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Steam reforming integrated with oxidation of methanol in a micro-channel reactor with different micro-baffle shapes

E. Omidbakhsh Amiri

Department of Chemical Engineering, University of Mazandaran, Babolsar, Iran

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Abstract

A micro-channel heat exchanger reactor with different micro-baffle shapes has been studied numerically. Governing equations were solved base on the finite volume method with FLUENT software. In the upper section of the reactor oxidation reaction of methanol occurred, and in the lower section steam reforming of methanol took place. Two sections were separated by a solid part which played the role of a heat exchanger and transferred heat from the oxidation reaction to the steam reforming section. In addition to the straight micro-channel, other types of micro-channel with different micro-baffle shapes on both sides of the solid section were studied. Micro-baffles are thought to act as static mixers, inducing further mixing and improving the convective heat transfer coefficients which eventually expedite heat transfer which increases conversion efficiency. Also, the hydrogen yield obtained by a rectangular micro-baffle is 17% higher, on average, than that obtained with by a straight configuration. Five different micro-baffles with Rectangular, Triangular, Triangular 90, Trapezoidal and Trapezoidal 90 shapes were studied and the highest conversion efficiency for the micro-baffles was achieved in the Trapezoidal shape.

1. Introduction

In hydrocarbon fuel reformers the fuel, such as gasoline, ethanol, methane or methanol, can be converted into hydrogen-rich gas through steam reforming (SR) or auto thermal reforming (ATR), following by a water gas shift (WGS) reaction and a partial oxidation (POX) reaction. Among these available hydrocarbon fuels, methanol is a unique fuel because it is sulfur-free and can be activated at a relatively low temperature (under 300°C). Micro-channel reactors have been introduced to miniaturize a complicated steam reformer. This was done mainly because the micro-channel reactors merit a higher surface to volume ratio, which might be several orders of magnitude higher compared to the traditional steam

*Corresponding Author's E-mail address: e.omidbakhsh@umz.ac.ir

reformer; and hence, the heat and mass transfer inside the micro-reformer can be greatly enhanced [1].

These features of micro-channels allow for effective use of the catalysts and coupling of exothermic and endothermic reactions in spatially-segregated configurations , i.e., the reactive streams are unmixed and the only mode of transport is heat exchange through the walls [2]. The concept of coupling exothermic and endothermic reactions in micro-channel systems has been thoroughly covered in the literature using computational models [3-10].

Andisheh Tadbir and Akbari [3] studied methanol reforming in a micro-reactor. They investigated autothermal reforming of methanol in a single rectangular micro-channel. The effects of steam/carbon ratio at the reformer inlet, gas space velocity of both channels, and the reformer catalyst thickness on methanol conversion and hydrogen production were investigated. Also, Stefanidis and Vlachos [4] studied hydrogen production of a multifunctional micro-device consisting of thermally coupled catalytic plate combustion and reforming micro-reactors for methane and methanol reforming in a high and low temperature process, respectively. The 3-D simulation work by Arzamendi et al. [5] involved investigating coupling of methanol combustion and reforming in multiple micro-channels. Different flow directions (co-current, counter-current and cross flow) of combustion and reforming streams were considered. Karakaya and Avci [2] investigated coupling of exothermic and endothermic reactions in parallel micro-channels through parametric variation of geometric and material properties in the context of hydrogen production by steam reforming of iso-octane, as a surrogate for gasoline. The influence of parameters, such as wall thickness, material, and channel-opening dimensions, on hydrogen yield, defined as number of moles of hydrogen produced per mole of iso-octane fed, was investigated. The use of non-straight textures, called micro-baffles, in the catalytic walls of the micro-channels was also explored. Use of micro-baffles enhanced the hydrogen yield, and was attributed to both the enhancement in heat transfer coefficients and axial conduction in the wall. Heidary et al. [6] studied heat transfer and flow field analysis in the anode side of direct methanol fuel cells. To enhance the heat exchange between the bottom cold wall and the core flow, the bottom wall of the fluid delivery channel was considered as a corrugated boundary instead of a straight one. Four different shapes of corrugated boundary are recommended here: rectangular shape, trapezoidal shape, triangular shape and wavy shape. The performed parametric studies showed that the corrugated channel with a trapezoidal, triangular and wavy shape enhanced the heat exchange up to 90%. With these boundaries, the cooling purpose of reacting flow in the anode side would be better than the straight one.

