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Design of Microreactor Flow Channel for Fischer Tropsch Synthesis Using Computational Fluid Dynamic

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Abstract

In this work, the numerical model was used to investigate the effect of microchannel geometries including channel aspect ratio and cross-sectional area of a microreactor for Fischer – Tropsch reaction performance. The comparative study was based on 20%Co-0.5%Re/γ-Al_2O_3 catalyst, a space time in a range of 0.0020 – 0.0060 g_{cat}·min/N·cm^3 and Reynolds number lower than 1. The base case model was set as a square cross-sectional area with a dimension of 0.7 mm×0.7 mm and a length of 20 mm. It gave 22.41% of CO conversion. The simulation results showed that changing of aspect ratio induced a little effect to reaction performance. The maximum conversion difference was 7.90% resulted from the most shallow flow channel of 1.225 mm × 0.175 mm compared to the base case model. When increasing microchannel cross-sectional area to 1.00 mm × 1.00 mm, it gave negative effect on Fischer – Tropsch reaction performance, while reducing the cross-sectional area, to a 0.300 mm × 0.300 mm with the length of 50.18 mm, it gave higher CO conversion and higher pressure drop when compared to other cases. Minimum channel length obtained from different cross-sectional area of microchannel were studied. The smaller cross-sectional area the shorter channel length was required. In this work, systematic analysis and interpretation of microchannel for FTS design were achieved.

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Keywords: Microchannel reactor; Fischer Tropsch Synthesis; computational fluid dynamic; flow channel

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1. Introduction

Gas to Liquid (GTL) process can be used for converting associated petroluem gas to longer chain hydrocarbons, such as gasoline or diesel fuel [1]. GTL process mainly comprises of two major steps: reforming of natural gas to produce synthetic gas (CO and H_2) and Fischer Tropsch Synthesis (FTS) to produce liquid fuels [2]. FTS has been addressed as a promising route for liquid fuel production from syngas [3, 4]. The synthetic fuels produced from FTS are considered as clean fuels because it has high cetane number and sulphur free [3]. In a conventional process, FTS have been operated in fixed bed, fluidized bed and slurry bubble reactor [3, 5]. However, the conventional reactors exhibit heat and mass transfer limitations [6]. Taking into account highly exothermic characteristic of FTS, a large amount of heat of reaction are released during the FTS. Recently, a concept of microchannel reactor has been widely studied. Microchannel reactors could provide predominant advantages over conventional reactor including remarkable improvement of mass and heat transfer, on – site production and safety in case of preventing runaway situation [4, 5]. Various design studies for microchannel device have been widely reported [7, 8]. However, in FTS technology, a few investigations have been carried out on microchannel reactor geometries study. The channel geometries play an important role on hydrodynamic behavior of fluids in a microreactor [9, 10]. In an actual design process, many decision variables, such as microreactor configurations, channel geometries, pressure loss (ΔP) and reaction performance are involved. However, fabrication restrictions in micro – scale device are also a concern. In this present study, channel shape and size were focused in order to provide specific perspective of microreactor design for FTS on reaction performance and fabrication possibility.

Nomenclature

k	reaction rate constant ($mol_{co}/kg_{cat}\cdot s\cdot atm^2$)
K_{CO}	equilibrium constant for CO adsorption
P	partial pressure (atm)
E_A	apparent activation energy (kJ/mol)
ΔH	heat of reaction (kJ/mol)
T	temperature (K)
R	gas constant (J/mol·K)

2. Computational Fluid Dynamic of microchannel reactor

As mentioned above, there are number of chemical reactions in FTS for converting a mixture of CO and H_2 into various hydrocarbons. The simplified set of FTS reactions can be shown in equation 1 – 6 [4].

