Integration of a Natural Gas Fuel Processor for Residential PEM Fuel Cell Based Micro Cogeneration Application

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Abstract

Fuel cell based cogeneration systems, which supply thermal energy and electricity simultaneously, are very promising for the near future commercial stationary applications, especially for residential power generation. The natural gas is the most suitable fuel for this application due to existing infrastructure and cost. The aim of this paper is to summarize the achievements of natural gas processor integration within "Fuel Cell based Micro Cogeneration System" project which is supported under TUBITAK Support Programme for Research Projects of Public Institutions. In this project 5 kW_e and 30 kW_t power are produced by fuel cell and catalytic combustion systems respectively. Sub systems such as fuel processing for hydrogen production, catalytic combustion, PEM fuel cell, power conditioning and control systems have been designed, manufactured and integrated for cogeneration application. Then a test program for the operation of the system has been performed. This paper presents the main experimental results on autothermal reforming of natural gas and gas clean-up systems concerning the requirements of PEM fuel cell system. It has been found that design parameters of the fuel processor were very important but operational parameters of the system were crucial for system performance.
Key words: Hydrogen, natural gas, reforming, micro cogeneration

1   Introduction

Coal and gas power plants which are the common centralized energy generation applications operate with 25–50% efficiency and cause high amount of greenhouse gas emissions [1, 2]. Decentralized power production, which is one of the most important research areas in advanced and clean energy technologies, supplies the energy demand by small scale systems established near the consumption area. The most important advantages of this method are the utilization of the alternative and renewable energy sources, availability for cogeneration, high efficiency (up to 90%), reduction in emission levels and avoidance of energy loss in transmission and distribution lines [3]. Micro turbines, internal combustion engines, Stirling engines and fuel cells are used for small scale electricity production [3]. Fuel cell systems are very efficient, reliable, silent and environment-friendly technologies and can be utilized in micro cogeneration systems on which intensive research has been conducted all over the world. Fuel cell based micro cogeneration systems, which supply thermal energy and electricity simultaneously, are very promising technologies for the near future stationary decentralized power generation applications. As well as the first prototypes were developed, the studies in this area still continue due to its importance from the point of clean energy [1-5].

2   Fuel Cell Based MiCro CoGeneraTIon SYstem

"Fuel Cell based Micro Cogeneration System" project, which has been conducted under TÜBİTAK Support Programme for Research Projects of Public Institutions by a consortium that includes TÜBİTAK Marmara Research Center Energy Institute, İstanbul Technical University, Kocaeli University and Türk Demirdöküm Fabrikaları A. Ş., has started on July 15, 2006 and is planned to be completed by the end of 2010. The aim of the project is to develop a 5 kWe Polymer Electrolyte Membrane (PEM) fuel cell based micro cogeneration system prototype to utilize the natural gas as a fuel to generate electricity and heat simultaneously for residential applications. The main units of the prototype are given below (Figure 1a):

- Hydrogen production unit via autothermal reforming of natural gas in a capacity range compatible with a 5 kW_e PEM fuel cell system,
- Gas clean up reactors for high and low temperature water-gas shift and preferential oxidation reactions,
- 5 kW_t catalytic burner unit for combustion of the anode-off gas of fuel cell,
- 30 kW_t natural gas burner for thermal power,
- 5 kW_e PEM fuel cell module and system,
- The control and power conditioning systems.

