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Modeling process intensified catalytic plate reactor for synthesis gas production

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HIGHLIGHTS

- Catalytic plate reactor model for co- and counter-flow configuration.
- Simulations using detailed surface reaction mechanism for reforming and oxidation.
- Process intensification studies to replace industrial steam reformer with catalytic plate reactor.

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ABSTRACT

This paper presents numerical study of co- and counter-flow arrangements for catalytic plate reactors (CPR). CH_4 steam reforming coupled with CH_4 oxidation is simulated using detailed surface reaction mechanisms. Effect of inlet velocities to the reforming channel, oxidation channel, and material properties of the plate on the resulting plate temperature and CH_4 conversions is studied. The simulation results agree very well with an industrial scale reformer unit and calculations are further carried out to evaluate the number of CPRs and stacks required to replace and industrial unit.

1. Introduction

Steam reforming (SR) of natural gas widely used for the production of synthesis gas in petrochemical and refinery units takes place in tubular reactors positioned in large gas fired furnaces. The reaction is highly endothermic and the desired reaction temperature is achieved by means of radiation from flue gas and furnace walls. Industrial scale applications generally use Ni as catalyst, however, Rh is also considered for experimental studies. As the process is bulky and has low thermal efficiency, SR at micro- and meso-scales has received increased attention over last decade (Stefanidis et al., 2009; Venkataraman, 2003; Kolb and Hessel, 2004; Stefanidis et al., 2008).

There are several advantages associated with micro-reactor technology. The small length scale makes the process safer, millimeter or sub-millimeter dimension improves heat and mass transfer and minimizes hot spot formation, and lower hold-up volume within micro-reactors reduces the magnitude of any unfortunate events like explosion or chemical release. Finally, for all processes where production rates are limited by transport, micro-reactor technology offers drastic reduction in equipment sizes leading to capital cost savings and enhanced energy efficiency.

Micro-reactors like catalytic plate reactor (CPR) for methane steam reforming reaction coupled with methane combustion reaction is discussed in this paper. There are many studies regarding steam reforming of fuel in CPR, however, most of them are simulation works barring a few.

A CPR essentially is a plate coated with a bed of catalyst material on either side with flow channel over it. A schematic representation of CPR is shown in Fig. 1. Rh or Ni is used as reforming catalyst and Pt is generally used as oxidation catalyst (Stefanidis et al., 2009; Janardhanan et al., 2011). Catalyst loading, plate thickness, plate thermal conductivity, inlet velocities, and inlet temperatures are some of the operational and design parameters that affects the performance of CPR (Zanfir and Gavriilidis, 2003). CPR may be arranged in co-flow or counter-flow configuration. Higher conversion and better utilization of overall heat are possible with counter-flow arrangement (Zanfir and Gavriilidis, 2004). However, the exit temperature can be high in counter-flow configuration compared to

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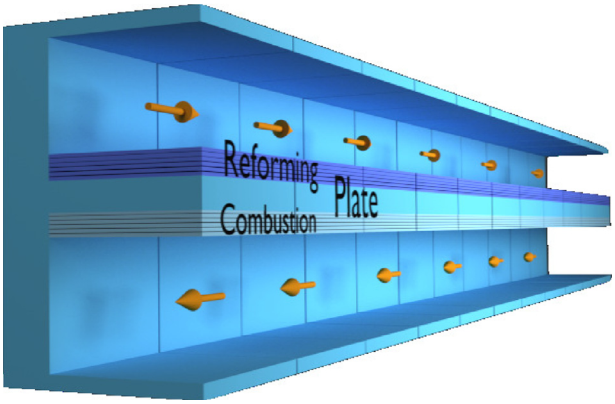

Fig. 1. Schematic representation of counter-flow arrangement.

co-flow configuration and in general co-flow configuration can mitigate hot spot formation (Zanfir and Gavriilidis, 2004). Since the heat requirement for reforming reaction in CPR is met by oxidation reactions occurring in a parallel channel, CPR is operated adiabatically. The presence of insulation not only increases the maximum plate temperature but also causes steep axial temperature gradients, which may be detrimental for the catalyst and mechanical properties of the plate (Stefanidis et al., 2009). Low steam reforming velocities and high-conductivity ceramics or metals are preferred to low conductivity ceramic as plate materials as they smooth out temperature gradients.

Experimental reports on CPR are very scanty, particularly using CH_4. Polman et al. (1999) experimentally achieved 99.98% and 97% conversions of combustion and reforming reactions, respectively, in a co-current plate reactor. Venkataraman (2003) studied combined catalytic methane combustion on Pt and steam methane reforming on Rh in meso-scale catalytic plate reactor. They reported stable operation for several hours without catalyst deactivation as well as high methane conversion (more than 95%), with a residence time of ~70 ms for three channel parallel plate reactor (two combustion channels and one reforming channel).

