



A review on methane transformation to hydrogen and nanocarbon: Relevance of catalyst characteristics and experimental parameters on yield



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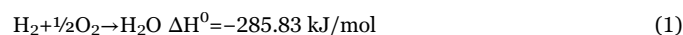
ABSTRACT

Co-synthesis of hydrogen and nanocarbon via methane cracking is a single step technique which meets ever growing need of greenhouse gas (GHG) free energy. Additionally, as produced multifunctional nano-carbon that have a variety of technological applications reduces the process cost. This review is intended to provide a critical and wide-ranging assessment of impact of metal catalyst characteristics and methane decomposing parameters on hydrogen and nanocarbon yield, as well as the alteration of characteristic properties of as-produced nanocarbon. The major factors influencing thermocatalytic decomposition of methane (TCD) includes catalyst support, porosity, surface area, particle size, metal loading, calcination temperature, feed flow rate, partial pressure, and reaction temperature. Literature survey emphasizes that higher temperature and partial pressure together with lower feed flow is the reliable experimental condition to yield high purity hydrogen. Furthermore, initial catalytic activity resembles to the chemical structure of the catalyst and long term activity corresponds to the physical characteristics of catalyst. The structural features of as-produced nanocarbon have inevitable association with catalytic characteristics, such as textural supporters, particle size and material dispersion by physical interactions or chemical interaction. The interaction of metal and support results in modification of electronic properties of metal particles and subsequently influence their catalytic characteristics. In addition to investigation of one-factor-at-a-time experiments, the latest studies with Design of Experiment are also thoroughly reviewed, which analyze the influence of each process variables and their interactions simultaneously. The manuscript, then, extended to the microscopic level understandings on TCD for synthesis of nanocarbon and hydrogen via computational study in the finishing section.

1. Introduction

Hydrogen and nanocarbon are the two most emerging research topics in the field of environmentally benign energy and material science, respectively. These two treasured products can be simultaneously produced by methane transformation in a single step chemical approach. There has been an intense research effort on this topic in recent years. Hydrogen, the most abundant element in the known universe and major constituent of earth, is a substantial raw material in the chemical and petroleum industries. Hydrogen is supposed to play a pertinent role as an energy vector in the near future mainly because of two reasons [1]. Firstly, renewable raw materials like water, biomass, or biogas are the major resources of hydrogen. Secondly, water is the only by-product during its combustion and oxidation as shown in Eq. (1) [2,3]. H₂-O₂ fuel cell combines hydrogen and oxygen to produce electricity, heat, and water. Hence, automobile industries, science

laboratories, and governments have focused greater attention toward hydrogen as a possible alternative fuel, in order to simplify both widespread production and distribution.



Despite its simplicity and abundance, hydrogen is not very common on the Earth as independent gas molecule, but existing mainly as compounds of oxygen and carbon, such as water (in the form of liquid, ice, or hydrated minerals), hydrocarbons, or biomass. The keen tendency of hydrogen to form compounds under atmospheric condition reflects its higher reactivity. While, the higher hydrogen density at high altitudes of Earth's atmosphere may be because of the solar wind, as well as the hydrogen that has diffused from below. Hence, hydrogen moves very fast with a greater mean speed than the escape velocity from the Earth as of its small size and light weight. The reaction of hydrogen with oxygen to form water is exothermic in nature as shown

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Table 1
Physical and chemical properties of hydrogen and methane [6].

Properties	H ₂	CH ₄
Molecular weight (g/mol)	2.016	16.04
Mass density (kg/N A m ³) ^a	0.09	0.72
Mass density of liquid H ₂ (kg/N Am ³) ^b	70.9	N/A
Boiling point (K)	20.2	111.6
HHV (MJ/kg) ^c	142.0	55.5
LHV (MJ/kg) ^d	120.0	50.0
Flammability limits (vol%)	4.0–75.0	5.3–15.0
Detonation limits (vol%)	18.3–59.0	6.3–13.5
Diffusion velocity in air (cm/s)	200.0	51.0
Ignition energy (mJ) ^e	0.02	0.29
Ignition energy (mJ) ^f	10	20
Flame velocity in air (cm/s)	265–325	37–45
Toxicity	Nontoxic	Nontoxic

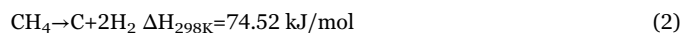
^a Mass density measured at a pressure of 1 atm and temperature of 0 °C. ^b Mass density of liquid H₂ measured at temperature of 20 K. ^c and ^d Includes water and steam production in higher heating value and lower heating value, respectively. ^e and ^f Ignition energy measured at stoichiometric mixture and lower flammability limit, respectively.

in Eq. (1) [4]. The high enthalpy associated with very low density of hydrogen in Eq. (1) fallouts in the exceptional energy-to-weight ratio of any fuel. However, its energy-to-volume ratio is poor [5]. The detailed physicochemical properties of hydrogen is given in Table 1.

