



Design of highly efficient coal-based integrated gasification fuel cell power plants

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ABSTRACT

Integrated gasification fuel cell (IGFC) technology combining coal gasification and solid oxide fuel cell (SOFC) is believed to be the only viable solution to achieving U.S. Department of Energy (DOE)'s performance goal for next generation coal-based power plants, producing electricity at 60% efficiency (coal HHV-AC) while capturing more than 90% of the evolved CO₂. Achieving this goal is challenging even with high performance SOFCs; design concepts published to date have not demonstrated this performance goal. In this work an IGFC system concept consisting of catalytic hydro-gasification, proven low-temperature gas cleaning and hybrid fuel cell-gas turbine power block (with SOFC operating at about 10 bar) is introduced. The system is demonstrating an electricity efficiency greater than 60% (coal HHV basis), with more than 90% of the carbon present in the syngas separated as CO₂ amenable to sequestration. A unique characteristic of the system is recycling de-carbonized, humidified anode exhaust back to the catalytic hydro-gasifier for improved energy integration. Alternative designs where: (1) anode exhaust is recycled directly back to SOFC stacks, (2) SOFC stack operating pressure is reduced to near atmospheric and (3) methanation reactor in the reactor/expander topping cycle is removed, have also been investigated and the system design and performance differences are discussed.

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1. Introduction

With increasing energy demand and growing concern of global climate change caused by greenhouse gas emissions, the U.S. Department of Energy (DOE) is promoting R&D on coal-based power plants with ultra-high thermal efficiencies and carbon capture capability. Integrated gasification fuel cell (IGFC) technology combining coal gasification and solid oxide fuel cell (SOFC) is believed to be the only viable solution to achieving DOE's performance goal of producing electricity at 60% efficiency (coal HHV-AC) while separating at least 90% of the evolved CO₂ amenable to sequestration [1]. This efficiency goal as set by the DOE is to account for any penalty associated with CO₂ separation but not that due to its compression to the sequestration pressure. Previous conceptual analyses of such IGFC plants have clearly shown the potential for improved efficiency and emissions relative to other system approaches [2–17], yet none of the research work published to date have demonstrated this performance goal, mostly due to the fact that even with highly efficient SOFC as the power block, CO₂ separation and the gasification process posed significant efficiency penalties on the system. Besides, integration of an SOFC power

block with a gasification system is very different from the integration of gas turbine and steam turbine power blocks that are typical of an integrated gasification combined cycle (IGCC) plant due to the unique operating and control features of SOFC stacks. As a result, novel IGFC system design concepts that capture the unique synergies between SOFC and gasification subsystems are required to achieve the aggressive performance goal.

In this work an IGFC system concept consisting of catalytic hydro-gasification, low-temperature gas cleaning and hybrid fuel cell-gas turbine power block (with SOFC operating at about 10 bar) is introduced. A unique characteristic of the system is recycling de-carbonized, humidified anode exhaust gas back to the hydro-gasifier for improved energy integration.

Alternative designs where: (1) anode exhaust is recycled directly back to SOFC stacks, (2) SOFC operation pressure is reduced to near atmospheric and (3) methanation reactor in the reactor/expander topping cycle is removed are also discussed.

2. IGFC development: a literature review

Integrating gasifiers with high-temperature fuel cell systems (MCFC or SOFC) to create power generation systems of ultra-high efficiencies and low emissions have been attracting significant research attention around the world since early 1990s.

Early conceptual designs mostly focused on MCFC-based systems due to the relative maturity of the MCFC technologies at that

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time. Jansen et al. [2] proposed an IGFC system featuring Shell oxygen blown dry feed entrained-flow gasifier, high-temperature gas cleaning with an operating temperature in the neighborhood of 350 °C and MCFC, and concluded that such a system could achieve electricity efficiency of 53.1% (LHV basis); he further investigated the feasibility of CO₂ capture downstream of the MCFC using shift reaction followed by ceramic CO₂ separation membrane and pointed out the system electricity efficiency would drop to 47.5% (LHV basis) due to the carbon capture. In a later paper Jansen et al. [3] compared the effects of different gas cleaning processes on the performance of an IGFC system employing Texaco oxygen blown slurry feed entrained-flow gasifier and MCFC (with no carbon capture), and demonstrated that the IGFC system with high-temperature gas cleaning could achieve electricity efficiency of 53.2% (LHV basis) while employing the proven low-temperature gas cleaning would decrease the electricity efficiency to 49.2% (LHV basis).

In Japan, as part of the Coal Energy Application for Gas, Liquid and Electricity project (the so-called “EAGLE” project), an IGFC system featuring oxygen blown dry feed entrained-flow gasifier, low-temperature gas cleaning and MCFC has also been designed and the system is expected to produce electricity at an efficiency of 53.3% (HHV basis) [4].

With the development of SOFC technologies, hybrid SOFC-GT technologies that combine SOFC with gas turbines began to demonstrate superior power generation efficiencies, which are important to IGFC systems. The capability of SOFC to internally reform hydrocarbon fuels (mostly CH₄) provides SOFC-based systems more flexibility in fuel input [5–8]. Thus, recently much research and development attention has been paid to SOFC-based IGFC systems.

In 1992, the Electric Power Research Institute (EPRI) performed an evaluation study on the Westinghouse SOFC technology for electricity utility applications in Japan [3]. In this system, two Shell oxygen blown gasifiers were proposed for coal conversion and the SOFC system was composed of 96 modules generating 60% of the total electricity. The system also included a high-temperature gas cleaning subsystem. The study showed net electrical efficiencies approaching 47% (HHV basis).

Lobachyov and Richter [9] proposed an IGFC system composed of Conoco CO₂ acceptor gasification process and SOFC-gas turbine hybrid system. The system efficiency was estimated to be quite high, 63% (HHV basis) but with no carbon capture. However, this proposed system relies heavily on the CO₂ acceptor gasification process for not only syngas production but also sulfur and other contaminants removal. This cleanup scheme does increase the system efficiency significantly because the syngas coming out of the gasifier is used directly in the SOFC with minimal thermal energy loss. However, the feasibility of producing a syngas with contaminants removed to meet the stringent specifications of an SOFC while utilizing such a gas cleanup process is not considered practical at the present time.

In the early 2000s, several European research institutes initialized a “BARAK” project to investigate the technical and economic feasibility of IGFC systems for combined heat and power (CHP) applications [10]. Both MCFC-based and SOFC-based systems were investigated and compared. The research work showed that SOFC technologies are more suitable for IGFC application due to several reasons.

