



A novel integrated thermally double coupled configuration for methane steam reforming, methane oxidation and dehydrogenation of propane



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ABSTRACT

The goal of this study is the simultaneous production of synthesis gas, hydrogen and propylene in a thermally double coupled steam reformer reactor. This reactor has three concentric tubes where the exothermic reaction of methane oxidation is supposed to occur in the middle tube and the inner and outer tubes are considered to be endothermic sides of steam reforming and propane dehydrogenation, respectively. The motivation is to combine the energy efficient concept of coupling one exothermic reaction with two endothermic reactions, enhancement of synthesis gas production, propylene and hydrogen production and also producing two different H₂/CO ratio streams of syngas. A steady state homogeneous model of fixed bed for three sides predicts the performance of this new configuration. The simulation results are compared with corresponding predictions of the conventional steam reformer. The results prove that synthesis gas production is increased in a thermally double coupled reactor in comparison with conventional steam reforming. In addition, the thermally double coupled reactor reduces the capital and operating costs by reducing the reactor size and consumption of energy.

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1. Introduction

Nowadays, there has been an enormous increase in energy demands due to fast growing of industrial development and population of the world. The shortage of fossil fuels as a main energy sources has brought energy crisis. In order to eliminate fossil fuel dependence, comprehensive research has been carried out on searching alternative energy resources. Natural gas is recognized as one of the cleanest and most abundant fossil fuels utilized in various industrial processes. Different technologies are today available to convert natural gas to future energy carriers such as synthesis gas of which chemical conversion is of great interest. Conversion increases the value of the raw material 3–10 times (Løvik, 2011). Synthesis gas can be an excellent candidate for future energy carrier due to its high potential energy.

1.1. Synthesis gas

Synthesis gas or syngas is a major route from hydrocarbons to many vital chemical products consisting primarily of H₂ and CO. Syngas can be produced from Natural Gas (NG), refinery off-gases, naphtha, heavy hydrocarbons and also from coal. Hydrogen has been considered as a clean and renewable energy carrier to support energy consumption. Hydrogen has characteristics of an environmentally friendly, efficient, safe and sustainable energy source (Heinzel et al., 2002; Lokurlu et al., 2003). The products of hydrogen combustion are water and a tiny amount of NO_x which can be reduced by proper methods. The use of hydrogen in energy sector enhances the security of energy supply and improves economic competitiveness (Muller-Langer et al., 2007). Hydrogen has the best energy-to-weight ratio of any fuel. A fuel which contains higher proportion of hydrogen provides more energy (Sun et al., 2012). Therefore, pure hydrogen would be the leading fuel, which can satisfy the increasing demand for many processes such as: methanol, electricity, ammonia, aniline, oil refining, fuel cell, vehicle engines, power plants, etc. (Itoh et al., 2008; Brown, 2001). Several processes have been widely investigated as primary processes for

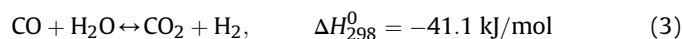
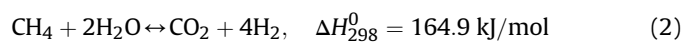
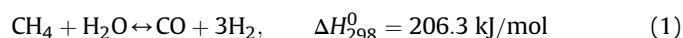
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converting hydrocarbons or alcohols into hydrogen, including steam reforming (Lee and Park, 1998; Ko et al., 1995; Rahimpour and Alizadehhesari, 2009; Rahimpour et al., 2010; Seo et al., 2008a,b; Yu et al., 2009), partial oxidation (Wang et al., 2009; Kim et al., 2004; Gao et al., 2008), auto-thermal reforming (Takeguchi et al., 2003; Youn et al., 2008a,b), and CO₂ reforming (Luna and Iriarte, 2008; Nandini et al., 2006). It must be noted that hydrogen production must be low in CO₂ emissions and other pollutants. Among all mentioned technologies, catalytic steam reforming of natural gas as a feasible process is widely used for hydrogen production, 80–85% of the world's total hydrogen production is provided by this method (Simpson and Lutz, 2007).

1.2. Steam reforming

Steam reforming technology is the most commercial method for synthesis gas (CO, H₂) production, the hydrogen cost is less than hydrogen produced by using renewable energy sources or from gasification of solid fossil fuel (Rostrup-Nielsen, 1993; Tugnoli et al., 2008). Steam reforming may involve several catalytic steps: desulfurization of the fuel, steam reforming of methane, a water gas shift reactor and purification of hydrogen using the PSA unit. This method is suitable for light hydrocarbons such as natural gas (mainly CH₄), naphtha, and liquefied petroleum gas (Ryden et al., 2006). Three main catalytic reactions involved in steam reformer reactor are steam reforming of methane (SRM) and the water–gas shift reaction.

