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Dry reforming of methane: Influence of process parameters—A review

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ABSTRACT

This review will explore the influences of the active metal, support, promoter, preparation methods, calcination temperature, reducing environment, particle size and reactor choice on catalytic activity and carbon deposition for the dry reforming of methane. Bimetallic (Ni–Pt, Ni–Rh, Ni–Ce, Ni–Mo, Ni–Co) and monometallic (Ni) catalysts are preferred for dry reforming compared to noble metals (Rh, Ru and Pt) due to their low-cost. Investigation of support materials indicated that ceria–zirconia mixtures, ZrO₂ with alkali metals (Mg²⁺, Ca²⁺, Y²⁺) addition, MgO, SBA-15, ZSM-5, CeO₂, BaTiO₃ and Ca_{0.8}Sr_{0.2}TiO₃ showed improved catalytic activities and decreased carbon deposition. The modifying effects of cerium (Ce), magnesium (Mg) and yttrium (Y) were significant for dry reforming of methane. MgO, CeO₂ and La₂O₃ promoters for metal catalysts supported on mesoporous materials had the highest catalyst stability among all the other promoters. Preparation methods played an important role in the synthesis of smaller particle size and higher dispersion of active metals. Calcination temperature and treatment duration imparted significant changes to the morphology of catalysts as evident by XRD, TPR and XPS. Catalyst reduction in different environments (H₂, He, H₂/He, O₂/He, H₂–N₂ and CH₄/O₂) indicated that probably the mixture of reducing agents will lead to enhanced catalytic activities. Smaller particle size (< 15 nm) had a significant influence on the suppression of carbon deposition and catalytic activity. Fluidized bed reactor exhibited the highest activity and stability, lower carbon deposition and higher conversion compared to a fixed-bed reactor. Moreover, membrane reactor, solar reactor, high-pressure reactor and microreactor were also investigated with specific features such as: pure H₂ production, detailed reaction information with enhanced safety, higher pressure applications and dry reforming reaction with/without catalyst under sunlight. The study of parameters would improve the understanding of various preparation and reaction conditions leading to various catalytic activities.

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1. Introduction

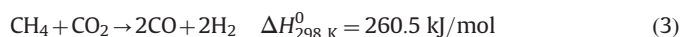
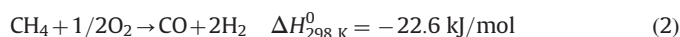
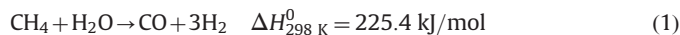
Rapid increase in population and high-energy consumption has been forecasted over the years [1]. Growing human demands have shifted the energy scenario over the years by industrialization [2] and energy demand will increase 57% from 2004 to 2030 [3]. The dependence on fossil fuels to meet energy demand has created environmental issues by the production of greenhouse gases (GHG) [4]. Methane and carbon dioxide constitute a major part of GHG and have the key contributions in climate-change [5], forecasted in terms of greater incidence and magnitude of hurricanes, floods, and droughts [6], affecting productivity, natural ecosystems, agriculture, rangelands, forestry and society [7,8]. An increase of 0.8 °C in global surface temperature occurred in the 20th century and further increase (1.4–5.8 °C) has been anticipated in twenty-first century [6,8]. The concentrations of methane and carbon dioxide in 2011 were 1.813 and 390.5 ppm, respectively, based on the assessment of the World Meteorological Organization (WMO) [9]. Even though the concentration of methane in the atmosphere was lower than carbon dioxide [10,11], it kicked in about 20% of overall global warming [12,13]. Two sources of methane emission were identified, firstly: from naturally occurring activities such as: termites, grasslands, coal beds, lakes, wetlands and wildfires and secondly: from anthropogenic activities such as: landfills, oil and gas processing, wastewater treatment plants, coal mining, rice production, cattle ranching and agricultural activities [14]. Methane was estimated to be 6875 million metric tons CO₂ equivalent (MMT CO₂e) from all anthropogenic sources in 2010 [15].

Methane is a major component of natural gas (NG) and also contains low balances of other hydrocarbons comprised on ethane (C₂H₆), propane (C₃H₈), and butane (C₄H₁₀). NG also contains inert diluents such as molecular nitrogen (N₂) and carbon dioxide (CO₂) [16]. The majority of NG reservoirs are located far from industrial complexes and often produced offshore. The transportation of NG to potential market by pipelines may not be available and liquefaction for shipping purpose by an ocean-going vessel is expensive [17]. In 2011, large volume of NG (140 billion cubic meters, (BCM)) has been flared globally and the flaring data for the top 20 nations have been shown in Fig. 1 in which Russia has the highest share in the global flaring data [18]. Two disadvantages occurred from flaring, first: the wastage of an important hydrocarbon source worth billions and second: global warming by the production of GHG [19].