- (1) A system based on SOFC might be less complex than a system based on MCFC where CO₂ needs to be transferred from the anode outlet to the cathode inlet.
- (2) The allowable temperature increase for the SOFC is much larger than the allowable temperature increase of the MCFC, indicating that the cooling need and thus the needed flow of air through the cathode is lower for the SOFC. Since the air pass-

ing through the cathode has to be compressed to the pressure level of the fuel cell, a lower airflow results in lower energy consumption for compressing the air.

- (3) The electrolyte in the SOFC is a solid and thus allows for greater deviating pressure levels between anode and cathode; the MCFC, on the other hand, has a carbonate melt as the electrolyte, and could cause a fateful gas crossover if operating with a high pressure differential across the anode and cathode.

The research work finally defined an IGFC CHP system featuring oxygen blown entrained-flow dry feed gasifier, low-temperature gas cleaning and SOFC capable of producing electricity at an efficiency of 46.7% (LHV basis) and an overall CHP efficiency that could be as high as 84.8% (LHV basis).

Kuchonthara et al. [11] proposed an IGFC system called “thermochemical recuperative coal gasification cycle”; the basic idea was to use the thermal energy in the gas turbine exhaust for the endothermic methane reformation reaction so that the thermal energy can be recovered both thermally and chemically. The system electricity efficiency was estimated to be 46.3% (HHV basis).

Ghosh and De [12] presented a conceptualized CHP IGFC system based on coal gasification and high temperature, pressurized SOFC in the topping cycle and a bottoming steam cogeneration cycle and demonstrated substantial efficiency gain due to CHP cogeneration. In a follow-up work Ghosh and De [13] also conducted exergy analysis of the proposed system and concluded that major part of total exergy loss occurs in the gasifier and the SOFC owing to combustion and electrochemical reactions; plus, sensitivity analysis showed that increasing the operation pressure of the SOFC would decrease exergy losses in most of the equipment thus yielding better cogeneration performance.

The U.S. DOE had been actively promoting R&D work on IGFC systems through research programs such as “Vision 21” and “Solid State Energy Conversion Alliance (SECA)”. The efficiency targets were first set to be 60% (HHV basis) for coal-fueled plants producing electricity only without CO₂ capture [14]. Under the sponsorship of the U.S. DOE/National Energy Technology Laboratory (NETL), the Advanced Power and Energy Program (APEP) of the University of California, Irvine investigated a series of IGFC system configurations featuring different production and emission control characteristics [15]:

- (1) IGFC power only case: an IGFC system consisting of air blown fluidized-bed gasifier, high-temperature gas cleaning with operating temperature in the neighborhood of 400 °C and pressurized tubular SOFC (operated at 18–19 bar) was conceptualized and analyzed to have an electricity efficiency of 60.1% (HHV basis) when no carbon capture capability was included.
- (2) IGFC near “Zero Emission” case: when carbon capture capability is required, the air blown gasifier needs to be replaced with oxygen blown gasifier, and a water gas shift conversion followed by a high-temperature H₂ separation membrane is included for CO₂ separation. The inclusion of carbon capture capability causes a significant system efficiency penalty and results in an electricity efficiency of 49.6% (HHV basis, 95% CO₂ captured).
- (3) IGFC “Advanced FutureGen” case: the characteristics of SOFC make it possible to cogenerate electricity and H₂ (which can be used as clean transportation fuel for automobiles powered by fuel cells) at the same time in an IGFC system. This cogeneration capability can be achieved by adding a water gas shift conversion with high-temperature H₂ separation membrane unit upstream of the SOFC in the system mentioned above. Depending on the extent of H₂ exported, the system overall thermal efficiency can be 61.10–54.83% (HHV basis, 95% CO₂ captured).

Table 1
Brief summary of IGFC analysis work performed to date.

Researcher	Gasifier type	Syngas cleanup ^a	Fuel cell	Carbon capture	Efficiency
Jansen et al. [2]	Shell gasifier (oxygen blown, entrained-flow, dry feed)	HTGC	MCFC	Downstream of the fuel cell, water gas shift reaction followed by ceramic membrane for CO ₂ separation	53.1% (LHV basis) without carbon capture; 47.5% (LHV basis) with carbon capture
Jansen et al. [3]	Texaco gasifier (oxygen blown, entrained-flow, slurry feed)	HTGC or LTGC	MCFC	No carbon capture capability	53.2% (LHV basis) with HTGC; 49.2% (LHV basis) with LTGC
EAGLE [4]	Oxygen blown, entrained-flow, dry feed	LTGC	MCFC	No carbon capture capability	53.3% (HHV basis)
EPRI [3]	Shell gasifier (oxygen blown, entrained-flow, dry feed)	HTGC	SOFC	No carbon capture capability	49% (LHV basis)
Lobachyov and Richter [9]	Conoco CO ₂ acceptor gasification	No syngas cleaning process	SOFC	No carbon capture capability	63.1% (HHV basis)
Kivisaari et al. [10]	Prenflo gasifier (oxygen blown, entrained-flow, dry feed)	LTGC	SOFC	No carbon capture capability	46.7% (LHV basis) electricity efficiency; 84.8% (LHV basis) overall efficiency for CHP
Kuchonthara, et al. [11]	Oxygen blown, fluidized-bed gasifier	Not specified	SOFC	Upstream of the fuel cell, water gas shift reaction followed by membrane for CO ₂ separation	46.3% (HHV basis)
Rao et al. [15]	Air blown, fluidized-bed gasifier	HTGC (warm gas cleaning)	SOFC	No carbon capture capability	60.1% (HHV basis)
Rao et al. [15]	Oxygen blown, fluidized-bed gasifier	HTGC (warm gas cleaning)	SOFC	Downstream of the fuel cell, water gas shift followed by H ₂ separation membrane	49.6% (HHV basis)
Verma et al. [16]	Oxygen blown, fluidized-bed gasifier	HTGC (warm gas cleaning)	SOFC	Downstream of the fuel cell, water gas shift followed by H ₂ separation membrane	50.3% (HHV basis)
Ghosh and De [12,13]	Oxygen blown, entrained-flow gasifier	HTGC	SOFC	No carbon capture capability	30% fuel energy savings (reference is electricity efficiency of 40% and boiler efficiency of 90%)
Gerdes et al. [17]	Catalytic hydro-gasifier	HTGC (humid gas cleaning)	SOFC	Downstream of the fuel cell, oxygen combustion followed by water condensation	56.2% (HHV basis) with carbon capture; 61.8% (HHV basis) without carbon capture

^a Due to lack of standardization of designations used in defining the operating temperature range of the various elevated temperature gas cleanup technologies, the cleanup technologies in the above summary have been divided into two categories: the high-temperature gas cleaning (HTGC) with operating temperature in excess of 250 °C; and low-temperature gas cleaning (LTGC) operating near ambient temperature. The designations used by the original authors are listed in parentheses.

