Numerical Simulation and Calculation of Carbon Catalytic CH₄-CO₂ Reforming Reactor

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A pilot carbon catalytic CH₄-CO₂ reforming reactor was developed and simulation was carried out with computational fluid dynamics simulation software. The reforming reactor structure, working principle and chemical reaction processes of the reforming reactor were introduced. The temperature distribution, flow field and species distribution in the reforming reactor were simulated. The simulation results were as follows: entrance content of 44 % H₂ in mixed gas, export content of 56.6 % H₂ in synthetic gas, entrance content of 23.2 % CO, export content of 29.5 % CO, entrance content of 14.71 % CH₄, export content of 1.62 % CH₄, entrance content of 7.97 % CO₂, export content of 1.4 % CO₂ and H₂/CO of 1.9-2.2. Based on the predicted simulation results, an actual carbon catalytic CH₄-CO₂ reforming reactor was developed. The material balance, heat balance and flow resistance were calculated. The calculation results showed that the goal was successfully achieved.

Key Words: Carbon catalyst, Numerical simulation, Computational fluid dynamics, Material balance, Heat balance, Flow resistance.
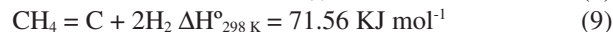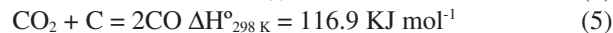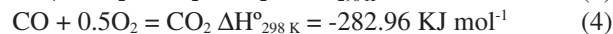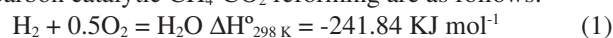
INTRODUCTION

CH₄-CO₂ reforming to synthesis gas (syngas) (H₂ + CO), as well as the production of methanol, F-T synthesis, ammonia and other chemical products, has been extensively investigated. Conventional catalytic reforming methods use Ni- or noble-metal-based catalysts[1,2]. Noble metal-based catalysts have high activity and selectivity, but they are expensive. Nickel-based catalysts have higher activity and selectivity, but are easily deactivated by carbon deposition. Therefore, the development of new, low-cost catalysts resistant to carbon deposition is imperative. Our group has previously proposed the double-gas multi-generation technology (973 items) of a pyrocarbon system under coke oven gas and gasification gas. Using coke oven gas (27 % CH₄) and gasification gas (20 % CO₂), CH₄-CO₂ is reformed to syngas at a high temperature with carbon catalyst further integrating the synthetic alcohol-ether fuels[3]. Based on this small-scale experimental study[4], a pilot carbon catalytic CH₄-CO₂ reforming reactor was developed and a numerical simulation was carried out in the current work. A simulation study for coke oven gas and mixed-intake gasification gas was performed. Partial oxidation of oxygen was achieved with a carbon catalyst for temperature field, flow field and component distribution in the reforming reactor to verify the performance of the designed pilot-reforming reactor. Based on simulation result predictions, an actual carbon catalytic CH₄-CO₂ reforming reactor was developed.

The material balance, heat balance and flow resistance were calculated. Given the irregular size distribution of the carbon catalyst, the fluid resistance in the reforming reactor was studied.

EXPERIMENTAL

Reaction process analysis: The main chemical reactions in carbon catalytic CH₄-CO₂ reforming are as follows:

$$H_2 + 0.5O_2 = H_2O \quad \Delta H^\circ_{298\,K} = -241.84\ KJ\ mol^{-1} \quad (1)$$
$$CH_4 + 0.5O_2 = CO + 2H_2 \quad \Delta H^\circ_{298\,K} = -27.32\ KJ\ mol^{-1} \quad (2)$$
$$CH_4 + 2O_2 = CO_2 + 2H_2O \quad \Delta H^\circ_{298\,K} = -802.60\ KJ\ mol^{-1} \quad (3)$$
$$CO + 0.5O_2 = CO_2 \quad \Delta H^\circ_{298\,K} = -282.96\ KJ\ mol^{-1} \quad (4)$$
$$CO_2 + C = 2CO \quad \Delta H^\circ_{298\,K} = 116.9\ KJ\ mol^{-1} \quad (5)$$
$$CH_4 + H_2O = CO + 3H_2 \quad \Delta H^\circ_{298\,K} = 206.15\ KJ\ mol^{-1} \quad (6)$$
$$CH_4 + CO_2 = 2CO + 2H_2 \quad \Delta H^\circ_{298\,K} = 247.27\ KJ\ mol^{-1} \quad (7)$$
$$CO_2 + H_2 = CO + H_2O \quad \Delta H^\circ_{298\,K} = 115.98\ KJ\ mol^{-1} \quad (8)$$
$$CH_4 = C + 2H_2 \quad \Delta H^\circ_{298\,K} = 71.56\ KJ\ mol^{-1} \quad (9)$$

Reactions (1-7) essentially reflected gasification. Reactions (1-4) represented the gas combustion reaction, which were strong exothermic processes comprising the primary set of reactions. These reactions occurred rapidly, with the complete reaction finishing within several dozens of milliseconds. Reactions (5-7) represented the control procedure of the entire process. These endothermic reactions were collectively called the secondary set of reactions. During catalyzed partial oxidation, the conversion temperature was maintained within

1200-1300 K. The oxidation process was finished within 1-2 s[5]. The appropriate H/D ratio had to be provided for the hearth to ensure the completion of the secondary reaction.