The present work deals with transient analysis of co- and counter-current configuration of CPR with porous catalyst applied on either side of the plate. The work differs from the above reported ones in various aspects. All the above reported work implements global reaction kinetics for reforming and oxidation, whereas in this work we implemented detailed kinetic models for the surface reactions. The porous media transport is implemented using modified Fick's Law. Counter-current arrangement of CPR is studied in detail. The model is also experimentally validated to obtain the conversions and residence time reported by Venkataraman (2003) for co-flow three channel arrangement.

2. Numerical model

2.1. Channel flow

The flow of gaseous mixture through the top and bottom channels is modeled as plug flow according to (Zhu et al., 2005)

$$\frac{\mathrm{d}(\rho u Y_k)}{\mathrm{d}z} = \frac{w_\mathrm{d}}{A_\mathrm{c}} J_k W_k, \quad k = 1, \ldots, K_g \tag{1}$$

Here w_d is width of catalyst layer and A_c is cross sectional area of channel, J_k is the molar flux of species k at the interface between the flow channel and the porous catalyst bed. Y_k is the mass fraction and W_k is the molecular weight of species k. Since the channel dimensions we consider here are quite small ~ 1 mm, the plug flow assumption is adequate to represent the species transport in the channels. The velocity in the channel is calculated from

$$\frac{\mathrm{d}(\rho u)}{\mathrm{d}z} = \sum_{k=1}^{K_g} \frac{w_\mathrm{d}}{A_\mathrm{c}} J_k W_k, \quad k = 1, \ldots, K_g. \tag{2}$$

The local density in the flow channel is calculated from the ideal gas equation by assuming constant pressure p as

$$\rho = \frac{p\overline{W}}{RT}. \tag{3}$$

The energy balance equation for the flow channels is written as

$$\rho u c_\mathrm{p} \frac{\mathrm{d}T}{\mathrm{d}z} = \frac{4}{D_\mathrm{h}} h (T_\mathrm{p} - T), \tag{4}$$

where T_p is plate temperature, T is fluid temperature, D_h is hydraulic diameter of the flow channel, c_p is the specific heat capacity of the gas mixture, and h is the heat transfer coefficient given by

$$h = \frac{N_\mathrm{u} \lambda}{D_\mathrm{h}}. \tag{5}$$

Here λ is thermal conductivity and the Nusselt number N_u is defined according to the Forzatti correlation (Hayes and Kolaczkowski, 1997)

$$N_\mathrm{u} = 3.657 + 8.827 \left(\frac{1000}{G_z} \right)^{-0.545} \exp\left(-\frac{48.2}{G_z} \right), \tag{6}$$

where G_z is the Graetz number defined as a function of Reynolds number Re and Prandtl number Pr according to

$$G_z = \frac{D_\mathrm{h}}{z} Re\, Pr \tag{7}$$

2.2. Porous media flow

Unlike the governing equations for channel flow, the species transport equations are written in the transient form. Species transport through porous bed is modeled in one dimension, but transverse to the direction of flow through the channel, given by

$$\epsilon \frac{\partial(\rho Y_k)}{\partial t} + \frac{\partial(J_k W_k)}{\partial y} = A_s \dot{s}_k W_k, \quad k = 1, \ldots, K_g, \tag{8}$$

where ϵ is the porosity of the porous bed, A_s is the specific catalyst area available for surface reaction and $\dot{s}_k$ is the heterogeneous production rate of the chemical species k. The species molar flux J_k in the porous bed is evaluated according to

$$J_k = -D_\mathrm{eff} \nabla [X_k] - \frac{B_\mathrm{g}[X_k]}{\mu} \nabla p. \tag{9}$$

The first term on the right hand side represents the flux due to concentration gradients and the second term on the right hand side represents the viscous flux. $[X_k]$ is the concentration of species k. The effective diffusion coefficient D_eff is calculated according to

$$\frac{1}{D_\mathrm{eff}} = \frac{1}{D_\mathrm{km}^\mathrm{e}} + \frac{1}{D_{k,\mathrm{Kn}}^\mathrm{e}}. \tag{10}$$

The effective mixture diffusion coefficient D_km^e is given by

$$D_\mathrm{km}^\mathrm{e} = \frac{\varepsilon}{\tau} D_\mathrm{km}, \tag{11}$$

and the effective Knudsen diffusion coefficient is given by

$$D_{k,\mathrm{Kn}}^\mathrm{e} = \frac{\varepsilon}{\tau} \frac{d_\mathrm{p}}{3} \sqrt{\frac{8RT}{\pi W_k}}. \tag{12}$$

In the above equations ε is the porosity and τ is the tortuosity of the porous catalyst bed. D_km is the mixture diffusion coefficient for the species k calculated according to

$$D_\mathrm{km} = \frac{\sum_{j \neq k}^{K_g} X_j W_j}{\overline{M} \sum_{j \neq k}^{K_g} X_j / D_{jk}}. \tag{13}$$