Environmental impact assessment of sustainable hydrogen, steam and electricity trigeneration through integrated gasification and Cu-Cl cycle

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Abstract - This paper performs an environmental analysis of an integrated system that combines gasification technology with a gas turbine (Brayton cycle), steam turbine (Rankine cycle) and copper-chlorine (Cu-Cl) thermochemical water splitting cycle for trigeneration of hydrogen, steam and electricity. The paper focuses on the key environmental performance aspects through an integrated process model of an Integrated Gasification Combined Cycle (IGCC) and thermochemical Cu-Cl cycle, including assessment of CO_2, CO, NO_X, SO_2 emissions and hazardous air pollutant discharge into the atmosphere, release of aqueous effluent that contains hazardous species into water bodies and handling of large quantities of solid ash residues and their potential for leaching toxic substances into the soil and groundwater. Based on the analysis in this study, it is revealed that the proposed trigeneration system is capable of providing sustainable, high-efficiency hybrid energy-hydrogen supply with reduced environmental impact, compared with other commercial technologies.

Keywords — Enviromental assessment, Gasification, Copper-Chlorine (Cu-Cl) cycle, Sustainable, Trigeneration

1 INTRODUCTION

The world energy demand is increasing at a very rapid rate, amidst a crucial need for a significant reduction of greenhouse gas emissions. The concentration of conventional fossil fuel reserves in geopolitically unstable regions of the world represents a challenge to reliability and security of energy supplies [1]. Furthermore, there is a drastic need to decarbonize global energy systems to mitigate the risks associated with climate change [2, 3]. A limited supply of fossil fuel reserves for heat, power, and other usage is leading to an increasingly growing exploration and development of alternative fuels [4]. Hydrogen is a promising clean energy carrier of the future, and potentially major solution to climate change. It is widely used in numerous industry applications [5].

An Integrated Gasification Combined Cycle, IGCC, is a successful integrated process concept that combines modern gasification technology with both gas turbine and steam turbine power generation for the production of electricity, steam and hydrogen. Gasification technology can help address the challenges of the rising global energy demand, while reducing greenhouse gas emissions, and improving energy security by converting domestic low-grade feedstock such as coal, Alberta's oil sands and refinery heavy residues into high-value synthesis gas. Syngas is composed primarily of hydrogen (H_2) and carbon monoxide (CO), which is a precursor, to several energy and chemical plants [6, 7]. The concentration of hydrogen in synthesis gas is variable and strongly depends on the chemical composition of the feedstock, steam and air input, and the gasification temperature [8].

In view of its vast usage in industry, the production of hydrogen with lower cost, more efficient and environmentally benign methods is vital for greenhouse gas reductions. One of the promising emerging technologies is thermochemical water splitting with the copper-chlorine (Cu-Cl) cycle. A thermochemical cycle is a process consisting of a closed loop of thermally driven chemical reactions, where water is decomposed into hydrogen and oxygen, and all other intermediate compounds are recycled. All of the Cu-Cl compounds are recycled internally, with no emissions to the environment [9-12]. The Cu–Cl cycle has numerous advantages over other existing methods of hydrogen production, particularly much lower environmental impact than carbon-based technologies. In comparison to conventional electrolysis, it has a significant margin of superior overall conversion efficiency, with more than one-third improvement over electrolysis. It has much lower operating temperatures than other thermochemical cycles, thereby potentially reducing material and maintenance costs [13]. Also, it can effectively utilize low-grade waste or process heat, thereby improving cycle and power plant efficiencies.

On the one hand, approximately 71.1% of the net heat required for the Cu-Cl cycle is in the form of external heat input, while the remainder is provided by internally recycled heat from exothermic processes [14]. On the other hand, using oxygen instead of air for IGCC is an effective way to improve combustion efficiency in order to reduce the economic cost and environmental aspects of gasification processes, through a significant reduction in NO_X and other greenhouse gas emissions [8]. The innovative

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integration of IGCC and the Cu-Cl cycle confirms their synergetic and complementary roles through a trigeneration system to improve upon both their individual and integrated electricity and hydrogen production efficiencies. In this process integration, the major portion of external heat required for the Cu-Cl cycle is provided by the IGCC plant. The oxygen produced by the Cu-Cl cycle is used instead of air for the IGCC gasification process [15].

