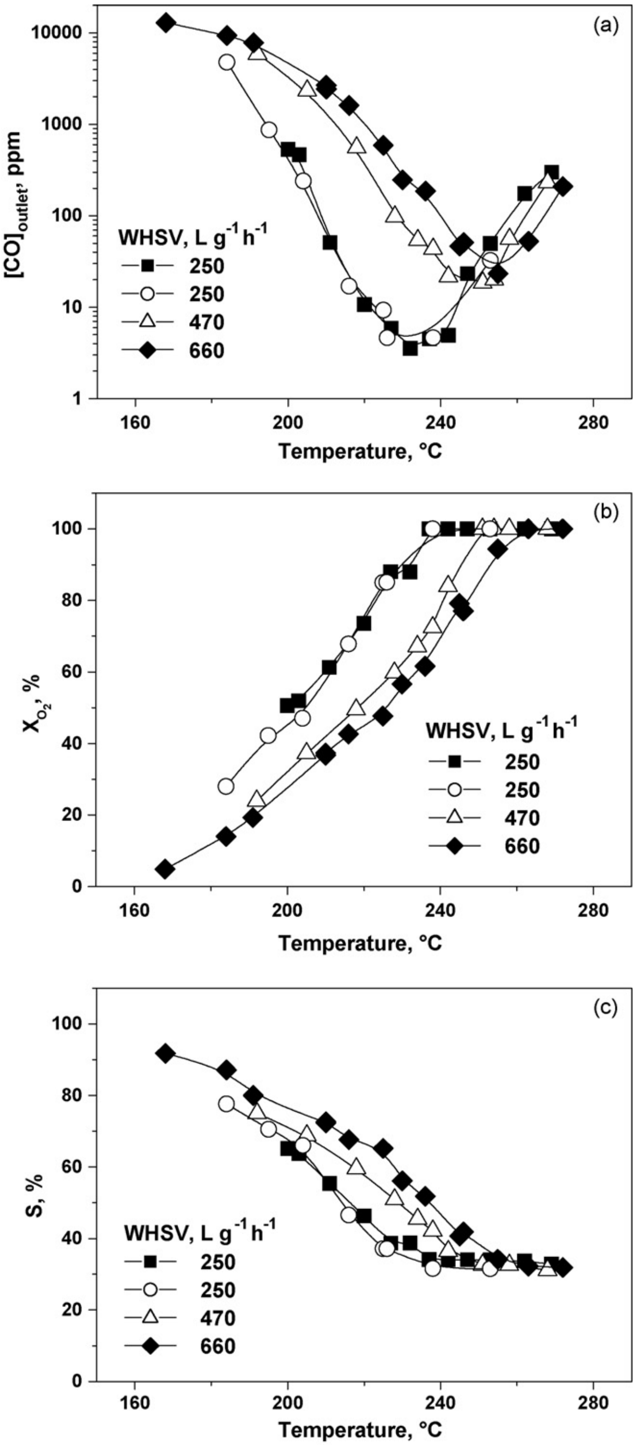

Fig. 3. The CO outlet concentration (a), the O_2 conversion (b), and the selectivity (c) in the preferential CO oxidation as a function of the reactor temperature at a WHSV of 250, 470, and 660 $L\,g^{-1}\,h^{-1}$. Filled symbols—microreactor with 23 mg of catalyst, open symbols—microreactor with 31 mg of catalyst The reformate composition is 1.5 vol.% CO, 2.25 vol.% O_2, 57 vol.% H_2, 10 vol.% H_2O, 20 vol.% CO_2, with He as balance.

