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Bench-scale methanol autothermal reformer for distributed hydrogen production

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Abstract

A bench-scale methanol autothermal reformer (ATR) for distributed proton exchange membrane fuel cell (PEMFC) power system has been developed. A coating of $ZnO-Cr_2O_3/CeO_2-ZrO_2$ mixed oxides on ceramic honeycombs was employed as the reforming catalyst. In order to avoid non-uniform distribution of the reactants at the inlet of the reforming catalyst bed, a distributor has been designed by computational fluid dynamics (CFD) simulation. Thus, uniform distribution of the reactants can be achieved, which can lead to a good performance of the reformer. Based on the optimized reformer, a fuel processor comprised of an ATR unit, a water gas shift (WGS) unit, a CO preferential oxidation (PROX) unit and a fuel evaporator unit has been developed and successfully integrated with a 75 kWe class PEMFC stack. The test shows that 120 $Nm^3\ h^{-1}$ H_2-rich reformate can be provided by the methanol fuel processor, with 53 vol% H_2 and less than 20 ppm CO content, and the peak power output of the PEMFC system can attain 75.5 kWe during the 3-h operation of the integrated system. By using the anode offgas from the PEMFC stack to evaporate the fuel, the lower heating value (LHV) efficiency of the fuel processor can reach 96.5%.

Keywords: Bench-scale; Methanol autothermal reformer; Distributor; CFD; Integration; PEMFC

1. Introduction

In the past decade, hydrogen production from a fuel processor, to be used in the proton exchange membrane fuel cell (PEMFC), has been developed rapidly for both mobile and stationary applications. Many international companies and research organizations have tried to develop fuel processors for converting fossil fuels to hydrogen on-board the vehicles, and accordingly, several fuel cell vehicles have been demonstrated [1]. However, it is difficult to achieve two technical targets simultaneously, i.e. a start-up time of less than 1 min at 20 °C and start-up energy of less than 2 MJ for a 50 kWe system. The fuel processor is currently a more practical means for stationary fuel cell systems and distributed hydrogen generation stations than for on-board applications [2,3]. The start-up performance of the integrated system is vital for on-board application. Furthermore, it can be affected by the fuel composition used for the fuel processing.

Gasoline, natural gas, methanol, dimethyl ether and ethanol are often considered as promising fuels to supplying reforming hydrogen for fuel cell stacks [4–8]. Semelsberger et al. [9,10] have performed extensive research of various fuels in start-up parameters analysis and given a detailed description of the fuel effects on the fuel processor size, the start-up energy and the efficiency. Gasoline and natural gas have the advantage of well-established fuel infrastructures, while there are also some technical difficulties for their real applications, such as the reactor volume, the catalyst durability, the start-up energy and the high operating temperatures. Their total fuel processor volume is much larger than that of the methanol due to the reduction of CO [10–12]. Furthermore, the sulfur in the gasoline has to be removed, or sulfur-free gasoline and diesels (referred to as reformulated gasoline and diesel) have to be employed. Ethanol, as a renewable energy source, has similar energy requirements for the start-up, and it is prone to form carbon deposition, leading to durability problems similar to natural gas and gasoline. Dimethyl ether has significantly lower light-off requirements than ethanol, gasoline and natural gas. Thus, non-toxic dimethyl ether [13–15] is another potential fuel, and in reference [16] detailed comparisons between methanol and dimethyl ether have been made.

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A fuel processor of methanol has lower start-up energy than that of the dimethyl ether. From the points mentioned above, methanol is considered to be the more practical option due to its relatively easiness for reforming [17], although there still exist some shortcomings such as its toxicity and its resources which are essentially based on the reforming of non-renewable fossil fuels (natural gas and coal).

