



Design, fabrication and performance evaluation of an integrated reformed methanol fuel cell for portable use

Shubin Zhang^b, Yufeng Zhang^{a,b,*}, Junyu Chen^b, Congwen Yin^b, Xiaowei Liu^{a,b}

^a Key Laboratory of Micro-Systems and Micro-Structures Manufacturing of Ministry of Education, Harbin Institute of Technology, Harbin 150001, China

^b MEMS Center, Harbin Institute of Technology, Harbin 150001, China

HIGHLIGHTS

- A dynamic system model for reformed methanol fuel cell (RMFC) is established.
- A portable RMFC system is designed, fabricated and assembled.
- Measured performance of the RMFC is in good agreement with simulation.
- Maximum energy conversion efficiency of 36.2% is achieved.
- The RMFC is able to power a laptop.

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ABSTRACT

In this paper, an integrated reformed methanol fuel cell (RMFC) as a portable power source is designed, fabricated and tested. The RMFC consists of a methanol steam reformer (MSR), a high temperature proton exchange membrane fuel cell (HT-PEMFC) stack, a microcontroller unit (MCU) and other auxiliaries. First, a system model based on Matlab/Simulink is established to investigate the mass and energy transport characteristics within the whole system. The simulation results suggest a hydrogen flow rate of at least 670 sccm is needed for the system to output 30 W and simultaneously maintain thermal equilibrium. Second, a metallic MSR and an HT-PEMFC stack with 12 cells are fabricated and tested. The tests show that the RMFC system is able to function normally when the performances of all the components meet the minimum requirements. At last, in the experiment of successfully powering a laptop, the RMFC system exhibits a stable performance during the complete work flow of all the phases, namely start-up, output and shutdown. Moreover, with a conservative design of 20 W power rating, maximum energy conversion efficiency of the RMFC system can be achieved (36%), and good stability in long-term operation is shown.

1. Introduction

Micro-fuel cells have been considered as a promising alternative power source for batteries for decades [1]. Proton exchange membrane fuel cells (PEMFC) using pure hydrogen and oxygen have demonstrated electrical efficiencies up to 70% without any pollutant emission [2]. Besides, PEMFC stacks also have the advantages of silent operation and structural simplicity. However, when it comes to portable use, low volumetric and gravimetric efficiency of auxiliary devices for rigid hydrogen storage and control become a major technical shortcoming.

Reformed methanol fuel cell (RMFC), which refers to a system mainly contains a PEMFC stack and an on-board hydrogen supplying device where methanol reforming take place, arises as a solution.

Compared to auto-thermal reforming and partial oxidation reforming, methanol steam reforming has the lowest reaction temperature, longest catalyst lifetime and easiest fuel flow control. Thus, methanol steam reforming technology is preferred in portable RMFC. One of the features of RMFC is that it uses high temperature proton exchange membrane with high carbon monoxide tolerance in order to directly feed the reformate gas into the fuel cells without any pre-treatment for hydrogen purification. Polybenzimidazole (PBI) high temperature PEMFC (HT-PEMFC) operates at higher temperatures (120–180 °C) than the conventional, low temperature, Nafion-based fuel cell. This makes it easier to integrate the HT-PEMFC stack with the methanol steam reformer (MSR), in which the core temperature is above 240 °C. Recently, extensive studies have been done on HT-PEMFCs both numerically and

* Corresponding author. MEMS Center, Harbin Institute of Technology, Harbin 150001, China.
E-mail address: yufeng.zhang@hit.edu.cn (Y. Zhang).

experimentally. Development of HT-PEMFC stack technology for road vehicles is nicely reviewed by Liu [3]. The output power of the fuel cell stacks is ranged 1 kW–60 kW. In the design for such high-power HT-PEMFCs, temperature control is pointed out to be crucial to stack performance and stability. First, the HT-PEMFC stacks need at least several minutes to start up. Then cooling system must be enabled to keep the stack from being overheated. Wannek et al. has demonstrated that higher temperature will cause fast MEA degradation [4]. Other problems, like gas sealing and short circuit risk [5], are also need to be carefully dealt with, while they are rarely mentioned in portable HT-PEMFC design. Nevertheless, portable HT-PEMFC design requires more energy on flow field design and calculation of optimal operating parameters, such as flow speed and temperature. Weng et al. [6] fabricated a small-scale HT-PEMFC stack with less than 100 W output power. Based on graphite bipolar plates, 4 types of flow field were designed in their study. During discharging, stack operating temperature was successfully maintained at 160–180 °C using digital heater. Wu et al. [7] demonstrated an air-breathing, single-cell HT-PEMFC with the power density of up to 220 mW cm⁻² at 200 °C. Based on experimental works, numerical models were also established to investigate mass and energy transport in HT-PEMFCs. Effects of gas composition, flow field structure, and temperature were mostly discussed in the simulations [8–11].