It can be seen in Fig. 5 that the outlet CO concentration, the O_2 conversion, and the selectivity in the microreactor assembly demonstrated similar trends to those for each of the single microchannel reactors at different WHSVs. In these experiments, the inlet temperature of the reformate stream was 160 °C and the assembly was heated by an electrical heater. As the reactor temperature increased, the CO concentration decreased, reached a minimum at an optimum temperature and then increased. At a

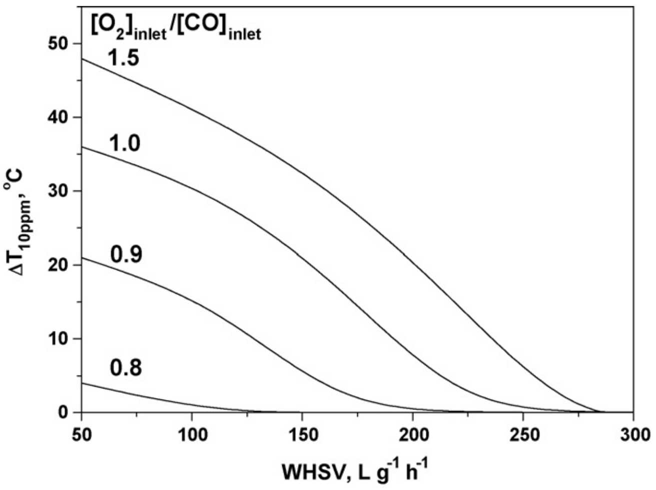

Fig. 4. PrOx operational window ($\Delta T_{10\,ppm}$) as a function of WHSV and $[O_2]_{inlet}/[CO]_{inlet}$ ratio. The reformate composition is 0.5–3.0 vol.% CO, 0.4–4.5 vol.% O_2, 57 vol.% H_2, 10 vol.% H_2O, 20 vol.% CO_2, with He as balance.

WHSV of 80, 160, and 240 L g^{-1} h^{-1}, the optimum temperatures were 210, 220, and 230 °C corresponding to CO concentrations of 3, 3, and 10 ppm, respectively. Full oxygen conversion was observed at these temperatures with a selectivity of 33.3%. The operation windows ($\Delta T_{10\,ppm}$) were 200–235, 205–235, and 225–235 °C in these three cases, respectively. It can be seen from a comparison of Figs. 4 and 5 that the PrOx operational window of the microreactor assembly is close to that of each of the single microchannel reactors. Insignificant narrowing of the operational window can be attributed to a non-uniform catalyst distribution in the single microchannel reactors which corresponded to a non-uniform WHSV distribution between the individual reactors. Nevertheless, the experimental results show that the microreactor assembly is capable of reducing the CO concentration in a realistic reformate gas from 1.5 vol.% to below 10 ppm.

Finally, the feasibility of steady-state operation of the assembly without an external heater was tested. The CO outlet concentration, the microreactor assembly and inlet reformate temperatures, the O_2 conversion and selectivity are plotted in Fig. 6 as a function of time. For the reactor assembly start-up, the procedure described in Section 2 was applied. The microreactor assembly temperature increased for 1 h, while the outlet CO concentration decreased. As the O_2 conversion attained 100% and the reactor temperature reached 237 °C, the outer insulating layer was removed, while the inner 0.5 mm thick layer of glass fiber was kept. The reactor temperature rapidly decreased to 233 °C and remained unchanged for the next 4 h. Under these conditions, the PrOx heat release compensated the heat losses to the environment. The outlet CO concentration attained 10 ± 2 ppm at 100% O_2 conversion at a selectivity of 33.3%. This stream can be fed to power a 100 W_e PEM FC.

3.3. Comparison with other PrOx microreactor devices

A detailed comparison of the performance of the microreactor assembly with other microreactor devices reported in the literature is problematic owing to various reaction conditions in different works. Therefore we selected for comparative analysis only those works which used realistic reformate stream (containing CO_2 and H_2O) supplied at space velocities sufficient for feeding a portable fuel cell (up to 100 W_e power).

The catalyst loading, the volume and the mass of the microreactor assembly of this work and the reaction conditions are compared in Table 2 with other configurations as reported in the literature.

Table 2
Comparison of performance of microreactor devices.

