



## Review

## Recent advances in dry reforming of methane over Ni-based catalysts

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## ABSTRACT

A steady increase in atmospheric carbon dioxide (CO<sub>2</sub>) and methane concentrations in recent decades has sparked interest among researchers around the globe to find quick solutions to this problem. One viable option is a utilization of CO<sub>2</sub> with methane to produce syngas via catalytic reforming. In this paper, a comprehensive review has been conducted on the role and performance of Ni-based catalysts in the CO<sub>2</sub> reforming of methane (sometimes called dry reforming of methane, DRM). Coke-resistance is the key ingredient in good catalyst formulation; it is, therefore, paramount in a choice of catalyst supports, promoters, and reaction conditions. Catalyst supports that have a strong metal-support interaction created during the catalyst preparation exhibit highest stability, high thermal resistance and high coke resistance. In addition, the outlook of the Ni-based catalysts has been proposed to provide researchers with critical information related to the future direction of Ni-based catalysts in industrial settings. Among others, it has been a great interest among researchers to synthesize catalyst supports from cellulosic materials (plant-based materials). The unique properties of the cellulose which are a well-defined structure and superior mechanical strength could enhance the catalytic activity in the DRM reaction.

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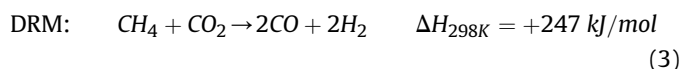
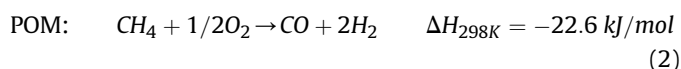
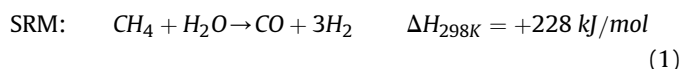
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## 1. Introduction

Over the past decade, there has been an increase in energy consumption, mainly due to a rapid growth in human population (Li, 2005). This growing demand for energy has shifted the energy scenario over the years by industrialization (Tanksale et al., 2010). Moreover, energy demand is expected to keep increasing in the future despite the current low oil price. At present, the dependence on fossil fuels which consist of oil, natural gas, and coal to meet energy demand have created environmental issues by the generation of anthropogenic greenhouse gases. Methane and CO<sub>2</sub> are the most abundant greenhouse gases and are the main contributors to the recent climate-change issues (Noor et al., 2013). Even though the concentration of methane in the atmosphere is lower compared to CO<sub>2</sub> (Talyan et al., 2007), surprisingly it has caused about 20% of the overall global warming (Wuebbles and Hayhoe, 2002). Traditionally, methane is produced from two sources; first, it comes from natural sources such as termites, grasslands, wildfires, lakes and wetlands and second, from human activities such as coal mining, landfills, oil and gas processing and agricultural activities (Yusuf et al., 2012). According to the U.S. Environmental Protection Agency (EPA) (Agency, 2011), the production of methane from landfill contributes to about one-third of all emitted methane in the US alone in which, landfill gas consists of 40–45% of methane and 55–60% of CO<sub>2</sub> by volume (Raco et al., 2010). Apart from that, methane is also a major component of natural gas but most natural gas reservoirs are located far from industrial areas and often produced offshore, and thus, the limitation in technology and cost for transporting this valuable natural gas from offshore to potential market has led to the flaring of a large volume of natural gas globally (Lunsford, 2000). These actions resulted in the wastage of an important hydrocarbon source and contributed to global warming by releasing a greenhouse gas to the atmosphere (Elvidge et al., 2009). Due to the pressure of fighting against the global climate change and ensuring the continuous energy sources, carbon dioxide capture and storage (CCS) was introduced around the world with the objective to minimize the carbon dioxide emissions (Yang and Wang, 2015). Moreover, to reduce the substantial dependency on crude oil and its undesirable influence on the atmosphere, renewable energy is needed immediately for substituting petroleum-based resources (Fayaz et al., 2016).