In this work, steam reforming (SR) of methanol integrated with oxidation (OX) of methanol is investigated in a micro-channel reactor for hydrogen production. An OX reaction occurred in the upper channel, while a SR reaction took place in the lower channel. The two sections were separated with a solid part which played the role of a heat exchanger and transferred the heat from the OX reaction to the SR section. Micro-baffles with different shapes were used and studied to determine which produced better heat transfer.

2. Model description

A micro-channel heat exchanger reactor has been studied. In upper section an OX reaction of methanol occurred and in lower section the SR of methanol occurred. The two sections were separated with a solid part which played the role of a heat exchanger and transferred heat from the OX reaction to the SR section. SR is an endothermic reaction, while the OX reaction is an exothermic reaction and supplies the heat for the SR reaction. The base model of Andisheh Tadbir's work [3] (Figure 1(a)) was used as a reference for our main work in geometry and boundary conditions. Figure 1 shows the different shapes of micro-baffle on a wall of the solid section. Figure 1shows the upper and lower sections where the OX and SR reaction occurred. The solid section is colored black. The boundary of the baffles is outlined in red for a clearer view of the baffle shape. The five micro-baffle shapes used in this work were: straight micro-channel (STR), Rectangular (RB), Triangular (TRGB), Triangular 90 (TRG90B), Trapezoidal (TRPB) and Trapezoidal 90 (TRP90B) shapes. The lengths of the OX and SR sections, in all models, were equal 20 mm. Also, the heights of the OX and SR sections were 0.7 mm. The thickness of the solid section, placed between the two sections was 0.3 mm. The heights and widths of all micro-baffles were 0.3 and 2 mm, respectively. For the trapezoidal micro-baffle, the width of the upper sections was 0.5 mm.

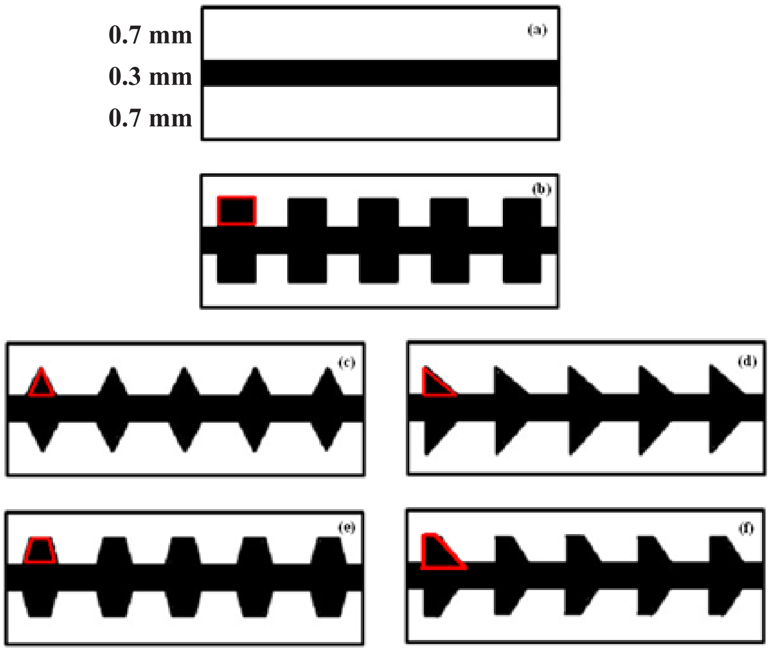

Fig.s 1. (a) Straight micro-channel (STR) and different shapes of micro-baffle, (b) Rectangular shape (RB), (c) Triangular shape (TRGB), (d) Triangular 90 shape (TRG90B), (e) Trapezoidal shape (TRPB), (f) Trapezoidal 90 shape (TRP90B).

