

Heat Transfer Characteristics and Performance Prediction of Methane Reforming in a Fluidized Bed Reactor

Johan Ko¹ Taebeom Seo² Guiyoung Han³

¹Dept. of Mech. Eng., Graduate school, Inha Univ., Incheon, Korea,
johansko@gmail.com

²Dept. of Mechanical Engineering, Inha University, Incheon, Korea,
seotb@inha.ac.kr

³Dept. of Chemical Engineering, Sungkyunkwan University, Suwon, Korea,
@

ABSTRACT: Hydrogen production from thermocatalytic decomposition(TCD) of methane in a fluidized bed reactor(FBR) with carbon-based catalyst is known to free-CO₂ hydrogen production process. Methane decomposition over a carbon-based catalyst requires reaction temperature over 1000K. At this temperature range, heat transfer characteristics of the FBR is complicated and highly affects the methane conversion efficiency. In the view point of heat exchanger design, it is essential to devise a optimized heat exchanger system to maintain stable reaction temperature. The FBR installed in Yecheon city is 0.288m i.d. and 2.0m in height. LPG and produced hydrogen is used to heat the FBR. In this paper, heat transfer characteristics of the FBR is simulated and thermal efficiency enhancement methods are considered. Hydrogen production from TCD of methane in the FBR is compared with experimental data and computational fluid dynamics(CFD) prediction.

Keywords: heat transfer, fluidized bed reactor, methane reforming, computational fluid dynamics

1. INTRODUCTION

At present, H₂ is produced largely from fossil fuels (48% from natural gas, 30% from refinery/chemical off-gases, 18% from coal, the rest from electrolysis for small pure H₂).[1] Hydrogen from natural gas and from coal are the most widely applied hydrogen production technology. For example, natural gas steam reforming and coal gasification. Natural gas steam reforming and coal gasification are technological most matured and economically cheapest.[2] These methods has been researched for a long time and meets technical readiness, stable hydrogen supply and low production costs.

However, these methods emit large amount of CO₂. In order to reduce CO₂ emission from present hydrogen production from natural gas and coal, many technologies are researched and developed.

Decomposition of natural gas has been also researched as an environmentally attractive approach to production of hydrogen. The process produces hydrogen without CO₂ by-products, which eliminates the need for water gas shift and CO₂ removal stages.

Non-catalytic thermal decomposition of methane requires high temperatures (exceed 1200 - 1700 °C), therefore, much research were focused on Thermocatalytic

decomposition (TCD) of methane via metal catalyst (e.g., Ni, Fe, Co, etc.). These studies reported deactivation of catalyst, decomposition of carbon on the catalyst surface. During the post process, however, in order to regenerate the catalyst, produces large amount of CO₂.

To satisfy CO₂ free hydrogen production at lower reaction temperature, using carbon based catalysts is suggested and for various kind of carbon based catalysts (activated carbon, carbon black etc.) are researched.[3,4]

The general reaction temperature of thermocatalytic decomposition of methane using carbon based catalyst is in the range of 800 - 1000 °C.[3,4,5,6,] In the aspect of practical approach of H₂ production from above method, reaction temperature obtained from combustion. Without proper supplement apparatus design or selection, total system efficiency and operating cost will deviate.

In this paper, to support the reactor design and further system development, the heat transfer characteristics and performance of the fluidized bed reactor is simulated using commercial CFD code, Fluent 6.3.26.