Based on feasibility studies on demands of distributed hydrogen energy, many multipurpose PEMFC generating systems of kW scales have been developed in recent years. The SK Corporation of Korea has developed a 25 kWe methanol fuel processor [18], consisting a methanol steam reformer and a metal membrane purifying module. Moreover, Chevron-Texaco Technology Ventures has aimed at demonstrating a 50 kWe absorption-enhanced reformer capable of providing nearly pure H_2 from the steam reforming of natural gas, but up to now, the development of its fuel processor is still in progress [19]. A team of three organizations, the Air Products & Chemicals Inc., the Plug Power Inc. and the city of Las Vegas, has achieved successful operation of a H_2-fueled stationary 50 kWe fuel cell using the technology of methane steam reforming [20]. Argonne National Laboratory has developed an integrated 25 kWe autothermal reformer (ATR) which was suitable for utilizing various fuels [21]. Besides, Los Alamos National Laboratory has also reported on their work about fuel cell research [22] such as the integrated 10 kWe methanol steam reformer [23] and the diesel reforming for fuel cell auxiliary power units [24]. However, few results have been reported on the detailed performance of integrated fuel processor and fuel cell in these investigations. Furthermore, all these studies have not investigated the transient operations, but only the steady-state operations.

With the development of the PEMFC system, some technical problems have appeared such as optimization of the units comprised of the reformer, the heat exchangers, controlling of the fuel cells for special requirements, as well as the possible interaction between the units when they are integrated into an overall system. Among them, one of the most serious technological points is the non-uniform distribution of the reactants in the catalyst bed of a scale-up reformer system, which will lead to an insufficient use of the catalyst. This problem has been mentioned in several investigations on the scale-up of the fuel processors, and correspondingly, special gas distributors have been designed to achieve a uniform distribution of the reactant gas before they enter the reactor, but fewer details have been mentioned [3,25].

In recent years, Dalian Institute of Chemical Physics has accumulated certain experiences in the fuel reforming technology, especially in the scaling-up of PEMFC power systems, [26,27]. Recently, based on the previous studies of 5 and 30 kWe prototypes of methanol reforming systems, a bench-scale methanol autothermal fuel processor with an output of 120 $Nm^3\,h^{-1}$ reformate (suitable for a 75 kWe PEMFC stack) has been developed. In this paper, we focus on the optimum design and performance of the methanol reformer. Prior to the reformer, the novel design of a reactant distributor and its influence on the temperature distribution in the reformer is reported. Besides, an overall fuel processing system and the main operating characteristics integrated with a 75 kWe PEMFC stack are also proposed.

2. Experimental

2.1. Monolithic reformer

The monolithic reformer mainly consisted of a reactant distributor and a high temperature stainless steel housing in which the catalyst was positioned. The catalyst used in the experiments was a coating of ZnO-Cr_2O_3/CeO_2-ZrO_2 mixed oxides on the honeycomb substrate (Φ241 mm × 241 mm × 75 mm, 400 cpsi, Corning). The cordierite honeycomb was firstly pretreated with diluted acid and coated with a CeO_2-ZrO_2 sol via the incipient wetness impregnation method. Then the coated substrate was dried at 120 °C for 2 h, followed by calcination at 800 °C for 2 h. The procedure was repeated until 45 wt% of the CeO_2-ZrO_2 mixed oxides were loaded onto the monolith. Subsequently, the monolith coated with CeO_2-ZrO_2 was dipped into a nitrate solution of Zn and Cr. By means of a proprietary hot impregnation method, 30 wt% loading of the active metal components of ZnO-Cr_2O_3 were coated onto the monolith, followed by drying and calcination procedures as described above. In the catalyst formula, the molar ratios of CeO_2 to ZrO_2 and ZnO to Cr_2O_3 were 4 to 1 and 3 to 1, respectively. And the weight percentage of the coating catalyst was calculated referring to the total catalyst weight.