$$CO + 3H_2 \xrightarrow{k_1} CH_4 + H_2O \quad (1)$$

$$2CO + 5H_2 \xrightarrow{k_2} C_2H_6 + 2H_2O \quad (2)$$

$$3CO + 7H_2 \xrightarrow{k_3} C_3H_8 + 3H_2O \quad (3)$$

$$4CO + 9H_2 \xrightarrow{k_4} C_4H_{10} + 4H_2O \quad (4)$$

$$14CO + 29H_2 \xrightarrow{k_{C_{5+}}} C_{14}H_{30} + 14H_2O \quad (5)$$

$$CO + H_2O \xrightarrow{k_{CO_2}} CO_2 + H_2 \quad (6)$$

The reaction scheme of FTS Langmuir – Hinshelwood – Hougen – Watson (LHHW) equations were considered as rate expressions for the elementary steps consuming CO and H_2 and producing hydrocarbons and water. Lumped kinetic equations based on 20%Co-0.5%Re/γ-Al_2O_3 catalyst used in this study were adopted from Almeida et al. [4] as shown in equation 7 – 10.

$$R_n = \frac{k_n P_{CO}^{\beta} P_{H_2}}{(1 + K_{CO} P_{CO})^2} \quad \begin{matrix} \beta = 1 (n < 5) \\ \beta = 1.2 (n = 5) \end{matrix} \quad (7)$$

$$k_{C_n} = k_{C_n,473} \cdot \exp\left[\frac{-E_A}{R} \cdot \left(\frac{1}{T} - \frac{1}{473} \right) \right] \quad (8)$$

$$K_{CO} = K_{CO,473} \cdot \exp\left[\frac{\Delta H_{CO}}{R} \cdot \left(\frac{1}{T} - \frac{1}{473} \right) \right] \quad (9)$$

$$R_{CO_2} = k_{CO_2} P_{H_2O}^{0.35} \quad (10)$$

The kinetic model can be applied to the simulation model of the microchannel reactor for all cases. The three – dimensional simulation model was conducted in a steady state mode. The system composes of reaction channel with 35.00 μm of wall coated catalyst. The commercial computational fluid dynamic (CFD) package, COMSOL Multiphysics 3.5a was used for the simulation. The momentum equations were solved to study influence of fluid hydrodynamics. The syngas mole ratio (H_2:CO:N_2) used in this study was 6:3:1. The three – dimensional simulation was conducted at steady state based on the conditions of 508 K and 10 atm with a space time of 0.0020 – 0.0060 g_{cat}·min/N·cm^3 and catalyst density of 29.29 g_{cat}/m^2 [4].

2.1. Base Case Model

In general, residence time distribution (RTD) is directly affected by aspect ratio (channel height/channel width) of the microchannel. Aubin et al. suggested that, the microchannel should be designed with low aspect ratio in order to minimize the RTD [11]. Burns and Ramshaw suggested that narrow channel could provide better performance in term of lower diffusion path length [12]. Hence, to study the effect of aspect ratio on FTS reaction performance, microchannel with upper limit of 0.700 mm×0.700 mm cross-sectional area with the aspect ratio of 1 was set as the base case model as showed in Fig. 1.

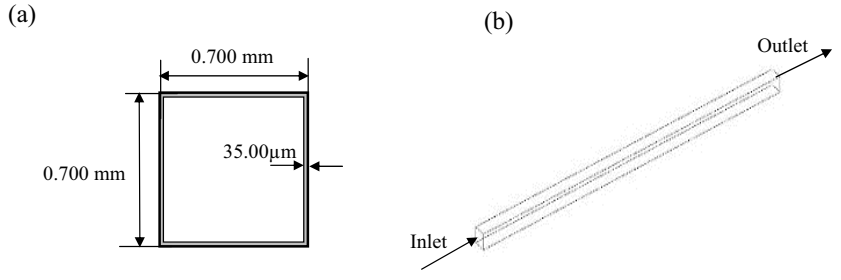

Fig. 1. Base case model of a square microchannel (case A) (a) front view; (b) overall view

To investigate effect of channel geometry on reaction performance, base case model of the microchannel was set according to Almeida et al. [4]. The dimension of square cross section channel was 0.700 mm×0.700 mm with the

length of 20.00 mm. The channel shape and size were focused in this work, structural microchannel of different cases can be shown in Table1. Structural parameters of the microchannel of different cases.