Firstly, requirements of main and sub-units of the hydrogen production and gas clean-up system were determined by process simulations. The most critical parameters for catalytic reactors were studied. Design and construction of reactors were realized according to the optimum operating conditions. Finally, reactors and sub-units were integrated to obtain a prototype unit (Figure 1b).
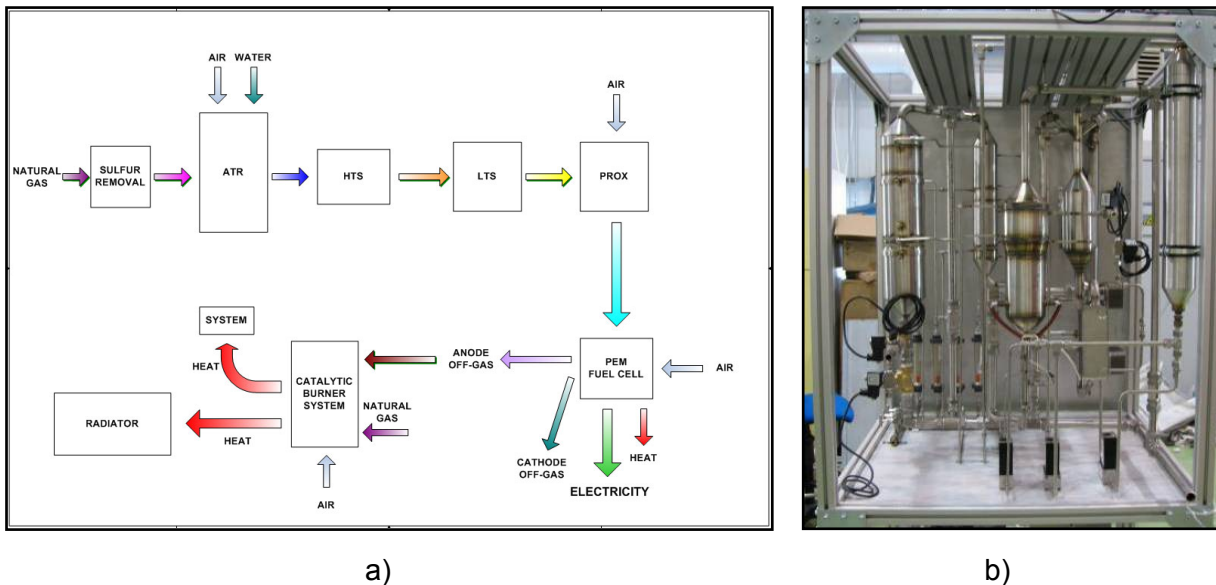

a)   b)

Figure 1:   a) Process flow diagram b) Process experimental set-up.

2.1 Autothermal reforming (ATR) unit

In this study, autothermal reforming of natural gas (NG) was studied in a catalytic reactor. NG consisted of ~ 90% methane (CH_4) and the main goal of this reaction was to convert CH_4 to obtain a syngas rich in hydrogen (H_2) with the lowest amount of CO formation. The selected primary operating variables in ATR reactor were temperature of the preheated fuel/air/steam mixture, steam-to-carbon (S/C) ratio, oxygen-to-carbon (O_2/C) ratio and reforming temperature (T_{ATR}). O_2/C ratio range was studied between 0.5–0.6, while a considerably wide S/C ratio range (2.0–3.0) was selected to see the effect on H_2 yield since lower S/C ratios could favor soot and coke formation which was not desired in catalytic operations. Figure 2 and Figure 3 show the effect of O_2/C ratio (at S/C = 3) and S/C ratio (at O_2/C = 0.55) on CH_4 conversion, H_2 and CO production rate at adiabatic conditions respectively.

