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Miniaturized Methanol Reformer for Fuel Cell Powered Mobile Applications

John C. Telotte*

Jesse Kern[†]

Srinivas Palanki[‡]

*Florida State University, telotte@eng.fsu.edu

[†]Florida State University, jesse.kern@gmail.com

[‡]University of South Alabama, spalanki@usouthal.edu

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Miniaturized Methanol Reformer for Fuel Cell Powered Mobile Applications*

John C. Telotte, Jesse Kern, and Srinivas Palanki

Abstract

In this paper, the design of a miniaturized methanol reformer is considered that can operate in two different modes to produce sufficient hydrogen for generating a net power of 24 W and 72 W. The reformer is modeled as a radial flow packed bed reactor and the Ergun equation is used to model the pressure drop. Simulation studies are conducted to study the effect of steam to methanol ratio, inlet pressure and reactor temperature on the production of hydrogen. It is shown that a volume of 20 ml is required to produce sufficient hydrogen for generating the necessary power if an inlet pressure of 202 kPa and a steam to methanol ratio of 1.5 is used. A temperature of 500 K is required for the lower power application while a temperature of 550 K is required for the higher power application.

KEYWORDS: methanol reformer, modeling, fuel cell

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Introduction

The demand for power sources with superior performance has increased as a result of rapid growth of portable electronics [Kundu *et al.* 2007]. This has led to interest in developing alternatives to conventional batteries, which are thought to be approaching their limits in energy storage capacity [Service, 2002]. One promising approach involves the use of small fuel cell stacks (< 25 ml) that utilize hydrogen to produce power in the 10-100 W range. There are several applications in this power range where fuel cells are a viable alternative to conventional batteries. Some of these applications are radios (6-7 W), computer displays (3-7 W), Sensors (9.5 W), and computers (17-18 W) [NRC, 2004]. Furthermore, several unmanned air vehicles such as BATCAM, Hornet and Wasp require about 24 W of power for cruising and a peak power of around 72 W for climbing [UAS Roadmap, 2005].

Several researchers have utilized conventional microfabrication techniques to develop micro-scale and small-scale fuel cells that utilize pure hydrogen to produce power in the 10-100 W range [Kelley et al., 2002; Ormerod, 2003]. However, the difficulties and hazards involved in the storage and handling of hydrogen fuel in either compressed gas or liquid form have been a major hurdle for successful commercialization of both large as well as miniature hydrogen proton exchange membrane (PEM) fuel cells [Pattekar and Kothare, 2005]. Furthermore, the stored energy density of liquid hydrocarbons, which can be reformed to produce hydrogen on demand, is significantly higher as compared to hydrogen. For these reasons, the development of miniaturized reformers that provide hydrogen to power small fuel cell stacks has been a subject of considerable research activity [Holladay, 2002; deWild and Verhaak, 2000]. In particular, reformers that utilize methanol to produce hydrogen have attracted considerable attention. Methanol is sulfur-free and offers high hydrogen-to-carbon ratio, high energy density (5600 Wh/kg), ready availability, and low boiling point [Choi and Stenger, 2002]. Pattekar and Kothare [2005] fabricated a radial flow micro-packed-bed reactor via deep reactive ion etching that utilizes methanol to generate sufficient hydrogen for a 20 W power application. Kundu *et al.* [2007] developed a serpentine patterned micro-reformer that converts methanol to hydrogen via steam reforming. Shah and Besser [2008] developed an integrated silicon microreactor-based methanol steam reformer that produces sufficient hydrogen for 0.38 W of power. Sohn *et al.*, [2007] developed a plate-type integrated fuel processor-PEM fuel cell where methanol is reformed to produce upto 150 W of power.