Verma et al. [16] later used the "IGFC 'Near Zero Emission' case" described above as a baseline case and conducted sensitivity analyses to investigate the impacts of SOFC pressure, SOFC voltage, SOFC fuel utilization and the gasifier carbon conversion on the overall system performances. Sensitivity analyses revealed that SOFC pressure is the most significant factor, followed by SOFC fuel utilization, gasification conversion and SOFC voltage. Based on the analysis results, an improved system was proposed which featured an electricity efficiency of 50.3% (HHV basis). The efficiency improvement does not appear very significant compared with a 49.6% (HHV basis) for the baseline case but the impact of pressure on system efficiency is reduced with the new design; the higher efficiency being realized under relatively moderate SOFC operation pressure (this is desirable given the challenges associated with developing the required seals as well as the materials for fuel cells operating at high pressures).

With increasing awareness of the risk of climate change due to greenhouse gas emissions, U.S. DOE has modified the electricity efficiency targets of next generation coal-fueled power plants to be 60% (HHV basis) with more than 90% carbon capture [1]; and IGFC systems are believed to be the only viable path to this aggressive goal. In a recent report published by DOE [17], an IGFC system featuring catalytic hydro-gasifier, high-temperature gas cleaning

with operating temperatures in some of its subsections in excess of 400 °C and pressurized SOFC has been analyzed. The system is estimated to be capable of producing electricity at efficiency of 56.2% (HHV basis) with 90% CO₂ being captured and compressed for sequestration; if the IGFC system is operated without CO₂ capture capability, the plant efficiency can be pushed up to 61.8% (HHV basis). The use of a catalytic gasifier is an important contributing factor to the high efficiency: the catalytic gasification process operates at relatively low-temperature and produces high methane content syngas, which benefits both the gasifier efficiency and the SOFC performance.

Table 1 summarizes and compares the major IGFC systems proposed and analyzed to date. It can be seen that:

- (1) IGFC systems are capable of producing electricity at very high efficiency. This is due to the high electrochemical conversion efficiency of the fuel cell and the fact that elevated operation temperature of SOFC and MCFC enables combined cycles to produce even more electricity. SOFC generally has characteristics more suitable for IGFC application than MCFC.
- (2) Efficient as IGFC is, the inclusion of carbon capture and the inefficiency in gasifiers will inevitably pose significant efficiency penalties on IGFC systems. So far, an IGFC system incorporat-

ing the catalytic hydro-gasifier [17] comes closest to realizing DOE's IGFC efficiency goal, but the target of 60% efficiency (HHV basis) with more than 90% CO₂ separated has not been achieved yet.

- (3) Higher operation pressure for the SOFC is generally beneficial to the IGFC system performance [13,16], but this has always been a challenge for the SOFC designers.
- (4) The syngas cleaning processes also affect IGFC system efficiency significantly. High-temperature gas cleaning generally results in higher IGFC efficiency, but the technologies are highly developmental and are not mature and proven technologies as is low-temperature (near ambient temperature) gas cleaning to produce a syngas that can meet the purity specifications of an SOFC system.

To sum up, although many exciting efforts have been made in the field of IGFC system development, it is still challenging to achieve the goal of producing electricity at >60% (HHV basis) efficiency while separating >90% of the evolved CO₂. Innovative designs to better integrate coal gasification and fuel cell stacks for highly efficient electricity generation are still in urgent need.

3. Design basis and methodology

3.1. General plant design basis

The design ambient conditions consist of utilizing ISO ambient conditions of 15 °C (59F) dry bulb temperature, 60% relative humidity and sea level barometric pressure. Mechanical draft cooling towers are utilized for plant heat rejection with a 3.9 °C (7 F) approach to the wet bulb temperature. An 11.1 °C (20F) temperature rise is assumed for the cooling water while a 5.6 °C (10F) approach temperature is utilized in the steam turbine surface condenser when included in the plant.

The coal utilized in this study is a high rank bituminous Pittsburgh No. 8 coal containing 6% moisture and with a HHV of 28,959 kJ kg⁻¹ (as-received basis). The design basis for the overall plant and the gas turbines when included in the plant are summarized in Table 2.

3.2. Coal gasifier

The oxygen blown entrained-flow gasifiers are by far the most proven and matured gasification technologies and have been most frequently chosen to pair with SOFC stacks. Due to the higher oxygen content in the gasifier and higher operation temperature, the syngas produced by such gasifiers generally features high CO and H₂ content, with only small amount of CH₄. However, in IGFC applications an anode inlet gas rich in CH₄ is favored [17–19]. This is mainly because CH₄ can undergo the steam methane reformation in SOFC stack channels; thermodynamically the endothermic reaction has the potential to act as a chemical heat sink for the SOFC and can reduce the air flow rate to the fuel cell stacks for the purpose of cooling, thus saving parasitic energy consumption for air compression. Furthermore, higher CH₄ content in the syngas generally results from gasifiers operating at relatively lower temperatures, and with such gasifiers less fuel-bound energy is degraded to heat to maintain the gasifier operation temperature, resulting in cold gas efficiencies higher than those operated at higher temperatures. Concerns, however, are that the reaction rates are relatively slow and the carbon conversion is low due to a lower operating temperature.

Methane can be produced by directly reacting coal with H₂ through the methanation reaction:

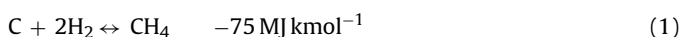
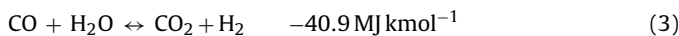
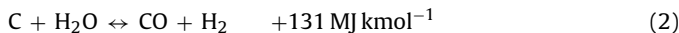


Table 2
Summary of plant design basis.