In Figure 2, effect of O_2/C ratio on CH_4 conversion, CO and H_2 formation at constant S/C ratio of 3 can be seen. Increasing O_2/C ratio resulted in increasing CO and decreasing H_2 formations at constant S/C ratio. If O_2/C ratio was increased from 0.5 to 0.6, the H_2 concentration decreased from 45% to 40% at the reactor outlet stream. The CH_4 conversion was also positively affected by increasing O_2/C ratio at constant S/C ratio of 3. The conversion of CH_4 was observed around 99% at O_2/C ratio of 0.6, while it was decreased to 93% as O_2/C ratio decreased to 0.5.
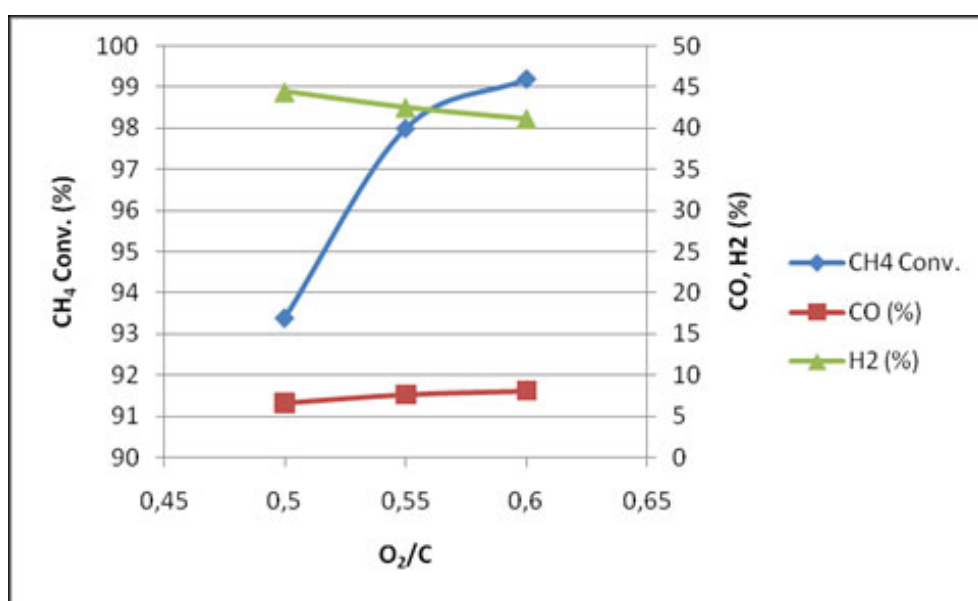

Figure 2: Effect of O_2/C ratio on CH_4 conversion, CO and H_2% composition (S/C = 3).

In Figure 3, the effect of S/C ratio on CH_4 conversion, CO and H_2 formation at constant O_2/C ratio of 0.55 is seen. The effect of S/C ratio on H_2 production was more pronounced at moderate O_2/C ratio of 0.55 and increasing the S/C ratio favored H_2 formation. On the other hand CO formation was depressed at higher S/C ratios and decreased as S/C ratio was increased. CH_4 conversion seemed to be slightly increased as S/C ratio was increased.
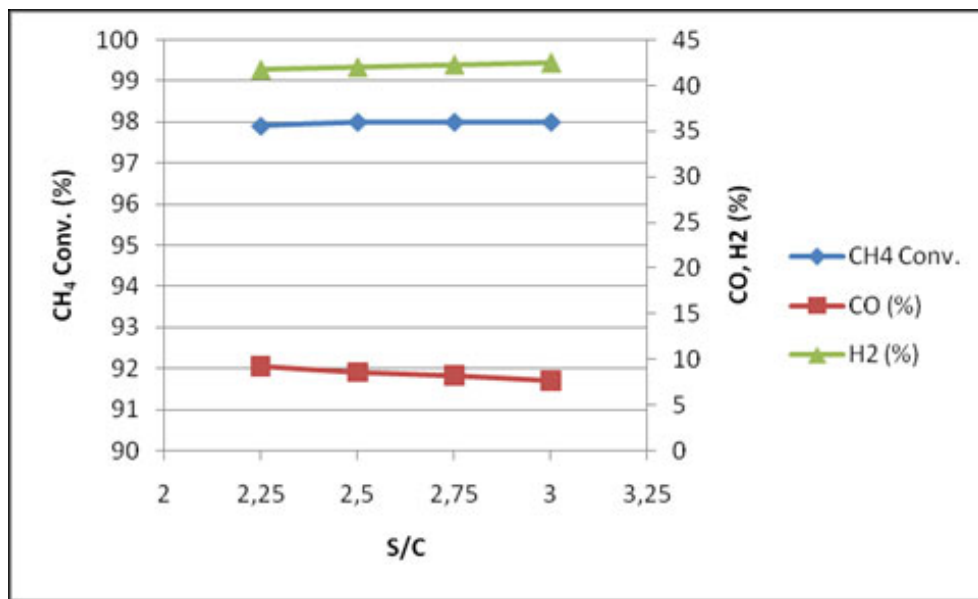
Figure 3:   Effect of S/C on CH$_4$ conversion, CO and H$_2$% composition (O$_2$/C = 0.55).