During the experiment, two sections of the reforming catalyst of 75 mm height were positioned in the reformer. Temperatures of the catalyst bed were measured with K-type thermocouples inside the honeycomb channels. A mixture of methanol and water was pre-heated up to 280 °C by a feed evaporator before they were blended with air and then fed into the reformer. In order to obtain a good performance of the catalyst, a reactant distributor designed by computational fluid dynamics (CFD) simulation was placed in front of the catalyst bed. All the data reported in this study were obtained under the following operating conditions: the entire gas hourly space velocity of 14500 h^{-1}, molar ratios of H_2O to CH_3OH and O_2 to CH_3OH in the reactants being kept at 1.5 and 0.3, respectively. Product gases of H_2, N_2, CO, CH_4 and CO_2 were analyzed by an HP4890 gas chromatograph equipped with a TCD detector.

2.2. Simulation studies of the reactants distributor of the reformer

To optimize reactor design as well as to provide a sufficiently uniform mixture over the component cross-sections, three dimensional CFD simulation for upstream reactant injection and mixing distributors was carried out using FLUENT, a commercially available CFD package. A cold-state model was adopted in this study, therefore, the energy equation was not considered.

Dimensions of the reforming unit used in this study are shown in Fig. 1. The two sections of the 75 mm catalyst beds were simplified as one unit of 150 mm in the simulation. An isotropic view of the geometry and grid system is also shown in Fig. 1.

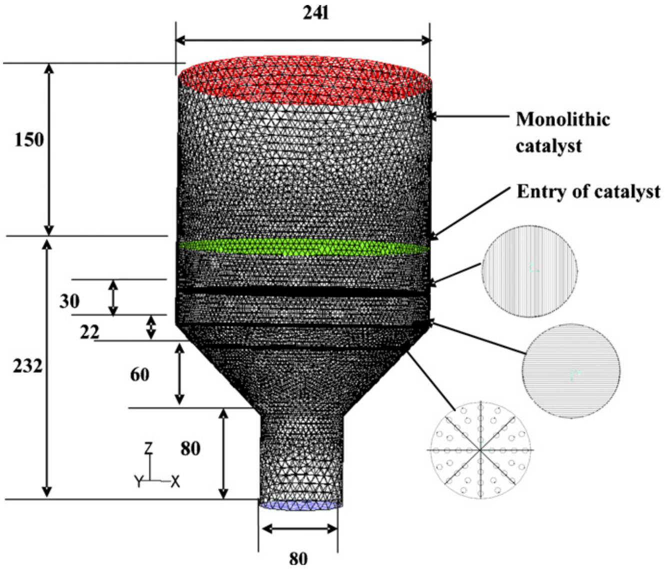

Fig. 1. Configuration and grid of the distributor at the head of the reformer (unit: mm).

GAMBIT—a FLUENT pre-processor [28] was used for geometry creation and meshing.

2.3. Methanol fuel processor system

Based on the data of the optimized reformer, our research group has developed a fuel processor that consisted of an ATR unit, a CO water gas shift (WGS) unit, a CO preferential oxidation (PROX) unit and a fuel evaporator unit for distributed power supply. The fuel processor and the PEMFC stack were coupled to form a process chain, starting from methanol as the input to electric power as the output.