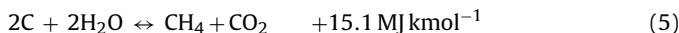
Parameters	Setting value
<i>Plant general settings</i>	
Location	Midwest
Site conditions	ISO
Coal	Pittsburgh No. 8
Coal heating value (as received HHV)	28,959 kJ kg ⁻¹
Plant heat rejection	Mechanical draft cooling towers
Air separation unit	Cryogenic
NO _x emission	<0.5 ppmvd (15% O ₂ basis)
SO _x capture	>99% of sulfur present in coal
Hg capture	≥90% of Hg present in raw syngas
Liquid wastes	Class 1 waste water injection well
<i>Gasifier design basis</i>	
Gasifier type	Catalytic hydro-gasifier
Gasifier exit temperature	691 °C (1275 F)
Gasifier exit pressure	70 bar (1015 psi)
<i>CO₂ separation design basis</i>	
CO ₂ separation	90% of carbon present in coal and flux less carbon in slag
CO ₂ pressure at plant battery limits	~1 bar ^a (14.7 psi)
CO ₂ composition	Meet requirements for adjacent geologic sequestration
<i>SOFC design basis</i>	
Maximum single pass fuel utilization	75%
Working cell voltage	0.8 V
Maximum ΔT at anode side	200 °C
Maximum ΔT at cathode side	200 °C
Maximum operating temperature	850 °C
Anode exhaust temperature	850 °C
Cathode exhaust temperature	850 °C
Maximum operating pressure	~10 bar
ΔP at anode side	48.3 kPa for pressurized SOFC, 20.7 kPa for near atmospheric SOFC
ΔP at cathode side	62.1 kPa for pressurized SOFC, 27.6 kPa for near atmospheric SOFC
Minimum O ₂ mole fraction in cathode exhaust	10 mol%
Inverter efficiency	97.0%
<i>Turbomachinery design basis</i>	
Compressor polytropic efficiency	91.4%
Turbine isentropic efficiency	90% per stage
Combustor ΔP	4% of inlet air pressure

^a Specified by U.S. DOE/NETL while calculating the overall plant thermal efficiency.

Alternatively, methane can also be produced by reacting coal with steam through the combination of the following reactions:



The overall reaction of the above three can be combined to be:



Reaction (1) is exothermic and prefers higher pressure and lower temperature from an equilibrium standpoint. The net effect of coal reacting with steam (reaction (5)) is generally endothermic. It is thus preferred to use both H₂ and steam for the coal gasification: the mix of hydrogen and steam allows for temperature control and heat balance in the gasifier.

Research work on developing gasifiers capable of producing a high CH₄ content syngas which can be further cleaned and methanated to produce substitute natural gas (SNG) attracted significant research interest in 1970s when there was a severe escalation of natural gas price. The R&D projects were later placed on

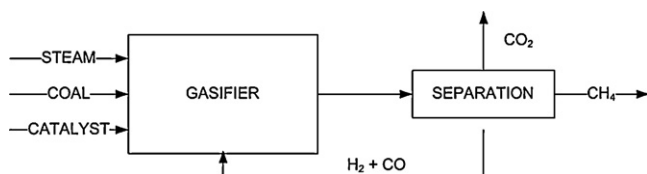


Fig. 1. Exxon catalytic coal gasification process.

the shelf mostly because the prices for fossil fuels, especially natural gas, did not increase as expected by forecasts.

The catalytic hydro-gasification process development work conducted by Exxon in 1970s [20,21] was among the most extensive and systematic. In Exxon's process, gasification agent consisting of steam, H_2 and CO was fed to a fluidized-bed gasifier to react with crushed coal. In the gasifier, coal is gasified and converted to CH_4 through a combination of steam gasification, water gas shift reaction and methanation reaction. Due to equilibrium limits, the product gas also contains some H_2 and CO; these components are separated and recycled back to the gasifier to help maintain thermodynamic equilibrium and heat balance in the gasifier. The gasification process is schematically shown in Fig. 1. Because the gasification process takes place at relatively low temperature, potassium catalyst (KOH and K_2CO_3) is employed to promote reaction rates and carbon conversion. The Exxon catalytic gasification process is used as a reference for gasifier performances in the following design work.

In more recent years, gasification processes capable of producing CH_4 rich syngas are attracting much R&D interests again as an important building block of future clean coal technologies. GreatPoint Energy is developing a "hydromethanation" technology called "bluegasTM"; the reaction process is similar to that of the Exxon's, but using a proprietary catalyst. The company is planning to produce 1 billion cubic feet per day of equity gas from multiple large-scale bluegasTM facilities operating or under construction by 2022. Arizona Public Service (APS) [22] has been developing a non-catalytic hydro-gasification based SNG and electric power co-production process capable of capturing CO_2 for sequestration. Instead of recycling H_2 from downstream separation unit, the APS concept proposes to produce H_2 from renewable sources.

The characteristics of SOFC can actually enable even bigger synergy when combining such hydro-gasifiers with SOFC. For example, SOFC can internally reform hydrocarbon fuel, yet not all the hydrogen formed inside SOFC is consumed due to Nernst potential limits. Thus the SOFC can work as a gas separation unit for the gasifier and provide H_2 that can be recycled back to the gasifier; this can be more efficient than conventional gas separation processes, thus further improving the system performance.

3.3. Gas cleanup and CO_2 separation strategy

It has been shown that high-temperature gas cleaning is more beneficial to the system's efficiency [3,15,17] but these technologies as pointed out previously are considered highly developmental for SOFC applications. In this work, conventional low-temperature gas cleaning technologies consisting of water scrubbing for removal of alkalis, halides and ammonia, activated carbon bed for removal of Hg and other volatile metals and SelexolTM process followed by ZnO guard bed for removal of sulfur compounds are employed due to their proven status.

Unlike some of the previous designs where the entire CO_2 separation occurs upstream of the SOFC ("pre-anode CO_2 separation"), the CO_2 separation is accomplished downstream of the SOFC ("post-anode CO_2 separation") from the depleted anode gas by keeping the anode and cathode streams exiting the SOFC separate. Post-anode

CO_2 separation has the advantage of higher system efficiency. Typically, pre-anode CO_2 separation requires a water gas shift reactor upstream of the CO_2 separation unit to convert the CO in the syngas into CO_2 in order to meet the carbon capture requirements. The exothermic water gas shift reaction degrades chemical energy contained in the syngas into thermal energy and this portion of the energy bypasses the SOFC when the syngas is cooled for desulfurization and decarbonization at lower temperatures. Since it is always preferable to put more energy into the topping unit of a cascading power system for higher overall system efficiency, it is advantageous to locate the CO_2 separation unit downstream of the SOFC so that more chemical energy contained within the syngas can be conserved for conversion in the SOFC stacks. Furthermore, when the syngas contains a high concentration of CH_4 , for pre-anode CO_2 separation it becomes necessary to reform the CH_4 first to form CO and ultimately to form CO_2 in order to meet the carbon capture requirement. With such a scheme, the advantages of having a high CH_4 content syngas providing a heat sink in the SOFC stack are lost.

The methyl diethanol amine (MDEA), a chemical absorption process, is selected for the CO_2 separation since the anode exhaust gas coming out of the SOFC stacks contains CO_2 at relatively low partial pressure (compared to syngas upstream of the SOFC stacks), making chemical absorption process more energy efficient than a physical absorption processes such as SelexolTM which is driven by partial pressure of the species being separated.

