

Available online at www.sciencedirect.com**SciVerse ScienceDirect**

APCBEE Procedia 3 (2012) 17 – 22

**Procedia
APCBEE**www.elsevier.com/locate/procedia

ICCCP 2012: 5-6 May 2012, Kuala Lumpur, Malaysia

Novel Heat Integration in a Methane Reformer and High Temperature PEM Fuel Cell-based mCHP System

Ademola M. Rabiu^{a,*}, Nkosikho Dlangamandla^a, Øystein Ulleberg^b^aDepartment of Chemical Engineering, Cape Peninsula University of Technology, Cape Town 8000, South Africa^bInstitute for Energy Technology, Instiuttveien 18, 2027, Kjeller, Noerway

Abstract

A highly integrated and optimized heat recovery system is essential for efficient operation of a HTPEM fuel cell-based mCHP system. The main challenge with such system is to design energy efficient systems with low start-up time. Recent studies have shown that this can be achieved by proper heat integration of the balance of plant (BOP) components. This study proposed a novel scheme that optimally integrated the sub-components of a mCHP system consisting of a 2 kW_{el} HTPEM fuel cell integrated with methane processing units, for recovery and use of the process and waste heat. A steady state system modeling and simulation of the complete mCHP system was implemented in Aspen Plus v7.2. The design of the heat recovery system was achieved with pinch analysis techniques. Heat integration results show that external cooling utility is not required and result in a 5% increase in the overall system efficiency.

© 2012 Published by Elsevier B.V. Selection and/or peer review under responsibility of Asia-Pacific Chemical, Biological & Environmental Engineering Society Open access under [CC BY-NC-ND license](https://creativecommons.org/licenses/by-nc-nd/4.0/).

Keywords: Heat integration, HTPEM fuel cell, micro CHP system, Pinch analysis, methane reformer

1. Introduction

In recent years, combine heat and power systems (CHPs) that generates both heat and power with high efficiency have received considerable attention. Small scale CHPs also referred to as domestic (dCHP) or micro CHPs (mCHPs) have found niche markets in domestic and office application due to the ability to rapidly vary their electrical load to reduce or increase the thermal output [1-3]. Although small in terms of

* Corresponding author. Tel.: +27-73-192-1434; fax: +27-21- 460-3854.

E-mail address: RabiuA@cput.ac.za.

power output (usually less than 5 kW_{el}), the portability and simplicity of mCHP allowed for installation in millions of homes, particularly where there is a huge market for heating fuel. In such applications, the mCHP serves as the central heating unit providing heat and hot water and additionally supplies the electrical power requirements, for domestic appliances [1, 4]. Globally, pre-commercial projects have already been initiated for the implementation of mCHP systems on the larger scale.

A fuel cell-based CHP (FC-CHP) system converts on-site, the chemical energy in the fuel into electrical power and heat with an overall efficiency of around 80-90% [5]. This system can operate on the existing natural gas distribution network using a reformer to convert methane to hydrogen thus minimizing infrastructure requirements. The ultimate goal is to use existing sustainable energy source such as wind, solar or tidal wave power to produce the hydrogen from electrolysis of water. These advantages make FC-CHP systems more attractive in the recent years, as the issue of energy sources reliability and availability become more pressing. Such systems are designed to provide high-quality electrical and thermal energy to a site regardless of what might occur on the power grid, thus ensuring improved power quality and minimize the impact of power outages on sensitive equipment. Most fuel cells use platinum-group-metals (PGM) as the electrodes catalyst. The PGMs are also essential to achieve low-temperature reforming to improve the efficiency of CHP systems. South Africa, with its vast PGM resources will benefit immensely from the development of FC-based CHP systems. The first generation of such fuel cell based CHP-systems were based on low temperature PEM fuel cells[6]. The challenge with LT-PEMFCs is their low operating temperature (<80°C), making it difficult to utilize the waste heat from the fuel cell for high temperature heating applications, and their low tolerance for CO (<10 ppm) in the feed [6]. Hence, new systems based on high temperature PEM fuel cells, operating at 140-160°C and capable of running on reformat with a much higher CO content (<300 ppm) are being developed [7]. The cell losses are lower at higher temperatures and compensate substantially for the decrease in the theoretical cell potential [4].