2.2   Gas clean up unit

Since fuel cell anode catalyst is deactivated by the CO concentration in the downstream of ATR, CO concentration should be gradually decreased to below 1% by high and low temperature water-gas shift reactors (HTS, LTS) and then to below 10 ppm by preferential oxidation (PrOx) reactors [6, 7].

Commercial catalysts were used for gas clean up (HTS, LTS, PrOx) reactors. Since the steam feed flow rate was optimized in order to maximize ATR performance and the amount of the unreacted steam in ATR downstream was enough for the HTS and LTS reactions, there is no additional water feeding to these reactors. So the only studied operating parameter for HTS and LTS was the feed temperature of the reactor. During the HTS tests, the effect of feed temperature on CO conversion was studied and the optimum operating temperature was determined to be 400°C at which maximum CO conversion of 69% was obtained as seen from Figure 4.
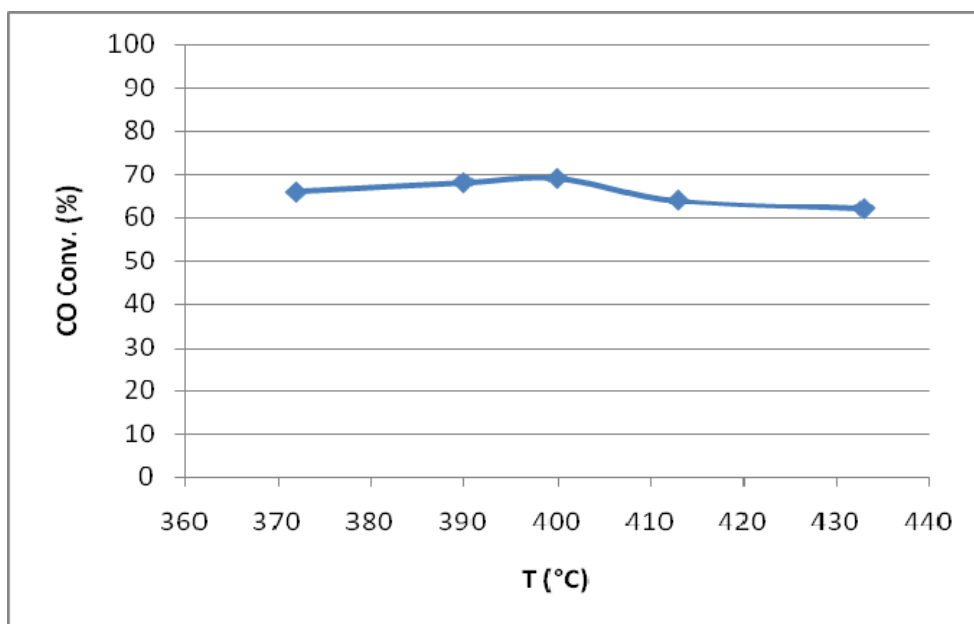
Figure 4:   Effect of feed temperature on CO conversion in HTS reactor.

The effect of feed temperature on CO conversion in the LTS reactor was studied and the maximum CO conversion of 60.2% was obtained at 298°C as seen in Figure 5. At these conditions, CO concentration was ~ 0.5%, which was suitable to be fed to PrOx reactors.
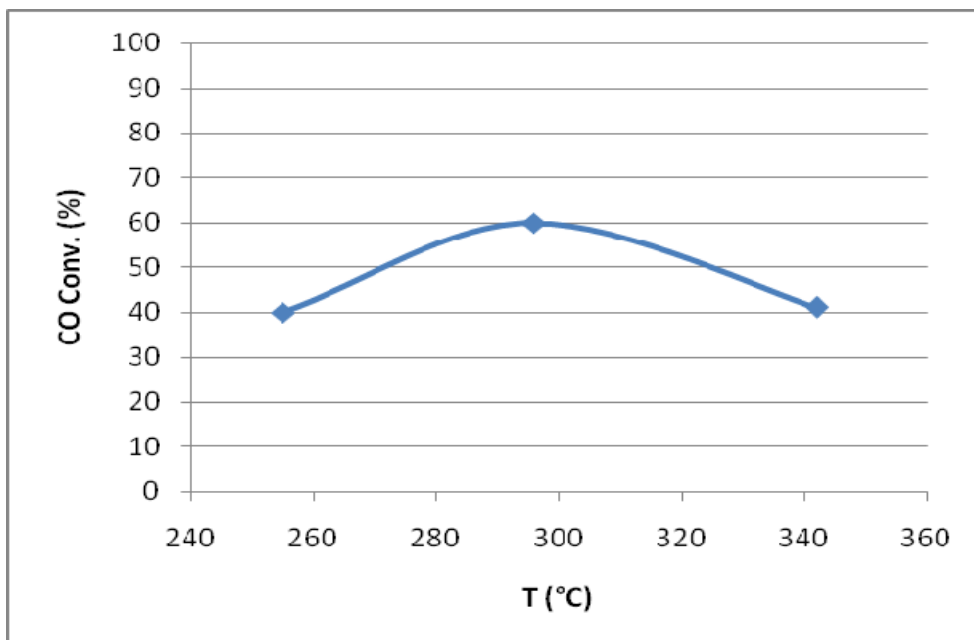