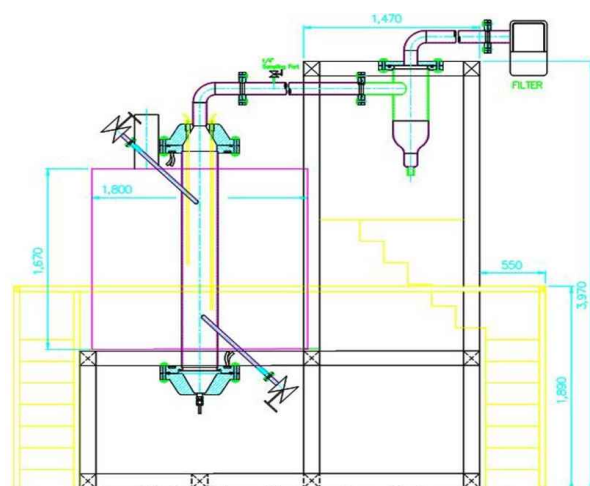


Figure 1: drawings of FBR and combustion gas heating chamber

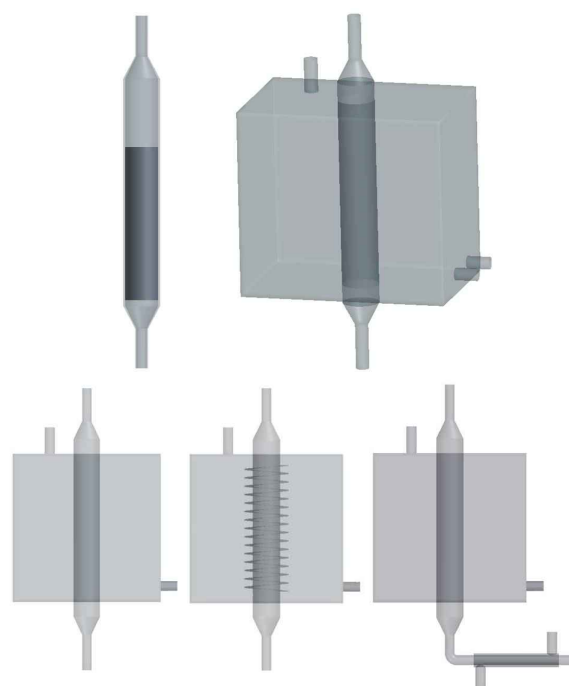


Figure 2: geometry of FBR and combustion gas heating chamber. original model, annular fin, heat recovery model (from left).

2. MODEL DESCRIPTION

2.1 Geometry

The fluidized bed reactor geometry for simulation is based on the reactor which is installed in the experimental station, Yochon city. The geometry is divided into two parts, outer chamber of the bed for combustion bed heating, and inside the bed where the decomposition reaction occurs. The scheme of experiments equipments is shown in fig.1 and fig.2.

The cylindrical body is 288mm inner diameter and 2000mm in height with diverging inlet and converging outlet. Inlet and outlet nozzle of the FBR is extended compare to real bed size.

The combustion gas heating chamber is 1670mm height and 1800mm in width. Two gas burners are attached on the left side of the chamber wall in a low, and an exhaust pipe is on the upper wall side the FBR exit.

2.2 Meshing Scheme

Gambit version 2.4 is used for the pre-processor of CFD solver, Fluent. Gambit 2.4 support both type of tetrahedral mesh and hexahedral mesh. With complicated equations like chemical reaction (e.g., combustion and thermal decomposition) and with mesh quality sensitive equations like radiative transfer equation(RTE), using hexahedral mesh is essential. Entire region of FBR and combustion gas heating chamber area is discretized with hexahedral meshes.

2.3 Boundary and initial conditions

Experimental data is applied to the boundary and initial conditions for simulation. In combustion gas heating simulation, heat of absorption in the FBR due to the endothermic reaction is set as a boundary condition obtained from experiment. And the inlet condition of two gas burners are also from experimental data.

Inlet conditions of the FBR is equal to the experiment values. The bed wall temperature and heat flux comes from the result of the combustion gas heating simulation. After calculating the combustion gas heating, the bed wall temperatures and heat fluxes are profiled. And this profiled data is used for the FBR boundary conditions.

3. SIMULATION

The Simulation geometry is divided into two part. The gas chamber zone where the heat is transferred from the combustion gas and the bed zone where decomposition reaction occurs. In the view point of the simulation, it is not easy to solve this conjugated phenomena at once. In this reason the simulation is performed in 2 step.