3. Numerical modeling

3.1. Governing equations

In this model it is assumed that the flow is laminar and steady state, gravitation force is neglected, the inlet flow is an ideal gas, and there are wall surface reactions on the top and bottom walls of the OX and SR sections. Governing equations for these systems in the gas phase consisted of continuity, momentum, energy transport and Species equations which are listed in Equations (1-4), respectively.

Continuity Equation:

$$\rho_{gmix}\nabla.(\vec{v}) = 0 \tag{1}$$

Momentum transport Equation:

$$\rho_{gmix}\vec{v}.\nabla\vec{v} = -\nabla P + \mu_{gmix}(\nabla^2\vec{v}) \tag{2}$$

Energy transport Equation:

$$\rho_{gmix}c_{pmix}\nabla.(\vec{v}T) = k_{gmix}\nabla^2 T + \Delta H_R \tag{3}$$

Species mass balance Equation:

$$\rho_{gmix}\nabla.(\vec{v}y_i) = -\nabla.\vec{J}_i + R_i \tag{4}$$

In equations (1-4), v, P, T, y_i, J_i are velocity, pressure, temperature, mass fraction of species i, and diffusion flux of species i, respectively. Also, ρ_{gmix}, μ_{gmix}, c_{pmix} and

k_{gmix} are density, viscosity, specific heat capacity and thermal conductivity of the gas mixture and are defined in Table 1. The thermal conductivity of the gaseous mixture k_{gmix} was calculated in the same manner as μ_{gmix}. More explanations are defined in [11]. Also, ΔH_R is the heat of reaction for both reactions (OX and SR) and are also defined in [11]. R_i is the net rate of production of species *i* by chemical reaction and has a non zero value only on the reaction wall and is listed in Table 1. The heat released in the OX reaction (Eq. 5) in the SR section and the required heat of the SR reaction (Equation (6)) was accounted and

$$CH_3OH + 1.5O_2 \rightarrow 2H_2O + CO_2 \tag{5}$$

$$CH_3OH + H_2O \rightarrow 3H_2 + CO_2 \tag{6}$$

And r_{OX}, r_{SR}, C_i and P_i are the oxidation reaction rate, steam reforming reaction rate, concentration and partial pressure of species *i*, respectively.
In the solid section between the two SR and OX channels, only the energy equation is solved. The energy equation for this section is written as Equation (7):

$$\nabla^2 T = 0 \tag{7}$$

3.2. Boundary Conditions and Solution Method

The top and bottom walls of the OX and SR sections are the wall surface reaction, respectively. The inlet flow, temperature, composition and mass flow rate of feed is specified as in Andisheh Tadbir's work [3].

Table 1. Physical Properties and Chemical Reaction Rates

Property	Formula
Density	$\rho_{gmix} = \dfrac{P}{RT\sum_i \dfrac{y_i}{Mw_i}}$
Specific heat capacity	$c_{Pmix} = \sum_i y_i c_{P,i}$
Viscosity	$\mu_{gmix} = \sum_i \dfrac{x_i \mu_i}{\sum_j x_i \varphi_{ij}}$ $\varphi_{ij} = \dfrac{\left[1 + \left(\dfrac{\mu_i}{\mu_j}\right)^{1/2} \left(\dfrac{Mw_j}{Mw_i}\right)^{1/4}\right]^2}{\left[8\left(1 + \dfrac{Mw_i}{Mw_j}\right)\right]^{1/2}}$
Steam Reforming Reaction Rate	$r_{SR} = 5.3*10^{12} \exp(-105/RT) P_{Methanol}^{0.26} P_{H_2O}^{0.03}$
Oxidation Reaction Rate	$r_{OX} = 2.761*10^{5} \exp(-13.1/RT) C_{Methanol}^{1.3}$

In Table 1, *R* and *p* are the universal gas constant and operating pressure, respectively. Also, y_i, x_i and M_{wi} are the mass fraction of species *i*, mole fraction of species *i*, and the molecular weight of species *i*, respectively.