While there is considerable literature on fabrication of micro reformers that demonstrate the feasibility of utilizing methanol to produce hydrogen for micro

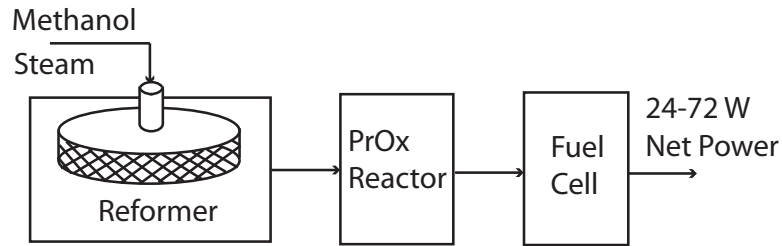


Figure 1: Schematic of Process

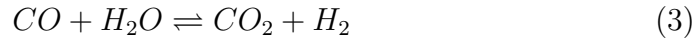
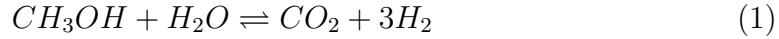
and small scale applications, there are very few papers in modeling and analysis of these reactors, which is necessary for optimizing performance. In particular, the literature is very sparse in papers where reaction kinetics are utilized to determine the size and performance of micro reformers. In this paper, the design of a reformer is considered that has the capability to generate 24 W of net continuous power and 72 W of net peak power. This device would have potential applications for both micro UAVs as well as mobile electronics equipment.

System Description

A schematic of the process under consideration is shown in Figure 1. The reformer is a disc-shaped radial-flow packed bed reactor similar to the one fabricated by Pattekar and Kothare [2005]. Methanol and water enter the reactor from the center of the disc where they are vaporized and brought to the reaction temperature via an external heat source. The reactor is packed with $Cu/ZnO/Al_2O_3$ catalyst particles. The reacting and product gases flow outward radially. The product gas stream is then passed through a preferential oxidation reactor to convert carbon monoxide to carbon dioxide and the resulting hydrogen and carbon dioxide mixture is sent to a fuel cell where an electrochemical reaction occurs to generate power.

Reformer Modeling and Design

The following reactions take place in the reformer:



Kinetic expressions for the above reactions were developed by Peppley *et al.* (1999a, 1999b) and corrected in Peppley (2006) and are shown below:

$$r_1 = \frac{k_R K_{CH_3O(1)}^* \left(\frac{P_{CH_3OH}}{P_{H_2}^{0.5}} \right) \left(1 - \frac{P_{H_2}^3 P_{CO_2}}{K_R P_{CH_3OH} P_{H_2O}} \right) C_{S_1}^T C_{S_{1a}}^T S_C \rho_b}{\left(1 + K_{CH_3O(1)}^* \left(\frac{P_{CH_3OH}}{P_{H_2}^{0.5}} \right) + K_{HCOO(1)}^* P_{CO_2} P_{H_2}^{0.5} + K_{OH(1)}^* \left(\frac{P_{H_2O}}{P_{H_2}^{0.5}} \right) \right) \left(1 + K_{H(1a)}^{0.5} P_{H_2}^{0.5} \right)} \quad (4)$$

$$r_2 = \frac{k_D K_{CH_3O(2)}^* \left(\frac{P_{CH_3OH}}{P_{H_2}^{0.5}} \right) \left(1 - \frac{P_{H_2}^2 P_{CO}}{K_D P_{CH_3OH}} \right) C_{S_2}^T C_{S_{2a}}^T S_C \rho_b}{\left(1 + K_{CH_3O(2)}^* \left(\frac{P_{CH_3OH}}{P_{H_2}^{0.5}} \right) + K_{OH(2)}^* \left(\frac{P_{H_2O}}{P_{H_2}^{0.5}} \right) \right) \left(1 + K_{H(2a)}^{0.5} P_{H_2}^{0.5} \right)} \quad (5)$$

$$r_3 = \frac{k_W K_{OH(1)}^* \left(\frac{P_{CO} P_{H_2O}}{P_{H_2}^{0.5}} \right) \left(1 - \frac{P_{H_2} P_{CO_2}}{K_W P_{CO} P_{H_2O}} \right) (C_{S_1}^T)^2 S_C \rho_b}{\left(1 + K_{CH_3O(1)}^* \left(\frac{P_{CH_3OH}}{P_{H_2}^{0.5}} \right) + K_{HCOO(1)}^* P_{CO_2} P_{H_2}^{0.5} + K_{OH(1)}^* \left(\frac{P_{H_2O}}{P_{H_2}^{0.5}} \right) \right)^2} \quad (6)$$