Our current energy system produces billions of tons of greenhouse gas (GHG) and spend huge money and effort to dispose the produced GHG. However, the engine that consume pure hydrogen does not produce any polluting material, but water together with high energy. The quantity of energy produced during hydrogen combustion is at least three times higher than that produced by any other energy sources on mass basis [7]. The constructive environmentally benign characteristics of hydrogen results in its huge demand in recent years, especially in industrial applications and merchant uses [8–12]. An approximate quantity of 3×10¹² kg hydrogen would have to be produced annually to satisfy the world's total energy requirements, which is currently fulfilling by fossil fuels. As hydrogen is not a primary fuel, it should be extracted from water, biomass, or other hydrocarbons without contaminating the environment [13–15]. Water splitting, gasification of coal and biomass, and thermochemical cracking of methane are the commonly adopted hydrogen production methods [16,17]. Photocatalytic water splitting is environmental friendly and highly reproducible, however, which needs suitable semiconductor to produce hydrogen and oxygen at substantial atmospheric conditions [18,19]. Fujishima and Honda conducted a revolutionary experiment on photocatalytic water splitting using TiO₂ electrodes under ultraviolet light [20]. In the case of electrochemical water splitting, hydrogen evolution reaction occurs at cathode and oxygen evolution reaction at the anode [21,22]. Noble metals and metal oxides are state-of-the-art electrocatalysts for water splitting, which exhibit efficiency very close to thermodynamic potentials [23,24]. Sun et al. [25] developed a novel CoSe₂ nanocatalyst homogeneously disseminated on carbon fiber paper via pyrolysis and selenization of in situ grown zeolitic imidazolate framework-67, which presented high and stable catalytic activity in both hydrogen and oxygen evolution reaction. Comprehensive studies on low cost hydrogen production via photocatalytic water splitting and oxygen evolution reactions are reviewed elsewhere [26–28].

Gasification and reforming of biomass and biogas components were broadly investigated, though the drawback of such technologies are the requirement of hydrogen purification stage [29,30]. However, methane, major content of natural gas, is the best resource of hydrogen, which has the highest hydrogen – carbon ratio among all hydrocarbons. The physicochemical properties of methane is compared with hydrogen in Table 1. Furthermore, the value added nanocarbon is the only byproduct in methane cracking as shown in Eq. (2). Hence, the gas purification process could be eluded in methane decomposition.

The hydrogen produced by methane cracking, which is completely free from GHGs, could directly be applied in H₂–O₂ fuel cell and internal combustion engine [31]. Additionally, the storage and consumption of solid nanocarbon is easier and safer compared with any gaseous byproducts.



The utilization of methane as a resource for the production of hydrogen and nanocarbon in industrial scale is economically reliable because of the availability of huge natural reserves of methane in the planet, such as natural gas, petroleum-associated gases, and methane hydrates [32]. At present, more than 240 Billion cubic meters of hydrogen is extracting from natural gas, which is nearly 50% of global hydrogen production [33].

A higher temperature (> 1200 °C) is necessary in methane cracking to achieve a rational hydrogen yield [9,34]. Hence, methane molecule is highly stable with a tetrahedral geometrical structure supported with extremely strong four C–H bonds with a bond energy of 434 kJ/mol. Hence, the implementation of catalysts is essential to provide a pathway with a lower activation energy. Such methane cracking in the presence of a catalyst is known as thermocatalytic methane decomposition (TCD). Metal based or carbon based catalyst were introduced in order to operate at lower temperature and improve the process kinetics of methane cracking. Establishment of cost effective production of hydrogen and nanocarbon is deliberately depends on the development of highly active catalyst which maintain longer activity amid huge carbon deposition, optimized experimental parameters and development of proper reactors. The literature survey clearly evidenced that the catalyst support influence the activity and stability of catalyst [35,36]. Additionally, the factors such as catalyst material, the catalyst textural properties, and the operating conditions also clearly influencing the TCD process as well as the characteristics of as-produced nanocarbon [35,37–41]. This review would provide a panoramic view on the influence of metal catalyst characteristics and experimental parameters on the yield of hydrogen and nanocarbon during TCD process (Section 4). Additionally, the review also discusses the optimization of the TCD process with Design of Experiment (Section 5) and process kinetics (Section 6) for in depth understanding. Final section (Section 7) explains the micro-level findings on methane decomposition using computational technologies.

2. TCD for hydrogen production

Production of pure hydrogen by TCD process can be considered as a milestone towards the development of hydrogen economy. The catalytic activity and stability of the catalyst and the characteristics of as-produced nanocarbon are very relevant in TCD, since both play a vital role in determining the overall product yield [32]. The upgrading of catalytic characteristics, in the sense of improving methane conversion, is very essential on the overall hydrogen production efficiency of the process. The enhancements in catalytic characteristics is not only limited to the better reaction rate and minimal operating temperatures achieved, but also the competence to retain the thermochemical stability amid huge nanocarbon deposition. Accordingly, various metal and carbon based catalysts were introduced [8,9]. Metal-based catalysts are superior to carbonaceous catalysts in terms of their hydrogen production percentage and reaction rate. Transition metals, particularly Ni-, Fe-, and Co-based catalysts were given extensive attention in TCD [42,43]. Ni-based catalysts are distinguished among the metal based catalysts because of their relatively low-cost, low-toxicity, superior activity, stability, and environmental friendly characteristics [7,44]. Metal based catalysts preserve their activity by upholding a nanocarbon formation mechanism which retains the active site of the metal on the top of the filament towards the reaction medium. The growth mechanism of nanocarbon filament involves the diffusion of deposited carbon

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