Journal of Power Sources 271 (2014) 392–400

Contents lists available at ScienceDirect

Journal of Power Sources

journal homepage: www.elsevier.com/locate/jpowsour

A miniature fuel reformer system for portable power sources

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HIGHLIGHTS

- A miniature fuel reformer was developed and fabricated.
- Technology level reached exceeds laboratory prototype.
- The complete system was built: reactors, a combustor, evaporators and control.
- Engineering aspects focused: miniature design, integration, process control.
- Energy efficiency and other parameters were estimated experimentally.

ARTICLE INFO

Article history:
Received 26 May 2014
Received in revised form
18 July 2014
Accepted 4 August 2014
Available online 12 August 2014

Keywords:
Methanol reforming
Miniaturisation
Integration
Process control

ABSTRACT

A miniature methanol reformer system has been designed and built to technology readiness level exceeding a laboratory prototype. It is intended to feed fuel cells with electric power up to 100 W and contains a complete setup of the technological elements: catalytic reforming and PROX reactors, a combustor, evaporators, actuation and sensing elements, and a control unit. The system is engineered not only for performance and quality of the reformate, but also for its lightweight and compact design, seamless integration of elements, low internal electric consumption, and safety. In the paper, the design of the system is presented by focussing on its miniaturisation, integration, and process control.

1. Introduction

The combination of a miniature hydrocarbon fuel reformer and fuel cell represents a portable electric power source for various portable electric devices for civil and military use. Thanks to the high energy density of hydrocarbons, this type of power source can have a much higher density of stored energy (stored electric energy per mass) than modern batteries. An important benefit is almost instant recharging by refilling the fuel.

There are two types of fuel cell-based power sources using methanol as a fuel: direct methanol fuel cells (DMFC) and reformed methanol fuel cells (RMFC) combining a fuel reformer and a fuel cell [1]. The first type (DMFC) works with liquid methanol or methanol water solution and has a simpler structure because methanol is fed onto the anode of fuel cell directly and therefore fuel reformer is not needed. However, the efficiency and resulting energy density are lower due to high anode and cathode overpotentials. Power sources of this kind are available on the market, e.g. Ref. [2]. Polymer electrolyte membrane fuel cells (PEMFC) are usually used in DMFC power supplies. Solid oxide fuel cells (SOFC) are also very appropriate for direct methanol operation, however, they operate at a relatively high temperature (600–1000 °C) and have a long start-up time, which limits their use in miniature and portable (especially man-wearable) systems for civil or military applications. In any case, they have been adopted well in large-scale power generation systems [1]. The second type of power source (RMFC) is a combination of a fuel reformer and a fuel cell. A fuel reformer decomposes the fuel into hydrogen, carbon monoxide and carbon dioxide. In the category of portable miniature (and especially in man-wearable) RMFCs, PEM fuel cells have the widest use,

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http://dx.doi.org/10.1016/j.jpowsour.2014.08.021
0378-7753/

mainly due to solid phase electrolyte and relatively low operating temperatures (50–80 °C for low temperature PEMFCs, and 100–200 °C for high temperature PEMFCs). Low temperature PEMFCs do not tolerate carbon oxide, so it must be removed. For this type of power source the efficiency and energy density are higher, but the overall system complexity increases. The fuel reformer is a complex unit composed of at least a reforming reactor, a combustor, an evaporator and possibly also a CO removal reactor. The most widely used process for producing hydrogen from methanol is steam reforming, with the highest yield of hydrogen obtained at nearly complete conversion at temperatures lower than 250 °C. A comprehensive setup of peripheral and control equipment is also needed to operate the system. The miniaturization of the reformer and its integration with the fuel cell has been a major research activity for the successful development of PEMFC-based systems over the last fifteen years.