Table 1. Structural parameters of the microchannel of different width and height ratios.

Cases	W(mm)×H (mm)	Aspect ratio	L (mm)
A	0.700×0.700	1.00	20.00
B	0.875×0.525	0.60	20.00
C	1.050×0.350	0.33	20.00
D	1.225×0.175	0.14	20.00
E	1.00×1.00	1.00	13.78
F	0.500×0.500	1.00	28.60
G	0.300×0.300	1.00	50.18

2.2. Effect of aspect ratio on FTS reaction performance

A few investigations on the effect of aspect ratio on RTD and Reynolds number (Re) have been proposed [11, 12]. The lack of investigation about the effect of aspect ratio on a particular reaction performance was noticeable. In this section, different ratios of channel height and width resulting in different shapes of the microchannels were focused. As the FTS operation in microchannel reactor was in the laminar flow scheme with the Reynolds number approximately lower than 1, it can be anticipated that, with thinner flow channel, Reynolds number may increase due to reduction of channel hydraulic diameter. The resulting shape and structural parameter of the channel are summarized in Fig. 2 and Table 1 respectively. Perimeter and length of the channels of each case were fixed to keep catalyst loading of 1.64 mg constant for every case.

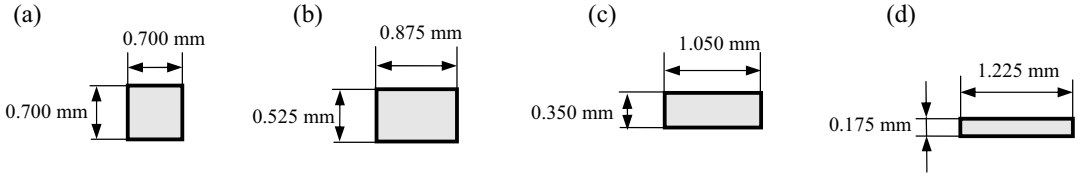

Fig. 2. Sketch of microchannel with different channel width and length (a) case A; (b) case B; (c) case C; (d) case D

2.3. Effect of crossectional area on FTS reaction

For industrial application of microstructured reactor, scale up is often considered in microscale and increasing capacity by numbering up. Thereby, a large number of small channels may be required. Adjusting size of the microchannel would be an alternative approach, if it benefits the design and characteristics of small scale operation can be preserved. In order to study the effect of microchannel size, aspect ratio was fixed to eliminate the effect of aforementioned parameters. Consequently, channel length of microchannel were changed to maintain catalyst loading. The structural parameters are shown in Fig. 3 and Table 1. In this section, space time and catalyst density were fixed at 0.002 – 0.006 g_{cat}·min/N·cm^3 and 29.29 g_{cat}/m^2, respectively [4].

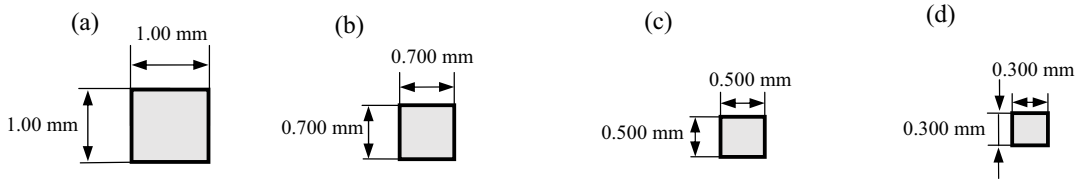

Fig. 3. Sketch of microchannel with different perimeter (a) case E (b) case A (c) case F (d) case G.