Numerical simulation of reactor

Control equation of numerical simulation: For steady, incompressible flow, the following forms of mass, momentum, energy and species conservation equations considering the turbulence effects are well-known. However, several considerations were made for the particular case being studied. Momentum equations for porous media in FLUENT, treatment of energy equations in porous media and P1 radiation model are reviewed in the literature[6]

Turbulence model: The standard k-ε model equitation is the most popular turbulence model applied in hydromechanics simulation. For uncompressed fluids, the standard k-ε model equitation is as follows[7] (Eqns. 10-12):

$$\frac{\partial(\rho k)}{\partial t} + \frac{\partial(\rho k u_i)}{\partial x_i} = \frac{\partial}{\partial x_j}\left[\left(\mu + \frac{\mu_t}{\sigma_k}\right)\frac{\partial k}{\partial x_j}\right] + G_k - \rho\varepsilon \quad (10)$$

$$\frac{\partial(\rho\varepsilon)}{\partial t} + \frac{\partial(\rho\varepsilon u_i)}{\partial x_i} = \frac{\partial}{\partial x_j}\left[\left(\mu + \frac{\mu_t}{\sigma_\varepsilon}\right)\frac{\partial\varepsilon}{\partial x_j}\right]$$
$$+ \frac{C_{1\varepsilon}\varepsilon}{k}G_k - C_{2\varepsilon}\rho\frac{\varepsilon^2}{k} \quad (11)$$

$$\mu_t = \rho C_\mu \frac{k^2}{\varepsilon} \quad (12)$$

Model constants

$$C_{1\varepsilon} = 1.44, C_{2\varepsilon} = 1.92, C_\mu = 0.09, \sigma_k = 1.0, \sigma_\varepsilon = 1.3$$

Component conservation equation:

$$\frac{\partial(\rho C_s)}{\partial t} + \text{div}(\rho\vec{u}C_S) = \text{div}(D_S\text{grad}(\rho C_s)) + S_S \quad (13)$$

where C_S is the volume concentration of the components, ρC_s is the mass concentration of the components, D_S is the component diffusion coefficient and S_S is the system per unit time per unit volume generated by a chemical reaction of the component quality (*i.e.*, productivity) (eqn. 13).

Mass transfer equation eqn. 14:

$$N_A = k_G a_m (C_{AG} - C_{AS})$$
$$= \frac{C_{AG} - C_{AS}}{1/k_{Ga}}\left(\frac{m}{s}\right)\left(\frac{m^2}{kg}\right)\left(\frac{kmol}{m^3}\right) \quad (14)$$

where K_G is the mass transfer coefficient and a_m is the ratio surface area of the particles (outer surface).

Heat transfer equation:

$$q = h_S a_m (T_S - T_G) = \frac{T_S - T_G}{1/(h_S a_m)} \quad (15)$$

where h_S is the heat transfer coefficient, a_m is the particle surface area, G represents a particle body and S represents the particle surface. The calculation of q is based on gas body as a benchmark, when q > 0, which means that the vapor is mainly endothermic[8] eqn. 15.

Pilot reforming reactor geometry: Fig. 1 shows a pilot carbon catalytic CH₄-CO₂ reforming process diagram for simulation study. GAMBIT geometry model established the non-structural grid as shown in Fig. 2.
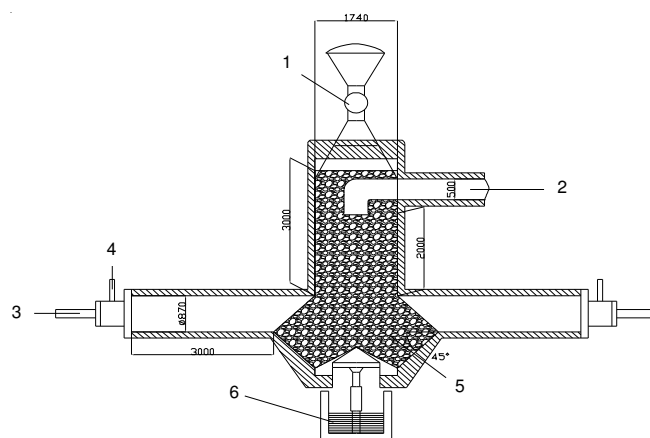

Fig. 1. Structure of the pilot-test reforming reactor. 1. catalyst feeding mechanism, 2. synthesis gas outlet, 3. mixed gas inlet, 4. O₂ inlet, 5. catalyst layer, 6. water seal device
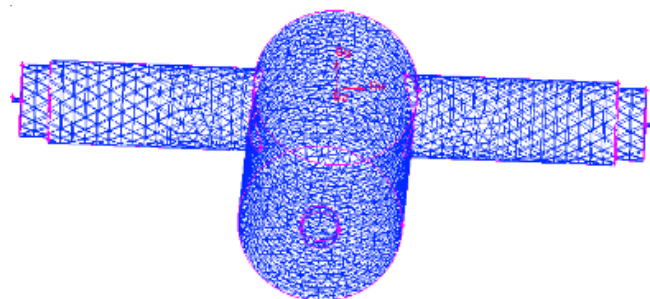

Fig. 2. Simulation geometry model and the unstructured grid

Working conditions of simulation: The wall temperature: 500 K; O₂ inlet velocity: 25.9 m/s; mixture gas inlet velocity: 0.2 m/s; M (gasification gas/coke oven gas) = 1.