Commercial IGCC power plants have proven capable of exceeding the most stringent emissions regulations currently applicable to comparable combustion-based power plants. They have achieved the lowest levels of NOx, SOx and CO pollutant air emissions, of any coal-fuelled power plants in the world [16, 17]. Emissions of trace inorganic and organic hazardous air pollutants (HAPs) are extremely low, compared with those from coal combustion-based plants that use advanced emission control technologies [18, 19]. Discharge of solid by-products and wastewater is reduced by roughly 50% compared with combustion-based plants [20]. Another significant environmental benefit is a reduction of carbon dioxide (CO_2) emissions, by at least 10% for an equivalent net production of electricity, due to higher operating efficiency, compared to existing coal-fuelled, combustion-based power generation technology [21]. If more significant CO_2 reduction is required in the future, gasification technology has major operating advantages that can be exploited to capture CO_2 more efficiently than currently possible with combustion technology.

This paper presents an environmental performance evaluation of integrated IGCC and Cu-Cl cycle trigeneration and compares its performance with other competing coal-based technologies. It is revealed that the IGCC - Cu-Cl trigeneration system demonstrated environmental performance capabilities that allow it to exceed current environmental standards for coal-based combustion systems.

2 INTEGRATED IGCC AND CU-CL CYCLE

2.1 Gasification-based power generation system

Gasification is the conversion of either a solid (coal, coke, biomass, solid waste) or a liquid fuel (oil, tar, pitch) into a gas, often identified as syngas, in which the major components are hydrogen (H_2) and carbon monoxide (CO) [22]. Unlike a combustion process that produces only carbon dioxide (CO_2) and water, gasification is a partial oxidation process that occurs in an oxygen-limited environment. The resulting synthesis gas (syngas) is more useful than combustion flue gas and it has the potential to generate electricity more efficiently.

Modern gasification technologies can be integrated with power generation cycles, acting as a link between coal or heavy fuel oils and gas turbines. Syngas from gasification can be cleaned to very low levels of contaminants, such as sulphur compounds and particulates [23]. After cleaning, syngas can be utilized in gas-steam turbine combined cycle power plants, namely IGCC, which generate electricity more efficiently than traditional combustion-based plants [24]. Fig. 1 depicts the schematic of a generic IGCC power plant.

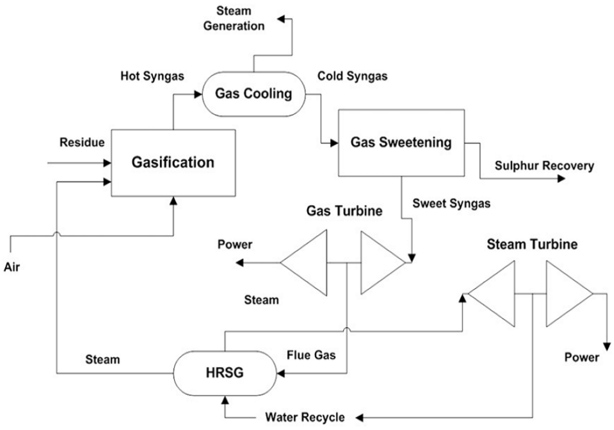

Fig. 1: Schematic of generic IGCC power plant

The resulting process configuration of IGCC is the only power generation technology, including burning coal or high sulphur residues, that can approach the technical and environmental performance of natural gas-fired systems. The environmental impact of IGCC systems can be minimized even further when coupled with carbon capture and storage techniques. The oxygen-fired gasification systems can be designed to produce a material stream composed almost entirely of CO_2, which can be captured and sequestered [25]. The resulting syngas is cooled and cleaned of impurities before combustion in a gas turbine. The resulting hot exhaust gases are then used in a heat recovery steam generator (HRSG) to boil high-pressure steam for expansion in a steam turbine. The low-pressure exhaust from the steam turbine is then recycled back into the HRSG and reused.