The principal design of the overall system is described in Fig. 2. The reactants for the processor were methanol, deionized water and air, which were controlled by the mass flow controllers and pumps. By means of an ignition system, the methanol and water feed mixture was firstly pre-heated in the heat exchangers between all the component sections, then the warmed-up feed was ultimately heated up to about 280 °C in the following evaporator, where a portion of the anode offgas containing unconverted hydrogen was mixed with air and catalytically combusted over a combustion catalyst for supplying heat. Afterwards, the vaporized feed mixture and air were well blended with each other and entered the reformer, where the monolithic catalyst of the methanol autothermal reforming, $ZnO\text{-}Cr_2O_3/CeO_2\text{-}ZrO_2$ supported on cordierite honeycomb, was positioned. And the temperature of the reforming reaction was about 500 °C. In the subsequent WGS reactors the CO content of the reformate was reduced to less than 1.5 vol% in two separate temperature steps (the high temperature shift at about 320 °C and the low temperature shift at about 250 °C). Three modified Pt/CeO_2-based mixed oxides monolithic catalysts ($\Phi 241\,mm \times 241\,mm \times 150\,mm$, 400 cpsi) with different Pt loadings (0.5–1.5 wt%) were prepared by the impregnating method and used for the WGS reaction. Residual CO was further removed in a four-stage PROX purification unit with 0.5 wt% $Pt\text{-}Co/Al_2O_3$ monolithic catalysts ($\Phi 241\,mm \times 241\,mm \times 150\,mm$, 400 cpsi). After passing through the CO purification unit, the reformate gas was fed to the PEMFC stack, where electrical power was generated. The H_2-rich reformate gas composition of each unit in the flow could be measured in order to monitor and adjust the CO content for meeting the need of the PEMFC. Among them, H_2, N_2, CO, CH_4 and CO_2 were measured by an HP4890 gas chromatograph equipped with TCD and FID, while the content of CO was accurately measured by an on-line Infrared Gas Analyzer made by SIMADZU CGT-7000. Furthermore, almost no methanol or other hydrocarbons were monitored in the reformate gas. To maintain the required gas flow of the PEMFC stack, the whole system was operated under a pressure of 2 bar.

3. Results and discussion

3.1. Performance of the ATR catalyst

The monolithic $ZnO\text{-}Cr_2O_3/CeO_2\text{-}ZrO_2$ catalyst employed in this study has been previously investigated in lab-scale under the same operating conditions in our laboratory [29]. The results showed that nearly 100% methanol conversion could be obtained even after a long-term test of 1000 h. The same results were also achieved in the present work when the catalyst dimension was scaled up to about 2000-fold. Fig. 3 shows the catalyst perfor-

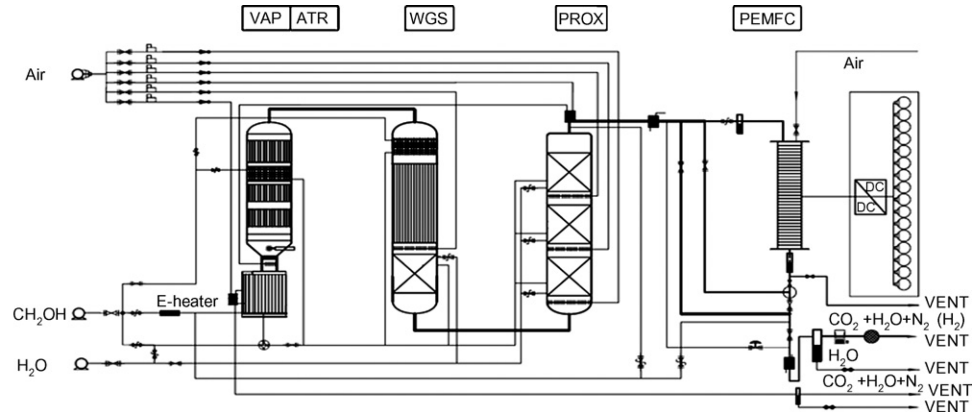

Fig. 2. Flow sheet of the bench-scale methanol ATR hydrogen generation system integrated with a PEMFC stack.

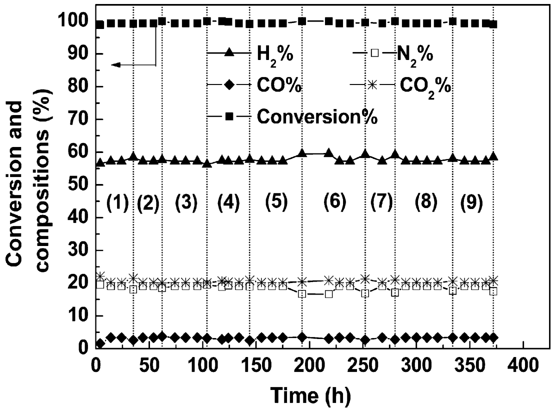

Fig. 3. Activity test of the monolithic ATR catalyst in the bench-scale methanol reformer. $O_2/CH_3OH = 0.3$, $H_2O/CH_3OH = 1.5$, $GHSV = 14500\,h^{-1}$, $T = 500\,°C$.