A combination of low-temperature reforming with a high-temperature HTPEM fuel cell (operating at 120°C – 180°C) opens a unique perspective in thermal management for domestic CHP systems, with a total efficiency yield of primary energy above 80% [1]. The high temperature operation of the PEMFCs tolerates relatively high fuel impurity levels [3, 6], thereby simplifies the reforming system with better fuel cell and reformer synergy [8]. Integrated systems consisting of a reformer that converts natural gas to hydrogen and a HTPEM fuel cell that converts the hydrogen to heat and power can provide an energy efficient option for small-scale (5-50 kW_{el}) natural gas based distributed CHP systems. The main challenges with HT-PEMFC based CHP-system is to develop durable materials (e.g. MEAs) that can last several thousand hours (up to 60 000 hours) and secondly to design energy efficient and low-cost integrated CHP-systems with low start up time [9]. The first challenge is mainly material science related. The latter however is mainly engineering related and arises because of the need to raise the temperature of the feed stream to the stack operating temperature. Recent studies [5, 10] have shown that this can be resolved by proper thermal integration of the system. The advantages of such system are only realizable with maximum recovery of process and waste heat co-produced with power in the fuel [11]. Developing efficient thermal management and integration strategies is therefore very essential to obtain the high overall system efficiency predicted.

The main thrust of this project is to employ the technique of pinch technology to evolve a novel heat integration scheme that will result in an optimal heat management in a domestic CHP system consisting of a 2 kW_{el} HTPEM fuel cell integrated with a methane steam reformer. This will be done by perform steady state system modelling and simulation of the complete mCHP system relating the balance of plant subcomponents. In achieving this broad objective, the study covers process modelling and simulation and heat integration. The first part developed detailed thermodynamic and kinetic models for the fuel processing that relate the various process conditions to hydrogen yield and CO content. The second part deals with the use of the pinch analysis methodology to obtain a heat integration scheme that will minimize the demand for external utilities and

hence improve the economics of the CHP system.

2. Systems Description

The proposed mCHP consists of the fuel processor, HTPEM fuel cell, after burner (to utilize the unreacted fuel) and the thermal recovery unit. The fuel processor sub-system consists of the reformer, and the additional units for CO removal: the water-gas shift, preferential oxidation and the methanator. A pre-heater is used to preheat methane and water to the steam methane reformer (SMR) temperature. The mixture is sent to the SMR filled with a catalyst, under adiabatic conditions. The dominant reforming reactions are highly endothermic favouring high reforming temperatures. The heat necessary for the endothermic reaction is supplied to the SMR via a burner. The reformat is purified in the water gas shift (WGS) and the preferential oxidation reactors. The hydrogen yield is increased in the 2-stages (high- and low-temperature) WGS reactor where the CO reacted with steam to form more hydrogen and carbon dioxide. The preferential oxidation (PrOx) is used to further reduced the reformat CO content with minimal hydrogen conversion to water.

The fuel processing simulation was carried out in AspenPLUS v7.2 environment to obtain optimal fuel processing conditions that maximizes hydrogen yield, reduce CO concentration to about 5 ppm and make high quality heat available for recovery and use. The SMR reactor is modelled as an isothermal reactor using the RPLUG model. The kinetics and equilibrium expressions developed by Xu and Froment [12] were used to account for materials flow. The HT-WGS and LT-WGS reactors are modelled as equilibrium reactor (REQUIL) while the RSTOIC model is used for the PrOx reactor. The rate expressions of Kim [13] and Choi [14] were used for the HT and LT WGS reactors respectively. In the PrOx reactors, it is desired that the CO content should be reduced to below 10ppm with minimal hydrogen consumption. A 95% of CO is therefore stipulated. The hydrogen rich reformat is fed to the anode side of the HTPEMFC operated at 160°C and 1 atm. The air for cathode side is filtered and pre-heated to 160°C, to reduce the start-up time of the stack. The anode and cathode stoichiometry were set at 1.25 and 2 respectively. The detail specifications of the HTPEM stack are described in Rabiu *et al.* [15]. The 2 kW_{el} stack consists of 96 cells of area 0.01m². The anode off-gas is sent to a flash drum to condense the steam before entering the afterburner where unreacted methane is combusted and the heat recovered. Heat integration was implemented to reduce the processes utilities requirement by setting feasible energy targets and achieving them through optimizing the heat recovery systems. ASPEN Energy Analyzer was used for the pinch analysis. To generate more feasible schemes, the remaining problem analysis was used to design the MER network.

3. Results

Figure 1 (a) present the effect of steam-to-carbon ratio (S/C) and temperature on the reformat compositions and yield of hydrogen. The methane conversion plot follows the same trend as Fig 1 (b). It could be seen from Figure 2, that excessive S/C only leads to increase moisture content of the reformat. The effect of pressure on methane is not profound (Figure 3). From these, a temperature of 780 °C and S/C = 3 was chosen, given a methane conversion of 98% in the SMR. The HTWGS at 400 °C gives a CO conversion of 76.5% while the LTWGS at 200 °C reduced the CO content to 5 ppm. Similar results have been reported [16-21]. On these bases, the simulation results were exported to Aspen Energy Analyzer module for heat integration study. Using a ΔT_{\min} 10°C, a threshold case (with zero cooling requirements) was obtained as expected due to the chosen SMR temperature, with minimum heat duty of 0.15kW. This present significant savings on utilities compared to the heating and cooling load of 1.239 kW and 1.09 kW respectively for existing (un-integrated) system.