2.4. Effect of channel length on FTS reaction performance

To obtain optimal length of a particular perimeter (case A, E, F and G), the effect of channel length was studied. The simulation model of microchannel was set with constant aspect ratio, space time and catalyst density of 1.00, 0.0020 – 0.0060 g_{cat}·min/N·cm^3 and 29.29 g_{cat}/m^2, respectively. In this section, perimeter and channel length were defined to investigate performance of the microchannel on FTS.

3. Results and discussion

3.1. Influence of height and width ratio

For the base case model, obtained CO conversion were 10.27%, 22.40% and 29.36% at space time of 0.0020, 0.0045 and 0.0060 g_{cat}·min/N·cm^3, respectively. These results are in good agreement with Almeida et al. showing 22% CO conversion at a space time of 0.0045 g_{cat}·min/N·cm^3 [4]. Thus, the base case model was satisfied. Fig. 4 shows results from varying space time in range of 0.0020 – 0.0060 g_{cat}·min/N·cm^3 indicating that the lower aspect ratio the higher CO conversion at every space time. The change of height and width ratio might affect diffusion time of the reactants from bulk phase into catalytic coated wall. Diffusion time (τ_D) of the reactants can be calculated by using equation 11 [13]. The obtained diffusion time for all cases could be ranked as case A > case B > case C > case D. Thus, the reactants might be faster to access to the catalytic surface. Subsequently, case D gave the highest CO conversion compared to other cases showing 4.17%, 5.90% and 5.90% higher than that of case A at space time of 0.0020, 0.0045 and 0.0060 g_{cat}·min/N·cm^3, respectively. It could be concluded that shallow channel possibly achieve effective diffusion in microchannel. Moreover, reducing aspect ratio of microchannel also affects to Reynolds number and pressure drop along the channel. The Re can be calculated by equation 12 [14]. From Table 2, the base case revealed much lower Re and ΔP compared to case D. However, Re and ΔP of all cases are in very low magnitude.

$$\tau_D = \frac{d^2}{D_{CO,H_2}} \tag{11}$$

$$Re = \frac{\rho d v}{\mu} \tag{12}$$

Where diffusion coefficient of CO and H_2 (D_{CO,H_2}) at FTS reaction conditions is 1.963×10^{-5} (m^2/s), d is hydraulic diameter of the microchannel, ρ is density of gas mixture (kg/m^3) and μ is viscosity of gas mixture (kg/m·s).

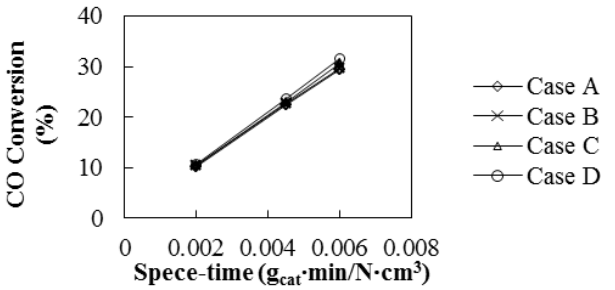

Fig. 4. Effect of channel aspect ratio on CO conversion.

Table 2. Reynolds number and pressure drop of microchannel at space time of 0.0045 g_{cat}·min/N·cm^3.

Cases	A	B	C	D
Re	0.02	0.03	0.04	0.07
ΔP (Pa)	0.14	0.18	0.41	2.42

3.2. Influence of crossectional area

According to the base case, square channel of 0.700 mm × 0.700 mm, the comparative study of cross-sectional area was investigated in this section. As mentioned above, constant aspect ratio and catalyst loading of the channel leads to different channel length. In this particular study, when changing the width and height of a square flow channel, the channel length would be changed accordingly as catalyst weight was kept constant. Therefore, the flow channel gave the shorter channel length and vice versa. The simulation results illustrates that CO conversion was affected by change of cross-sectional area. The highest CO conversion was obtained from the channel with a smallest cross-sectional area of 0.300 mm × 0.300 mm (case G) at every space time as shown in Fig. 5. The biggest flow channel gave CO conversion lower than that of the base case over 24% at a space time of 0.0045 g_{cat}·min/N·cm^3. This might be a result from increasing cross-sectional area increases diffusion time of the reactants from bulk flow to catalytic surface. Table 3 presented the result from the effect of cross-sectional area of flow channels on Re and ΔP. The results also showed that the smallest flow channel and longest channel (case G) exhibited the highest Re and ΔP. Moreover, the obtained ΔP from case G was approximately 100 times higher than that of the base case.

