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A power cycle of natural gas decarbonization and dual fuel cells with inherent 100% carbon capture

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ABSTRACT

An integrated natural gas decarbonisation and dual fuel cells power cycle (NGDDFC) was evaluated. The hydrogen and solid carbon produced in the natural gas decarbonisation process are fed to hydrogen and carbon fuel cells respectively. A fraction of the produced hydrogen is combusted to supply the heat required for natural gas decarbonisation. A pure carbon dioxide stream is produced from the carbon fuel cell, suitable for sequestration. The power cycle shows an efficiency of about 61% with near 100% carbon capture, 15 to 20% points higher than natural gas combined cycle (NGCC).

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Introduction

Natural gas (NG) decarbonisation by means of thermal cracking, decomposition, or pyrolysis to produce hydrogen and solid carbon has been explored for mitigating carbon emissions [1–4]. Hydrogen is an ideal fuel for hydrogen fuel cells (HFC). For the produced carbon, one published suggestion is to produce nano-carbon materials or carbon fibers [5]. However, in energy or power production processes, the quantity of fossil fuel is massive, and the material manufacturing industry cannot consume that much carbon. The large quantity of the carbon produced must be utilized to generate power in smarter ways other than straight combustion in conventional steam boiler power plants. The direct

carbon fuel cell (DCFC) sometimes called carbon fuel cell (CFC) [6,7] could be a better choice to realize power generation from natural gas decarbonisation: via a system with both hydrogen and carbon fuel cells. In this application, the enormous merits of a CFC are high electrical efficiency and a pure CO₂ stream produced from the anode, suitable for compression, transportation and sequestration [8]. There is no need for any additional carbon capture technology such as amine solvent systems, resulting in a significantly low energy-penalty, and potentially a low cost, carbon sequestration compared to other carbon capture technologies, e.g., post combustion capture for steam boiler power plant.

There is an industrial *Thermal Black* process for producing carbon black by natural gas thermal cracking at a scale with a feeding rate of several tonnes of NG per hour [9,10]. It could be

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viable to modify, reengineer and scale up the existing thermal black technology for power applications. The thermal black process uses two reactors, each operating in an intermittent mode. Inside the reactors are ceramic heat storage blocks. One reactor is heated up to 1300 °C by burning part of the produced hydrogen. Then natural gas is switched to flow through the reactor and gets thermally cracked to hydrogen and carbon black, which is cooled and collected by a bag filter. Due to endothermic cracking of the natural gas, the temperature of the furnace decreases to 800 °C, the reactor is switched back to heating mode again. The two reactors are shifted alternatively between heating and cracking mode, resulting in an overall continuous production. There are no detailed performance reports publically available. It is very interesting to know the hydrogen purity from the thermal black process. An analysis was carried out on the product composition of methane thermal cracking by using FactSage 7.0, 2015. The results are shown in Fig. 1.

The main products are hydrogen and the unconverted methane. At 800 °C, the methane conversion is about 92%. The next abundant species besides hydrogen and methane is ethylene with a negligible yield at a level of $\sim 10^{-6}$. Suelves, et al., reported methane thermal cracking through a carbon bed at 850 °C [11]. Only hydrogen and the unconverted methane were detected by gas chromatography, which is in accord with the above calculation. At 1100 °C, the conversion is greater than 99%. At even higher temperatures up to 1300 °C, it reaches close to 100% conversion. The average hydrogen purity from the thermal black process would likely be about 98% by integrating the chart in Fig. 1. Moreover, it is likely that as the temperature cools down from 1300 °C to 800 °C during the cracking mode, the feeding rate would need to be reduced to accommodate the ever-lowering thermal cracking kinetics. If the decrease of feeding rate during the cracking mode is factored in by assuming it to be reduced half from 1300 to 800 °C, the hydrogen purity would be about 99%. We assume the hydrogen produced by natural gas (NG) decarbonisation is pure enough to be fed to a solid oxide fuel cell (SOFC).