In our work we focused on the design and implementation of a miniature reformer of methanol, aimed at feeding a low-temperature PEM fuel cell with electric power up to 100 W. Several prototypes of miniature scale reformers have been reported in the literature. For example in Refs. [3], [4] and [5] several prototypes of reforming reactors are presented, but they are not thermally self-sustained and they do not provide CO removal. In Refs. [6], [7] and [8] more complete and integrated prototypes are reported, containing also fuel evaporators and catalytic combustor, which means that they do not depend on external (electric) heating sources. Most of the presented systems are early prototypes demonstrated in a laboratory environment using laboratory peripheral and control equipment, such as laboratory pumps, stand-alone gas mass flow controllers (MFCs) or meters (MFMs). This kind of equipment cannot usually be embedded into a miniature system due to its size, price, power consumption, and many other parameters. Several miniature systems that aim at a higher technology level by being thermally self-sustained and in some cases also containing a CO removal reactor have been presented in the recent literature, a literature overview can be found in Refs. [9] and [10]. One such system is reported in Ref. [11] and is implemented as a single structure with a combustor, preheaters, a reformer reactor and a water gas shift reactor, but it does not include a final CO removal reactor. Similarly, in Ref. [12] a tubular quartz reactor structure composed of two concentric tubes is presented. Combustor, evaporator, reforming reactor and CO removal reactor (methanation) are all integrated into one quartz structure. The problem with the integration of the reforming reactor and CO removal reactor is that they require different operation temperatures, and this is difficult to achieve within a miniature and monolithic reactor structure. Operating at the same temperature may lead to non-optimal operation of one or both reactors. The system, presented in this paper, is designed to maintain the reformer and CO removal reactors at different temperatures, which leads to a more complex design, but makes it possible to achieve more optimal operation. In Ref. [13] another interesting integrated reactor concept is presented that is composed of a ceramic micro-channel monolith with several parallel channels, some employed for combustion and heating and some for reforming. Thanks to the mixed position of the combustion and reforming channels, good internal heat exchange and self-insulation is achieved. For the supply and distribution of reagents and the collection of products, relatively complex and precisely machined metal distributor elements and seals are required, which is somewhat similar to the approach used in our system. The system does not include a CO removal reactor and does not include evaporators, as the water/methanol mixture is evaporated and supplied to the reformer reactor by passing inert carrier gas through the bubbler, which is filled with a mixture of methanol and distilled water. The use of additional inert gas may be acceptable for research but it is less convenient for final implementation. All three mentioned systems depend on laboratory equipment for the supply of input flows, measurements and control. In our case all the equipment (the manifold, pumps, tanks, sensors, electronic control unit and thermal insulation elements) is integrated into the system. In addition, our system was also designed for easy maintenance and servicing by providing the possibility of disassembling and replacing the main parts including the catalyst. This is usually not possible in the case of monolithic structure systems, as presented in the literature. Some similar systems are already available on the market, e.g. Refs. [14] and [15]. These are complete RMFC power sources, but they use high temperature PEM fuel cell, which means that CO removal reactor is not present in the system since this kind of fuel cell can tolerate higher concentration of CO (up to 1%).

Our goal was to design a complete reformer system reaching technology readiness level 6-7 [16], which represents a fully functional system prototype that has been demonstrated in an operational environment. This entails building the complete reformer system, including the peripheral and control equipment. The basic technologies of the reactor design and catalyst preparation and deposition are essential for the optimal operation of the system. This part of the design has been addressed in detail and was presented in our previous papers [17–21]. However, to upgrade the basic technologies and prototypes to operational systems, basic manufacturing and control technologies must be integrated and coordinated. This is particularly challenging in miniature scale systems.

For optimal reactor operation, the relevant process parameters (the flow rates of the reactants, the temperatures of the reactor) must be controlled. The closed loop control concept is traditionally used to control process parameters, but this approach requires sensing, actuation, and control elements. While this can be readily applied in medium- and large-scale processes, there are limitations at miniature scale. The reason is that miniature sensors and actuators are not easily available and their use expands the setup and makes the miniature system more complex. To keep the system miniature and lightweight, all system components, including control elements, have to be optimised not only for performance, but also for small dimensions, lightweight and compact design, and it must be possible to elegantly integrate them into the process. One preferable option is the seamless integration of sensors with the process equipment [3], e.g. "printing" the sensors on the reactors.

To achieve acceptable energy efficiency of the overall system, the internally consumed electric power should be far lower than the generated electric power. Therefore, it is necessary to minimise electricity consumption by designing reactors with a low pressure drop, by designing/selecting low power control and actuation elements, and minimising or even omitting the control elements by using passive techniques [22].

In the paper we summarise the system design, emphasising its miniaturisation, integration, process control, and raising the technology readiness level.

2. Background

In the presented system, conversion of methanol into hydrogen is performed by a steam reforming process. The output reformate gas, which is rich in hydrogen, can then be fed into a fuel cell and converted into electricity. The chemistry of steam reforming is well known and can be described by the following equations:

$$CH_3OH + H_2O \leftrightarrow CO_2 + 3H_2\ (\Delta H = 49.5\ \text{kJ mol}^{-1}) \quad (1)$$

$$CH_3OH \leftrightarrow CO + 2H_2\ (\Delta H = 90.7\ \text{kJ mol}^{-1}) \quad (2)$$