The original composition of gasification gas and coke oven gas and the simulation results (Table-1).

TABLE-1
ORIGINAL COMPOSITION OF GASIFICATION GAS AND COKE OVEN GAS AND THE SIMULATION RESULTS

	CO	H₂	CO₂	CH₄	H₂O	O₂	N₂
Gasification gas (%)	39.9	31.0	11.94	1.42	15.74		
Coke oven gas (%)	6.5	57.0	4.0	28.0		0.5	4.0
Mixed gas inlet (%)	23.2	44.0	7.97	14.71	7.87	0.25	2.0
Export simulation results (%)	29.5	56.6	1.4	1.62	1.38	0.1	2.31

RESULTS AND DISCUSSION

Fig. 3 shows the coke oven gas and gasification gas mixture with partially oxidized oxygen combustion reaction to form two symmetrical jet flames in the nozzle entrance. A temperature up to 3000 K, CH₄, H₂ combustion generating CO₂, H₂O in the vertical section of the carbon catalyst layer and unburned CH₄ reforming reaction were needed to produce syngas (H₂ + CO). The catalyst layer had a temperature of 1500 K, which was suitable for reforming. Fig. 4 shows the
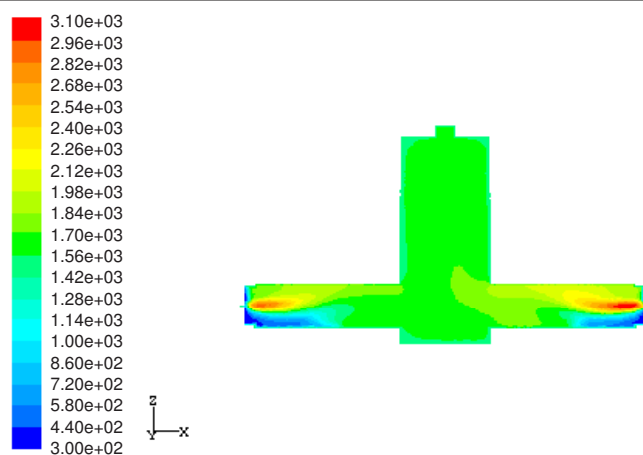
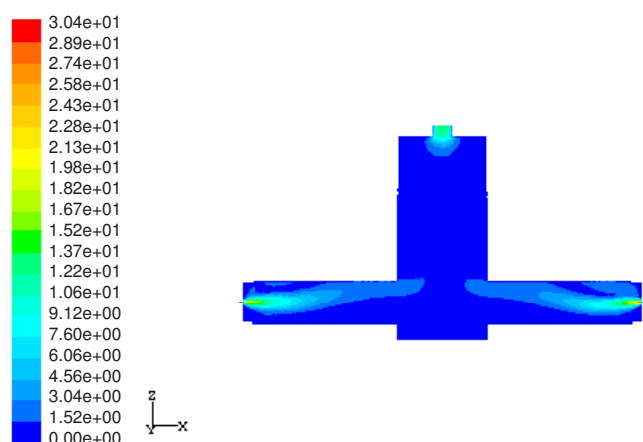Fig. 3. Temperature distribution in pilot reforming reactor

Fig. 4. Flow field in pilot reforming reactor

Fig. 5. Distribution of major components in reforming reactor (a-CO_2, b-CO, c-H_2, d-CH_4)

Fig. 6. Schematic of the actual reforming reactor

distribution of two shares of symmetric turbulent jets. The flow rate was small in the catalyst layer. Thus, catalyst reforming gas stays for a long time and is suitable for reforming.

Fig. 5 shows the distribution nephogram of main components within a pilot reforming reactor. The different colours represent different component contents. H_2 content from 44 % increased to 56.6 %. CO content from 23.2 % increased to 29.5 %. CO_2 content from 7.97 % decreased to 1.4 %. CH_4 content from 14.71 % dropped to 1.62 %. Exporting to H_2/CO ratio was around 2.0. Through carbon catalytic reforming, CH_4 and CO_2 effectively converted into syngas (H_2 + CO). Syngas is a high-quality synthetic methanol material.

Calculation of actual reforming reactor: Based on the simulation schematic of the pilot carbon catalytic CH_4-CO_2 reforming reactor, an actual reforming reactor (Fig. 6) was developed by Taiyuan University of Technology. According to the properties of the carbon catalyst, the compositions of raw gas and syngas, size parameters of reforming reactor, other basic research data of carbon catalytic CH_4-CO_2 reforming, the material balance equation, the heat balance equation and the flow resistance model of the carbon catalyst bed layer were established[9]. Furthermore, the material balance, heat balance and flow resistance in the system were calculated.

