Computer-Aided Design of Micro-scale Fuel Processors

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Introduction

Recent advances in fuel cells and fuel reforming have enabled the efficient use of methanol and other liquid fuels as energy sources for small electronic devices, with the potential to replace the lower-energy density batteries currently in use [1, 2]. Development of a micro-scale fuel processor is particularly significant in the applied areas of intelligence, military, industrial sensoring and meteorology for high energy density and long duration.

Such a fuel processor is expected to be self-sustained without external heating. Thus design of the reactor with thermally high efficiency is important. Understanding the temperature distribution within the catalytic reactor, which is usually not measurable due to the size of the fuel processor, helps to optimize the reactor design and operating conditions. This paper describes transport phenomena and reaction kinetics in a micro methanol steam reformer and a selective carbon monoxide methanator with a three-dimensional pseudo-homogenous model. Numerical simulation provides local temperature distribution as well as carbon conversion in the fuel processor. Computer-aided reactor design tailors temperature profiles in the catalytic beds to achieve their maximum performance in producing low-CO-content hydrogen gas stream.

Materials and Methods

. The micro fuel processor is incorporated of three catalytic sections: methanol steam reforming, methanol combustion to supply the heat, and methanation. The total volume is less than 0.3cm^3 and the device weighs less than 1g. The reactor uses premixed liquid methanol and water with H_2O/C of 1.2. Supported Pd/ZnO on Al_2O_3 was used as methanol steam reforming catalyst [3-5] and 3%Ru on γ-Al_2O_3 as methanation catalyst. The mean diameter of the catalyst particles was 200μm. Methanol steam reformer has 14mg catalyst loading and the methanation bed consists of 13mg catalyst. Methanol and water mixture flowrate was varied from 0.03 to 0.2 cc/hr. The gaseous products were analyzed with an on-line gas chromatograph (Agilent QuadH) as well as an infrared CO gas analyzer (ZRH, California Analytic Instruments).

A three dimensional pseudo-homogeneous model was formulated based upon the first-principles of material and energy balance for the catalytic reactions of methanol steam reforming and selective CO methanation. The model was constructed on the FEMLAB platform where the parameters for different processes, such as steam reforming, methanation, and heat transfer in the insulation layer etc., were assigned within their geometrical sub-domains. Convection-diffusion and convection-conduction modules in the FEMLAB library were used as two multiphysics representing mass and heat transfer. The following general partial differential equations govern all the process domains but with specific coefficients in it.

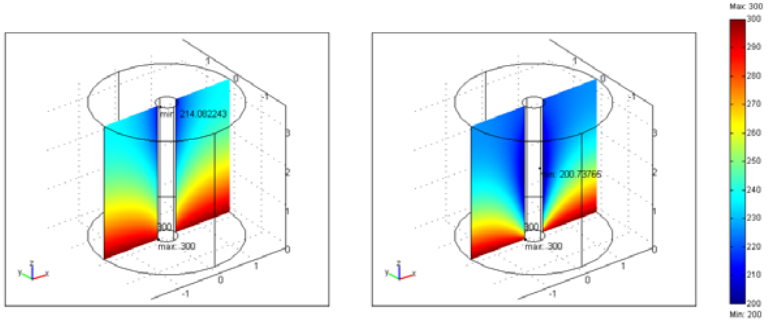
$$\nabla \cdot (-D_{eff} \nabla c) + \vec{u} \cdot \nabla c = R \quad (1)$$

$$\nabla \cdot (-k_{eff} \nabla T) + \rho C_p \vec{u} \cdot \nabla T = R(-\Delta H_r) \quad (2)$$

where R is the disappearing rate of the key component (MeOH or CO), which is function of component concentration c (or partial pressure p) and temperature T; ΔH_r is the heat of reaction; k_{eff} is the effective conductivity of the catalyst bed; D_{eff} is the effective diffusivity; and $\vec{u}$ is linear velocity.