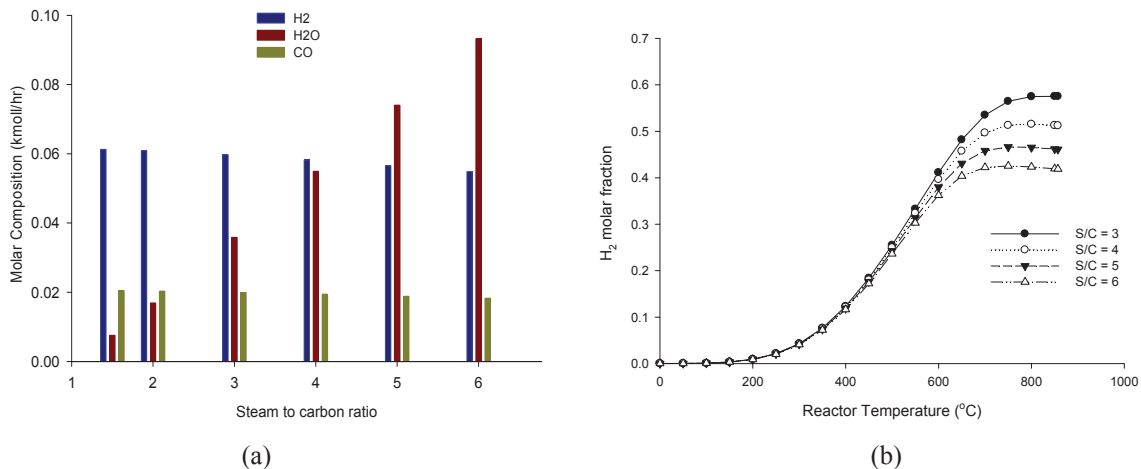


Fig. 1. Effects of S/C on (a) SMR products composition at T=780°C (b) on hydrogen production in the SMR at different T.

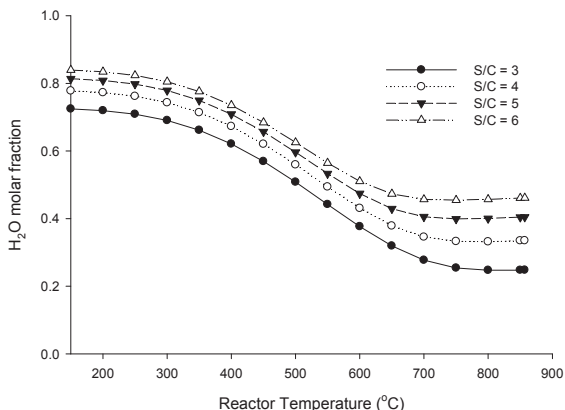


Fig. 2. Reformate water content as a function of S/C

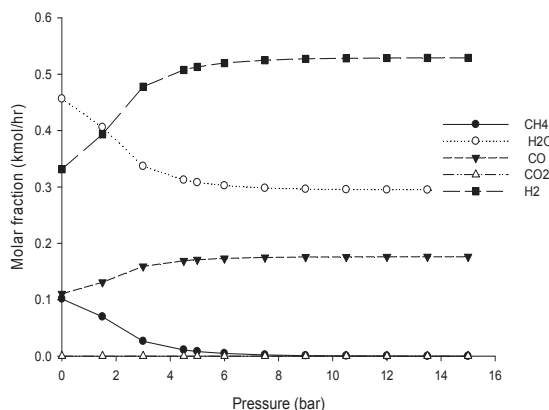


Fig. 3. Effect of pressure on reformate compositions

The pinch technology algorithms, together with the remaining problem analysis were used to obtain the grid network representation of the heat exchanger network design for maximum energy recovery in Figure 4. Heat exchangers are employed to recover the product heat content to preheat the feed stream into different reactors (Figure 5).

4. Conclusions

The simulation results show that the SMR favours the high temperature range of 650°C to 780°C and high S/C ratio at low operating pressure to maximize the high production of hydrogen with low CO concentration. For the CHP system a 47.5% and 38% electrical and thermal efficiency is obtained given an overall system efficiency of 85.5% for the 2 kW_{el} stack before heat integration. The integrated design features an overall

system efficiency of 88.91% an improvement of about 5%. Similar results have been obtained by [12]. The results show that the model developed has correctly predicted that effective heat integration of the plants components will improve the system efficiency.