These expressions were developed by considering surface mechanism for the main reforming reactions on $Cu/ZnO/Al_2O_3$ catalyst particles. Progressive improvements were made to the mechanisms based on experimental observations. A set of Langmuir-Hinshelwood rate expressions were derived based on steady-state analysis of the final surface mechanism. Rate data were collected for a large range of experimental conditions using a fixed-bed differential reactor (Peppley *et al.*, 1999). The kinetic and thermodynamic parameters for the rate expressions were obtained via nonlinear least squares regression. The resultant model was validated by comparing with experimental observations over a wide range of temperatures and pressures and it was observed that the model predictions were in good agreement with experimental observations

(Peppley *et al.* 1999b; Asprey *et al.* 1999). Given this experimental validation, these rate expressions can be used with a high degree of confidence for designing the miniaturized reformer under consideration in this paper.

The reformer is modeled as a radial-flow packed bed reactor. The pressure drop is modeled via the Ergun Equation [Fogler, 2006]. It is assumed that the reactor is jacketed and sufficient heat is provided for the reactor to operate isothermally. This heat will be generated using power from the fuel cell. Details of proposed operation of such a system can be found in Palanki *et al.* [2007] where mass and energy balance calculations were performed to determine the additional flow rate of hydrogen required to produce sufficient power for heating the reformer.

The steady-state model equations for each species are given as follows:

$$\frac{dF_{CH_3OH}}{dz} = (-r_1 - r_2)A_c \quad (7)$$

$$\frac{dF_{H_2O}}{dz} = (-r_1 - r_3)A_c \quad (8)$$

$$\frac{dF_{CO_2}}{dz} = (r_1 + r_3)A_c \quad (9)$$

$$\frac{dF_{H_2}}{dz} = (3r_1 + 2r_2 + r_3)A_c \quad (10)$$

$$\frac{dF_{CO}}{dz} = (r_2 - r_3)A_c \quad (11)$$

where F_{CH_3OH} , F_{H_2O} , F_{CO_2} , F_{H_2} , F_{CO} are the molar flow rates of methanol, steam, carbon dioxide, hydrogen, and carbon monoxide respectively and z is the radial dimension of the reactor. The term, A_c , which represents the area of cross section of the reactor that is perpendicular to the flow, varies with z for the radial flow reactor.

The pressure drop equation is given by:

$$\frac{dP}{dz} = -\frac{G}{\rho D_p} \left(\frac{1 - \phi}{\phi^3} \right) \left[\frac{150(1 - \phi)\mu}{D_p} + 1.75G \right] \quad (12)$$

where P is the reactor pressure, ϕ is the void fraction, D_p is the diameter of the catalyst particle in the reformer, μ is the viscosity of the gas mixture, ρ is the gas mixture density and G is the superficial mass velocity. Pattekar and Kothare [2005] used an alternative approach to model the flow characteristics of the system by using a modified form of the Navier-Stokes equation in conjunction with Darcy's law, which is valid for laminar flow conditions as would most likely exist in the device fabricated by Pattekar and Kothare

[2005]. However the approach described in this paper is valid for a wider range of flow rates that include both laminar and turbulent flow.

The Ergun equation requires the computation of the gas mixture density, ρ , as well as the gas mixture viscosity, μ , as a function of reactor length. The mixture density is estimated by computing the molar average density of the reacting gas mixture at each integration step. Due to the presence of hydrogen in the gas mixture, use of a simple mole fraction average method can lead to significant errors in the overall gas mixture viscosity [Bird *et al.*, 2006]. For this reason, a more rigorous kinetic theory expression from Reid *et al.* [1977] is utilized to estimate the gas viscosity at each integration step.

Table 1 lists the values of the kinetic parameters used in simulation studies in this research. The reaction rates r_1 , r_2 and r_3 are functions of the partial pressure of the various species involved in reactions 1-3. The partial pressures were written in terms of molar flow rates as follows by assuming that the species follow ideal gas behavior:

$$P_j = \frac{F_j}{\sum F_j} P \quad (13)$$

where $j = CH_3OH, H_2O, CO_2, H_2, CO$.

The reactor system under consideration must be designed to operate in two distinct modes. For a short period of time, a large hydrogen generation rate is required (to generate a net power of 72 W) and for a longer period of time, a considerably lower hydrogen generation rate is required (to generate a net power of 24 W). These design constraints are met by a combination of choice of operating conditions (temperature, pressure, steam to methanol ratio) and size of the reformer. There are trade-offs in these parameters. For instance, operating at higher temperature yields smaller volumes; however significantly higher power needs to be generated to get a net power of 24 W and 72 W respectively to keep the reactor isothermal at the higher temperature.