Figure 5:   Effect of feed temperature on CO conversion in LTS reactor.

CO conversion, selectivity, H_2 and CO composition at the outlet of the first stage of PrOx reactors (PrOx-1) at various air to fuel ratios (λ) were investigated. For the commercial PrOx

catalyst used, λ values between 2–2.8 were studied for PrOx-1 during the tests. Since the PrOx-1 reactor outlet contained trace amounts of CO (ppm level), an online infrared CO analyzer was used to measure CO amount and a gas chromatography was used to measure the other components. The outlet CO content was reduced to 250 ppm at the determined optimum value of λ.

During the tests of the second stage of PrOx reactors (PrOx-2), the air feed to the PrOx-1 reactor was adjusted to provide the determined optimum value of λ. CO conversion, selectivity, H_2 and CO composition at the outlet of the PrOx-2 reactor at various λ values were investigated. During the tests, in order to reduce the CO content of the PrOx-2 reactor outlet which would be fed to the PEM fuel cell, excess air was fed to the PrOx-2 reactor compared to the PrOx-1. Although the CO conversion was very high, the CO selectivity was estimated to be low since the excess air also reacted with some H_2 beside CO in the PrOx-2 reactor. However, the excess air feed (above the stoichiometric ratio) was not resulted in an unacceptable H_2 loss since the CO content at the inlet of the PrOx-2 reactor was very low. The outlet CO content was reduced down to 4 ppm which was acceptable to be fed to the PEM fuel cell.

3   Conclusion

In this paper, the main experimental results on autothermal reforming of NG and gas clean-up systems concerning the requirements of PEM fuel cell system have been presented. It has been found that design parameters of fuel processor were very important but operational parameters of the system were crucial for system performance and PEM fuel cell requirements.

In this study commercial catalysts were used in all the reactors. For autothermal reforming of NG, O_2/C ratio was studied between 0.5–0.6, while S/C ratio was studied between 2.0–3.0 to observe their effects on CH_4 conversion, CO and H_2 formation. As a result of the experiments, it was observed that increase in S/C ratio at constant O_2/C ratio increased the CH_4 conversion and H_2 production, while decreased the CO production. On the other hand, increase in O_2/C ratio at constant S/C ratio increased the CH_4 conversion and CO production, while decreased the H_2 production. For the HTS reactor, the feed temperature was optimized to be 400°C at which maximum CO conversion of 69% was obtained. In the downstream of the LTS reactor, the amount of CO was reduced down to 0.5% at 298°C at which maximum CO conversion of 60.2% was obtained. Even though the effect of inlet temperature on CO conversion can be obviously seen, the efficiencies of HTS and LTS reactors were not high enough. After the catalysts were replaced with the new ones, CO conversion was increased for both reactors. CO conversion, CO selectivity, H_2 and CO composition at the outlet of the PrOx-1 and PrOx-2 reactors at various air to fuel ratios (λ) were investigated. The optimum values of λ were determined for both reactors that resulted in 250 ppm CO content in the PrOx-1 outlet and 4 ppm CO content in the PrOx-2 outlet which was acceptable to be fed to the PEM fuel cell.

Acknowledgements

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