3.4. SOFC technology

The U.S. DOE have been supporting the development of SOFC stacks for large-scale stationary application through the SECA program and encouraging progress have been reported recently. Single planar SOFC cell performance under atmospheric pressure has reached the level of 500 mW cm^{-2} with 0.8 V working voltage [23]. Kerr of Delphi [24] has reported stack power density of 500 mW cm^{-2} with mean cell voltage of 0.87 V; Borglm of Versa Power [25] has also reported SOFC stack performance on a similar level. Based on this information and the expectation that SOFC stack performance will continue to improve in the following years, it is reasonable to use an operation voltage of 0.8 V for SOFC stacks in this design work. The maximum O_2 utilization is set to $\sim 52\%$, resulting in a minimum O_2 mole fraction in the cathode exhaust stream of 0.1. The maximum air temperature rise is restricted to 200°C (less than the full operating range of the SOFC) to constrain thermo-mechanical stresses on the SOFC. The design parameters for the SOFC stacks are also summarized in Table 2.

4. Modeling approach

The IGFC systems were modeled in Aspen Plus[®] 2006 process engineering flow sheet software package [26].

Conventional high-temperature oxygen blown gasifiers are generally simulated based on the minimization of Gibbs free energy, due to their high reaction rates resulting from the high gasifier operation temperature. In this work, the operating temperature of the catalytic hydro-gasifier is relatively low and the assumption that the gaseous products have reached chemical equilibrium is not guaranteed. To account for this non-equilibrium effect, the approach temperatures of individual reactions in the "Gibbs reactor" in Aspen Plus[®] were varied so that the compositions of the syngas produced closely matched the experimental results of the Exxon research work [20].

Separator blocks were used to depict the gas cleanup processes within the Aspen Plus[®] flow sheet. The separator model parameters were derived from unit performance data supplied by the various vendors.

5. Baseline case: IGFC with pressurized SOFC and anode exhaust recycled to gasifier

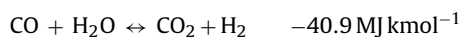
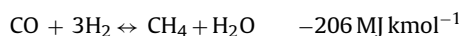
Based on literature review, previous work and brainstorming sessions, several IGFC design concepts were proposed and then evaluated. The most promising concept is the plant consisting of a high pressure catalytic hydro-gasifier system integrated with a pressurized SOFC (about 10 bar pressure)–gas turbine combined cycle with de-carbonized, humidified anode exhaust recycled back to the hydro-gasifier. The system will be used as a baseline case in this work and will be covered in this section. The overall process scheme is depicted in Fig. 2 while the major stream data are presented in Table 3.

5.1. Description of the baseline plant

The gasification agent containing steam, H₂ and CO is provided primarily from the SOFC anode exhaust after decarbonization and humidification. The “as received” coal is impregnated with potassium catalyst in the form of KOH and K₂CO₃ (mostly recycled catalyst along with some fresh makeup catalyst) and fed to the gasifier. The catalyst requirement is 15 wt.% of the dry coal input on a K₂CO₃ basis [20]. The unconverted carbon along with the accompanying ash (“fine ash”) after catalyst recovery is supplied to a high-temperature slagging entrained-bed oxygen blown gasifier to increase the overall carbon conversion of the IGFC plant while converting the ash into a vitrified non-leachable solid form (carbon conversion in the catalytic hydro-gasifier is typically about 90%). Some lime is used to regenerate the catalyst.

The raw catalytic hydro-gasifier effluent at a temperature and pressure of 690 °C and 70 bar enters the heat recovery and gas cleanup system after dry particulate removal. The raw syngas is initially cooled against a humidified H₂ recycle stream utilized in the gasifier. The raw syngas after providing additional heat for the steam system is then supplied to the syngas cleanup/low temperature gas cooling/heat recovery system, which includes water-wash to remove particulates, alkalis, chlorides and NH₃ followed by a Selexol™ solvent wash unit to remove the sulfur compounds. A carbonyl sulfide (COS) hydrolysis reactor and a sulfided activated carbon bed for capture of mercury (Hg) and other volatile metals such as arsenic (As) are provided upstream of the Selexol™ unit.

Next, water vapor is introduced into the clean CH₄-rich syngas leaving the Selexol™ unit. The added moisture prevents deposition of carbon in the downstream reactors and the SOFC anode. The water vapor is introduced by directly contacting the syngas with liquid water flowing down through a counter-current column. Counter-current humidification allows for recovery of low temperature heat generated within the plant while minimizing the use of high pressure steam. Clean process condensate collected from within the plant is used as the makeup water for this humidifier. The humidified syngas is then supplied to the reactor/expander topping cycle [18]. In this topping cycle additional CH₄ is formed and the gases are heated up by the exothermic methanation reaction followed by expansion through a turbine to recover power, as depicted in Fig. 3. The major thermodynamic reactions occurring within the reactor are:



A guard bed is included upstream of the methanation reactor as a final cleanup step to remove trace amounts of chlorides and sulfur compounds to the level required by the methanation catalyst (and the reforming catalyst within the SOFC system), which is 0.1 ppmV for each of these impurities. The guard bed consists of

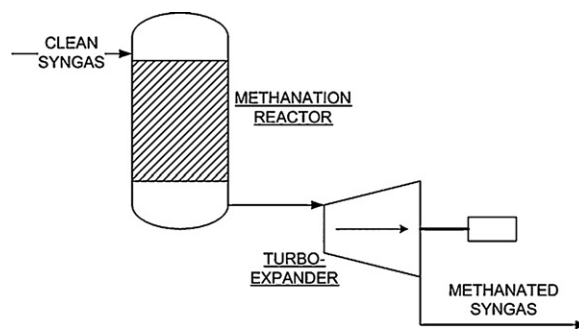


Fig. 3. Reactor/expander topping cycle.

alternating layers of COS hydrolysis catalyst such as a Co, Mo, or a Ni–Mo catalyst and ZnO for capture of the H₂S and the chlorides.

The CH₄-rich gas (36 mol.% CH₄ on a wet basis) is then supplied to the SOFC system, which includes a pre-reformer to chemically recuperate the sensible heat contained in the anode exhaust gas. The remaining CH₄ (28 mol.% CH₄ on a wet basis) present in the syngas is reformed within the SOFC channels utilizing heat generated within the stack. The large heat sink associated with the steam methane reformation reaction makes it possible to reduce the excess air required by the SOFC to the minimum set by the design basis (corresponding to 10 mol.% O₂ in cathode exhaust) and thus decrease the parasitic load of air compression. The ratio of pre-reformation to internal reformation is established so as to achieve the maximum allowable air temperature rise (200 °C) across the SOFC at the minimum excess air flow.