Material balance of the reforming reactor system: The raw gas was drained out from the heating furnace into the reforming reactor and oxygen gas was streamed through the oxygen buffer tank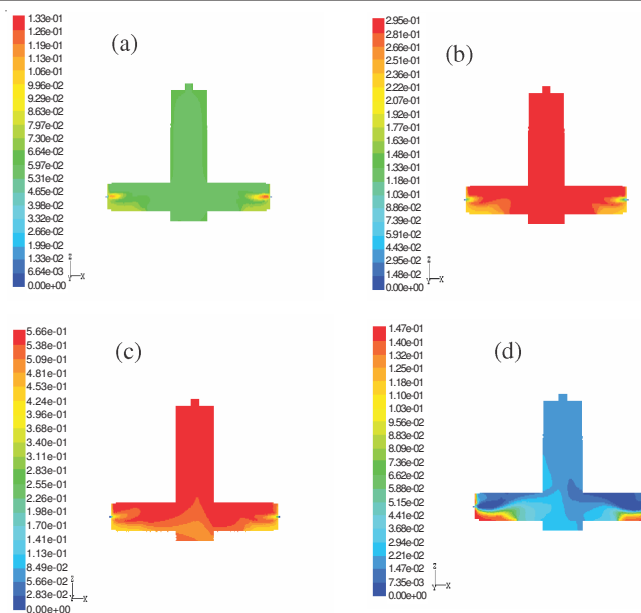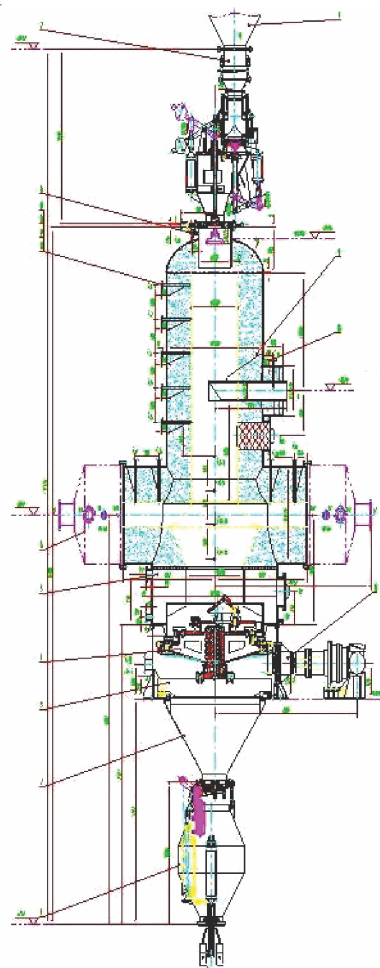 and oxygen heater, where the temperature further increased to 450 ℃ into the reforming conversion reactor. Carbon catalyst was loaded from the top of the reforming conversion reactor, wherein the gas underwent heat exchange through some chemical reactions. After the reaction,

the solid residue was discharged from the furnace bottom. Based on the reforming reactor technique, the material balance and heat balance equations were established. The relevant parameters are shown in Table-2.

TABLE-2
MAIN PARAMETERS OF THE REACTOR

Parameter	Value
Carbon catalyst consumption (kg h^{-1})	50
Carbon catalyst bed layer height h (m)	1.5
Cross-sectional area of the reactor interior S (m^2)	0.785
Entrance raw material gas flow (Nm^3 h^{-1})	588
Entrance oxygen flow (Nm^3 h^{-1})	132
Synthesis gas export flow (Nm^3 h^{-1})	920
Carbon catalyst particle size (mm)	5-35
Carbon catalyst residence time t (h)	13.32
Inner diameter of the reactor d (m)	1.0
Import raw gas temperature (ºC)	600
Temperature of the inlet oxygen (ºC)	450
Temperature of the outlet synthesis gas (ºC)	900
Ash discharging temperature (ºC)	1050

In addition, the simulation prediction determined the operating conditions at different gas volume fractions X_i (H_2, CO, CO_2, CH_4, N_2, H_2O and O_2) into the furnace to be 46.52, 7.73, 5.04, 16.03, 3.65, 3.43 and 17.54 %, respectively; those for the outlet volume fractions Y_i (H_2, CO, CO_2, CH_4, N_2, H_2O and O_2) were 56.19, 31.34, 1.71, 0.83, 2.61, 6.76 and 1.02 %, respectively. The content of the main components (H_2 and CO) and the aforementioned simulation results had percentage errors of 0.7 % H_2 and 5.9 % CO because of the raw gas fluctuation and slightly different simulation conditions. The trace error can guarantee the accuracy of the subsequent calculations.

Material balance model: A small amount of dust was found in the import and export gas of the reforming conversion reactor. However, the quantity was negligible. Using the reforming conversion reactor system as the research object and based on the balance of the material income and material expenses, the material equilibrium model was established (eqn. 16):

$$G_1 + m_1 = G_2 + G_3 + m_2 \qquad (16)$$

where G_1 is the loaded carbon catalyst quality (kg/h), G_2 is the clinker quality of the reforming conversion reactor (kg/h), G_3 is the carbon catalyst volatile quality (kg/h), m_1 is the gas quality into the reforming conversion reactor (kg/h) and m_2 is the gas quality of the outlet in the reforming conversion reactor (kg/h). Main parameters of the reactor (Table-2).