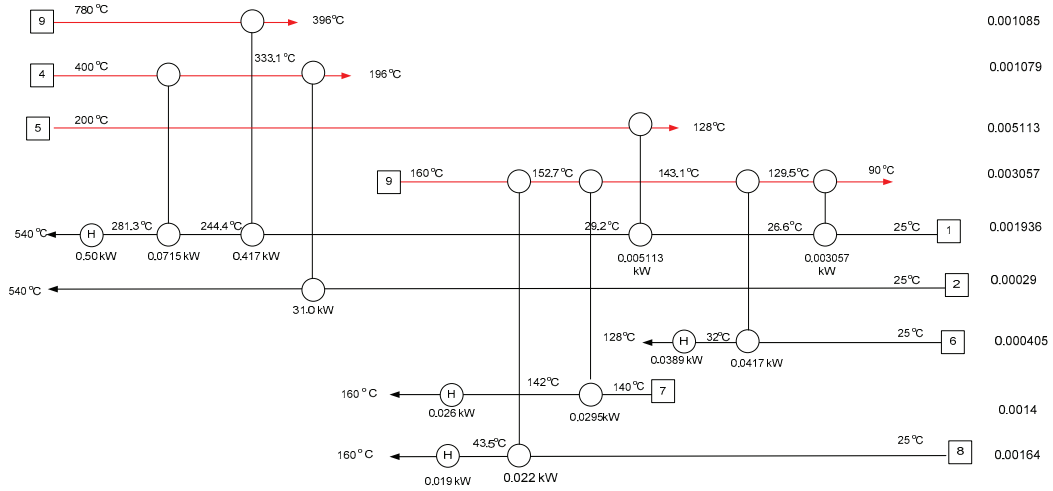


Fig. 4. Grid representation of the HEN design for MER

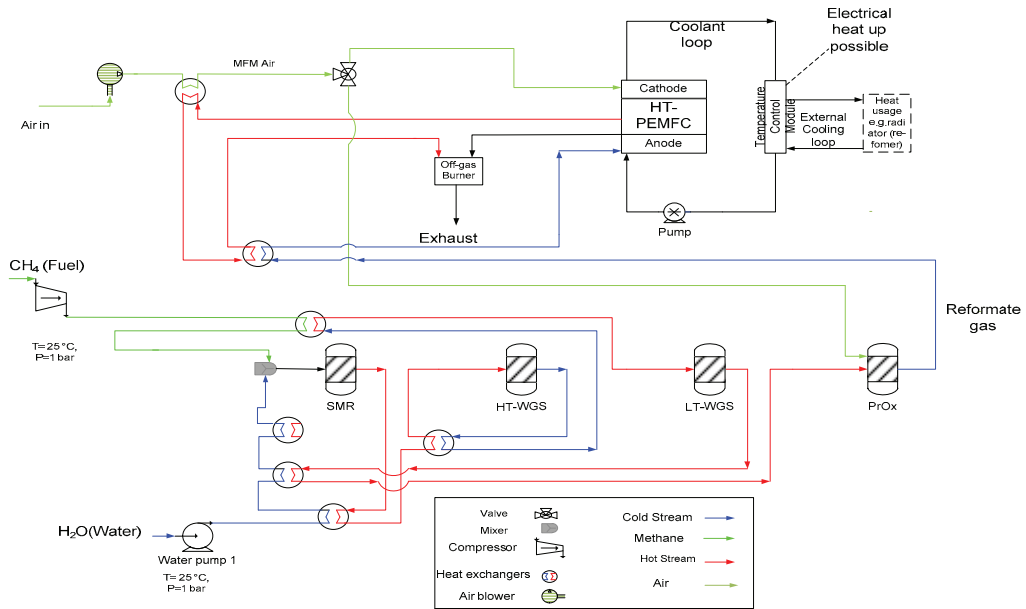


Fig. 5. The proposed integrated CHP System

Acknowledgements

The study is supported by the fund provided by the South African Departments of Science and Technology under the Hydrogen South Africa (HySA) program.