The H₂ demand of the catalytic hydro-gasification is met partially by supply of syngas from the oxygen blown gasifier and partially by the recycle of the anode exhaust gas after heat recovery, CO shifting, CO₂ separation and compression. Humidification of this stream after compression in a counter-current column is conducted in order to minimize the high pressure steam demand of the gasifier. Process condensate collected from the low-temperature gas cooling subsystem is used as the makeup water for this humidifier. This humidifier then also serves as a sour water stripper.

The per pass fuel utilization in the SOFC is limited to about 73% in order to recycle the balance of H₂ required by the catalytic hydro-gasifier. The overall fuel utilization is much higher, however, at about 98% due to the recycle. A small fraction (about 6%) of the anode exhaust gas is purged from the recycle loop and combusted with the cathode exhaust gas, in order to limit the concentration buildup of N₂ and Ar within the system.

After providing heat for preheating and partially pre-reforming the methanated syngas, the anode exhaust gas is further cooled while generating steam and then supplied to a catalytic reactor for the water gas shift reaction.

The effluent from the shift reactor, after further heat recovery/cooling, is fed to a MDEA unit for CO₂ separation. The CO₂ stripped from the solvent is combined with the oxidized acid gas leaving the Selexol™ unit. This combined stream may be dehydrated and compressed for sequestration.

The purge gas combustor exhaust is partially expanded in a turbine, fed to a recuperator to preheat the cathode inlet air supplied by a compressor, and then further expanded to near atmospheric pressure in the turbine to generate additional power before entering the heat recovery steam generator. These two turbines which are mounted on the same shaft as the cathode air compressor provide bulk of the power required by the compressor, the balance of the power being met by an electric motor. Excess steam generated within the plant is expanded in a condensing steam turbine.

All the required general facilities for a stand-alone plant such as water treatment, instrument air supply, flares, cooling tow-

Table 4
IGFC plant thermal performance summaries.

	Pressurized SOFC, with anode exhaust recycled to gasifier (baseline)	Pressurized SOFC, with anode exhaust recycled to SOFC	Near atmospheric SOFC, with anode exhaust recycled to gasifier	Pressurized SOFC, without methanation reactor before syngas expander
Coal energy input (GJ h ⁻¹) (HHV)	1397	1397	1397	1397
SOFC operation pressure (bar)	10.1	10.1	1.1	10.1
<i>Gross power output (major)</i>				
SOFC electrical power (MW)	247.77	227.96	229.91	247.75
Cathode exhaust expander (MW)	63.38	65.65	–	63.78
Steam turbine ^a (MW)	2.57	3.09	1.43	2.80
Syngas reactor/expander topping cycle (MW)	9.34	7.95	15.97	7.58
Total gross power generated (MW)	323.30	304.99	247.55	322.13
<i>Auxiliary power consumption (major)</i>				
Coal milling and coal handling (MW)	0.60	0.60	0.60	0.60
Coal pump, MW	1.60	1.60	1.60	1.60
Coal drying air blower (MW)	–	–	4.91	–
ASU air compressor (MW)	1.55	3.15	1.55	1.55
ASU O ₂ compressor (MW)	0.55	1.62	0.55	0.55
ASU auxiliary consumption (MW)	0.08	0.17	0.08	0.08
SOFC air compressor/blower (MW)	66.91	65.99	11.07	66.92
Recycled H ₂ compressor (MW)	8.24	5.03	18.87	8.22
Selexol™ unit (MW)	0.98	1.40	1.02	0.98
MDEA unit (MW)	0.62	0.59	0.60	0.62
Gas cooling humidifier pump (MW)	0.09	0.09	0.09	0.09
Recycled H ₂ humidifier pump (MW)	0.27	–	0.28	0.27
Cooling tower fan (MW)	0.36	0.42	0.50	0.37
Cooling tower pump (MW)	0.68	0.78	0.93	0.69
Condensate recycle pump (MW)	0.29	0.30	0.37	0.29
BFW feed pump (MW)	0.16	0.55	0.24	0.17
Transformer losses (MW)	0.70	0.66	0.54	0.70
Miscellaneous BOP and lighting (MW)	1.00	1.00	1.00	1.00
Total internal power consumption and losses (MW)	84.68	83.95	44.87	84.70
Net electric power (MW)	238.62	221.04	202.69	237.43
Overall thermal efficiency (%HHV)	61.5	56.9	52.2	61.2

^a The steam turbine would actually be used as a driver turbine to run one of the large compressors in the plant such as the recycled H₂ compressor, with motor assistance to makeup the difference in power.

ers, etc. are also accounted for in the overall plant performance estimation.

5.2. Performance of the baseline plant

The plant's thermal performance is summarized in Table 4 along with the other cases investigated.

The IGFC plant with pressurized SOFC and anode exhaust recycled to gasifier is estimated to generate power at a net efficiency of 61.5% on a HHV basis, with more than 90% of the carbon present in the syngas separated as CO₂ amenable to sequestration. Several unique design features make it possible to achieve the very high system efficiency:

- (1) Using gasifiers capable of producing a syngas with high CH₄ content at high cold gas efficiency (low syngas exit temperature). Thermodynamically the SOFC cooling associated with internal methane reformation is sufficient to reduce the air flow requirement to the minimum allowed by the imposed oxygen utilization constraint, thus decreasing the parasitic load of cathode air compression.
- (2) Recycling humidified, de-carbonized SOFC anode exhaust back to the catalytic hydro-gasifier. This is a very desirable synergistic integration between the catalytic hydro-gasifier and the SOFC stacks. It has been introduced in Section 3.1 that certain amount of H₂ is required for gasification process and energy balance in the catalytic hydro-gasifier. Instead of using a conventional gas separation process to separate H₂ and CO from CH₄ product (like in the Exxon process), the SOFC and the downstream carbon separation units fulfill the function of gas separation at relatively high efficiency. Also with the recycle

strategy, the per-pass fuel utilization in the SOFC stacks can be kept low (~70%), yielding high Nernst potential in the SOFC and correspondingly higher power density while simultaneously achieving very high (>95%) overall fuel utilization. This can be further demonstrated through Fig. 4, the Sankey diagram [29] showing a quantitative account of the major energy flows associated with the plant. As can be seen, a significant fraction of the input energy is recycled from the power block back to gasification system, enabling even bigger energy input to the SOFC stacks for highly efficient energy conversion.