First, heat transfer from combustion gas to the bed is solved at the gas chamber zone. Then, the obtained data at the bed wall from the first simulation is used for the second simulation, the hydrogen production from

the thermocatalytic decomposition reaction inside the FBR.

The purpose of the first simulation is to find the efficiency of the heat transfer from the combustion gas to the inside of the FBR and from this, to suggest the appropriate heat transfer enhancing method.

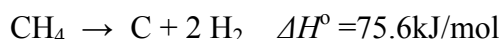
In the second simulation, using heat flux data from the 1st simulation result, the performance of the hydrogen production is estimated.

Basic settings for the two simulations are the same. Pressure-based segregated method is chosen for the solver algorithm which calculates velocity, continuity, energy and turbulence successively. 2nd order upwind method is set for interpolation discretization. and pressure-velocity coupling algorithm is Semi Implicit Method for Pressure Linked Equations-Consistent(SIMPLEC) method.

Standard k-ε model is chosen for turbulence and P-1 model is chosen for radiation. Both models are known for economic and robust convergence with reliable result in the heat transfer calculation.

Thermal decomposition of methane is consists of several reactions. From the kinetic modeling of hydrogen production by thermal decomposition of methane, 8 reaction steps are considered.[7]

In computational simulation, only overall reaction is considered:



The Arrhenius rate and constants are employed to estimate the hydrogen production rate at the highest conversion efficiency appears. Rate constants, activation energy, pre-exponential factors and other reaction constants used in the simulation refer to Lee et al[5] and Kim et al[6].

4. EXPERIMENTAL

The experiments are proceed at the experimental station located in Yecheon

city. The brief schemes and components of experimental apparatus are shown in fig.1. Shapes and the size are the same as that of the simulated geometry. A thermocouple is fixed inside the FBR. This thermocouple is connected to the controller which controls the gas burner flow to maintain the FBR temperature. LPG and a portion of produced hydrogen is consumed as a fuel of gas burner. Two pipes are attached to insert and exhaust the catalyst powder. FBR exit is connected to the cyclone separator to collect the powder catalyst.

Combustion chamber which covers the FBR is in height of 1670mm and 1800mm in width. Two gas burners are attached on the side wall of the combustion chamber. Standard capacity of a single burner is 50000 kcal/hr.

Temperature measured at the center of the FBR ($T_{b,center}$) from the thermocouple is maintained around 925°C. Catalyst used in this experiments is Raven900 and the amount is 20kg. Gas velocity is $2 U_{mf}$ which corresponds to the space velocity of 0.1691 l/g_{cat} hr.

5. RESULT AND DISCUSSION

5.1 Gas heating simulations

In the first simulation, which solves heat transfer between the gas burner to fluidized bed reactor wall, the FBR in Yecheon is calculated. To increase the heat transfer rate and thermal management, Finned wall and heat recovery system (HRS) is also considered.

The average temperature inside the FBR ($T_{b,avg}$) is calculated for various conditions. Fig 4. shows the average bed temperature changes as the bed inlet temperature rises. The average bed temperature shows inelastic behavior against the inlet temperature change. The gas burner capacity (heating value; HV) is constant with 102262.1kcal/hr.

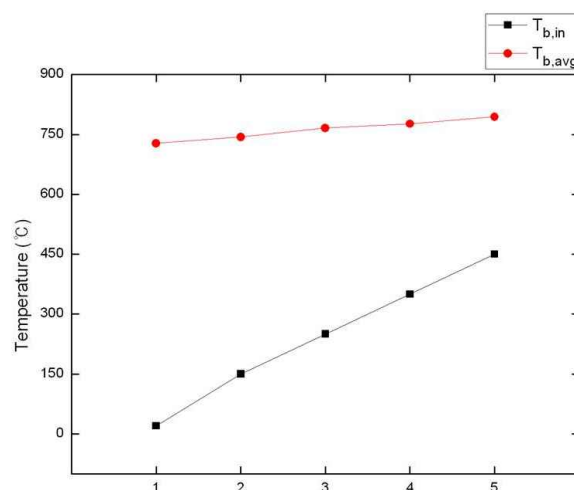


Figure 4: average bed temperatures of the original model.

