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Methane decomposition over iron catalyst for hydrogen production

Ahmed A. Ibrahim, Anis H. Fakeeha, Ahmed S. Al-Fatesh*,
Ahmed E. Abasaed, Wasim U. Khan

Chemical Engineering Department, College of Engineering, King Saud University, P.O. Box 800, Riyadh 11421, Saudi Arabia

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ABSTRACT

Methane is the most stable hydrocarbon and the highest component of natural gas. The occurrence of vast resources of natural gas promotes the catalytic decomposition of methane to produce hydrogen and valuable carbon. Hydrogen is an excellent energy carrier and can play a very significant role in energy systems. The present study explored the decomposition of methane by iron catalyst to produce hydrogen and carbon. The catalysts were calcined at 700 °C and reduced with pure H₂ at 650 °C. The reaction was carried out at 700 °C. The study involved the use of different iron loadings (15–100%) supported on alumina catalysts obtained by co-precipitation technique. The yield of hydrogen production was investigated. Spent and fresh catalysts were characterized by BET, XRD, H₂-TPR, TGA, EDS and HRTEM. The catalyst characterization revealed the formation of multi-walled nanotubes from alumina supported iron catalysts. Alternatively, time on stream tests of the supported catalyst for about 4 h at 700 °C showed the relative profiles of hydrogen production and hydrogen yield increased as the % loading of Fe was increased. The maximum H₂ yield of 77.2% was obtained using 60% Fe/Al₂O₃ catalyst. Higher Fe loadings decreased the surface area of the catalyst.

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Introduction

Methane, chief component of natural gas, is the most used hydrogen source. Hydrogen production from methane is usually preformed with following reactions by steam reforming or partial oxidation respectively [1,2].



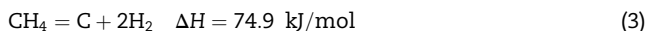
Though, the reforming of natural gas has industrially been used for hydrogen production [3,4], the reaction is highly endothermic and an enormous energy input is needed and furthermore substantial quantity of carbon oxides emissions takes place [5]. Particularly, in partial oxidation the generated carbon monoxide is commonly eliminated by further

* Corresponding author. Tel.: +966 11 4676859; fax: +966 11 4678770.

E-mail addresses: aidid@ksu.edu.sa (A.A. Ibrahim), anishf@ksu.edu.sa (A.H. Fakeeha), aalfatesh@ksu.edu.sa (A.S. Al-Fatesh), aba-saeed@ksu.edu.sa (A.E. Abasaed), wasimkhan49@gmail.com (W.U. Khan).
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oxidation to CO₂ - a greenhouse gas. Therefore, thermo-catalytic cracking of methane becomes an attractive method to produce pure hydrogen.