Figure 2 shows boundary condition for straight micro-channel (Figure 1(a)); other models have the same conditions. Also, in Table 2 boundary conditions are listed.

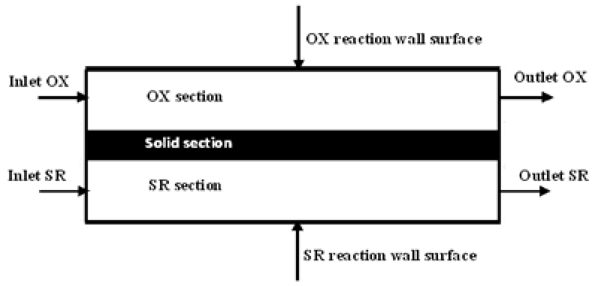

Fig. 2. Boundary conditions of STR model.

shown in [11]. The comparison between the results of our model and Andisheh Tadbir's work [3] is shown in Figure 3. Excluding the first point, there is good agreement and the average error is 4.5%.. So, the model is acceptable.

4.2. Effect of micro-baffle

In the first section the STR model was compared with the RB model to consider the effect of using micro-baffles. Two parameters were defined for this comparison. The first, methanol conversion efficiency is defined as follows:

Table 2. Boundary conditions of STR model.

Boundary	Condition	
Inlet OX (or Inlet SR)	Mass flow rate,	$\dot{m} = \dot{m}_{in}$
	composition and	$y_i = y_{i,\,in}$
	temperature are specified.	$T = T_{in}$
OX (or SR) reaction wall surface	No slip condition	$v=0$
	reaction (OX or SR)	
	was occurred on this wall	

Governing equations (1-4) as well as equation (7) are solved with the finite volume method over the control volume with FLUENT software. A first-order upwind discretization scheme was used for the governing equations expect for pressure which is discretized by standard method. The SIMPLE algorithm was employed to solve the convection-diffusion equations. In thorough of control volume, Quad or Quad-Pave grids were used. Utilizing these different considerations, the optimal size and type of grids were achieved for any model (either "were achieved for the model" or "can be achieved for any model").

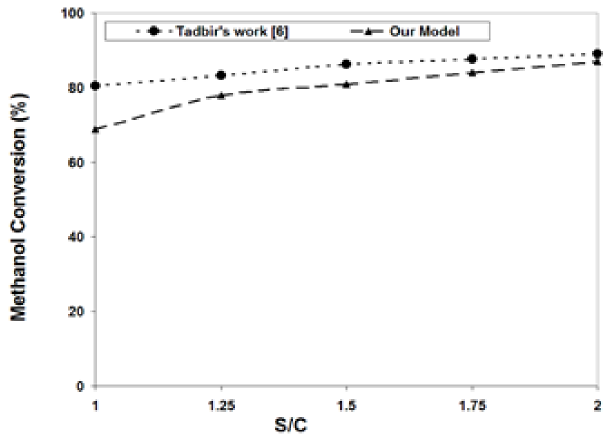

Fig. 3. Comparison of simulation results with Andisheh Tadbir's work [3].

4. Results and discussion

4.1. Model validation

In the first step, the model was validated. Andisheh Tadbir's work [3] was used for the validation. A rectangular micro-channel, with a cross section of 0.7 * 0.7 mm^2 and a length of 10 mm, was studied in their work. Also, the boundary conditions of their work were used. More explanations of model validation are

$$\eta = \frac{X_{MeOH,in} - X_{MeOH}}{X_{MeOH,in}} * 100 \qquad (8)$$

Where $X_{MeOH,in}$ and X_{MeOH} are the mole fraction of methanol at the inlet flow and along the micro-channel length. The second parameter was hydrogen yield and is defined as follows:

$$Y_{H_2} = \frac{\dot{n}_{H_2}}{\dot{n}_{MeOH,in}} \tag{9}$$

In equation (9), n˙ is the molar flow rate.