Calculation of material balance

Calculation of material income: The loaded carbon catalyst mass is expressed as eqn. 17

$$G_1 = \rho_v S \frac{h}{t} = 0.65 \times 0.785 \times 1.5 \times \frac{13.32}{57.47} \text{ kg/h} \qquad (17)$$

where ρ_v is the carbon catalyst bulk density (t/m^3) and t is carbon catalyst residence time (h).

The gas quality into the reforming conversion reactor is expressed as eqn. 18

$$m_1 = \sum \frac{M_i X_i V_1}{22.4} = 486.30 \text{ kg/h} \qquad (18)$$

where M_i is the molar mass of the component i (g/mol), X_i is the volume fraction of the component i and V_1 is the gas flow rate into the reformer conversion reactor (Nm^3/h).

Calculation of material expenditure: The ash quality of the reforming conversion reactor is expressed as eqn. 19

$$G_2 = G_1 \times \frac{10 \%}{t} = 57.47 \times \frac{10 \%}{13.32} = 5.75 \text{ kg/h} \qquad (19)$$

The gas quality of the outlet of the reforming conversion reactor is expressed as eqn. 20

$$m_2 = \sum \frac{M_i Y_i V_2}{22.4} = 536.32 \text{ kg/h} \qquad (20)$$

The carbon catalyst volatilization mass is eqn. 21

$$G_3 = G_1 \times \frac{3 \%}{t} = 57.47 \times \frac{3 \%}{13.32} = 1.72 \text{ kg/h} \qquad (21)$$

Table-3 lists the calculated results of material balance in the reforming conversion reactor. The data indicated that the import and export materials of the reactor were balanced.

TABLE-3
RESULTS OF MATERIAL BALANCE OF THE REACTOR

Material income	Quality of carbon catalyst loaded to the reactor G_1	57.47
(kg h^{-1})	Gas quality into the reformer reactor m_1	486.30
Total		543.77
Material expenditure	Gas quality of the outlet of the reforming reactor m_2	536.32
(kg h^{-1})	Ash quality of the reforming conversion reactor G_2	5.75
	Carbon catalyst volatilization mass G_3	1.72
Total		543.79

Element balance: The gas elements involved are mainly C, H, O and N. The quality of various elements was determined according to the gas flow rate, composition and relevant data of carbon catalyst in the import and export. The results are as follows:

C income: (7.73 + 5.04 + 16.03) × 720 × 12/100 × 22.4 + 57.47 = 168.56 kg/h

C expenditure: (31.34 + 1.71 + 0.83) × 920 × 12/100 × 22.4 = 166.98 kg/h.

H income: (46.52 × 2 + 16.03 × 4 + 3.43 × 2) × 720 × 1/100 × 22.4 = 52.72 kg/h.

H expenditure: (56.19 × 2 + 0.83 × 4 + 6.76 × 2) × 920 × 1/100 × 22.4 = 53.07 kg/h.

O income: 7.73 + 5.04 × 2 + 3.43 + 17.54 × 2) × 720 × 16/100 × 22.4 = 289.65 kg/h.

O expenditure: 31.34 + 1.71 × 2 + 6.76 + 1.02 × 2) × 920 × 16/100 × 22.4 = 286.25 kg/h.

N income: 3.65 × 2 × 720 × 14/100 × 22.4 = 32.85 kg/h

N expenditure: 2.61 × 2 × 920 × 14/100 × 22.4 = 30.02 kg/h.

The aforementioned results were allowed within the range of material balance. C, H, O and N appeared in response to the balance of import and export.

Heat balance equation and its calculation in the reforming conversion reactor

Heat balance equation in reforming system: Using the reforming conversion reactor as the system, the heat balance

was calculated. The heat income equals the heat expenditure as expressed in the following formula eqn. 22:

$$Q_1 + Q_2 + Q_3 = Q_4 + Q_5 + Q_6 + Q_7 \qquad (22)$$

where Q_1 is the heat of gas into the reformer conversion reactor (kJ/h), Q_2 is the heat of carbon catalyst into reformer reactor (kJ/h), Q_3 is the heat released by the chemical reaction (kJ/h), Q_4 is the heat of gas outlet of the reformer conversion reactor (kJ/h), Q_5 is the heat of ash in the reforming conversion reactor (kJ/h), Q_6 is the heat absorbed by the chemical reactions (kJ/h) and Q_7 is the system heat loss (kJ/h).