Microreactor device/type of catalyst	Overall catalyst weight (g)	Volume of device (cm^3)	Weight of device (g)	Reaction temperature (°C)	CO_{in} (vol.%)	CO_{out} (ppm)	O_2/CO	Flow rate (STP) (L min^{-1})	Productivity, mol_{CO} (g_{cat}^{-1} h^{-1})	Ref.
5 wt%Cu/CeO_{2-x}	0.732	88	520	230–240	1.5	10	1.5	3	0.16	This work
10 wt%Pt–Rh/γ-Al_2O_3	0.25	175	–	160	0.94	10	4	1.1	0.11	[12]
Pt–Co/α-Al_2O_3	0.98	175	980	160	0.5	7 (300[a])	3.2	0.8	0.01	[14]
Pt–Ru/α-Al_2O_3	1.5	60	150	130–170	0.5	10–20 (30[b])	1.75	3	0.03	[15]

[a] Outlet CO concentration after a period of 4 h time on stream.

[b] Outlet CO concentration after a period of 15 h time on stream.

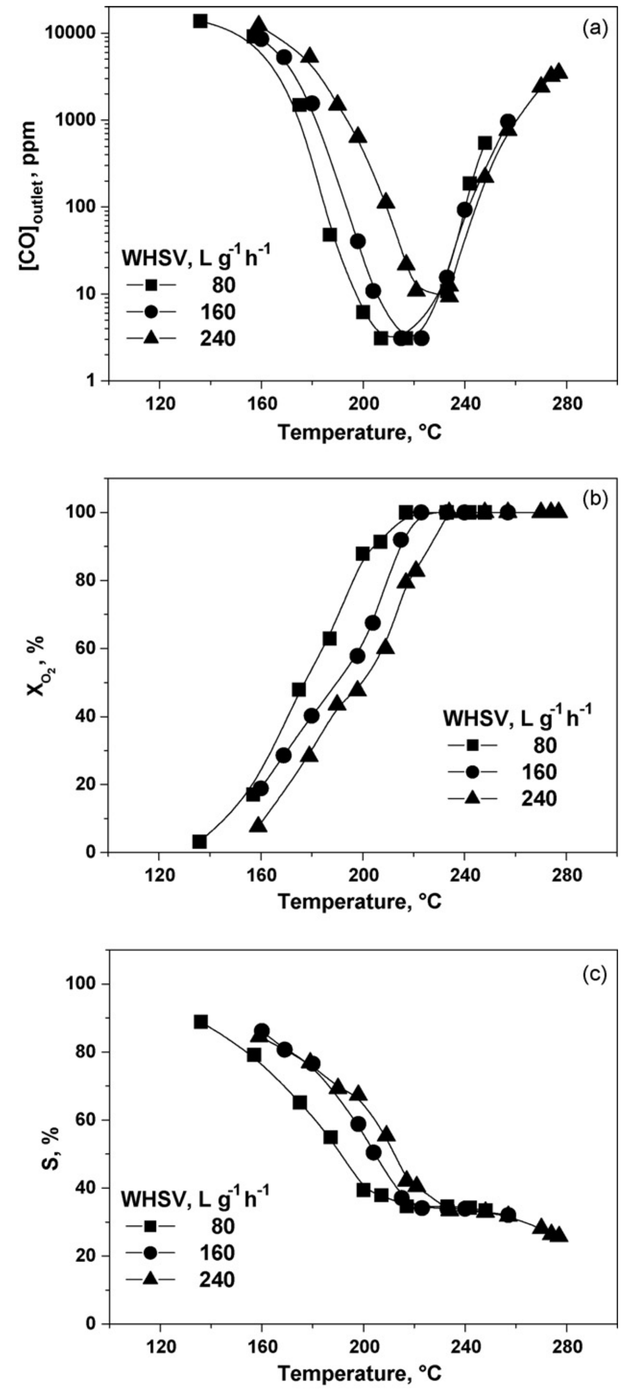

Fig. 5. The CO outlet concentration (a), the O_2 conversion (b), and the selectivity (c) in the preferential CO oxidation as a function of the reactor temperature at a WHSV of 80, 160, and 240 L g^{-1} h^{-1}. The gas inlet temperature is 160 °C. The reformate composition is 1.5 vol.% CO, 2.25 vol.% O_2, 57 vol.% H_2, 10 vol.% H_2O, 20 vol.% CO_2, with He as balance.