Figures 4 (A) and (B) show the methanol conversion efficiency and hydrogen yield at y=0.2 mm (y is the distance from the SR reaction wall surface) along the channel lengh for STR and RB models. Conversion efficienctу in the RB model was higher than the STR model. Also, hydrogen yield obtained with the RB model was 17% higher, on average, than that obtained with the straight configuration.

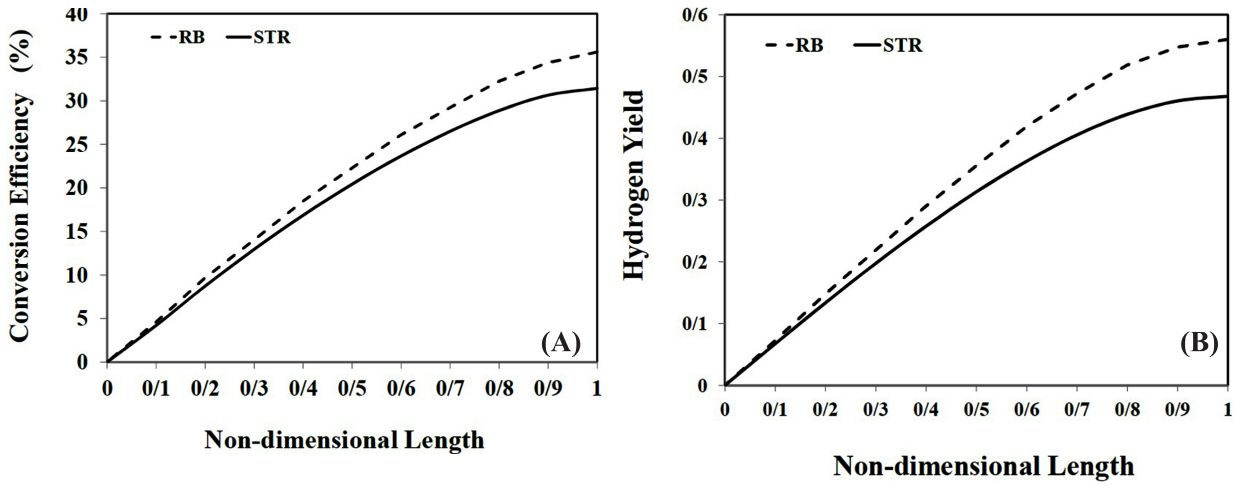

Fig. 4. (A) Methanol conversion efficiency along the channel length for STR and RB models, (B) Hydrogen Yield along the channel length for STR and RB models (Re=900).

Figure 5 shows the methanol conversion efficiency in the outlet flow for STR and RB models at different Reynols numbers. At all Reynolds numbers, efficiency of the RB model was higher than that of the STR model; although, when the Reynolds number increases the difference between two models decreases. Figures 6 (A) and (B) show the streamline for STR and RB models along the channel length. It can be seen, that the fluid flows in a straight path in STR model, , while the flow meanders, due to the disturbance of micro-baffles, in the RB model. It seems the micro-baffles act as static mixers, inducing further mixing and improving the convective heat transfer coefficients which eventually expedites heat transfer, so the conversion efficiency is more than one. Micro-baffles servea dual purpose of enhancing the heat transfer coefficient and axial heat conduction. Figure 7 shows the temperature variations of the bulk of fluid (in the SR section) for STR and RB models along the channel length. It can be seen that the temperature increased on the baffle positions.