Numerical models are also widely used in designing MSRs. Kinetic models were reported by Peppley et al. [12] and Mastalir et al. [13]. Generally, two- and three-dimensional models have higher accuracy of predicting the reactor performance than one-dimensional models because they can be coupled with more realistic boundary conditions. Chein's numerical studies [14,15] indicated that effective heat transfer characteristics in mini-scale reactors are essential to higher hydrogen productivity. Others found uniform flows are another important factor that affects the performance of microreactors, so micro-channel reactor was recommended by Hsueh et al. [16]. Nevertheless, with reasonable assumptions, one dimensional models are also able to determine methanol conversion and hydrogen productivity. A one-dimensional model built by Kawamura et al. [17] successfully helped them to design a Si-based methanol reformer.

In the system level, A highly integrated RMFC with an electric output power of 30 W was firstly developed by Wichmann et al. [18]. Highlights of the research included an innovative heat exchanger design that allowed the integration of all relevant fuel processing steps such as catalytic combustion, vaporization, reforming, and heat exchange into a single component and also the integration with the HT-PEMFC. In the system, fuel utilization in the fuel cell between 60% and 70% was obtained from the experiments. RMFC systems with higher electric power outputs were also reported. Andreasen et al. [19] showed the development of a cascade control strategy to control temperature for a RMFC system. A series of tests were conducted on the methanol reformer used in the Serenergy H3 350 W mobile battery charger. Desired reference reformer temperature was successfully obtained using the proposed cascade control structure by introducing two control loops. One is the inner loops to ensure proper and safe control of the burner air/fuel mixing and temperature and the other is the outer loop to vary the burner temperature. More importantly, the prediction of the critical internal states within the RMFC system could be used in common RMFC design. An HT-PEMFC stack from Serenergy was also referred by Sahlin et al. [20] in their recent work on system model development of a 5 kW RMFC system. The model, predicting an efficiency of 27–30%, created an efficient and fast way to evaluate different control strategies and gave insight into the dimensions of the cooler, methanol pump and blowers. In the model, however, simulation for methanol steam reforming was neglected and the input parameters for the system were narrowed to current and fuel flow rate, showing only an early initial base for future work to improve the system efficiency. In the work of Wu et al. [21], numerical investigations on a 5 kW PEMFC stack with a fuel reforming system including methanol

steam reforming, water gas shift and partial oxidation of methanol were presented. Furthermore, multiple heat exchangers were connected to improve energy efficiency. In the study by Schuller et al. [22], a heat exchanger methanol steam reformer was designed, which used the waste heat from a 400 W HT-PEMFC stack. Stamps et al. [23] developed a dynamic system model of a RMFC based on a low-temperature PEMFC stack for vehicular application. The reformed gas and oxygen in the cathode were compressed to form high pressure so that the performance of PEMFC can be greatly improved. The study on the interconnections between sub-units and the formulation of control loops to operate the system to meet desired power targets were of great value to RMFC system design. However, in portable application, the method of using high pressure gas could hardly be adopted. A MEMS-based small-scale RMFC was developed by Morse et al. MEMS fabrication process highlighted the design of the microfluidic fuel processor and fuel cell platform [24]. The compact RMFC enabled rapid start-up operation, but system design strategy and energy efficiency were never discussed in the study. In the works by Besser [25] and Ribeirinha et al. [26,27], the integration of MSR and HT-PEMFC were numerically and experimentally studied. The results they achieved were promising and the novel design strategies could be used for portable devices. However, these conceptual designs still show a certain distance to practice.

A literature review reveals that few efforts have been devoted to the work of designing and fabricating a portable RMFC system. Yet there are still some critical issues of mass and energy transport in the whole system to be understood, numerical techniques can be utilized to form a universal design method. In addition, the realization of a portable RMFC system should be complete and able to exhibit a robust performance. In this paper, a mathematic model for RMFC is established from a systematic point of view to investigate the mass and energy transport characteristics. Through the system model, fuel feeding rate and temperature control strategies are able to be tested and optimized in the first place. Following the simulation results, a RMFC prototype is designed, fabricated and tested step by step. The emphasis of this paper is on: (1) how the system model works and helps to comprehend the mechanism and operation patterns of the system, (2) fabrication of the compact MSR and the HT-PEMFC stack with a new thin bipolar plate, and (3) finally making the portable RMFC work stably as a complete system with necessary auxiliary devices.

2. Numerical

A numerical model for RMFC system is built in this section. We consider a RMFC structure, as illustrated in Fig. 1, which consists of MSR, HT-PEMFC stack, pumps, valve, control unit and electronic loads. The MSR includes a vaporizing room (VR), a burning room (BR, or combustor) and a reforming room (RR, or reformer). The whole system are connected by two flow lines: one is for air to be blown into the cathode of the HT-PEMFC stack and then to the combustor, the other is for fuel to be pumped from the fuel cartridge to VR, followed by RR, the anode of the HT-PEMFC and combustor.

The self-build model is time-based, aiming at simulating the whole work process of the RMFC system, namely startup phase, output phase and shutdown phase. On behalf of simplicity, assumptions are made as follows:

- (1) Temperature is uniformly distributed in either MSR or HT-PEMFC stack.
- (2) It is plug-flow in each reactor and the gasses obey the ideal gas law.
- (3) Oxygen and hydrogen are the only reactants in the HT-PEMFC.
- (4) Heat capacity for each gas is regarded as constant.
- (5) CO, CH₃OH, H₂ are fully combusted in BR

2.1. MSR model

The model for MSR includes the descriptions for chemical reactions,

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