2.2 Thermochemical Cu-Cl water splitting cycle

The copper-chlorine cycle (Cu-Cl) cycle is a sequence of processes for hydrogen production by thermochemical water splitting. This cycle has been identified by Atomic Energy of Canada Ltd. (AECL) [26, 27] at its Chalk River Laboratories (CRL) as a highly promising cycle for thermochemical hydrogen production. The Cu-Cl cycle involves four chemical reactions for water splitting, whose net reaction decomposes water into hydrogen and oxygen. All other chemicals are recycled. The Cu-Cl cycle can be linked with nuclear plants and/or other heat sources such as solar and industrial process/waste heat (i.e. incinerators, chemical plants or lost energy from furnaces) to potentially achieve higher efficiencies, lower environmental impact and lower costs of hydrogen production than other conventional technologies [28]. The Cu-Cl cycle is a hybrid process that employs both thermochemical and electrolysis steps with an estimated overall efficiency of 43% [29], excluding the additional potential gains of utilizing waste heat in the cycle. Fig. 2 represents a schematic representation of the Cu-Cl cycle.

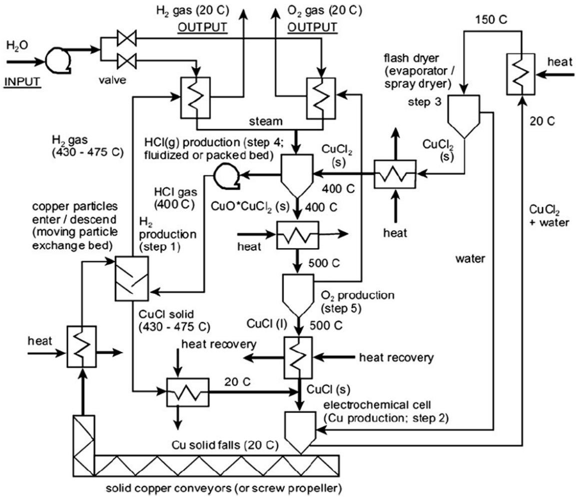

Fig. 2: Schematic representation of a copper-chlorine (Cu-Cl) cycle

In order to increase the efficiency and reduce economic and environmental costs of gasification processes, oxygen can be used instead of input air to improve the combustion efficiency, increase the hydrogen content of produced syngas, and reduce the NOx and other greenhouse gas emissions [15]. This can be achieved by linkage of the gasification and Cu-Cl processes. The proposed approach is to use heat provided by the syngas cooling section of an IGCC plant, previously used for low-pressure steam generation, as the major input of external heat required for the Cu-Cl cycle. Fig. 3 shows a schematic diagram of this integrated process.

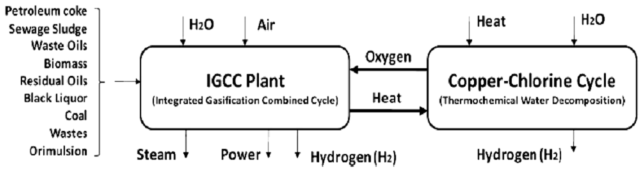

Fig. 3: Schematic representation of an integrated IGCC and Cu-Cl cycle trigeneration system

At a fixed turbine inlet temperature, the IGCC overall efficiency will change if the steam bottoming cycle capacity is modified by integration of the gas cooling unit with the Cu-Cl cycle. The sensitivity analysis of the gasification process shows that by using oxygen from the Cu-Cl cycle, instead of air in the amount of 1.546 × 10^4 (kg/h), it can effectively improve the combustion efficiency and increase the hydrogen content of produced syngas by about 20%. At the same time, it significantly reduces the NOx emissions by around 12.4% [15]. Fig. 4 shows the effect of using oxygen from the Cu-Cl cycle on syngas composition and NOx emissions of gasification processes.