The design equations described by equations 7-12 were integrated numerically in MATLAB using the stiff ordinary differential equations routine *ode15s*. Preliminary calculations were performed for high inlet feed rates to determine the approximate reactor volume required to achieve sufficient hydrogen production to generate a net power of 72 W. The reformer size parameters determined from this calculation are shown in Table 2 along with details of the catalyst. Once the reactor size was fixed, two different flow rates of methanol entering the reformer were considered: $9.333 \times 10^{-5} \text{ mol/s}$ and $3.5 \times 10^{-4} \text{ mol/s}$. If the methanol conversion is high ($\sim 95\%$), these flow rates will produce hydrogen flow rates of $2.6 \times 10^{-4} \text{ mol/s}$ and $1 \times 10^{-3} \text{ mol/s}$ respectively. These flow rates are sufficient to generate 24 W and 72 W of net power respectively

Parameter	Value
k_R	$7.4 \times 10^{14} \exp(-102800/(RT)) \text{ m}^2/\text{mol.s}$
k_D	$3.8 \times 10^{20} \exp(-170000/(RT)) \text{ m}^2/\text{mol.s}$
k_W	$5.9 \times 10^{13} \exp(-87600/(RT)) \text{ m}^2/\text{mol.s}$
K_R	$10^{(1.4142 \times 10^{-13} T^5 - 4.2864 \times 10^{-10} T^4 + 5.3993 \times 10^{-7} T^3 - 3.6385 \times 10^{-4} T^2 + 1.4096 \times 10^{-1} T - 2.0258 \times 10^1)} \text{ bar}^{-2}$
K_D	$10^{(2.9463 \times 10^{-13} T^5 - 8.8919 \times 10^{-10} T^4 + 11.1130 \times 10^{-6} T^3 - 7.4160 \times 10^{-4} T^2 + 2.7969 \times 10^{-1} T - 4.4944 \times 10^1)} \text{ bar}$
K_W	$10^{(-1.4936 \times 10^{-13} T^5 + 4.5026 \times 10^{-10} T^4 - 5.6216 \times 10^{-7} T^3 + 3.7206 \times 10^{-4} T^2 - 1.3726 \times 10^{-1} T + 2.4537 \times 10^1)}$
$K_{CH_3O(1)}^*$	$\exp(41.8/R - (-20000/(RT))) \text{ bar}^{-0.5}$
$K_{CH_3O(2)}^*$	$\exp(30/R - (-20000/(RT))) \text{ bar}^{-1}$
$K_{HCOO(1)}^*$	$\exp(-179200/(RT)) \text{ bar}^{-1.5}$
$K_{OH(1)}^*$	$\exp(-44.5/R - (-20000/(RT))) \text{ bar}^{-1.5}$
$K_{OH(2)}^*$	$\exp(30/R - (-20000/(RT))) \text{ bar}^{-1}$
$K_{H(1a)}$	$\exp(-100.8/R - (-50000/(RT))) \text{ bar}^{-1}$
$K_{H(2a)}$	$\exp(-46.2/R - (-50000/(RT))) \text{ bar}^{-1}$
$C_{S_1}^T$	$7.5 \times 10^{-6} \text{ mol/m}^2$
$C_{S_{1a}}^T$	$1.5 \times 10^{-5} \text{ mol/m}^2$
$C_{S_2}^T$	$7.5 \times 10^{-6} \text{ mol/m}^2$
$C_{S_{2a}}^T$	$1.5 \times 10^{-5} \text{ mol/m}^2$

Table 1: Kinetic Parameters

Reactor Parameter	Value
Catalyst particle diameter, D_p	0.00005 m
Catalyst density, ρ_b	1300 kg/m ³
Specific surface area, S_C	102000 m ² /kg
Void fraction, ϕ	0.38
Reactor height, h	0.01 m
Reactor inner radius, r_1	0.0001 m
Reactor outer radius, r_2	0.02523 m

Table 2: Reactor Parameters

[Palanki *et al.*, 2007]. The effect of varying operating temperature, pressure and steam to methanol ratio on hydrogen production were studied.