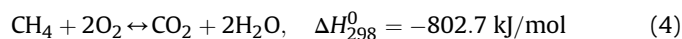


Conventional steam reformer consists of vertical tubes packed with Ni-based catalyst located inside the huge furnace. The required heat for endothermic reforming reaction is provided by direct combustion of fuel in the furnace. Therefore, the reformer tubes are under very high thermal stress (Brown, 2001). In order to solve this issue, the recuperative coupled reactor has been suggested in which an appropriate exothermic reaction is used as a heat source. Hunter and McGuire (1980) were pioneers in coupling endothermic and exothermic reactions without direct heat transfer. A review on the process intensification for methane steam reforming in a thermally coupled membrane separation technology was studied by Bhat and Sadhukhan (2009). Patel and Sunol (2007) suggested a thermally coupled membrane reactor that is composed of the three channels for methane steam reforming. A numerical model for natural gas, steam reforming and coupling with a furnace was developed by Ventura and Azevedo (2010). In an interesting idea, Ryden and Lyngfelt (2006) studied steam methane reforming coupled with a chemical looping combustion reactor in order to enhance H₂ production with CO₂ capture.

1.3. Catalytic oxidation of methane

Today, steam reforming and partial oxidation are the main technologies to produce synthesis gas from natural gas. However, there are two main operational issues in producing synthesis gas by using these methods; Firstly, obtaining the suited H₂/CO ratio and the second is reaching to high methane conversion and avoiding coke deposition (Jiang et al., 2007). For the characteristics of undesired H₂/CO ratio of steam methane reforming for production of methanol and Fischer–Tropsch Synthesis and also high energy cost

due to its endothermic reaction, alternative routes have been investigated. In this way, many other methods have been studied, including: carbon dioxide reforming (Mark et al., 1997; Bradford and Vannice, 1999), the partial methane oxidation (Mallens et al., 1997; Nakagawa et al., 1998), combinations of these reaction routes (Choudhary et al., 1995, 1998; Ruckenstein and Hu, 1998) and Tri reforming (combination of CO₂ reforming, steam reforming, and partial oxidation) (Song, 2001). An effective option is to use catalytic oxidation of methane, which combines steam methane reforming and total combustion of methane in a single reactor. Four catalytic reactions of (1)–(4) are used to describe the oxidation of methane:



This reaction is exothermic, so that no need for an external heat supply and leads to significant reduction of the total costs. In addition, the heat of reaction can be a source of energy supply for one or two suitable endothermic reactions like steam reforming and propane dehydrogenation in a thermally coupled reactor.

1.4. Dehydrogenation of propane

In today's scenario, due to rapidly growing demand of alkenes, industrial procedure of corresponding alkanes dehydrogenation has received much attention. Alkenes, especially ethylene and propylene, are indispensable raw materials for numerous petrochemical products (Thapliyal and Deo, 2003). Propylene has wide application as the major feedstock for the production of diverse products, ranging from solvents to plastics like (Sahebdehfar et al., 2012; Budavari, 1996):

- Petrochemicals such as plastic polypropylene, acrolein and acrylic acid.
- Films, packaging, caps and closures.
- Isopropanol (Propan-2-ol), acrylonitrile, propylene oxide (epoxy propane) and epichlorohydrin.

Moreover, the propylene market reached the average growth rate of 5–6% per year (Tullo, 2003).

Propene is basically produced from fossil fuels, petroleum and natural gas via steam cracking and direct dehydrogenation (Warren and Oyama, 1996; Vitry et al., 2004). However, propane dehydrogenation is the economical and shortest route to propylene (Nawaz and Wei, 2011). The dehydrogenation reaction is a highly endothermic and equilibrium controlled; therefore, acceptable equilibrium conversion and reaction rate achieved with higher temperatures and lower pressure (Chin et al., 2011).

1.5. Thermally double coupled reactors

Recently, coupling exothermic and endothermic reactions are more interested in order to improve the thermal efficiency of the process and consequently enhances the production. This type of reactors aims to use energy released by exothermic reaction for proceeding endothermic reaction. In general, the coupled reactors exist into three main classes: direct coupling, recuperative coupling and finally regenerative coupling. At present, recuperative coupling has attracted the most attention of many researchers (Song et al., 2003). Coupling one endothermic and one endothermic reaction are more usual in this field. A double integrated reactor for dimethyl ether synthesis and H₂ production from the cyclohexane dehydrogenation has been suggested and mathematical model by Vakili et al. (2011). Rahimpour et al. (2011) also studied the methanol synthesis and the dehydrogenation of cyclohexane to benzene

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