Calculation of heat balance in reforming system

The heat of gas into the reformer conversion reactor is expressed as eqn. 23

$$Q_1 = \Sigma V_i C_i t_i = 6.323 \times 10^5 \text{ kJ/h} \qquad (23)$$

The heat of carbon catalyst into the reformer reactor is expressed as eqn. 24

$$Q_2 = G_1 C_1 t_1 = 57.47 \times 20 \times 0.738 = 8.483 \times 10^2 \text{ kJ/h} \quad (24)$$

The heat of gas outlet of the reformer conversion reactor is expressed as eqn. 25

$$Q_4 = \Sigma V_i C_i t = 1.163 \times 10^6 \text{ kJ/h} \qquad (25)$$

The heat of ash in the reforming conversion reactor is expressed as eqn. 26

$$Q_5 = G_2 C_2 t_2 = 5.75 \times 0.902 \times 1050 = 5.443 \times 10^3 \text{ kJ/h} \quad (26)$$

The system heat loss is expressed as eqn. 27

$$Q_7 = (K_\pi + K_k)(t_n - t_B)F \qquad (27)$$

where, K_π and K_k are the convective and radioactive heat transfer coefficients [W/(m² K)], respectively. They are expressed as eqns. 28-30:

$$K_\pi = \frac{K\left[\left(\dfrac{T_n}{100}\right)^4 - \left(\dfrac{T_B}{100}\right)^4\right]}{T_n - T_B} \qquad (28)$$

$$K_K = 1.43 \times \sqrt[3]{\Delta t} \qquad (29)$$

$$\Delta t = t_n - t_B \qquad (30)$$

where K is the emissivity coefficient of the material in the outer surface of the reforming conversion reactor [W/(m² K)] taken as 5.5, $t_n(T_n)$ is the surface temperature of the reforming conversion reactor [°C (K)], $t_B(T_B)$ is the average air temperature [°C (K)] and F is the outer surface area of the reforming conversion reactor (m²) taken as 21.58.

Calculations showed that $Q_7 = 2.953 \times 10^5$ kJ/h at $t_n = 150$ °C, $t_B = 20$ °C.

Endothermic and exothermic reactions of the system: A complex chemical reaction and its extent in the reforming reactor are difficult to determine in the course of the reaction, particularly when only expansion works are performed in the process. Simultaneously, under constant pressure, the heat of reaction Q is ΔH. According to the first law of thermodynamics, the heat of reaction is decided by the complete state and has nothing to do with the process. In this study, based on the substance of enthalpy $\Delta_f H_m^\Theta$, the endothermic and exothermic reactions were calculated in the system. Various heats of reaction are given in Table-4.

TABLE-4 LIST OF HEATS OF REACTION				
	CO	CO_2	CH_4	H_2O
Feeding (Nm³ h⁻¹)	55.67	36.29	115.42	24.70
Out material (Nm³ h⁻¹)	288.33	15.73	7.64	62.19
Material difference (mol h⁻¹)	10 387.14	-917.68	-4811.61	1673.93
$\Delta_f H_m^\Theta$ (kJ mol⁻¹)	-110.5	-393.5	-74.8	-241.8
Q × 10⁵ (kJ h⁻¹)	-11.48	3.611	3.599	-4.048

The heat released by the chemical reaction is expressed as eqn. 31

$$Q_3 = -(Q_{CO} + Q_{H_2O}) = 1.553 \times 10^6 \text{ kJ/h} \qquad (31)$$

The heat absorbed by the chemical reactions is expressed as eqn. 32

$$Q_6 = Q_{CO_2} + Q_{CH_4} = 7.210 \times 10^5 \text{ kJ/h} \qquad (32)$$

The heat balance summary in the reforming reactor is shown in Table-5. The data indicated that the expenditure and income of heat were balanced in the reforming conversion process. Results of heat balance of the reactor are given Table-5.

TABLE-5 RESULTS OF HEAT BALANCE OF THE REACTOR	
	Heat income (kJ h⁻¹)
Heat of gas into the reformer reactor Q_1	6.323×10^5
Heat of carbon catalyst into the reformer reactor Q_2	8.483×10^2
Heat released by the chemical reaction Q_3	1.553×10^6
Total	2.186×10^6
	Heat expenditure (kJ h⁻¹)
Heat of gas outlet of the reformer conversion reactor Q_4	1.163×10^6
Heat of ash in the reforming conversion reactor Q_5	5.443×10^3
Heat absorbed by the chemical reaction Q_6	7.210×10^5
System heat loss Q_7	2.953×10^5
Total	2.185×10^6

Calculation and study on fluid resistance: Multi-component gas and carbon catalysts were acceded into the reforming conversion reactor. The factors that required consideration were the mechanical motion of the gas, the exchange of physical heat between gas and carbon catalyst, the chemical reaction between gas and gas, as well as the chemical reaction between gas and carbon catalyst when the gas flows through the reforming conversion reactor. For the calculation of fluid resistance, the following hypotheses were made: (1) the fluid flow was one-dimensional and (2) the fluid environment was under constant pressure.

Calculation of fluid resistance in the reforming reactor

Resistance of carbon catalyst bed layer: When the material layer thickness was 1 m, the formula of the resistance calculation was as follows (eqn. 33):

$$\Delta P = \frac{5.9 v^{0.45} \omega^{1.55} S^{1.45} \rho'}{V_{CB}^3} \qquad (33)$$

where v is the gas viscosity (m²/s), ω is the free-section average velocity (m/s), ρ' is the gas density at the average temperature

(kg/m^3), S is the free-section specific surface area (m^2/m^3) and V$_{CB}$ is the free volume (m^3/m^3).