mance in the bench-scale equipment during a nearly 380-h test. The catalyst underwent nine cycles of start-up and shutdown during the test. It can be seen that in both the steady-state test and the transient test, the catalyst showed 100% of the initial activity and no obvious deactivation was observed. The reformate flowing from the reformer contained about 57 vol% H_2 and 3.4 vol% CO. The good stability, lower CO content, no pre-activation as well as monolithic configuration requirements make the reformer very suitable for applications in fuel processing system supplying H_2-rich fuel for scaling-up of PEMFC stack.

3.2. Results of simulation studies of reactant distributor

Fig. 4 shows the contour plot of air velocity distribution in the entry of the catalyst bed without any reactant distributors. It can be seen that the feed is distributed mainly in the center of the catalyst and scarcely in the zone near the wall of the reformer. Non-uniform velocity distribution will lead to a non-uniform distribution of temperature and cause some hotspots in the reactor bed, which will be unfavorable for the efficient utilization of the catalyst. In our real experiment, it was also shown that without any distributors a 100% of methanol conversion could not be achieved, no matter what measures we have adopted, e.g., adjusting the molar ratios of O_2 to CH_3OH or H_2O to CH_3OH, even reducing the flow velocity.

An optimal design of the mixing zone is essential in order to gain a uniform distribution of the entire cross-section of the catalyst. In this study, sieve plates and grid baffles were adopted as efficient gas distributors. Parameters such as width of the grid intervals, diameter of the holes, space between the holes, punched ratio on the baffle and axial location of the mixing plate, were comprehensively investigated as influence factors for a distributed performance. As a result, a novel combination of the distributor was designed and placed at the head of the reformer, as shown in Fig. 1. Two grid baffles were settled vertically. The sieve plate with uniform holes was in the front in order to realize sufficient mixing of the reactants and a uniform distribution of velocity.

After adopting the design of the inlet gas distributor, the velocity distribution was greatly improved. As shown in Fig. 5, an acceptable uniform distribution was obtained in the cross-section of the catalyst inlet. As a result, this design of distributor was used in the following bench-scale experiments in order to realize uniform distribution of the temperature of the reactor bed and a nearly complete methanol conversion.

3.3. Temperature distribution of methanol reformer

In order to meet the efficiency goals, it is required to maintain the catalyst temperature to be very steady and close to the optimum temperature. Twenty-one pairs of thermocouples were installed at various spatial positions of the first section reforming catalyst for gaining detailed temperature profiles of the reactor

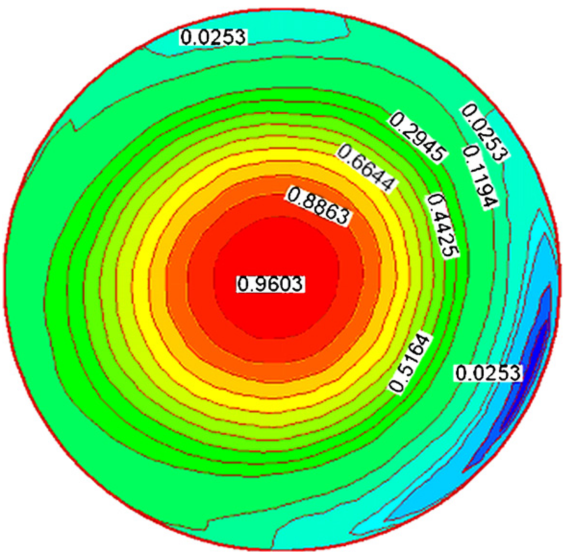

Fig. 4. Velocity distribution at the inlet of the bench-scale reformer without distributor.