In order to reduce the amount of methane and CO<sub>2</sub> in the atmosphere, extensive research has been conducted to find effective ways to convert methane and CO<sub>2</sub> into other valuable products. The most common option is the conversion of CO<sub>2</sub> and methane to syngas owing to a low cost and relatively established technology (Bahari et al., 2016). It is an important process to transform the hydrocarbons, usually in the chemical industries for the production of syngas (Alirezaei et al., 2016). Syngas is considered a building block that can be used as reactants for other applications such as Fischer-Tropsch (F-T) oil, methanol, and other valuable liquid fuels and chemicals (Penã et al., 1996). Reforming is the most common method used in industries to produce syngas, via one of the three reforming processes, via steam reforming of methane (SRM), partial oxidation of methane (POM) and dry reforming methane (DRM) (Asencios and Assaf, 2013). SRM is the conventional technology for production of hydrogen from hydrocarbon fuels due to the highest

hydrogen yield compared to the other two methods (Palma et al., 2016). Approximately 75% of hydrogen produced is derived from SRM process (Fan et al., 2016). The differences between these techniques are based on the oxidant used, the kinetics and energetics of the reaction, and the ratio of the syngas produced (H<sub>2</sub>/CO). The details of the main reactions for reforming processes are summarized as followings:



From Rxn. (1), SRM reaction produces a higher H<sub>2</sub>/CO ratio which is 3:1 (Gangadharan et al., 2012) compared to the ratio required for F-T synthesis which is 2:1 (Oyama et al., 2012). SRM requires intensive energy input due to the endothermic nature and caused it is very expensive (Nieva et al., 2014). In addition, a higher H<sub>2</sub>O/CH<sub>4</sub> ratio is required to attain a higher yield of H<sub>2</sub> which makes SRM process energetically unfavorable and accelerates catalysts deactivation (Carvalho et al., 2009). Moreover, SRM faces corrosion issues and requires a desulphurization unit (Djinović et al., 2012). In the case of POM, this process is suitable for the production of heavier hydrocarbons and naphtha (Larimi and Alavi, 2012). Typically, the POM process has very short residence time, high conversion rates and high selectivity (Ruckenstein and Hang Hu, 1999). However, the exothermic nature of the reaction causes the induction of hot spots on the catalyst and makes the operation difficult to control (Asencios and Assaf, 2013). Besides, POM requires a cryogenic unit to separate oxygen from the air (Djinović et al., 2012).

Of all other technologies, DRM is the most promising one as it utilizes two abundant greenhouse gases (CO<sub>2</sub> and methane) to produce syngas that is important for industries, and at the same time can reduce the net emission of greenhouse gases to the environment (Selvarajah et al., 2016). In addition, the DRM process is also cheaper than other methods since it eliminates the complicated gas separation of end products (San-José-Alonso et al., 2009). DRM produces a H<sub>2</sub>/CO ratio of unity that can be used for the synthesis of oxygenated chemicals (Wurzel et al., 2000) and higher hydrocarbons for F-T synthesis (Nieva et al., 2014). Moreover, DRM can be extended to biogas (CO<sub>2</sub>, CO and CH<sub>4</sub>) as a feedstock to produce clean and environmentally friendly fuels (Xu et al., 2009). Besides that, syngas from DRM is considered as solar or nuclear energy storage (Fraenkel et al., 1986). Table 1 shows the comparison between three processes in the CO<sub>2</sub> reforming of methane.

The use of catalysts in DRM reaction is important to maximize the production of syngas as it helps to alter and enhance the rate of reaction without being used up in the process. Catalyst works by providing an alternative mechanism that lowers the activation energy resulted in less energy required to reach the transition state. Even though DRM requires high temperature to operate due to its endothermic nature, the presence of catalysts could lower the temperature of the reaction significantly.

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