Table 3. Reynolds number and pressure drop of microchannel at space time 0.0045 g_{cat}·min/N·cm^3.

Cases	A	E	F	G
Re	0.02	0.02	0.06	0.16
ΔP (Pa)	0.14	0.02	0.79	10.74

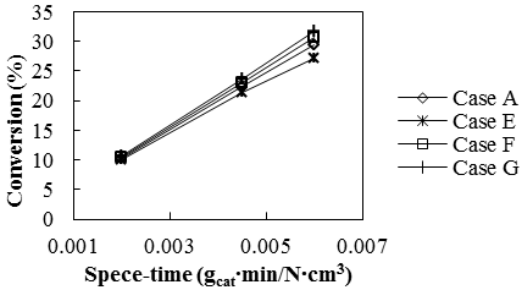

Fig. 5. Effect of cross-sectional area of square flow channel on CO conversion.

3.3. Minimum length of microchannel

Following the study from the previous part where the flow channel cross-sectional area was varied from 1.00 mm ×1.00 mm to 0.300 mm × 0.300 mm. This section focused on investigation of the minimum channel length and the simulation was conducted from varying space time in a range of 0.0020 – 0.0060 g_{cat}·min/N·cm^3. In this case, channel length for each cross-sectional area was increased to 100 mm and the conversion of CO was reported along with the change of channel length. The results were shown in Fig. 6. – Fig. 7. and the minimum channel length can be summarized in Table 4. It can be seen that, in every case at all three space times, the conversion of CO became insignificantly changed and became unchanged at a certain length. Thus, the minimum length of a flow channel was defined as a length where the conversion of CO was different from that at 100 mm no more than 5%. For the base case model with a space time of 0.0045 g_{cat}·min/N·cm^3, the minimum length where CO conversion became insignificant changed was at 20.26 mm as shown in Fig. 6. This was in a good agreement with Jermkwan et al.[15] where they found an optimal length when operated at the same operating condition with the same design of the reactor to be around 28.49 mm. From other results, it can be seen that, when the reactor operated at low space time, 0.002 g_{cat}·min/N·cm^3, bigger flow channel required longer flow channel. For the case with the biggest cross-sectional area flow channel of 1.00 mm × 1.00 mm (case E), CO conversion became insignificant changed were at 19.64, 25.30 and 26.13 at space time of 0.0020, 0.0045 and 0.0060 g_{cat}·min/N·cm^3, respectively. As for smaller cross-sectional area flow channel, 0.500 mm × 0.500 mm and 0.300 mm × 0.300 mm, they required shorter channel length as can be seen that the minimum channel length were 14.57 mm and 12.15 mm at a space time of 0.002 g_{cat}·min/N·cm^3 which was shorter than that of 1.00 mm × 1.00 mm reactor.

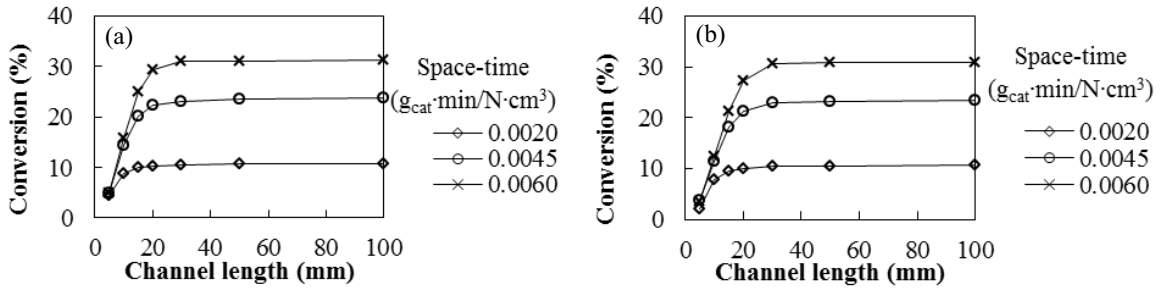

Fig. 6. Effect of channel length on CO conversion of base case of (a) case A; (b) case E