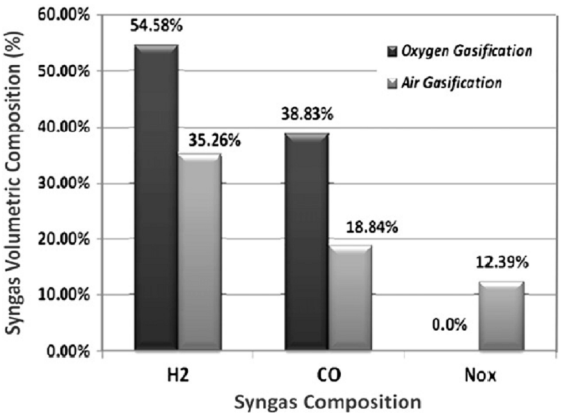

Fig. 4: Effect of using oxygen from Cu-Cl cycle on syngas composition and NOx emissions

By providing the required oxygen for the gasification process, based on simulated operating conditions, production capacity, and total heat required for the Cu-Cl cycle, about 1,933 (kg/h) of hydrogen is produced and 42,515 × 10^4 (kJ/h) of external heat is required. Heat recovery from the syngas cooling unit, which is mainly used for low pressure steam generation of the IGCC plant, can provide 59.3% of the external heat required for the Cu-Cl cycle. This decreases the capacity of the LP steam power generation and steam cycle efficiency, which reduces 2.9% of the overall IGCC plant efficiency. This provides the opportunity of clean sustainable hydrogen production with high efficiency through the Cu-Cl cycle [15].

3 ENVIRONMENTAL PERFORMANCE ASSESSMENT OF INTEGRATED IGCC AND CU-CL CYCLE TRIGENERATION SYSTEM

The environmental performance evaluation of an integrated IGCC and Cu-Cl cycle trigeneration system is presented in this section results for each of the key environmental topics previously mentioned. These results are primarily based on operating experience with IGCC plants that use entrained flow and slagging gasifier technology. The information does not necessarily account for different operating outcomes that may result from the use of other gasification reactor types like a moving-bed and fluidized bed gasifier in IGCC systems. For example, moving-bed gasifiers are much more likely to generate higher levels of organic emissions, such as tars and oils, than entrained-flow and fluidized bed gasifiers, which consequently will impact environmental control requirements and possibly emissions [30]. Therefore, these results cannot cover all IGCC designs, configurations, and feedstocks. While there are similarities between alternative gasification technologies and the manner in which they integrate into IGCC systems, there are also important differences that must be considered when evaluating their operating and environmental performance.

Nevertheless, all that use different gasifier designs have clearly demonstrated that they can be designed to achieve low environmental impact. Since the external required heat for the Cu-Cl cycle is provided by heat integration with an IGCC power plant, it could be considered that Cu-Cl cycle is environmentally benign through the proposed integrated trigeneration system.

3.1 General Environmental Aspects

The main element of any gasification-based system is the gasifier, which can process a wide variety of feedstocks, including coal, biomass, petroleum coke, refinery residues, and other wastes. Minerals in the feedstock (ash) separate and leave the bottom of the gasifier as inert slag or ash, a potentially marketable solid product. Only a small fraction of the ash is typically entrained with the syngas, which requires removal downstream in particulate control equipment.

Potential gaseous pollutants, such as sulphur and nitrogen compounds, form species that can be readily extracted. Hydrogen sulphide (H_2S) and carbonyl sulphide (COS), once hydrolyzed, are removed by dissolution in an organic solvent and converted to valuable by-products, such as elemental sulphur or sulphuric acid. Nitrogen is converted to NH_3, as well as some cyanide and thiocyanate, in the gasifier's reducing environment. It is readily removed via water scrubbing [31].

Most trace pollutants are removed with the slag/bottom ash or the particulate control equipment. Since some pollutants end up in the wastewater, proper water treatment facilities are important for overall environmental performance. Additionally, because CO_2 can readily be recovered in concentrated form with oxygen-blown gasification, CO_2 capture technology can be integrated into IGCC as part of a strategy to reduce greenhouse gas emissions [25].