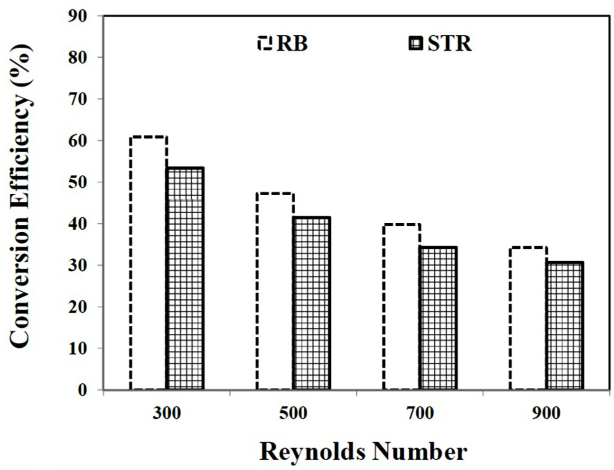

Fig. 5. Methanol conversion efficiency versus Reynolds number for STR and RB models.

4.3. Effect of micro-baffle shape

In this section, five different micro-baffle shapes are compared. Rectangular (RB), Triangular (TRGB), Triangular 90 (TRG90B), Trapezoidal (TRPB) and Trapezoidal 90 (TRP90B) shapes.

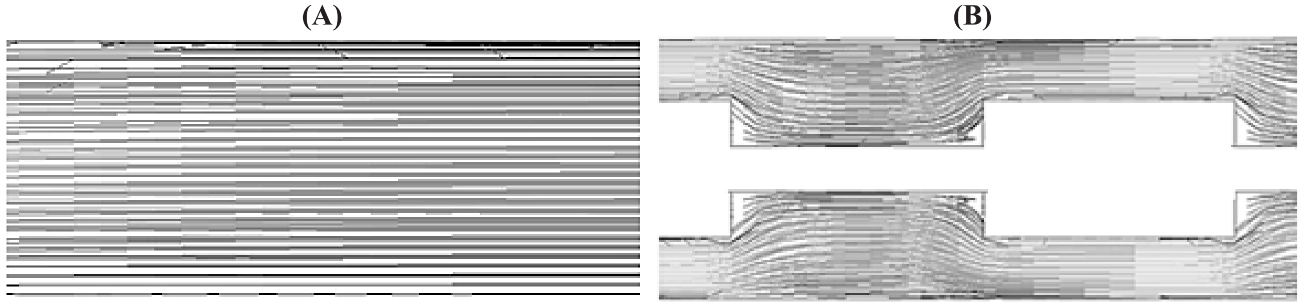

Fig. 6. Streamline for (A) STR and, (B) RB models along the channel length (Re=900).

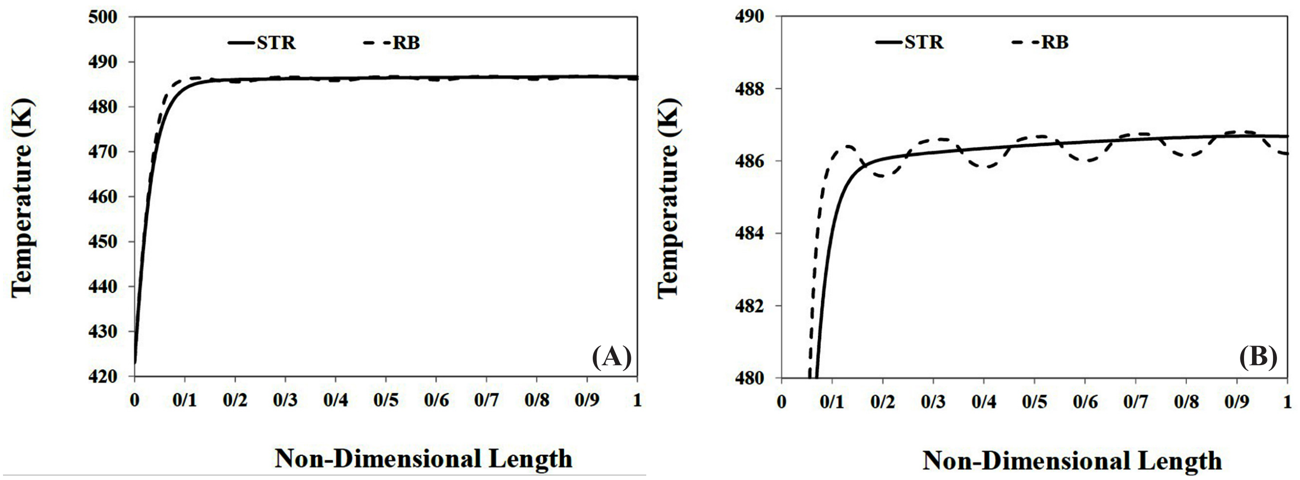

Fig.7. (A) Temperature variations versus non- dimensional length for STR and RB models, (B) zoomed on the baffle positions (Re=900)