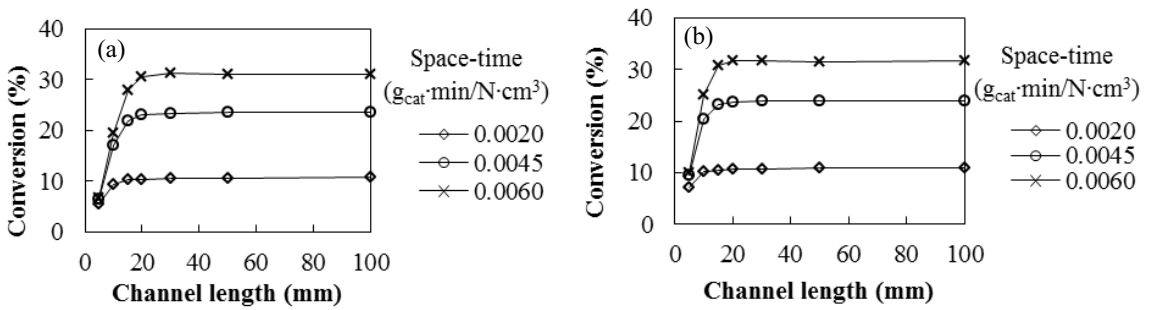

Fig. 7. Effect of channel length on CO conversion of (a) case F; (b) case G.

Table 4. Minimum length of microchannel reactor.

Space time (g_{cat}·min/N·cm^3)	Minimum length (mm)			
	Case A	Case E	Case F	Case G
0.0020	18.38	19.64	14.57	12.15
0.0045	20.26	25.30	15.63	14.17
0.0060	21.90	26.13	17.99	14.32

4. Conclusions

The channel geometry of microchannel reactor plays an important role on hydrodynamic of fluids within the microchannel reactor. From the design perspective, there are no universal threshold method. Designers have to design a reactor based on application of the device. In actual design process not only the channel geometry should be concerned but also fabrication tolerance. The geometry of microchannel should be selected in appropriate value depending on the reaction performance and fabrication possibility. This work provided a comparative study of the effect of microchannel aspect ratio and perimeter on FTS performance based on 20%Co-0.5%Re/γ-Al_2O_3 catalyst, space time in a range of 0.0020 – 0.0060 g_{cat}·min/N·cm^3 and Reynolds number lower than 1. The obtained CO conversion of base case model with channel dimensions of 0.700 mm × 0.700 mm × 20.00 mm was in a good agreement with Almeida et al. [4]. The base case model gave 22.41% of CO conversion. From the influence of channel aspect ratio, the simulation results indicated that the shallow and wide channel gave lower diffusion time for diffusion of reactants from bulk phase onto the catalytic wall. However, the change of aspect ratio induced a little effect to reaction performance in term of CO conversion. For the effect of square flow channel cross-sectional area investigation, the catalyst loading was kept constant. Thus, the smaller cross-sectional area the longer channel length was obtained. The results also showed that microchannel with the largest cross-sectional area of 1.00 mm × 1.00 mm (case E) gave negative effect on reaction performance compared to base case model. Moreover the obtained channel length and minimum length were not in the same range. The channel with small cross-sectional area and long channel length (case G) gave very high CO conversion and ΔP compare to other cases. Taking into account fabrication process, high fabrication cost might be required for very small channel size. From this study, it should be noted that designers might come up with groups of structural parameters. In decision process, operational design and manufacturing restrictions should be considered as a designing criteria.