- (3) The additional oxygen blown gasifier not only increases the overall plant carbon conversion and converts the fine ash into a non-leachable vitrified ash but also provides control flexibility in the event of changes in the catalytic hydro-gasifier H₂ demand caused by variability in coal characteristics, plant load factor and transients. About 80% of the H₂ and CO demand of

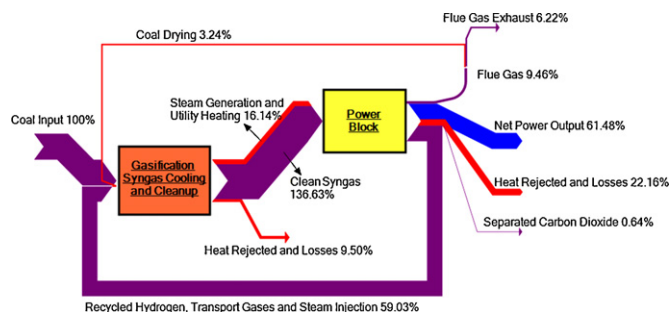


Fig. 4. Sankey diagram—IGFC power only plant with pressurized SOFC and anode exhaust recycled to gasifier.

The overall auxiliary power consumptions of the two cases are similar: in this design, the load of the air separation unit is more than twice of the baseline design because much more O_2 is required for the oxygen blown gasifier; but the work for compressing the recycled anode exhaust is also significantly smaller since the majority of the anode exhaust is only required to be compressed to about 17 bar for use in SOFC rather than 75 bar for use in gasifier.

However, because more coal is now split to the oxygen blown gasifier (for a given total coal input to the IGFC system), more chemical energy has to be utilized in the gasifier subsystem to maintain the higher operation temperature in oxygen blown gasifier. As a result, the overall efficiency of the gasification subsystem in this design is significantly lower than that of the baseline design. This results in smaller total energy input to the SOFC stacks, and although the two cases have similar overall fuel utilization around the SOFC stacks, the power produced by fuel cells in this case is significantly smaller than in the baseline case.

6.2. IGFC with atmospheric SOFC and anode exhaust recycled to gasifier

To demonstrate the effect of SOFC operation pressure, an alternative design where SOFC stack operation pressure was reduced to near atmospheric pressure was investigated. The process scheme is shown in Fig. 6. The plant process scheme is similar to the previously described baseline case. Major differences in the configuration are identified as follows:

- (1) The expansion ratio of the turbine in the reactor/expander topping cycle upstream of the SOFC system is significantly higher since the required anode inlet pressure is much lower for this case.
- (2) Additional equipment is added between the anode exhaust gas leaving the SOFC stacks and the shift reactor in order to raise the

gas pressure such that the shift reactor is operated at a pressure high enough to limit its size from becoming excessive. The gas is cooled to near ambient temperature after heat recovery while condensing out the water, compressed to a pressure of about 20 bar, and humidified to re-introduce water vapor required by the shift reaction prior to preheating and feeding it to the shift reactor.

- (3) The cathode air is supplied by a motor driven blower, while the combustor exhaust is fed directly to the heat recovery unit where the cathode inlet air is preheated and steam is generated. Because the combustor exhaust is at relatively low pressure, an additional blower is needed to pressurize the portion of hot exhaust leaving the heat recovery unit for the coal drying operation.
- (4) A small fraction of the de-carbonized gas leaving the MDEA CO_2 separation unit is provided as fuel gas to the combustor to satisfy the heat requirement of the system.

The per pass fuel utilization in the SOFC is limited to about 68% in order to recycle the balance of H_2 required by the catalytic hydro-gasifier. The overall fuel utilization is again much higher, at about 93% due to the recycle.

This case with the near atmospheric SOFC has a net thermal efficiency that is about 9 percentage points lower than the baseline case with the pressurized SOFC, which corresponds to an increase of about 18% in heat rate. Both the gross power generation and the internal power consumption of the pressurized system are higher than those of the atmospheric system. The cathode air compressor in the pressurized case consumes a significant amount of power while the turbine downstream of the SOFC stack produces a significant amount of power and supplies majority of the power required by the compressor. The higher output of the reactor/expander topping cycle for the atmospheric system nearly compensates for the power generated by the steam turbines in the pressurized system.

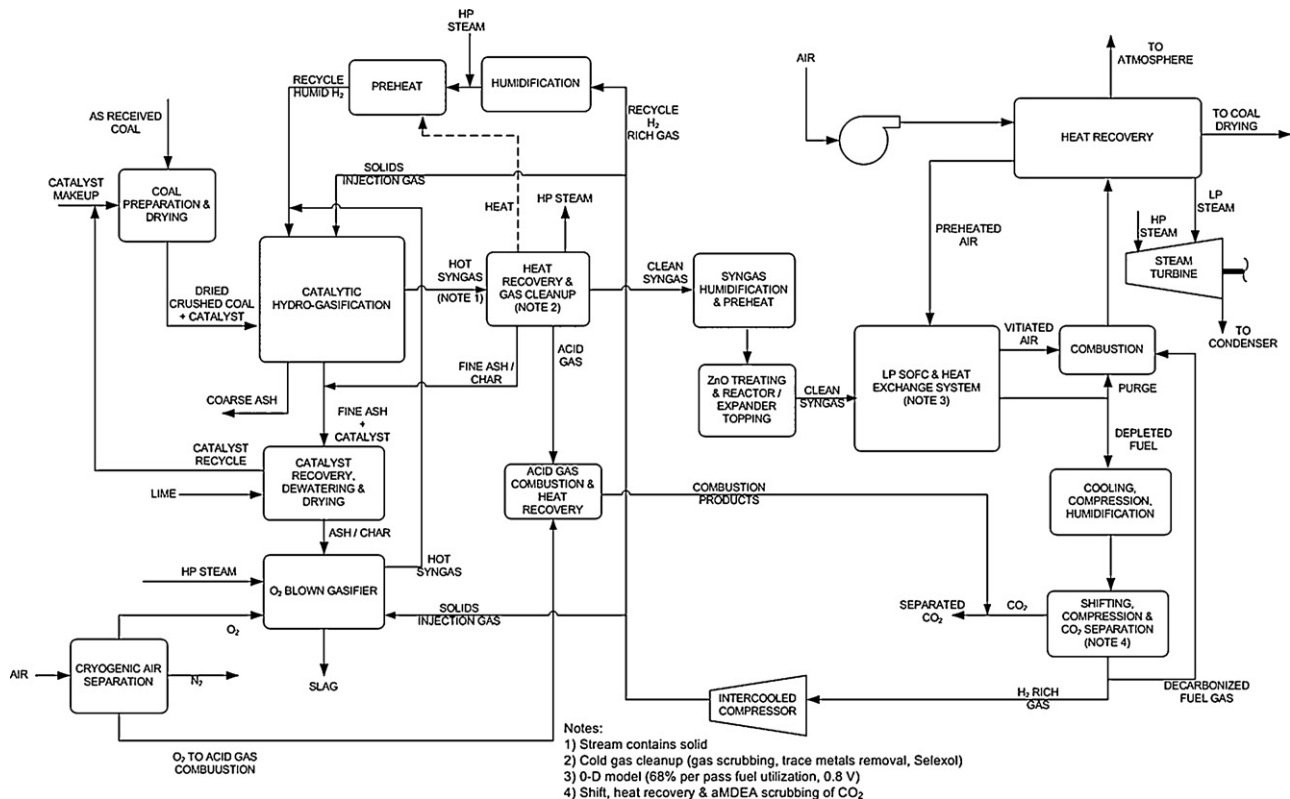


Fig. 6. Block flow sketch—IGFC power only plant with near atmospheric SOFC and anode exhaust recycled to gasifier.