Figures 8(A) and (B) show methanol conversion efficiency and hydrogen yield along the channel length for different micro-baffle shapes. From Figure 8, we see there is a higher conversion efficiency and hydrogen yield for the TRPB model than that for the other models. In the TRPB model the Trapezoidal shape with two crooked lengths may induce more mixing, so a higher conversion efficency was achieved. Figure 9 shows that this trend is valid for other Reynolds numbers. Although, increasing the Reynolds number decreases the residence time of the reactant and methanol conversion efficiency decreases.

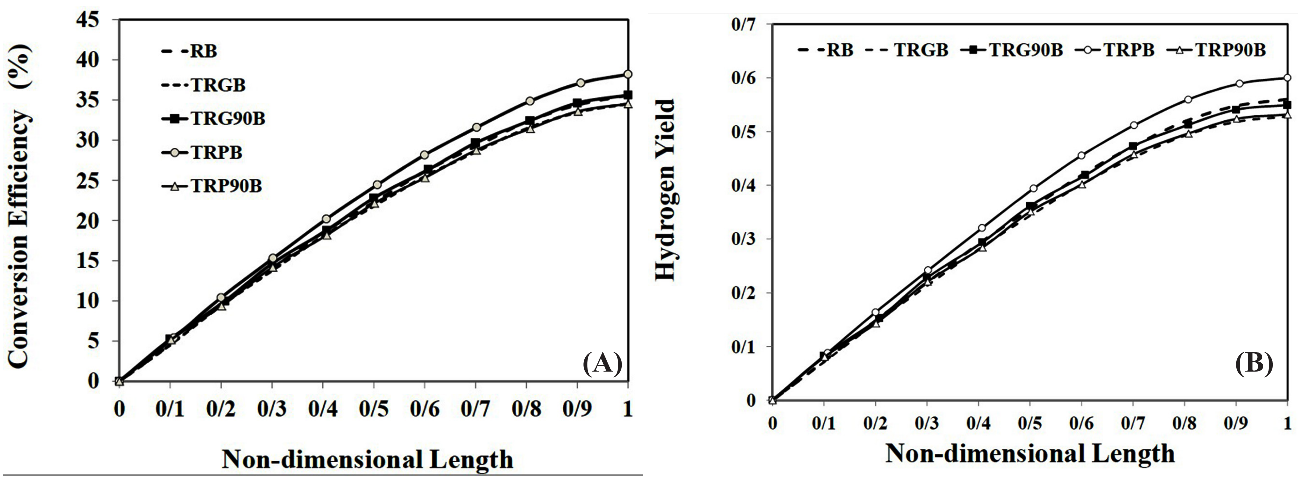

Fig. 8. (A) Methanol conversion efficiency along the channel length for different micro-baffle shapes, (B) Hydrogen Yield along the channel length for different micro-baffle shapes.

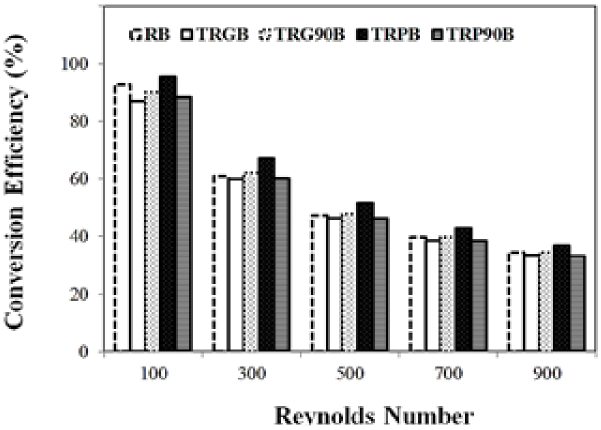

Fig. 9. Methanol conversion efficiency versus Reynolds number for different micro-baffle shapes.

