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A comparative study between operability of fluidized-bed and fixed-bed reactors to produce synthesis gas through tri-reforming

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ABSTRACT

Fluidized-bed reactor has been suggested as a suitable candidate for syngas production due to its special advantages such as elimination of pressure drop problem and bad temperature profile distribution. In this study the fixed-bed and fluidized-bed tri-reformer configurations to produce syngas are modeled heterogeneously based on the conservation mass and energy balances. The obtained data are validated by a pilot plant. Then the operability and performance of considered configurations is compared at steady state condition. This comparison reveals 1.2% and 6% enhancement in the methane conversion and CO_2 consumption in fluidized-bed tri-reformer reactor, respectively, which mainly result from the fact that fluidized-bed reactor presents more effective temperature management and smaller pressure drop in comparison with the fixed-bed reactor. An increase in methane conversion, hydrogen yield and CO_2 consumption as well as a decline in hot spot temperature in the catalytic bed shows the superiority of fluidized-bed tri-reformer reactor for producing synthesis gas.

1. Introduction

Methane reforming is a well-known process that is widely used in producing hydrogen and/or synthesis gas for ammonia, methanol and other chemicals manufacturing. Due to its key role, this process has been studied extensively (Dybkjaer, 1995; Bharadwaj and Schmidt, 1995; Ma and Trimm, 1996; Aasberg-Petersen et al., 2003; Heinzel et al., 2002; Pena et al., 1996; Mleczko and Baerns, 1995). Methanol is a vital multipurpose component. Although this simple chemical can be recovered from a wide variety of resources, it is mainly produced from synthesis gas which is obtained from natural gas in the reformer process (Choudhary and Mamman, 2000; Choudhary and Mondal, 2006; Feng et al., 2009; Ahmed and Gupta, 2009, 2011).

1.1. Process and model

Obtaining a suitable H_2/CO ratio with acceptable methane conversion along with decreasing coke formation is considered as the foremost operational issues in syngas production through natural gas. The suitable H_2/CO varies according to syngas usage intentions. This ratio should be 1.5–2 for the purpose of methanol production (Jiang et al., 2007). In order to improve synthesis gas production process, Song suggested tri-reforming of methane (TRM) (Song, 2001). In this process, steam reforming, CO_2 reforming and partial oxidation of methane take place in one reactor. O_2 and H_2O addition bring about a reduction in coke formation on the catalysts. What is more, a H_2/CO ratio close to 2 is of benefit to synthesis gas production. Tri-reforming includes a combination of reforming reactions and oxidation reaction. Considering the fact that the former reactions are endothermic and the later reaction is exothermic, this combination contributes to a reduction in energy consumption. Additionally, in tri-reforming, CO_2 from industrial effluent gases is consumed which is another merit of this process. Because of the significant advantages of tri-reforming of methane (TRM), it has been investigated extensively in laboratory scale (Song et al., 2002; Hong-tao et al., 2007; Lee et al., 2003a,b; Tomishige, 2004). In recent years, this process has been used at pilot plant and demonstration unit for DME synthesis in Korea (Cho et al., 2009). García-Vargas et al. (2013) studied the effect of the feedstock composition on methane conversion, the H_2/CO molar ratio of the synthesis gas obtained by tri-reforming of methane and the heat released or supplied to the system with a Ni/β–SiC catalyst. Lo Faro et al. (2013) optimized the coupling of a

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biogas-fed solid oxide fuel cell (SOFC) with an external tri-reforming system over ceria supported Ni catalyst (1.75-wt.% Ni/CeO_2). They showed that this process is a promising process for small and medium sized stationary power systems. Karimi-Golpayegani et al. (2005) presented a model for fluidized-bed steam reformer. Arab Aboosadi et al. (2011) applied differential evolution technique to optimize tri-reformer reactor in which syngas was produced for methanol production. In their suggested configuration, methane conversion increased by 3.8% respect to industrial reformers in a single reactor.

1.2. Fluidized-bed reactor

Replacing the fixed-bed reactor with the fluidized-bed reactor is a suitable way of increasing the efficiency of conventional fixed-bed reactor. Fluidized-bed reactor is a good candidate for highly exothermic reactions due to their unparalleled advantages including decent heat management ability, appropriate mixing and excellent temperature control, small pressure drop and diffusion limitations removal (van der Laan and Beenackers, 2000).