Thermodynamically, the methane cracking reaction given by Eq. (3) can proceed at higher temperatures above 1000 °C [6]. The favored temperatures for the general chemical engineering processes are well below this temperature. Therefore, it is necessary to decompose methane at lower temperatures by using catalysts based on transitional metals such Fe, Ni and Co [7–14]. The use of metal-based catalysts produces carbon of high quality as an added value that lowers the high cost of the catalyst. Several investigators have shown intense interest to carbon materials obtained by cracking of various hydrocarbons over metal supported catalysts due to their unique structure built-up by nanotubes [15–18]. Nickel based catalysts are employed for decomposition of methane at moderate temperatures. Venugopal et al. examined methane decomposition at 600 °C and 30 wt.% Ni/SiO₂ [19]. They obtained methane conversion of 32%. They also highlighted the influence of preparation conditions on H₂ production rates. Similarly, Suelves et al. found a methane conversion of 67% and hydrogen concentration of 80% at 700 °C by using Ni-based catalyst [20]. Alternatively, Shah et al. studied catalysts of iron coupled with other metals such Pd, Mo and Ni supported on alumina [21]. The decomposing methane at 700 °C - 800 °C revealed low methane conversion even though the hydrogen concentration was high. On the other hand, Torres et al. investigated hydrogen production by catalytic decomposition of methane using Fe-based catalyst in a fluidized bed reactor [22]. They found that the operating conditions i.e. temperature and space velocity had strong effect on the catalyst performance. In addition methane conversions in the range of 25–40%, hydrogen production of 1.55 L/gcat/h and carbon formation of 2.28 g/gcat were obtained at 700 °C during a 4 h reaction. Carbon was deposited in the form of nanofilaments. The study of Jin et al. highlighted that the presence of Fe increased the catalytic activity of methane decomposition by the affecting the surface area and pore volume [23]. Avdeeva et al. reported the catalytic activity of iron containing catalysts using a vibrating flow quartz reactor at lower temperatures [24]. They established that catalyst preparation method and composition influenced the properties of the catalyst. The best performance was found with co-precipitated Fe–Co–Al₂O₃ catalysts. The methane decomposition over iron-containing catalysts was investigated at temperatures 800–900 °C with the purpose of deriving hydrogen [25]. However, the developed iron catalysts were soon deactivated at carbon yield of about 30%. During this process iron carbide was produced, which cannot find practical application and the pre-treatment atmosphere had a significant effect on the catalytic activity. Numerous researchers tested methane decomposition over Fe-based catalysts at temperatures lower than 800 °C [26,27]. Ermakova et al. examined the effects of using various supports such as SiO₂, TiO₂, Al₂O₃, and ZrO₂ into iron oxides on the catalytic activity for methane decomposition [28].

Therefore, a more detailed study on methane decomposition over Fe-based catalysts at 700 °C is essential, especially on the yield of hydrogen and the structure of carbon formed and iron species during the methane decomposition. Nowadays, catalytic methane decomposition focuses more on the development of iron-based catalysts process since they accomplish better results than Ni catalysts [19]. Therefore, it is possible to obtain higher methane conversions with Fe catalysts than with Ni catalysts as they can withstand temperatures of 700 °C and above, resulting in a positive shift in the equilibrium. On the other hand, higher temperature accelerates the deactivation of catalyst. In this paper, the performance of hydrogen production from catalytic decomposition of methane was studied. The effect of various loadings of Fe based catalyst promoted texturally with alumina was investigated at moderate temperature of 700 °C. Furthermore, a detailed characterization of the iron catalysts has been carried out in order to check and correlate their main properties with their activity.

Experimental

Catalyst preparation

Iron catalysts were prepared by the co-precipitation technique. Stoichiometric amounts of the precursors i.e., Fe(NO₃)₂·9H₂O and Al(NO₃)₃·6H₂O were dissolved in distilled water under constant stirring at 60 °C and 10 vol.% NH₃ solution was added until the pH reached 9. The obtained precipitates were filtered, washed with deionized water and acetone, dried over night at 120 °C and finally calcined at 700 °C for 3 h.

Iron catalyst characterization

The morphology and the composition of the catalysts were investigated using a scanning electron microscope (SEM), JEOL JSM-6360A, coupled to a Si/Li detector for energy-dispersive spectroscopy.

Textural properties and BET surface area of the catalysts were measured by N₂ adsorption/desorption at –196 °C, by using a Micromeritics Tristar II 3020 instrument. Prior to analysis, the samples were degassed at 200 °C for 4 h. The surface area was calculated by applying the BET method to the respective N₂ adsorption isotherms.

HRTEM measurements of spent samples were performed on a JEOL JEM-2100F transmission electron microscope operated at 200 kV accelerating voltage to analyze the morphology of the deposited carbon.

X-ray diffraction (XRD) patterns of the samples were measured by a Rigaku (Miniflex) diffractometer with a Cu K α radiation operated at 40 kV and 40 mA. The analysis was performed from 10° to 85° with a step size of 0.02°/min.

Hydrogen temperature-programmed reduction (H₂-TPR) was performed in an automatic chemisorption (Micromeritics Auto Chem II 2920) apparatus equipped with a thermal conductivity detector. About 0.05 g sample was preheated to 300 °C to remove the water for 60 min before cooling down to 120 °C under the N₂ of 60 ml/min, and then heated to 1000 °C with the heating rate of 10 °C/min under pure H₂ atmosphere.

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