Figure 10 shows the temperature variations of the bulk of fluid (in the SR section) for different micro-baffle shapes along the channel length. It can be seen that the temperature increased at the baffle positions, while the temperature for the TRPB model is higher than that for other models.

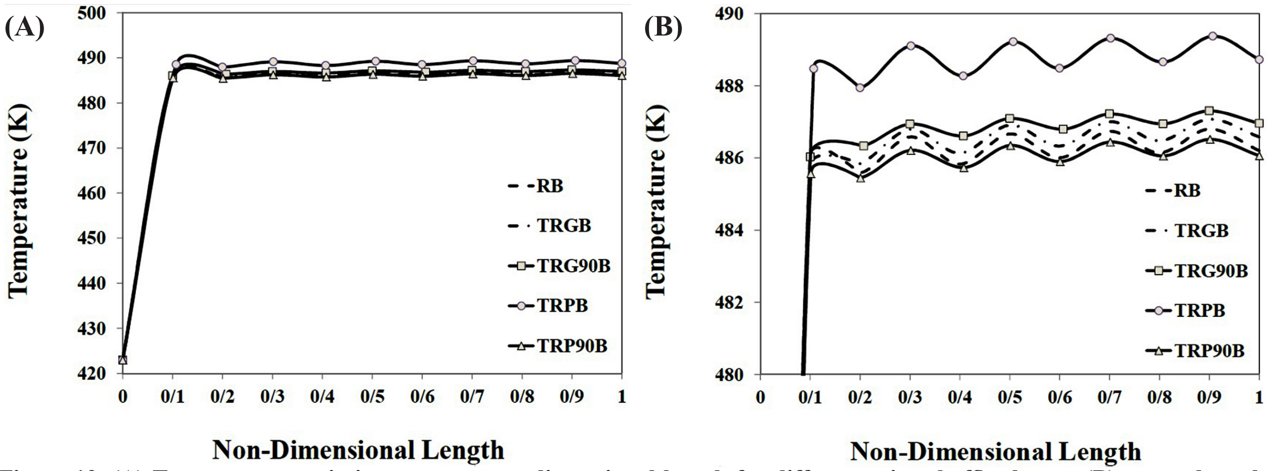

Figure 10. (A) Temperature variations versus non- dimensional length for different micro-baffle shapes, (B) zoomed on the baffle positions (Re=900).

5. Conclusions

Steam reforming (SR) of methanol integrated with oxidation (OX) of methanol was investigated in a micro-channel reactor with different micro-baffle shapes for hydrogen production. An OX reaction occurred in the upper channel, while in lower channel, a SR reaction took place. The two sections were separated by a solid section. The STR model was compared with the RB model to consider the effect of using micro-baffles. In the STR model a straight channel without baffles was used, and rectangular micro-baffles were used in the RB model. Two parameters are defined for this comparison, methanol conversion efficiency and hydrogen yield. Conversion efficiencty in RB models was higher than STR models. It seems the micro-baffles act as static mixers, inducing further mixing and improving the convective heat transfer coefficients which eventually expedite heat transfer, so the conversion efficiency is more than one. Hydrogen yield obtained with the RB model was 17% higher, on average, than that obtained with the straight configuration. Results showed higher temperatures on baffle locations than other positions. Five different micro-baffle shapes were compared, Rectangular, Triangular, Triangular 90, Trapezoidal (TRPB) and Trapezoidal 90 shapes. Methanol conversion efficiency along the channel length and at different Reynolds numbers were studied. Results showed higher conversion efficiency for the TRPB model than for the other models.

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