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#### An integrated natural gas power cycle using hydrogen and carbon fuel cells



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#### ABSTRACT

An integrated power cycle comprising natural gas decarbonization in conjunction with a hydrogen fuel cell and a carbon fuel cell is proposed, termed a natural gas decarbonization dual fuel cell system (NGDDFC). In this process hydrogen and solid carbon produced in a natural gas decarbonization step are fed to hydrogen and carbon fuel cells, respectively. A pure carbon dioxide stream is produced from the carbon fuel cell, suitable for sequestration. Process modeling of this cycle carried out using UniSim R430, for the non-CO<sub>2</sub> capture case, showed a cycle power efficiency of about 61%, which is 15 percentage points higher than the current natural gas combined cycle (NGCC) case. With 82% CO<sub>2</sub> capture, the NGDDFC had a net efficiency about 20 percentage points higher than the published NGCC option, albeit with a slightly lower capture percentage (82% vs. 90%).

#### 1. Introduction

In the past decade, North America has seen a significant shale gas boom, and an abundant natural gas supply that is expected to last well into the future. Natural gas (NG) is, and will be in the future, one of the major energy resources for power generation. Utility NG combustion systems provide appreciably less greenhouse gas (GHG) emissions compared to their coal predecessors. Natural gas combined cycles (NGCC) have high plant efficiencies (approximately 52%, [1]), low greenhouse gas emission rates (400 kg/MWh, [1]), and have been used as the standard for regulatory changes in Canada that dictate minimum requirements for future thermal power plants [2]. Although NGCC technology shows much higher power generation efficiency than conventional coal-fired technology, there is increasing demand for further improvements. Embracing a carbon-restrained economy, research on effective utilization of NG with the practice of low carbon emissions is ongoing. For example, in June 2016 the United Kingdom Energy Technologies Institute called for power technology developers to create a "template" for a carbon capture and storage (CCS) enabled gas-fired power plants [3]. Work is also being carried out to assess the option to replace commercial and residential natural gas with hydrogen from a steam-reforming process with CO2 capture [4]. The Leeds City Gate energy initiative proposes to transport the produced hydrogen through the existing NG pipeline network to end-users to realize low carbon energy production and distribution. Hydrogen is often considered to be the energy carrier of the future [5]; low carbon and innovative utilization of NG is therefore an important topic to be pursued. Recently, Brad Page, CEO of the Global CCS Institute, pointed out that "if we are serious about tackling climate change then we've got to reduce emissions from every possible sector of the global economy, urgently and without bias" [6]. The route of NG decarbonization (thermal cracking) to produce hydrogen and solid carbon has been explored for power generation and also for carbon sequestration [7,8]. The overall decarbonization reaction is endothermic and listed in Eq. (1):

$$CH_4 = 2H_2 + C \quad \Delta H^0 = 75.6 \text{ kJ/mol}$$
 (1)

The produced hydrogen is the cleanest fuel and can be used directly in furnaces, gas turbines, or fuel cells. The produced carbon could be made into carbon nano-materials or carbon fibers [9]. It has also been suggested to store this produced carbon as fuel for future consumption when the climate cycle turns into a cooling phase or when, for whatever reason the CO<sub>2</sub> in the atmosphere decreases [10]. Actually, there is an industrial-scale NG decarbonization process for producing carbon black as a pigment for ink, paint and tire production: the *thermal black process* can produce solid carbon at the significantly high rate of several tonnes per hour [10–13]. This process could be modified for this power generation application. The energy efficiency for hydrogen production by NG decarbonization is 58%, which is almost the same as NG steamreforming with carbon capture (60%) [8]. In steam reforming, excess steam is fed to the reformer, i.e., at least 2.5 times the stoichiometric

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Abbreviations: NGDDFC, natural decarbonization dual fuel cell; ASME, American Society of Mechanical Engineers; CEO, chief executive officer; CFC, carbon fuel cell; CCS, carbon capture and storage; DCFC, direct carbon fuel cell; FB-CFC, fluidized bed carbon fuel cell; GHG, greenhouse gas; HFC, hydrogen fuel cell; HRSG, heat recovery steam generator; HX, heat exchanger; IGFC, integrated gasification and fuel cell; MCFC, molten carbonate fuel cell; MC-CFC, molten carbonate carbon fuel cell; MIT, Massachusetts Institute of Technology; MW, megawatt; NG, natural gas; NGCC, natural gas combined cycle; OCV, open current voltage; SOFC, solid oxide fuel cell

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water has to be evaporated, which consumes a large amount of energy. Methane decarbonization via indirectly-heated solar reactors is a further option to reduce the energetic cost associated with producing carbon black, and has been studied recently at the laboratory scale in France [14,15]. The above research will open up more possibilities for the effective utilization of NG with low  $CO_2$  emissions.