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References

[1] A. Ghorbani, M. Jafari, M.R. Rahimpour, A comparative simulation of a novel gas to liquid (GTL) conversion loop as an alternative to a certain refinery gas flare. J Nat Gas Sci Eng 2013;11:23-38.
[2] K.J. Gabriel, M. Noureldin, M.M. El-Halwagi, P. Linke, A. Jiménez-Gutiérrez, D.Y. Martínez, Gas-to-liquid (GTL) technology: Targets for process design and water-energy nexus. Cur Opin Chem 2014;5:49-54.
[3] M.-S. Shin, N. Park, M.-J. Park, K.-W. Jun, K.-S. Ha, Computational fluid dynamics model of a modular multichannel reactor for Fischer–Tropsch synthesis: Maximum utilization of catalytic bed by microchannel heat exchangers. Chem Eng J 2013;234:23-32.
[4] L.C. Almeida, O. Sanz, D. Merino, G. Arzamendi, L.M. Gandía, M. Montes, Kinetic analysis and microstructured reactors modeling for the Fischer–Tropsch synthesis over a Co–Re/Al2O3 catalyst. Catal Today 2013;215:103-111.
[5] K.S.K. Ikhwan Jung, Seongho Park, Jonggeol Na, Yongkyu Lee, Jinjoo An, Seongeon Park, Chul-Jin Lee, and Chonghun Han, Computational Fluid Dynamics Based Optimal Design of Guiding Channel Geometry in U‑Type Coolant Layer Manifold of Large-Scale Microchannel Fischer−Tropsch Reactor. Ind & eng Chem R D 2016;55:505-151.
[6] L.C. Almeida, F.J. Echave, O. Sanz, M.A. Centeno, G. Arzamendi, L.M. Gandía, E.F. Sousa-Aguiar, J.A. Odriozola, M. Montes, Fischer–Tropsch synthesis in microchannels. Chem Eng J 2011;167:536-544.
[7] D. Mei, L. Liang, M. Qian, X. Lou, Modeling and analysis of flow distribution in an A-type microchannel reactor. Int J of Hydrogen Energ 2013;38:15488-15499.
[8] G. Agrawal, N.S. Kaisare, S. Pushpavanam, K. Ramanathan, Modeling the effect of flow mal-distribution on the performance of a catalytic converter. Chem Eng Sci 2012;71:310-320.
[9] M. Mohammadi, G.N. Jovanovic, K.V. Sharp, Numerical study of flow uniformity and pressure characteristics within a microchannel array with triangular manifolds. Comput Chem Eng 2013;52:134-144.
[10] E.R. Delsman, A. Pierik, M.H.J.M. De Croon, G.J. Kramer, J.C. Schouten, Microchannel Plate Geometry Optimization for Even Flow Distribution at High Flow Rates. Chem Eng Res Des 2004;82:267-273.
[11] J. Aubin, L. Prat, C. Xuereb, C. Gourdon, Effect of microchannel aspect ratio on residence time distributions and the axial dispersion coefficient. Chem Eng Process 2009;48:554-559.
[12] J.R. Burns, C. Ramshaw, Process and Product DevelopmentDevelopment of a Microreactor for Chemical Production. Chem Eng Res Des 1999;77:206-211.
[13] M. Zanfir, A. Gavriilidis, Catalytic combustion assisted methane steam reforming in a catalytic plate reactor. Chem Eng.Sci 2003;58:3947-3960.
[14] R.B. Bird, W.E. Stewart, E.N. Lightfoot, Transport Phenomena: Wiley; 2001.
[15] J. Waewsak, S. O-Thong, K. Sungkharak, N. Jermkwan, P. Inbamrung, P. Narataruksa, C. Prapainainar, 2015 International Conference on Alternative Energy in Developing Countries and Emerging EconomiesDesign Equations for Catalytic Microchannel Reactors: Fischer-Tropsch Synthesis. Energy Procedia 2015;79:772-777.