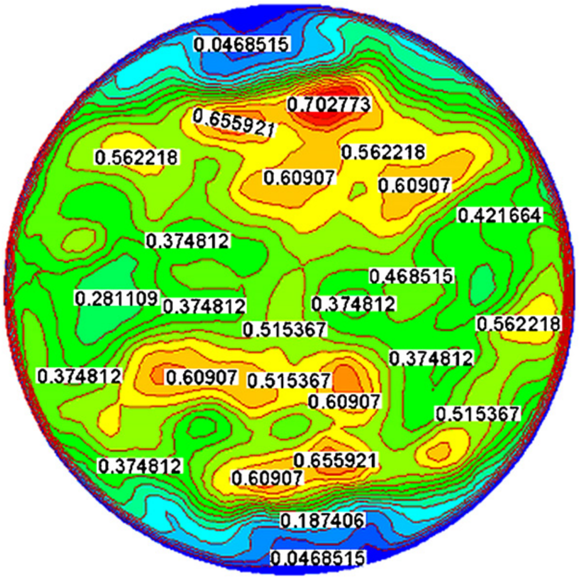

Fig. 5. Velocity distribution at the inlet cross-section of the bench-scale reformer with distributor.

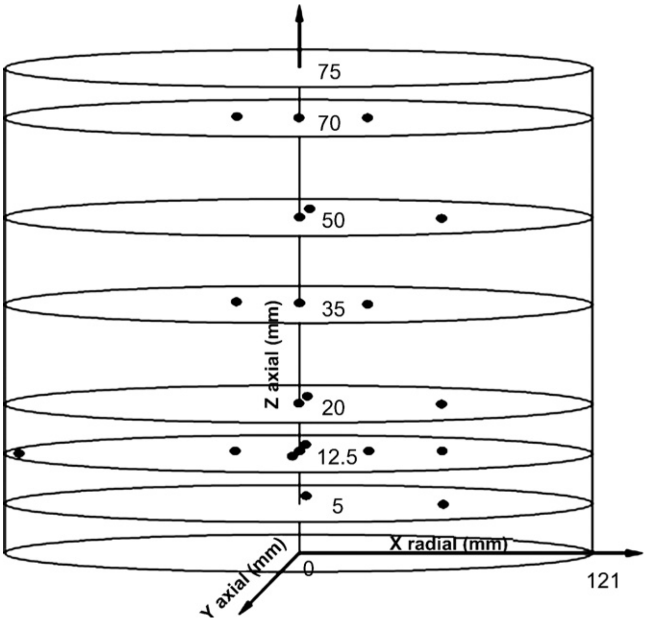

Fig. 6. Positions of thermocouples in the monolithic reforming catalyst.

bed, as shown in Fig. 6. The temperatures of all the positions were continuously monitored to maintain the conditions as close to the optimum ones as possible.

Fig. 7 presents the temperature distribution in the axial direction of the reforming catalyst bed, which corresponds to the central z-axis. It can be seen that the maximum temperature is in the initial part of the catalyst bed. Then the temperature drops along the flow direction. From these trends, it can be estimated that the methanol oxidation reaction mainly occurs in the very front part of the catalyst, which will release a large amount of heat. The endothermic reactions, such as the methanol steam reforming reaction, will dominate in the subsequent parts of the catalyst. The result is similar to those reported elsewhere in the literature [30–32].

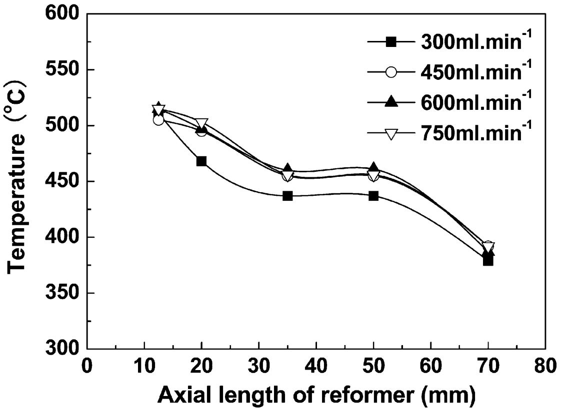

Fig. 7. Temperature distributions in the axial direction of the reformer.