3.2 Criteria of Air Pollutants

The US Environmental Protection Agency, EPA, designated air pollutants produced by the conversion of coal and other solid carbonaceous fuels (e.g., petroleum coke) in gasification-based power cycles are SO_2, NOx, particulates, CO, and lead. With the exception of lead, which may be introduced into the gasifier as a constituent of the solid fuel feedstock, these pollutants are formed from constituents of the syngas and air as the syngas is fired in the combustion turbine. Upon discharge from the combustor, the hot turbine exhaust gas is cooled in a heat recovery steam generator (HRSG) before being exhausted to the stack. Therefore, these air pollutants become part of the stack gas, and are discharged to the atmosphere. Criteria pollutants may also be emitted in much smaller amounts from equipment installed to treat the tail gas from the sulphur recovery process.

As presented in Table 1, the criteria pollutant emissions from a state-of-the-art IGCC plant will be well below the current Federal New Source Performance Standards (NSPS) for pulverized coal-fired (PC) power plants [16]. Brief evaluations of the criteria pollutant emissions, except lead, and controls are presented below.

Criteria Pollutant	**Expected IGCC Emission Levels (lb/10^6 Btu)**	**NSPS Limit (lb/10^6 Btu)**
SO_2	<0.15	1.2
NOx	<0.1	0.15
CO	<0.033	None

Table 1: IGCC expected emission levels of criteria pollutants [16]

3.2.1 SO_2 Emissions

During high-temperature, entrained flow gasification of coal, most of the sulphur in the coal matrix is released and converted to hydrogen sulphide (H_2S), as well as a small amount of carbonyl sulphide (COS), due to the reduced oxygen environment. These H_2S, COS and particulate contaminants are mostly removed from the syngas prior to combustion or other forms of fuel conversion. Acid gas removal equipment extracts 95-99% of the H_2S and COS from the fuel gas and converts it to a sulphur or sulphuric acid (H_2SO_4) by-product [16]. The small amount of residual sulphur that remains in the syngas is converted to SO_2 in the combustion turbine and released to the atmosphere in the primary stack gas or in the secondary stack gas from the sulphur recovery equipment.

3.2.2 NOx Emissions

The term "NOx" refers to the sum of the nitric oxide (NO) and nitrogen dioxide (NO_2) emissions from a combustion source. While most of the NOx produced during the combustion of syngas is in the form of NO, it is subsequently oxidized to NO_2 in the atmosphere. Unlike natural gas, coal contains chemically-bound nitrogen that forms most of the NOx emissions when it is fired in a typical excess-oxygen environment, such as a utility boiler. Fuel NO typically contributes over 80% of the total NOx emissions in a coal- fired combustion unit, and its formation is highly insensitive to the flame temperature [17]. The gasification process differs significantly from PC plants with respect to the impact of chemically-bound nitrogen in solid fuels, like coal. Gasification, because it operates with a deficiency of oxygen, converts most of the fuel nitrogen into harmless nitrogen gas (N_2). While a small portion is converted to ammonia (NH_3), as well as small amounts of hydrogen cyanide (HCN) and thiocyanate, these water-soluble species are removed during fuel gas cooling and cleaning. They are usually converted to nitrogen in the sulphur recovery process [32]. Therefore, the fuel gas produced is virtually free of fuel-bound nitrogen.

NOx formation is primarily the result of thermal NO produced at high temperatures in the combustion turbine. By maintaining a low fuel-air ratio (lean combustion) and adding a diluent like steam, the flame temperature can be lowered to reduce the potential for NOx formation.

3.2.3 CO Emissions

CO emissions are typically the result of incomplete combustion, but can also result from fugitive emissions. In an IGCC system, sources are typically the gas turbine, sulphur recovery unit tail gas incinerator, and the flare system and equipment leaks [20].

3.3 Hazardous Air Pollutants (HAPs)

Potential trace substance emissions from coal-fuelled power plants include ionic species, trace elements, and trace organic compounds. These trace substances can be emitted in the flue gas, aqueous discharges, and solid effluents. Ionic species of environmental concern in the effluent streams of coal-fuelled power plants include sulphate, nitrogen-containing ions like nitrate and ammonium, chloride, fluoride, phosphate and cyanide. The ionic forms of these species in stack gases are present only in the aerosol phase [33]. Chloride and fluoride, however, can exist as acids and, thus, can appear in the gas phase as well. Stack emissions of all ionic species are reduced to very low levels by using particulate and acid gas control equipment.