Results and Discussion

The use of a radial flow reactor was considered because of the potential for reduced pressure drop as compared to conventional tubular packed bed reactor [Pattekar and Kothare, 2005]. Figure 2 shows the effect of pressure drop through the radial flow reactor as a function of reactor volume. For this simulation, the operating temperature was 500 *K*, inlet pressure was 202 *kPa*, steam to methanol ratio of 1.5 and a methanol flow rate of 9.333×10^{-5} *mol/s*. It is observed that in this reactor geometry the pressure drop is negligible. The increase in moles due to reaction as well as the increasing area perpendicular to the flow are able to balance the pressure drop due to resistance to flow by the catalyst packing. These results were compared with the pressure drop calculations in a conventional tubular packed bed reactor with a diameter of 0.01 *m* and a length of 0.255 *m* that was operated under the same reaction conditions as the radial flow reactor. The reactor dimensions of the tubular reactor were chosen so that the volume of the tubular reactor was the same as that of the radial flow reactor (20 *ml*) and the diameter of the tubular reactor was equal to the height of the radial flow reactor. It is observed from Figure 2 that there is significant pressure drop in the tubular packed bed reactor. These results agree qualitatively with the experimental observations of Pattekar and Kothare [2005].

Figure 3 shows the effect of changing the inlet steam to methanol ratio. For this simulation, the operating temperature was 500 *K* and the inlet pressure was 202 *kPa*. Three different steam to methanol ratios were simulated: 1:1, 1.5:1 and 2:1. It is observed from Figure 3 that for an inlet flow rate of methanol of 9.333×10^{-5} *mol/s*, there is very little effect in the production of hydrogen for the three different ratios. Furthermore, the hydrogen flow rates obtained are close to the flow rates obtained if the reactor was at equilibrium, which indicates that the reactor is operating at close to optimal conditions for hydrogen production. Table 3 shows the percentage of carbon monoxide for the three different steam to methanol ratios. These values are also compared with equilibrium values. It is observed that as the steam to methanol ratio is increased, the percent carbon monoxide coming out of the reactor decreases, which results in a lower volume for the preferential oxidation reactor that is downstream of the reformer. However, increasing steam to methanol ratio comes at the cost of requiring more steam, which increases energy cost. For

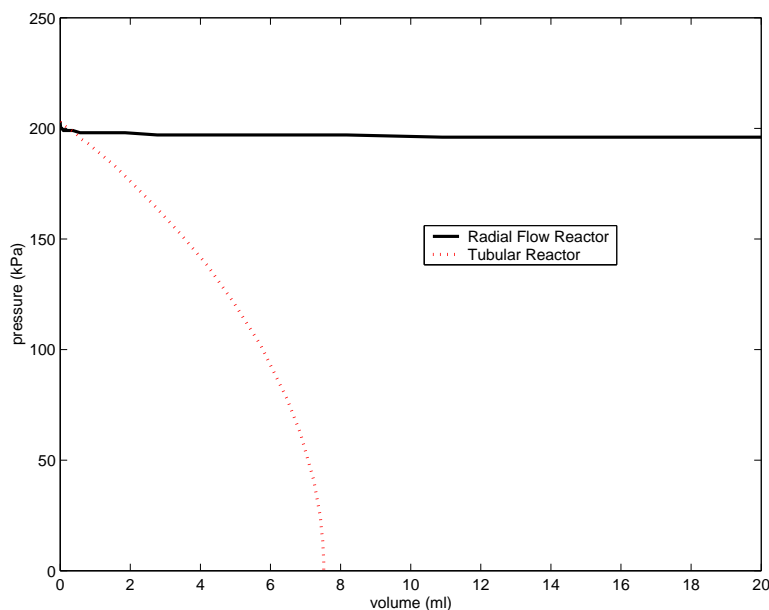


Figure 2: Effect of Reactor Geometry on Pressure Drop

Steam to Methanol Ratio, R	CO (kinetic)	CO (equilibrium)
1.0	2.39 %	2.86 %
1.5	0.76 %	0.93 %
2.0	0.39 %	0.44 %

Table 3: Effect of Steam to Methanol Ratio on Carbon Monoxide Production at Methanol Inlet Flow Rate of $9.333 \times 10^{-5} \text{ mol/s}$

this reason, a steam to methanol ratio of 1.5 was used in the remainder of this paper. A comparison with equilibrium values of carbon monoxide indicates that the carbon monoxide coming out of the reactor is much lower than the equilibrium values for the reactor conditions considered. This is a desirable result since a smaller percentage of carbon monoxide in the reformer outlet results in a smaller volume for the preferential oxidation reactor to convert carbon monoxide to carbon dioxide. Similar results were observed for an inlet flow rate of methanol of $3.5 \times 10^{-4} \text{ mol/s}$.