The population growth has resulted in the increased rate of waste generation and improper handling of waste created environmental issues [5]. In 2012, a study reported that currently there are about 3 billion urban resident generating 1.2 kg per person per day (1.3 billion tonnes per year) and it is estimated that in 2025 it will be likely to rise up to 4.3 billion residents generating 1.42 kg per person per day which will produce 2.2 billion tonnes per year [20]. Moreover, for developed countries, the rate of the increase was estimated to be 3.2–4.5% and for developing nations around 2–3% [21]. The disposal of MSW in the landfills lead to the generation of methane by anaerobic degradation of the organic content in the waste and biogas has been expected at different rates by various management methods employed [22]. Production of methane from landfill, constitute a major portion of all emitted methane in the US (about one third) [23]. Landfill gas consists of 40–45% methane and 55–60% carbon dioxide by volume [24] while some predicted 50–60% and 30–40% by volume of methane and carbon dioxide [25], respectively.

Keeping in view the above scenario, extensive research is being performed to convert methane into liquid fuels or higher hydrocarbons [26]. The researchers were investigating the production of methanol, formaldehyde, propanol, benzene and other aromatics by direct oxidative conversion of methane. Unfortunately, all the

forementioned processes have low yields or are unfeasible on an industrial-scale [27]. Various technologies are available to produce synthesis gas (syn-gas) from NG [28], as syn-gas is a building block for valuable liquid fuels and chemicals such as Fischer–Tropsch oil, methanol and dimethyl ether [17,29,30]. The three processes that draw industrial attention are steam reforming of methane (1), partial oxidation of methane with oxygen or air (2) and dry reforming of methane with carbon dioxide (3) [31].



Reforming of methane through steam or partial oxidation of methane with oxygen or air are well-established technologies with the advantages and disadvantages [32,33]. Steam reforming of methane produces a higher ratio of syn-gas (H₂/CO=3) [34] compared to that required for Fischer–Tropsch or methanol synthesis (H₂/CO=2) [35,36]. The process is energy intensive due to the endothermic nature and requires high investments of capital [37]. A higher H₂O/CH₄ ratio is required to produce higher yields of hydrogen, which makes steam reforming of methane energetically unfavorable leading to the deactivation of the catalyst [38]. Moreover, steam reforming faces corrosion issues and requires a desulphurization unit [32,33]. Partial oxidation of methane is suitable for the production of heavier hydrocarbons and naphtha [32,39]. The advantages of this process are high conversion rates, high selectivity and very short residence time [40]. The exothermic nature of reaction has certain drawbacks that it induces hot spots on catalyst arising from poor heat removal rate and makes operation difficult to control [31,33]. Desulphurization unit is not required in the partial oxidation of methane [33], but a cryogenic unit is necessary for the separation of oxygen from air [32].

Dry reforming of methane offers valuable environmental benefits: biogas utilization [41,42], removal of GHG (methane and carbon dioxide) and conversion of NG with a high carbon dioxide content to valuable syn-gas [17,26]. Dry reforming of methane yielded a lower syn-gas ratio (H₂/CO=1), which is suitable for the synthesis of oxygenated chemicals [43] and hydrocarbons from Fischer–Tropsch synthesis [35]. Syn-gas from dry reforming has also been considered for storage of solar or nuclear energy [44–46] through the chemical energy transmission system (CETS). Solar energy can convert feed gases (methane and carbon dioxide) to syn-gas that can be exported to places where energy sources are scarce. The energy stored in syn-gas is liberated by the backward reaction and utilized as an energy source [46].

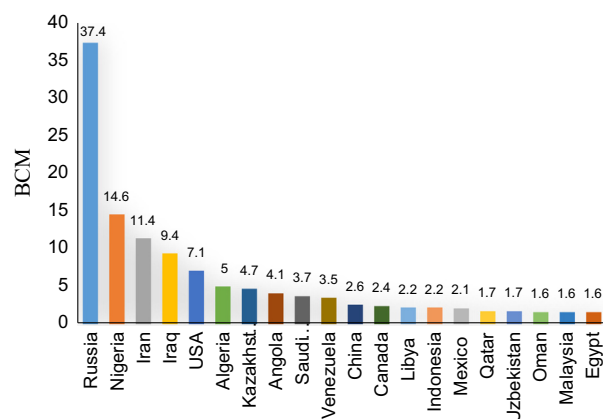


Fig. 1. Global natural gas flaring data [18].

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