The lower net power output of the atmospheric system is mainly due to the large compression load required for pressurizing the anode exhaust gas upstream of the shift reactor.

It should be pointed out that in this design the positive effect of high pressure on SOFC performance has not been fully demonstrated because a 0-D SOFC model is used and SOFC operating voltages have been maintained constant at 0.8 V for both the near atmospheric pressure case and the pressurized case. In this work the performance differences between the two cases are thus primarily due to the differences in the arrangement of the balance of plant (BOP) equipment as required by the SOFCs with the two significantly different operating pressures. It has been demonstrated experimentally that elevated pressure operation leads to enhanced SOFC stack performance which is more than can be accounted for by Nernst effects alone [30,31] and future research work employing a dimensional SOFC model will further investigate the pressure effects.

6.3. IGFC system without methanation reactor before syngas expander

Finally the baseline system was modified to remove the methanation reactor and leave in a shift reactor in the reactor/expander topping cycle to investigate the effect of this design. It was found that the system's thermal efficiency was reduced slightly to 61.2%. This is because the shift reactor by itself also provides benefit (the shift reaction also being an exothermic reaction) by raising the inlet temperature of the downstream expander to increase its power output [18] although not as much as the methanation reactor. The advantage of the reactor/expander topping cycle becomes more pronounced as the operating pressure of the SOFC is decreased since the pressure ratio of the expander is increased.

However, the inclusion of a methanation reactor in the reactor/expander topping cycle may still be preferred because it helps in controlling the amount of CH₄ in the syngas so that a relatively stable CH₄ content in the syngas fed to SOFC stacks can be maintained when the CH₄ content in the gasifier effluent varies due to variability in coal characteristics as well as plant load factor and transients.

7. Conclusions and recommendations

Coal-based power plants combining catalytic hydro-gasification and SOFC can be configured to achieve high efficiency approaching or exceeding 60% (HHV basis) while separating 90% of the carbon as CO₂. The most promising configuration identified thus far is the system with SOFC operated at elevated pressure and recycling de-carbonized, humidified anode exhaust back to the gasifier. The investigations into the various alternative designs have demonstrated that operating SOFC at elevated pressure provides the most significant efficiency benefit at a system level. The novel design of recycling de-carbonized anode exhaust gas back to the catalytic hydro-gasifier takes full advantage of the synergy between the hydro-gasifier and the SOFC and is also essential to achieving such high system efficiency. The methanation reactor in the reactor/expander topping cycle in the elevated pressure IGFC case does not provide a significant efficiency benefit but it may be a preferred design for better control of the syngas quality.

Each of the IGFC plant configurations introduced above exhibits significantly higher efficiency than an IGCC plant [32]. In addition to high thermal efficiencies, IGFC plants have other advantages over more conventional coal-based power plants. The raw water usage of an IGFC plant is about half that of an IGCC when wet cooling towers are employed for plant heat rejection, the baseline system presented in this work consuming only 0.7 m³ MW h⁻¹ of raw

water. Further, like most SOFC systems, the IGFC plants developed here produce essentially no nitrogen oxide (NO_x) emissions, since no high-temperature combustion of fuel with air takes place.

The catalytic hydro-gasifier and SOFC stacks are the two critical building blocks of such highly efficient IGFC plants. The remainder of the power plants may be configured with subsystems that have been commercially proven in similar services.

The desirable characteristics required of the catalytic hydro-gasifier, one of the two critical technologies required for such high efficiency IGFC plants are: (1) the ability to produce a syngas with high CH₄ content (in excess of 35 mol% on a dry basis), (2) high cold gas efficiency (low syngas exit temperature), (3) the ability to produce a tar- and oil-free syngas, (4) carbon conversion approaching 90%. As introduced in Section 3.2, such catalytic hydro-gasification technology was under development at Exxon in the 1970s and has been attracting significant R&D interest more recently.

The required characteristics for the SOFC stack, the other critical technology required for such high efficiency IGFC plants with CO₂ separation are: (1) operating pressure of approaching 10 bar, (2) separate anode and cathode exhausts, (3) SOFC internal reforming to minimize the excess air used for stack heat management (also without excessive SOFC temperature gradients). Meeting these requirements is not without challenges. The cathode and anode streams must be maintained at similar pressures, as the planar SOFC itself is very thin and cannot support large pressure differentials. Transient pressure spikes on one side of the cell have the potential to damage the SOFC. In the pressurized system presented here, the anode and cathode exhaust streams are kept separate (with the exception that the purge portion of the anode exhaust is combusted with the cathode exhaust), likely presenting a system control challenge. SOFC operating pressure as high as 6 bar has been demonstrated [30,31] but operating pressure around 10 bar has not been reported yet. Meanwhile, the thermal balance within the SOFC is complex and must be evaluated with a spatially resolved SOFC model in order to establish the true minimum air flow required such that maximum temperatures and temperature gradients are not exceeded. Such an analysis is underway and the system will be modified as necessary.

In the baseline case, the SOFC air compressor consumes more power than that generated by the exhaust gas expander and if the two are hooked up on a common shaft, then a motor is required rather than a generator as the case for a typical natural gas-based SOFC-GT hybrid. From a practical perspective, an easier design strategy might be to have the two on separate shafts, with the compressor hooked up to a motor and the expander (turbine) hooked up to a generator. The separation will greatly support and enable better control and dynamic operation of the system. The ability to independently manipulate and control these two components may even be essential to off-design operating conditions such as start-up and shut-down.

Attention should also be paid to the integration between SOFC and gasification subsystems through the recycle of anode exhaust. While this study has demonstrated the efficiency advantage of this design, future research work is recommended to investigate the system dynamic behavior given the large flow rates of the recycle flow.

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