Figures 4 and 5 show the effect of temperature on hydrogen production for inlet methanol flow rates of $9.333 \times 10^{-5} \text{ mol/s}$ and $3.5 \times 10^{-4} \text{ mol/s}$. The inlet pressure is 202 kPa . Tables 4 and 5 show the effect of temperature on methanol conversion for the lower and higher methanol inlet flow rates. At an

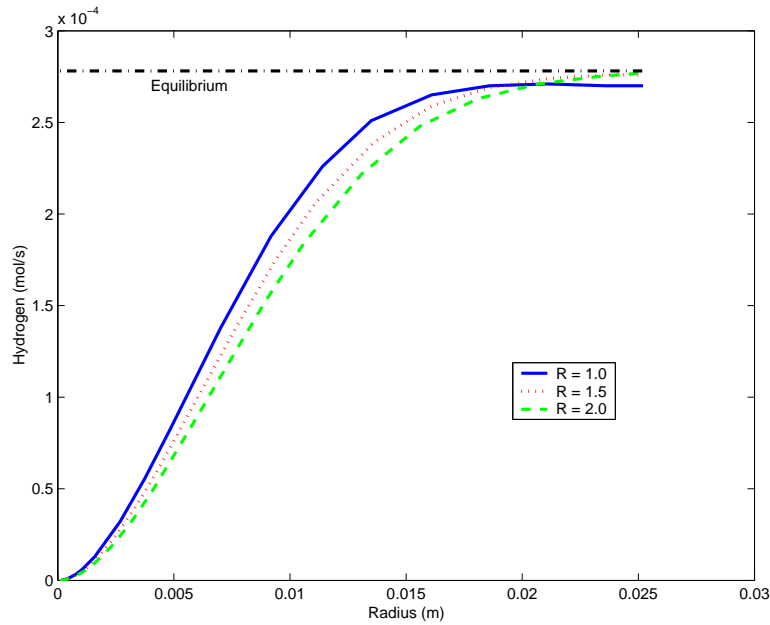


Figure 3: Effect of Changing Steam to Methanol Ratio on Hydrogen Production

operating temperature of 500 K, it is observed that the conversion of methanol is 99.8% for an inlet methanol flow rate of $9.333 \times 10^{-5} \text{ mol/s}$. However, when the inlet methanol flow rate is increased to $3.5 \times 10^{-4} \text{ mol/s}$, the conversion drops to 82.2%, mainly due to decreased residence time in the reactor. The conversion can be increased by either utilizing a larger reactor or by increasing temperature when the higher flow rate is used. Since increasing volume reduces energy density of overall reformer-fuel cell system, it is suggested that the reformer operate at a higher temperature when the higher flow rate is used (for 72 W of net power) and the temperature be reduced when the lower flow rate is used (for 24 W of net power). In Table 4, it is observed that as temperature is varied from 450 K to 500 K at the lower methanol inlet flow rate, methanol conversion increases, reaching almost 100% at 500 K. In Table 5, it is observed that as temperature is varied from 450 K to 550 K at the higher methanol inlet flow rate, methanol conversion increases, reaching almost 100% at 550 K.