Results and Discussion

Design options of the fuel injector were evaluated with the 3D model in an adiabatic methanol steam reforming reactor. Figure 1 shows the temperature profiles in the steam reforming catalyst pellet at the conditions correspondent to 80-mWe power output with two different fuel injection port locations. The significant endotherm of hydrocarbon steam reforming and the large latent heat of rapid liquid fuel vaporization are observed in both cases, which show cold spots in the center of the catalyst pellet. The maximum temperature difference in the reformer has been found to be 86°C throughout the entire reformer when the injection port is located at the entrance of the feeding tube. 100% overall methanol conversion was achieved at a weight hourly space velocity of 1.84g_{MeOH}/g_{cat}/hr in the above temperature field. As a design option, moving the fuel injection port downwards along the fuel feeding tube results in lower average temperature. Consequently, the overall methanol conversion was reduced to 90%. The fuel injection location was optimized in such a way that maximum carbon conversion can be achieved at a relatively uniform temperature distribution.



(a) (b)

Figure 1. Temperature distribution in a micro-scale methanol steam reformer with (a) an entrance fuel injection; (b) a lower injection port. (Temperature in °C, "adiabatic mode" ; Tcombustion=300°C, liquid fuel flowrate=0.05cc/hr, Steam/C=1.2)

The computational study was then applied to optimizing insulation thickness to ensure desired reaction temperature and high power density. Simulation shows that he maximum temperature difference within the reformer with half of inch insulation was 153°C that is very close to the

measured temperature difference 148°C with the two thermocouples. Compared to the ideal adiabatic steam reformer pellet, the temperature gradient in this realistically insulated reformer is larger due to the heat loss. The skin temperature of the insulation was found to be 50°C, and the heat loss of the entire reformer was 0.58W out of the methanol combustion heat generation source 1.39W. It is observed that energy loss to the environment via convective heat transfer and radiation is substantial comparing to the required heat supply of 71.5mW for fuel vaporization and steam reforming reaction. Subsequently, the thickness of the insulation was varied in the model for optimization. The simulation results indicate that the skin temperature of the insulated device non-surprisingly decreases with an increase of the insulation thickness. However, the total heat loss (W) shows a peak value at the point of 6.35mm insulation thickness. The less heat loss with thinner insulation (3.175mm) is due to its smaller heat transfer area, although it has larger driving force and heat transfer coefficient. Thick insulation also results in large surface area that increases the total heat loss. Additionally, the volume of the insulated device must be taken into account so that high energy density can be maintained.

As a methanator was used for onsite CO reduction, hydrogen consumption must be minimized by operating the catalyst at its desired temperature range. One typical design configures the methanation section of the fuel processor as a "cap" on top of the reformer. The simulated temperature profile shown in Figure 2 is correspondent to the conditions of the 80mWe output. It is observed that the steam reformer was operated at the temperature range from 270 to 300°C, and the CO methanator at 240-250°C. It has been experimentally demonstrated that less than 100ppm CO was achieved under the above conditions, but the hydrogen consumption was approximately 15%, which is higher than the anticipated 7-10% [5]. This indicates that methanation of CO_2 might have also occurred in the catalyst bed which is possibly at higher actual temperature. The methanation reactor temperature profile could be further tailored using such a CAD approach to achieve the maximum performance.

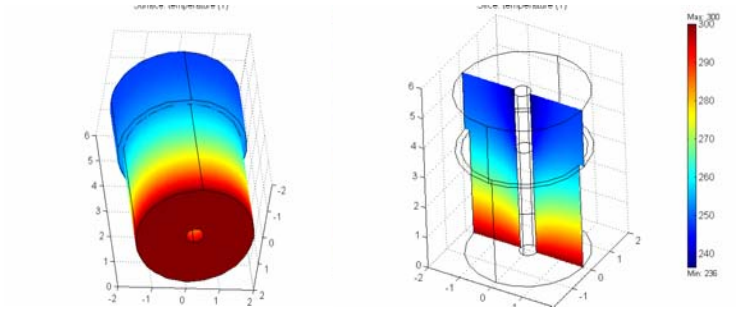


Figure 2. Temperature profiles in a micro-scale methanol steam reformer and selective CO methanator (Temperature in °C, "insulation mode", fuel injection point: tube inlet; Tcombustion=300°C, liquid fuel flowrate=0.05cc/hr, Steam/C=1.2)

Significance

This work provides a useful tool to reveal local temperature distributions within the complex of micro fuel processors. The simulation results were used for reactor design and optimization.

References

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