References

- [1] Barelli, L., Bidini, G., Gallorini, F., *et al.* Dynamic Analysis of PEMFC-Based CHP Systems for Domestic Application, *Applied Energy* 2012; 91:13-28.
- [2] Shaneb, O. A., Coates, G. and Taylor, P. C. Sizing of Residential μ CHP Systems, *Energy and Buildings* 2011; 43:1991-2001.
- [3] Wang, Y., Chen, K. S., Mishler, J., *et al.* A Review of Polymer Electrolyte Membrane Fuel Cells: Technology, Applications, and Needs on Fundamental Research, *Applied Energy* 2011; 88:981-1007.
- [4] Andreasen, S. J. and Kær, S. K. 400W High Temperature PEM Fuel Cell Stack Test, *Electrochemical Society* 2007; 5:197-207.
- [5] Arsalis, A., Nielsen, M. P. and Kær, S. K. Modeling and Parametric Study of a 1kW_e HT-PEMFC-Based Residential Micro-CHP System, *International Journal of Hydrogen Energy* 2011; 36:5010-5020.
- [6] Zhang, J., Tang, Y., Song, C., *et al.* Polybenzimidazole Membrane-Based PEM Fuel Cell in the Temperature Range of 120–200 °C, *Journal of Power Sources* 2007; 172:163-171.
- [7] Sousa, T., Mamlouk, M. and Scott, K. An Isothermal Model of a Laboratory Intermediate Temperature Fuel Cell Using PBI Doped Phosphoric Acid Membranes, *Chemical Engineering Science* 2010; 65:2513-2530.
- [8] Li, Q., Jensen, J. O., Savinell, R. F., *et al.* High Temperature Proton Exchange Membranes Based on Polybenzimidazoles for Fuel Cells, *Progress in Polymer Science* 2009; 34:449-477.
- [9] Department of Science and Technology (DST). Hydrogen and Fuel Cell Technologies Research, Development and Innovation Strategy. 5 February 2010 Available online at <http://www.dst.gov.za/fuelcells.html>.
- [10] Singdeo, D., Dey, T. and Ghosh, P. C. Modelling of Start-Up Time for High Temperature Polymer Electrolyte Fuel Cells, *Energy* 2011; 36:6081-6089.
- [11] Jiao, K., Alaefour, I. E. and Li, X. Three-Dimensional Non-Isothermal Modeling of Carbon Monoxide Poisoning in High Temperature PEM Fuel Cells with Phosphoric Acid Doped Polybenzimidazole Membranes, *Fuel* 2011; 90:568-582.
- [12] Xu, J. and Froment, G. Methane Steam Reforming, Methanation and Water-Gas Shift I: Intrinsic Kinetics, *AIChE J* 1989; 35:88-96.
- [13] Kim, K. Dynamic Proton Exchange Membrane Fuel Cell System Synthesis/Design and Operation/Control Optimization under Uncertainty. PhD Thesis, Virginia Polytechnic Institute and State University. Blacksburg, Virginia
- [14] Choi, W. New Approaches to Improve the Performance of the PEM Based Fuel Cell Power Systems. PhD Thesis, Texas A & M University. Houston, Texas
- [15] Rabiou, A., Nomnqa, M. and Ikhu-Omoregbe, D. Optimization Studies on the Performance of a High Temperature PEM Fuel Cell in *Proceeding of the International Science and Technology Conference*, Eds: A. Iqbal, *et al.*, ISTE 2011, Istanbul, Turkey, 2012:608-613
- [16] Harshini, D., Kwon, Y., Han, J., *et al.* Suppression of Carbon Formation in Steam Reforming of Methane by Addition of Co into Ni/ZrO₂ Catalysts, *Korean Journal of Chemical Engineering* 2010; 27:480-486.
- [17] Lima da Silva, A. and Müller, I. L. Towards H₂-Rich Gas Production from Unmixed Steam Reforming of Methane: Thermodynamic Modeling, *Journal of Power Sources* 2011; 196:8568-8582.
- [18] Oliveira, E. L. G., Grande, C. A. and Rodrigues, A. E. Methane Steam Reforming in Large Pore Catalyst, *Chemical Engineering Science* 2010; 65:1539-1550.
- [19] Sprung, C., Arstad, B. and Olsbye, U. Methane Steam Reforming over Ni/NiAl₂O₄ Catalyst: The Effect of Steam-to-Methane Ratio, *Topics in Catalysis* 2011; 54:1063-1069.
- [20] Urrejola, S., Sanchez, A., Cancela, A., *et al.* Study of Thermodynamic Factors for Equilibrium Reactions Involved in Steam Reforming of Natural Gas, *Chemistry and Technology of Fuels and Oils* 2011; 47:374-380.
- [21] Yue, B., Wang, X., Ai, X., *et al.* Catalytic Reforming of Model Tar Compounds from Hot Coke Oven Gas with Low Steam/Carbon Ratio over Ni/MgO–Al₂O₃ Catalysts, *Fuel Processing Technology* 2010; 91:1098-1104.