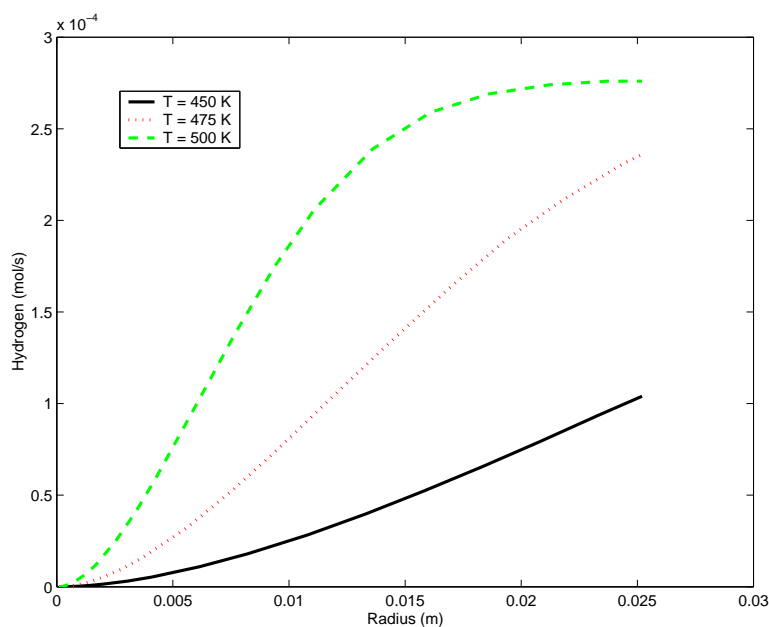


Figure 4: Effect of Temperature on Hydrogen Production at Methanol Inlet Flow Rate of $9.333 \times 10^{-5} \text{ mol/s}$

Temperature (K)	Methanol Conversion
450	37.3 %
475	80.8 %
500	99.8 %

Table 4: Effect of Temperature on Conversion at Methanol Inlet Flow Rate of $9.333 \times 10^{-5} \text{ mol/s}$

Temperature (K)	Methanol Conversion
450	30.7 %
500	82.19 %
550	99.9 %

Table 5: Effect of Temperature on Conversion at Methanol Inlet Flow Rate of $3.5 \times 10^{-4} \text{ mol/s}$

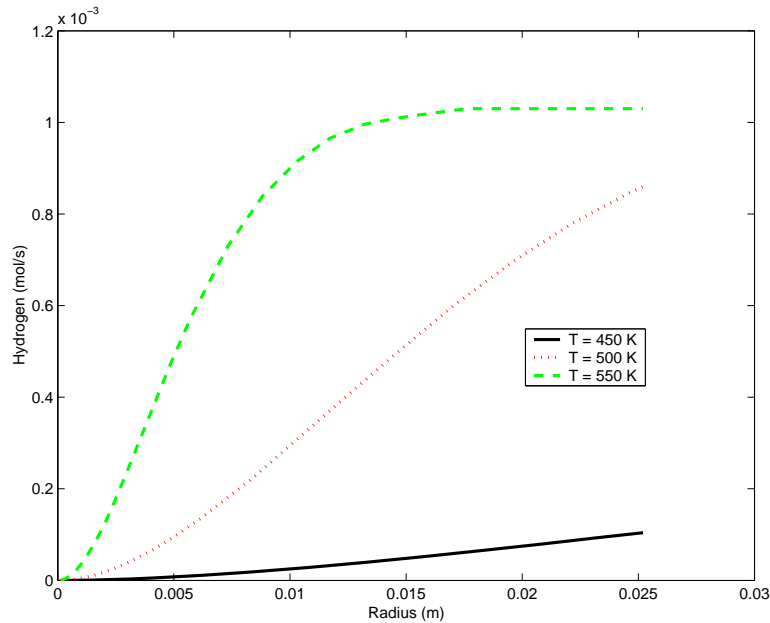


Figure 5: Effect of Temperature on Hydrogen Production at Methanol Inlet Flow Rate of $3.5 \times 10^{-4} \text{ mol/s}$

Conclusions

In this paper, a radial flow methanol reformer is designed for mobile power applications. It is shown that the radial flow geometry results in significant reduction in pressure drop as compared to a conventional tubular packed bed reactor. The effect of steam to methanol ratio and temperature are studied at two different methanol feeds for producing sufficient hydrogen to generate 24 W and 72 W of net power respectively. It is shown that a reactor volume of 20 ml is sufficient to generate the necessary hydrogen when a steam to methanol ratio of 1.5 and a pressure of 202 kPa are utilized. Furthermore, it is shown that for the lower power rating, an operating temperature of 500 K is sufficient while for the higher power rating, it is necessary to increase the operating temperature to 550 K.

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