3.4. Aqueous Effluents

The integrated IGCC plant has two principal water effluents that are similar to those in PC plants. The first is wastewater from the steam cycle, including blowdowns from the boiler feedwater, purification system and the cooling tower. Gasification processes typically purify and recycle raw process streams, and net water discharge is normally only a blowdown stream. These effluents contain salts and minerals that have been concentrated from the raw feedwater. The second aqueous effluent is process water blowdown, which is typically high in dissolved solids and gases along with the various ionic species washed from the syngas, such as sulphide, chloride, ammonium, and cyanide [17]. The process water blowdown is typically recycled to the feed preparation area for solid feedstocks, to the scrubber after entrained solids have been removed, to a zero discharge water system, or to a wastewater treatment system.

In general, water effluents may create fewer problems for IGCC than for PC power generation, because the steam cycle in an IGCC plant produces less than 40% of the power plant's output [15]. Therefore, effluents from boiler feedwater preparation and cooling-water blowdown are significantly less. However, the amount of process water blowdown is about the same for both gasification and PC combustion.

3.5 Solid Byproducts

In terms of quantities of waste material produced, as well as the potential for leaching of toxic substances into the soil and groundwater, IGCC power generation has demonstrated minimal environmental impact [32]. The largest solid waste stream produced by recent IGCC installations is slag, black, glassy, sand-like material that is potentially a marketable byproduct. Slag production is a function of ash content, so coal produces much more slag than an alternative fuel like petroleum coke. Regardless of the feed, as long as the operating temperature is above the fusion temperature of the ash, slag will be produced [16].

The other large-volume byproduct produced by IGCC plants is solid (or liquid) sulphur or sulphuric acid, both of which can be sold to help offset plant operating costs. In comparison, most coal combustion plants recover sulphur as wet scrubber sludge, dry or semi-dry spent sorbent. These sulphur forms have significantly larger mass and volume than pure sulphur. They are often more difficult to handle and market, and must usually be disposed in an appropriate landfill or surface impoundment [20].

3.6 CO_2 Emissions

The largest contributor to greenhouse gas (GHG) emissions from integrated IGCC and Cu-Cl cycle is the production of CO_2 from the carbon originally contained in the fuel fed to a gasifier. The production of other GHG emissions, such as N_2O and NH_3, are small compared with CO_2. Although CO_2 emissions are higher than natural gas-fired plants, IGCC's improved efficiency reduces CO_2 emissions relative to conventional PC plants [21].

The syngas has a high CO_2 concentration, which can be further increased by converting CO to CO_2 prior to combustion while simultaneously producing more hydrogen. IGCC gasifiers typically operate under relatively high pressure around ~400 psig [34]. This makes recovery of the CO_2 from the syngas much easier than capture from flue gas.

A recent study of one design concept concluded that 75% of the CO_2 could be captured from an IGCC plant with only a 4% loss in efficiency [21]. This does not account for transport of the CO_2 to a utilization or sequestration site and further processing.

4 CONCLUSIONS

In this paper, a proposed innovative integrated IGCC and Cu-Cl cycle trigeneration system shows its components have synergetic and complementary roles to improve upon both their individual and integrated electricity and hydrogen production efficiencies. Also, it is revealed that this trigeneration system can provide sustainable, affordable and high-efficiency energy production with minimal environmental impacts.

The integrated IGCC and Cu-Cl cycle systems can economically meet strict air pollution emission standards, produce water effluent within environmental limits, and produce environmentally benign hydrogen, with good potential to recover a valuable sulphur commodity by-product.

The outstanding environmental performance of the integrated system makes it an excellent technology for the "green" production of hydrogen, steam and electricity. It also provides flexibility in low-cost, widely available feedstocks from refinery wastes, to coal and Alberta's oil sands. The integrated IGCC and Cu-Cl cycle can provide an energy production alternative that is more efficient and environmentally friendly than other competing energy production technologies.

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