V$_{CB}$, S and ρ' are expressed as follows:

$$V_{CB} = 0.0005(7.9a_1 + 6.6a_2 + 6.4a_3) \tag{34}$$

$$S = 0.05(24.5a_1 + 38.5a_2 + 19a_3) \tag{35}$$

$$\rho' = \sum \frac{y_i \rho_i 273}{T_{CP}} \tag{36}$$

$$\mu_m = \frac{\Sigma y_i \mu_i M_i^{1/2}}{\Sigma y_i M_i^{1/2}} \tag{37}$$

$$\nu = \frac{\mu_m}{\rho'} \tag{38}$$

$$\omega \frac{V_r(t_{CP} + 273)}{273S \times 3600} \tag{39}$$

In the formula, a$_1$-a$_3$ are the sieve class compositions, respectively corresponding to 35-25, 25-15 and 15-5 mm. Their values were 25.70, 37.46 and 36.85, respectively. ρ$_i$ is a gas component density in the standard condition (kg/m^3), T$_{CP}$ is the average gas temperature in the furnace (K), μ$_i$ is the viscosity of a component (Pa s), V$_r$ is the circulating gas volume (m^3/h). The total resistance of the carbon catalyst bed layer can be calculated using the following equation (eqn. 40:

$$H = \Delta Ph \tag{40}$$

in the formula, h is the carbon catalyst layer height (m).

Resistance in front of gas into the carbon catalyst bed layer: Frictional resistance of intake gas (eqn. 41)

$$\Delta P_1 = \lambda \frac{L}{d} \frac{\rho_1 V_0^2}{2} \frac{T_1}{T_0} \tag{41}$$

At Re > 2300, $\lambda = \frac{0.175}{Re^{0.12}}$; at Re < 2300, $\lambda = \frac{64}{Re}$; where $Re = \frac{\rho_1 V_0 d}{\mu_1}$.

The local resistance of 90° turns and expands in the intake gas (eqn. 42)

$$\Delta P_2 = k \frac{\rho_1 V_0^2}{2} \frac{T_1}{273} \tag{42}$$

where k is the drag coefficient taken as 1.5.

Resistance of gas in the outlet of carbon catalyst bed layer: Local resistance of outlet gas in 90° turns and reduced (eqn. 43):

$$\Delta P_1 = k \frac{\rho_2 V_2^2}{2} \frac{T_2}{T_1} \tag{43}$$

Frictional resistance of outlet gas (eqn. 44):

$$\Delta P_2 = \lambda \frac{L'}{d} \frac{\rho_2 V_2^2}{2} \frac{T_2}{T_1} \tag{44}$$

Calculated data of fluid resistance in the reactor (Table-6).

Study of fluid resistance within the reforming conversion reactor: The calculation results above indicated that the resistance of the carbon catalyst bed layer was a main component of the fluid flow resistance in the reforming conversion reactor, comprising 90.6 % of the total resistance. The factor mainly affecting the carbon catalyst bed layer resistance was the sieve class composition. The most effective way to reduce the fluid resistance was to change the added carbon catalyst particle size distribution. Thus, the effects of different sieve class compositions on the carbon catalyst bed layer resistance at unit thickness were next examined.

Effects of sieve class composition on carbon catalyst bed layer resistance: a$_1$, a$_2$ and a$_3$ correspond to 35-25, 25-15 and 15-5 mm sieve class compositions and a$_1$ + a$_2$ + a$_3$ = 100 based on the a$_1$ and a$_2$ – ΔP relationship shown in Fig. 7.

Fig. 7 shows that when the added carbon catalyst particle sizes were 25-35 and 15-25 mm, the resistance in unit thickness of the carbon catalyst layer was smaller. On the other hand, when the carbon catalyst particle size was 25-35 mm, the degree of influence was the largest. Thus, the carbon catalyst bed layer resistance was most effectively reduced at 15-35 mm (sieve class composition).

Thus, the most appropriate sieve class composition of the carbon catalyst bed layer was a$_1$ = 80 %, a$_2$ = 15 % and a$_3$ = 5 %. Consequently, ΔP = 313.5 Pa/m.

Fluid flow resistance of the different locations within reforming conversion reactor: To study the flow resistance at different positions, six points representing specific locations were selected, as shown in Fig. 8. The fluid flow resistance is shown in Fig. 9.