On the other hand, small amount of the unconverted NG can undergo internal reforming to be fully consumed by the fuel cell [12]. This may be energetically even more beneficial

since the NG thermal cracking is endothermic, consuming hydrogen (heat supplied by burning hydrogen). By internal reforming, the waste heat generated by HFC is more effectively recuperated.

Along the idea of NG decarbonisation and carbon fuel cell, there have been several studies focusing on system electrical efficiency and overall performance. Hemmes, et al. studied a process of methane thermal cracking to produce hydrogen as a commercial product, and carbon to fuel a CFC for power generation [13]. The thermal energy source for methane thermal cracking was indicated as external. In their next paper [14], a similar system design is described by employing a concentrated solar power reactor through which methane (NG) is thermally dissociated to hydrogen and carbon. Steinberg evaluated a NG thermal cracking process by plasma, followed by a hydrogen fuel cell and a carbon fuel cell [15]. Liu, et al. [16], carried out an exergy evaluation on methane catalytic thermal cracking at 80% conversion. The hydrogen-rich mixture gas is fed to an HFC for power production. The unconverted methane in the hydrogen mixture undergoes *in situ* reforming to produce more hydrogen and carbon dioxide. The anode exhaust gases go to an after burner to burn out the remaining combustible. The heat from the after burner is used for the methane cracking. From their CFC, a pure CO₂ stream is produced. Thus, 80% carbon from the initial feed of methane is produced from the anode of the CFC, ready for carbon capture. It does not give a first law efficiency, which is always lower than the exergy efficiency. Zhuang, et al., proposed to burn a fraction of the natural gas feed to supply the heat for NG decarbonisation [17], just like in NG steam-reforming. The modeling results shows 18% of the initial NG feed has to be combusted, resulting in 82% carbon capture with a total power efficiency of about 62%.

This paper evaluated a system of NGDDFC by burning a fraction of the produced hydrogen for NG decarbonisation followed by an HFC and a CFC. In this process design, all the natural gas undergoes decarbonisation. Thus, all of the carbon in the natural gas is converted to carbon powder, which is then fed to a CFC for power production and forming CO₂ as anode exhaust, enabling 100% carbon capture with high power generation efficiency.

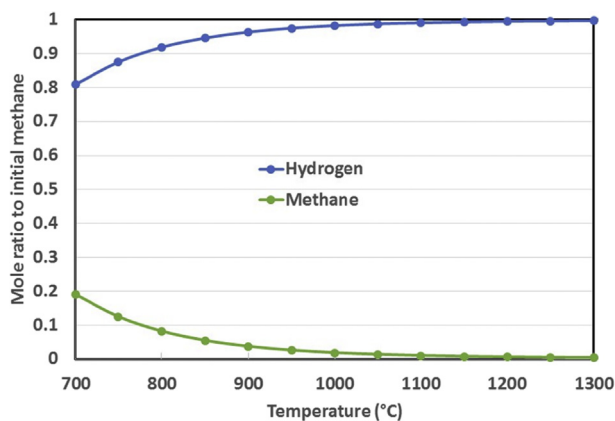
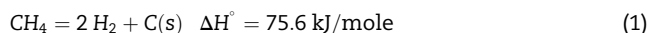


Fig. 1 – Gaseous products composition of methane thermal cracking at one bar.

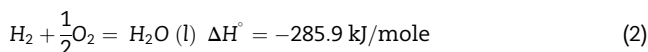
NGDDFC process and modeling

The three main chemical reactions involved in the NGDDFC are as follows

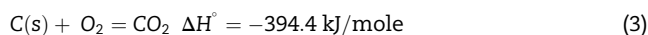
NG decarbonisation [18]:



The produced H₂ is sent to a HFC where it undergoes the following net reaction:



The produced carbon is sent to a CFC, with the following net reaction:



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