Through decades of development, fuel cell technologies have reached the commercial stage for distributed electricity generation as well as for large scale power generation [16,17] with demonstrations at the MW level, achieving high process efficiency for energy conversion to electricity, while maintaining low pollutant and GHG emissions. The most commercially-advanced fuel cell technology is the hydrogen fuel cell (HFC). To make use of this type of cell, fossil fuels or bio-fuels must first be converted to hydrogen or hydrogen rich gas in a fuel preparation step, such as NG steam-reforming, coal gasification, biomass steam reforming, or partial oxidation, etc. Recently, Massachusetts Institute of Technology (MIT) released a report that examined a hybrid system that combined a coal gasification process with a hydrogen fuel cell (IGFC). By using this approach, the power generation efficiency could increase to a level of 50-60% [18]. Compared to the conventional coal-fired power plants, which typically have electric efficiencies between 30 and 38%, this combination could double the electric efficiency, hence the carbon emissions could inherently be reduced by half. The high efficiency operation of a fuel cell is very significant. Fuel cell technology is regarded as a game changer for carbon capture [19].

Fuel cells for other fuels are also being developed to expand the possibility of using other energy sources to generate electricity with high efficiency, including methanol, ammonia and carbon fine powder (e.g., pulverized coal). CanmetENERGY, a division of Natural Resources Canada, recently carried out a carbon fuel cell (CFC) development project and achieved encouraging results. A bench-scale CFC was designed, fabricated, and successfully operated with feeds of coal, petroleum coke, and coke breeze [20]. The merits of CFCs are as follows: a) they offer a high fuel-to-electricity efficiency; b) carbon is widely available as a primary fuel resource; and c) a pure CO2 steam is produced from the anode and is ready to be compressed for transportation and sequestration [21]. There is no need for any additional carbon capture technology such as amine solvent systems, resulting in a significantly low energy-penalty and a low cost of carbon sequestration compared to other carbon capture technologies such as post combustion capture. The development of CFCs is still in the early stage. For coal- or petroleum coke-fueled CFCs, the main hurdle is the presence of ash and sulfur, which makes it a challenge to operate continuously and effectively [22].

Compared to the above-mentioned carbon fuels tested by CanmetENERGY [20] and others (which used coal or petroleum coke as a feed fuel), the solid carbon produced from NG decarbonization is highly pure and clean. It would be much more easily handled and converted through a CFC for power generation than other solid fuels. In this study, we propose a new route of power generation and CCS (carbon capture and sequestration) from NG. By integrating the NG decarbonization process with hydrogen and carbon fuel cells (NGDDFC), fuel-to-power efficiencies of 15–20 percentage points higher than a natural gas combined cycle can be realized with a low carbon capture cost.

#### 2. Methodology

In this study, the energy evaluation was carried out with the aid of UniSim Design R430. However, before this approach is described in detail, the operation of individual components, most importantly, the hydrogen and carbon fuel cells, is presented. This discussion outlines the method used to estimate fuel cell efficiencies, operating requirements, and losses, which are necessary inputs to the model. The main chemical reactions of the proposed integrated system are as follows:

NG decarbonization [8]:

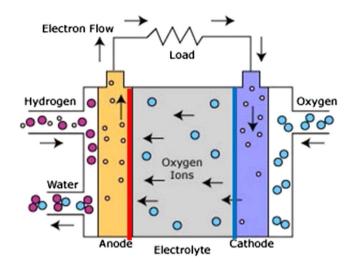


Fig. 1. Schematic of a hydrogen fuel cell [23].

$$CH_4 = 2H_2 + C(s)$$
  $\Delta H^0 = 75.6 \text{ kJ/mol}$  (1)

The produced  $\mathrm{H}_2$  is sent to an HFC where it undergoes the following net reaction:

$$H_2 + \frac{1}{2}O_2 = H_2O(l)$$
  $\Delta H^0 = -285.9 \text{ kJ/mol}$  (2)

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

$$C(s) + O_2 = CO_2 \quad \Delta H^0 = 394.4 \text{ kJ/mol}$$
 (3)

#### 2.1. Fuel cells

#### 2.1.1. Operation

Fuel cells are electrochemical devices that directly convert chemical energy in the fuel to electrical energy based on the principles of electrochemistry. Figs. 1 and 2 provide basic schematics of the hydrogen and carbon fuel cells, respectively. For each cell, oxygen is introduced at the cathode ends (with air), the fuel is introduced at or near the anode end. The electrolyte(s) separates the two electrodes and allows for the transport of ions between the two ends. The circuit is completed by connecting the electrodes externally with a load. The CFC configuration shown is a solid oxide and molten carbonate hybrid fuel cell (SO/MCFC) where carbon is fed directly into the electrolyte.

By avoiding the intermediate steps of producing heat, steam and mechanical work, typical of most conventional power generation methods, fuel cells are not limited by the thermodynamic limitations of heat engines defined by the Carnot efficiency. Also, because high temperature combustion is avoided, emissions of pollutants such as  $NO_x$ 

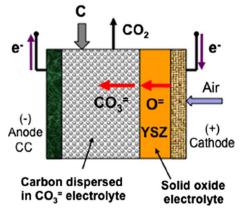


Fig. 2. Schematic of carbon fuel cell [24].

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