Fig 5. shows $T_{b,avg}$ changes as the gas burner capacity changes. $T_{b,avg}$ changes from 795, 809 and 836 °C as the gas burner capacity changes 94926.12, 102262.1314 and 109424.16 kcal/hr.

In both cases, a standard gas burner capacity is 50000kcal/hr. Consider produced hydrogen is consumed as a fuel, this simulation values are under estimated compare to the actual heating value.

This underestimated simulation input data explains the simulation result of the $T_{b,avg}$ is less than 925°C. Also there is a possibility of overestimation of heat losses from the combustion chamber.

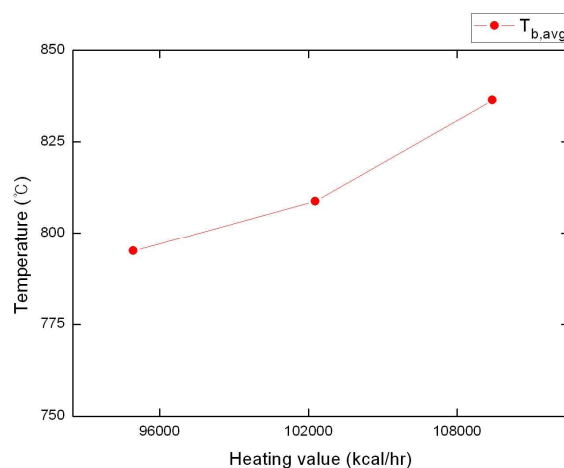


Figure 5: average bed temperatures of original model via various gas burner capacity.

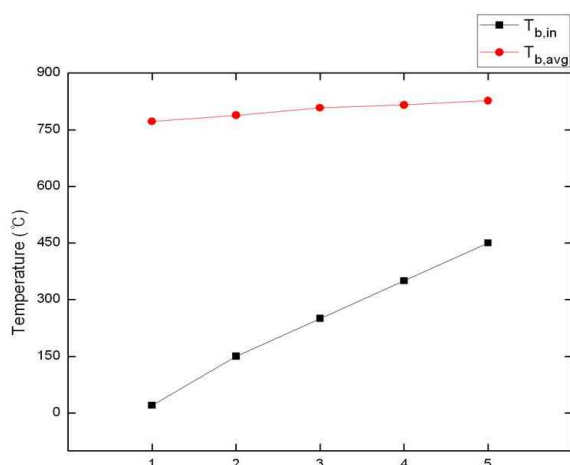


Figure 6: average bed temperatures of the annular finned model via various gas burner capacity.

To increase the average bed temperature, extended surface with annular fin is considered. In fig. 6 average bed temperatures of annular finned bed is shown. For the same inlet temperatures and the same gas burner capacity, the average bed temperature of finned bed is higher about 40°C.

The exhaust gas from the gas heating chamber is considerably high. Utilize this exhaust gas to pre-heat the reactant gas, introduce a cocentric tube. The reactant gas of ambient temperature flows through the center tube and the exhaust gas from the gas heating chamber exit flows the outer tube.

The average bed temperature is shown in fig. 7. The reactant gas temperature is 20°C at the heat recover cocentric tube inlet. The heating value changes from about 34000kcal/hr to 93000kcal/hr. The average bed temperatures are over 950°C.

Compare to the original model and annular finned model, the average bed temperatures with heat recover shows much higher for a less heating values. The average bed temperatures of the simulated models are shown in Table I for the same reactant gas inlet temperatures and gas burner capacity.