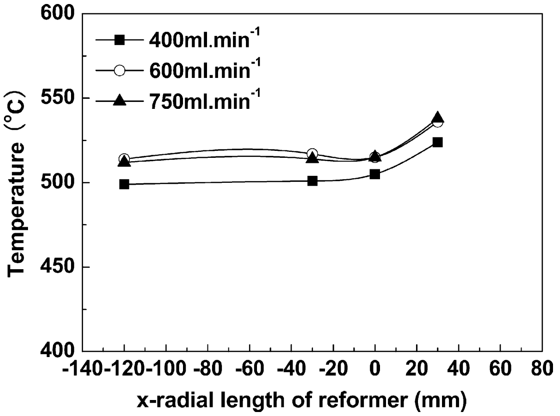

Fig. 8. Temperature distributions in the x-radial direction of the reformer.

For the bench-scale equipment, the radial temperature distribution of the catalyst bed is also a key factor that determines the reaction performance of the catalyst. The temperature distributions of the catalyst bed for the x-radial axis are shown in Fig. 8 and that of the symmetrical points in the plane of $z = 12.5$ mm are shown in Fig. 9. A comparatively uniform distribution of the radial temperature was achieved for various flow rates of the reactant with a maximum deviation of only about 30 °C, which is a tolerable error for such a large-scale system. It indicates that the design of the reactant distributor of the methanol reformer is necessary and feasible.

3.4. Integrated test of methanol fuel processor with a 75 kWe class PEMFC stack

An integrated test of the fuel processor with a 75 kWe class PEMFC stack has been conducted in order to determine whether the developed methanol processor was suitable for steadily supplying of H_2-rich fuel to the PEMFC stack. From the viewpoint of fuel purity, the CO content in the reformate is crucial because a trace amount of CO will deactivate the anode catalyst of the PEMFC and then drastically reduce its performance. The composition of the reformate supplied to the PEMFC stack during the integrated test is shown in Fig. 10. It can be seen that the fuel processor had maintained a steady-state operation for nearly 3 h, with a stable reformate composition of less than 20 ppm CO and more than 53 vol% H_2. The rather low and constant CO content so obtained was not only due to the employment of the CO purification system, but also due to the steady operation during the methanol reforming stage. And with the supplying of 120 $Nm^3 h^{-1}$ H_2-rich reformate, a high electric power output of

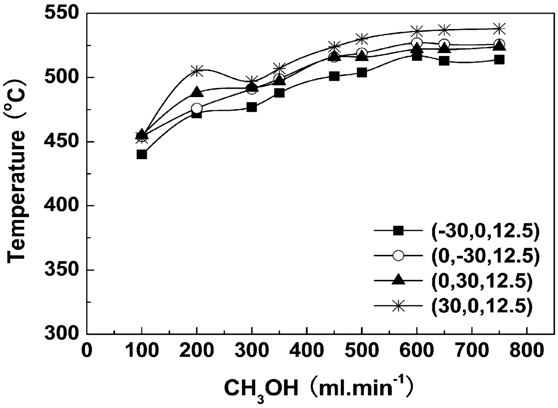

Fig. 9. Comparison of temperatures for symmetrical points in the reformer.

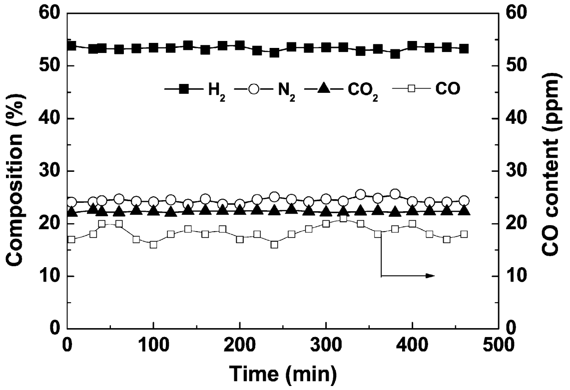

Fig. 10. Composition of the reformate supplied to the PEMFC stack during the integrated test. $O_2/CH_3OH = 0.3$, $H_2O/CH_3OH = 1.5$, GHSV = 14500 h^{-1}, $T = 500\,^{\circ}C$.