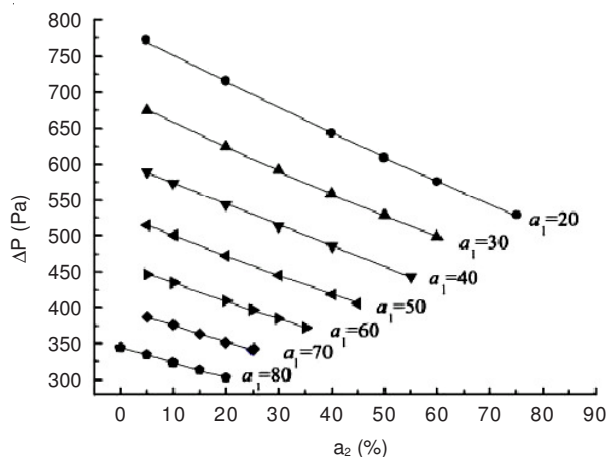| TABLE-6 CALCULATED DATA OF FLUID RESISTANCE IN THE REACTOR			
Parameter	Value	Parameter	Value
Gas density at the average temperature ρ' (kg m^{-3})	0.153	Gas average temperature in furnace t$_{CP}$ (°C)	750
Amount of recycled gas V$_r$ (m^3 h^{-1})	800	Kinematic viscosity v (m^2 s^{-1})	1.81×10^{-4}
Free cross section average velocity ω (m s^{-1})	1.06	Free volume V$_{CB}$ (m^3 m^{-3})	0.343
Specific surface area of the free cross-section S (m^2 m^{-3})	138.6	Pipe length of intake gas (L/m)	1.2
Density of intake gas under standard conditions (kg m^{-3})	0.632	Tube diameter of intake gas d (m)	0.2
Flow velocity of intake gas under standard conditions V$_0$ (m s^{-1})	6.28	Friction coefficient λ	0.0512
Tube diameter in the outlet gas d' (m)	0.273	Tube length in outlet gas L' (m)	1.49
Density of outlet gas under standard conditions (kg m^{-3})	0.518	Flow velocity of outlet gas under standard conditions V$_2$ (m s^{-1})	5.08
Carbon catalyst bed layer resistance of the unit thickness (Pa)	647.5	Friction coefficient λ'	0.0502
Total resistance H (Pa)	971.3	ΔP$_1$ (Pa)	12.24
ΔP$_2$ (Pa)	59.82	ΔP$_2$' (Pa)	7.56
ΔP$_1$' (Pa)	41.28	–	–

Fig. 7. Relationship curve of fluid resistance with the granulometric composition
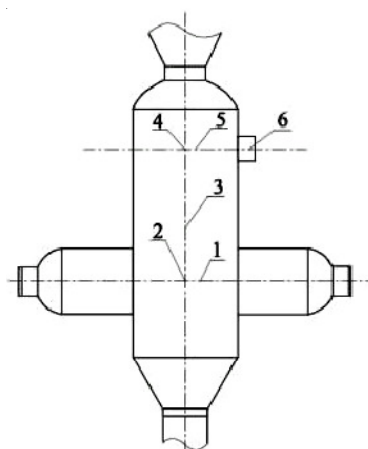

Fig. 8. Simplified diagram of the reforming reactor. 1-End of the gas inlet pipe, 2-Bottom of the carbon catalyst bed layer, 3-Carbon catalyst bed layer at 1 m, 4-Top of the carbon catalyst bed layer, 5-Beginning of the outlet pipe, 6-End of the outlet pipe
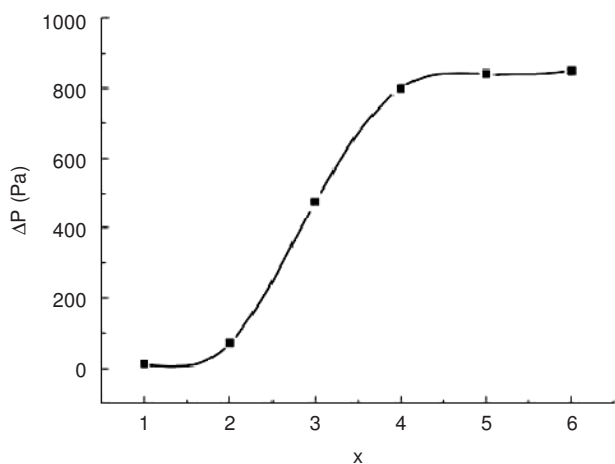

Fig. 9. Fluid resistance in the reforming reactor

Fig. 9 shows that a longer gas flow distance resulted in greater resistance of the fluid within the system. Furthermore, the resistance in the inlet and outlet pipes was minimal. The high resistance in the carbon catalyst bed layer increased with increased height of the catalyst bed.

Conclusion

The designed pilot carbon catalytic CH_4-CO_2 reforming reactor simulation results showed that the reactor temperature of the catalyst layer was 1500 K. This temperature was suitable for the reforming reaction. The catalyst layer velocity was sufficiently low to ensure adequate reforming reaction residence time. Component simulation results suggested that CH_4 and CO_2 were good materials for CO and H_2 gas synthesis. A description of the designed pilot reforming reactor indicated that its performance met pertinent requirements.

Based on the parameters of the reforming conversion reactor and relevant data of gas material, calculations showed that material balance was achieved.

A major factor influencing the carbon catalyst bed layer resistance was the sieve class composition. The carbon catalyst bed layer resistance in unit thickness decreased with increased carbon catalyst particle size. Thus, the carbon catalyst bed layer resistance was most effectively reduced when the carbon block size ranged from 25-35 mm. The most appropriate sieve class composition of the carbon catalyst bed layer was $a_1 = 80$ %, $a_2 = 15$ % and $a_3 = 5$ %, which resulted in $\Delta P = 313.5$ Pa/m.

In the reforming conversion reactor, the gas fluid resistance in the inlet and outlet pipes was low. The carbon catalyst bed layer resistance was the main component of reactor fluid resistance. Fluid resistance increased with increased height of the catalyst bed.

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