The microreactor assembly as developed in this work with the 5 wt% Cu/CeO_{2-x} catalytic coating showed the best performance and reduced the CO concentration from 1.5 vol.% to 10 ppm at a minimum $[O_2]_{inlet}/[CO]_{inlet}$ ratio in a realistic reformate gas flow of 3 L min^{-1}. A productivity of 0.16 mol$_{CO}$ g$_{cat}$$^{-1}$ h^{-1} was obtained that exceeded by an order of magnitude the performance of other PrOx microstructured reactors reported in the literature [14,15]. The reactor assembly has a slightly larger size and weight as compared with the microreactor studied in [15]. Those can be decreased by using geometrically optimized platelets (channel number and length) and a housing material with lower density, for example, aluminium instead of stainless steel. Note that both Pt–Co and Pt–Ru based catalysts rapidly lost activity [14,15], and the outlet CO concentration exceeded the level acceptable for PEM FC application, while the copper–ceria catalyst had stable activity.

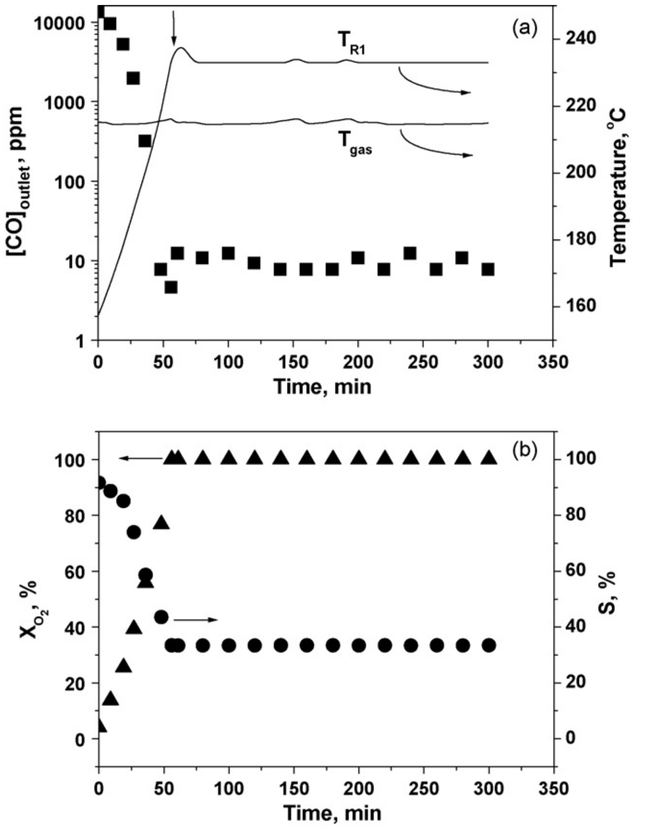

Fig. 6. The CO outlet concentration, and the microreactor assembly and inlet reformate temperatures (a), and the O_2 conversion and the selectivity (b) in the preferential CO oxidation as a function of time. The reformate composition is the same as those in Fig. 5. WHSV = 240 L g^{-1} h^{-1}. The vertical arrow indicates the time of the reactor insulation removing.

4. Conclusions

An authothermal preferential CO oxidation device consisting of an array of 26 microchannel reactors connected in parallel was designed for operation with a 100 W$_e$ PEM FC system. The carbon monoxide concentration in a realistic reformate gas was reduced from 1.5 vol.% to 10 ppm at an oxygen to carbon monoxide ratio of 1.5 within the temperature range of 230–240 °C, which is close to the temperature of the low-temperature water gas shift reactor. This facilitates the assembling of the CO PrOx and WGS microreactors without the need for an intermediate heat-exchanger to achieve higher efficiencies in the overall fuel processing. The washcoated copper–cerium oxide catalyst showed a higher activity and selectivity than platinum-based catalysts reported in the literature.

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