more than 70 kWe for about 1 h and a maximum power output of 75.5 kWe has been achieved. Fig. 11 shows the continually generated electric power of the PEMFC stack as well as the corresponding CO content in the reformate recorded by an on-line CO monitor during the 3-h test. The results prove further that the quality of the H_2-rich reformate derived from the methanol fuel processor is suitable for a stable operation of the PEMFC stack and can be used as a hydrogen source for the PEMFC power generation [33].

The energy efficiency (η) of the methanol fuel processor was also studied in the present work, which was defined as the quotient of the lower heating value (LHV) of the hydrogen supplied to the fuel cell to that of the methanol feed into the reformer, as shown in Eq. (1):

$$\eta = \frac{n_{H_2} \times \mathrm{LHV}(H_2)}{n_{CH_3OH} \times \mathrm{LHV}(CH_3OH)} \times 100 \qquad (1)$$

where n is the molar ratio of methanol or H_2. In this study, the energy efficiency corresponding to the peak output of the fuel processor was considered. When 750 ml min^{-1} methanol was fed into the reforming unit, 120 $Nm^3\,h^{-1}$ reformate gas was generated and fed to the fuel cell. Therefore, the energy efficiency of the fuel processor was calculated to be 96.5%, which was higher than other systems reported so far [6,18,34]. This is due to the utilization of an anode offgas containing unconverted hydrogen from the PEMFC stack as the fuel gas for the evaporator unit. Thus, an optimal energy balance of the whole fuel processor system was achieved in the present work. Furthermore, the optimal design of the heat exchanger system in this study might be another reason. In addition, the overall energy efficiency of the integrated reformer-fuel cell system was calculated to be 38.2% according to a maximum power 75.5 kWe.

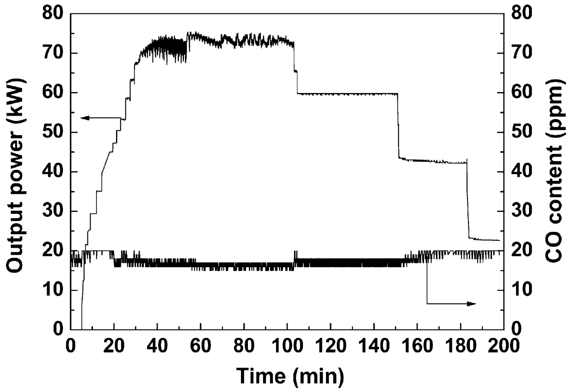

Fig. 11. Output power of the PEMFC stack and CO content in the reformate during the gratefully integrated test.

4. Conclusions

(1) A bench-scale methanol autothermal reformer for distributed hydrogen production has been developed. The monolithic reforming catalyst of $ZnO\text{-}Cr_2O_3/CeO_2\text{-}ZrO_2$ mixed oxides used in this study has shown a nearly 100% conversion of methanol in both the steady-state test as well as in the transient test.

(2) In the scaling-up of the reformer, a novel distributor was designed by three dimensional computational fluid dynamics simulations. A uniform distribution was achieved, leading to a comparatively uniform distribution of the radial temperature with a maximum deviation of only about 30 °C.

(3) The optimized ATR reformer was then combined together with a CO purification system into a methanol fuel processor, and then integrated with a 75 kWe class PEMFC stack. The integrated operation showed that 120 $Nm^3\,h^{-1}$ H_2-rich reformate could be supplied from the fuel processor, with a stable output of more than 53 vol% H_2 and less than 20 ppm CO.

(4) A steady electric power output could be maintained for 3 h, with a peak power output of 75.5 kWe. And due to the utilization of a H_2-contained offgas from the PEMFC anode, a high energy efficiency of 96.5% of the fuel processor was achieved.

Acknowledgements

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