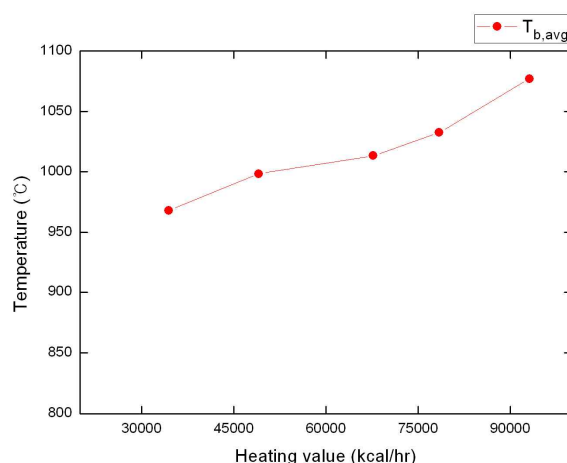


Figure 7: average bed temperatures of the heat recovery model via various gas burner capacity.

Table I: Comparison of average bed temperatures for considered models. ($T_{b,in} = 20^\circ\text{C}$, $HV = 102262.1\text{kcal/hr}$)

model	$T_{b,avg}$
original	728.084 °C
annular fin	778.986 °C
heat recovery	1077.052 °C

5.2 Reaction simulation

Heat fluxes calculated from the first gas heating simulation of the original model at the fluidized bed reactor wall is profiled (for, $T_{b,in}=450^\circ\text{C}$, $HV=102262.1\text{kcal/hr}$). This profile is applied to the boundary condition of the second simulation.

Instead of properties of catalyst Raven900, properties of the same carbon black catalyst BP-2000 is taken. Because the catalytic behaviors of BP-2000 is known.[6] Catalytic rate equation for BP-2000 is :

$$r_A = 2.55 \times 10^6 \exp(-143,000/RT) P_A \text{ mmol / min g}_{CB} \quad [6]$$

Among the reaction models available in the CFD program, Fluent6.3.26, Finite

Rate/Eddy Dissipation model is used to estimate the hydrogen production.

For a steady state simulation result, the methane conversion rate on a BP-2000 at the fixed bed condition ($T_{b,avg}=728.084^{\circ}\text{C}$) is 7.42%. The experimental result of methane conversion rate on a BP-2000 in the laboratory scale at the temperature range of 750°C is less than 3%.[6] The experimental result of the original model ($T_{b,avg}=950^{\circ}\text{C}$), the methane conversion rate on a Raven900 is 23.40%.

The estimation of methane conversion rate in the fixed bed at the simulated scale is much higher than that of laboratory scale.

5.3 Nomenclatures

$T_{b,avg}$	average bed temperature	$[^{\circ}\text{C}]$
$T_{b,in}$	bed inlet temperature	$[^{\circ}\text{C}]$
$T_{b,center}$	bed center temperature	$[^{\circ}\text{C}]$
r_A	methane decomposition rate	$[\text{mmol} / \text{min g}_{\text{CB}}]$
P_A	partial pressure of methane	$[\text{atm}]$

6. CONCLUSION

The simulation result of the average bed temperature is in the range near 750°C which is less about 200°C than the expected temperatures. This result comes from the underestimation of heating values and over estimation of the heat loss.

To increase the heat transfer from the combustion gas to the fluidized bed reactor annular fin is considered. With annular fin, the average bed temperature rise about 40°C .

To increase the thermal efficiency, the exhaust gas from the gas heating chamber is used to pre-heat the reactant gas. The simulation result of heat recover from the exhaust gas shows high average bed temperatures with less heating values. Which means that the overall system can be operates with less fuel consumption.

The methane conversion rate for the carbon black catalyst BP-2000 at the fixed bed condition is simulated. The methane conversion rate from the simulation result is higher than that of the laboratory scale. It is necessary to compare the simulation results for various reaction conditions to develop the accurate reaction model in the further researches.

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