1.3. Objectives

The main aim of this work is the assessment of fluidization concept application in the fluidized-bed reactor and its advantages over fixed-bed reactor. In order to validate the mathematical modeling, the obtained results are compared with the plant data. The comparison of the above mentioned configuration is obtained through comparing the methane conversion, the H_2 yield and temperature profile.

2. Process description

2.1. Fixed-bed tri-reformer reactor

In our previous work (Arab Aboosadi et al., 2011), steam methane reformer and auto-thermal reformer were replaced with a single fixed-bed tri-reformer reactor. It was also optimized with the help of DE method. The suggested reactor was packed with NiO–Mg/Ce–ZrO_2/Al_2O_3 catalyst which its size was the same as the catalyst size in steam methane reformer process. A schematic diagram of this reactor is depicted in Fig. 1.

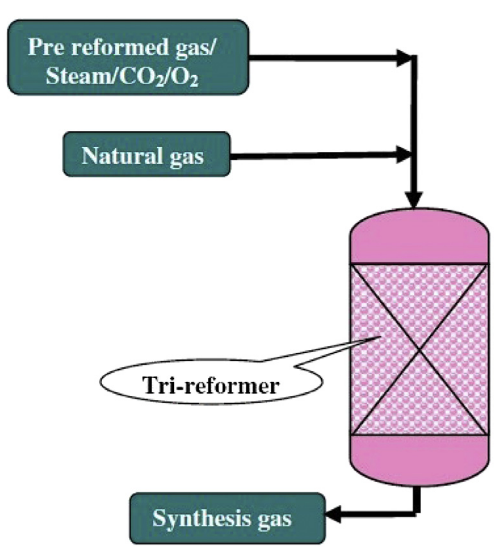

Fig. 1. Schematic diagram of fixed-bed tri-reforming process.

2.2. Fluidized-bed tri-reformer reactor

In this work, a single fluidized-bed tri-reformer reactor is substituted for the single fixed-bed tri-reformer reactor as a superior configuration. This reactor offers high methane conversion and proper H_2/CO ratio in one step. In this configuration, which is illustrated in Fig. 2, a mixture of pre reformed gas and natural gas stream along with a proper ratio of steam, CO_2 (which is obtained from cleaning up the syngas in a CO_2 absorber) and O_2 find their way into a fluidized-bed tri-reformer reactor. Combustion of methane takes place in the initial section of the reactor owing to the high temperature and pressure of reactant gases and provides the required heat for dry and steam reforming which occur in the remaining part of the bed. This brings about a reduction in energy consumption as the heating requirement could be removed or decreased. Therefore, this process not only consumes carbon dioxide, but it also reduces the emission of this gas by decreasing the fuel consumption. As the syngas leaves the reactor, its temperature and pressure reduce in waste heat boiler. In order to extract CO_2 from dewatered syngas stream, it should be compressed and sent to a CO_2 absorber. Table 1 presents the operating condition of both the fluidized-bed and fixed-bed tri-reformer reactors.

3. Mathematical model

In order to model the tri-reforming reactor, a set of equations is considered and a heterogeneous one-dimensional model is presented. The abovementioned set of equations includes: coupled energy balance, mass balance and kinetics equation.

3.1. Reaction network

Mostly, natural gas to synthesis gas reaction is expressed by three main catalytic reactions, include:

- methane steam reforming (SRM):

$$CH_4 + H_2O \leftrightarrow CO + 3H_2 \quad \Delta H^{\circ}_{298} = 206.3\ \mathrm{kJ/mol} \quad (1)$$

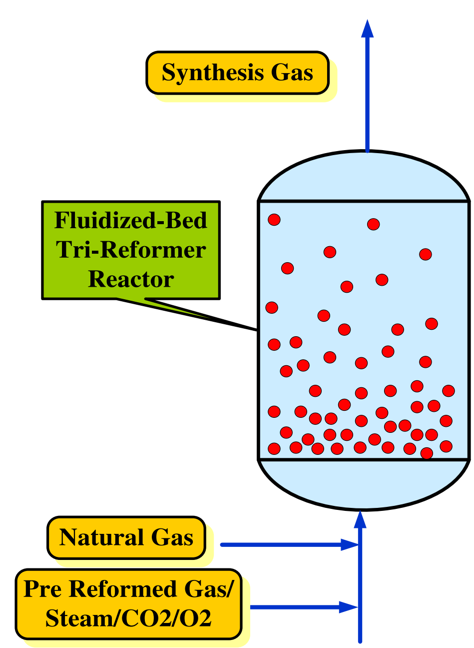

Fig. 2. Schematic diagram of proposed novel fluidized-bed tri-reformer reactor.