Natural gas decarbonization for low emissions bitumen recovery – Exergetic performance assessment

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Abstract:

We present in this paper an exergy-based performance assessment of a simple yet powerful method to reduce CO_2 emissions, water consumption and steam requirements of *in situ* bitumen recovery. This process involves natural gas decarbonization (NGD) prior to combustion, thus removing carbon from the process upfront – it is not a carbon and storage capture but a carbon sequestration technology. NG decarbonization requires high temperature heat, which can be supplied by oxycombustion of NG. A CO_2-rich flue gas stream from oxycombustion can be co-injected with steam into the reservoir to both sequester CO_2 and reduce *in situ* steam demand. Decarbonizing NG fuel to hydrogen avoids downstream CO_2 emissions by producing carbon black, a stable form of carbon. The hydrogen product of the process can be used as either a fuel for steam generation and/or hydrotreating. A similar method involves an auto-thermal NGD which uses a portion of the hydrogen product from NGD to provide heat required by the NGD reaction. In this paper, we use the produced hydrogen to generate steam by combustion, a process that produces water that offsets a significant fraction of process water losses from bitumen recovery. Processes that reduce CO_2 emissions could also exert significant energy penalties, and so, this process comes with some energy. We use exergy analysis (2nd Law of Thermodynamics) to determine the energy efficiency and performance of this low emissions steam generation process for bitumen recovery. The performance of the NGD-based processes is compared with that of the current steam generation method using once-through stream generators (OTSGs). We use the results of the analysis to quantify exergy losses and explore ways to reduce these losses. This study and its findings will be relevant to determine the thermodynamic minimum energy and operating cost requirements of novel approaches to decarbonize the oil sands industry.

Keywords:

Natural gas decarbonization, oil sands bitumen recovery, steam generation, exergy analysis, performance assessment.

1. Introduction

Even as evidence for global warming accumulates, the dependence of civilization on the use of fossil fuels for energy makes an appropriate response difficult [1]. Mankind's dependence on liquid transportation fuel produced from crude oil is a major contributor to climate change and this is heightened by a shift from conventional crude oil to unconventional heavy oil. To stabilize atmospheric greenhouse gas emissions (GHGs) to levels below a global temperature rise of 2°C, fast development and wide adoption of climate mitigation technologies is crucial [2, 3]. Climate mitigation technologies such as carbon capture and storage (CCS) could allow mankind to continue to use abundantly available fossil fuels in the short term and at the same time become a bridge to a sustainable energy future [2]. In this regard, there is a pressing need to reduce the energy intensity

and GHG footprint of liquid transportation fuels, especially those associated with oil sands (tar sands). High energy intensity and GHG emissions come mostly from bitumen extraction and its subsequent upgrading to synthetic crude oil (SCO). This is mostly due to thermal oil recovery operations where large amounts of steam are injected *in situ* to mobilize bitumen. Production of SCO results in 16.2-28.7 g CO_2 equivalents (CO_2eq) per MJ of GHG emissions whereas conventional crude oil production emits 4.5-9.6 g CO_2eq/MJ [4].

Several technologies, e.g. carbon capture and geological storage (CCGS) and mineral carbonation [2], have been proposed as viable options to decarbonize the energy system. In most processes, a pure stream of CO_2 must be separated from the steam generator or power plant flue gases. The produced CO_2, after it has been compressed to pipeline pressure (>120 bar), can be used for enhanced oil recovery (EOR) or sequestered underground in geological aquifers. However, the processes to capture CO_2 from flue gases, compressing it to >120 bar, and transporting it to storage sites are energy expensive and results in significant GHG emissions. For example, current state-of-the-art CO_2 capture – conventional amine-based processes – has an energy (mainly heat) penalty of 3-4 GJ/ton CO_2 [5].

The technology assessed here embodies a method to decarbonize oil sands operations by producing hydrogen and environmentally benign solid carbon (carbon black) from thermal decomposition of natural gas. The produced hydrogen can be burnt as fuel to generate steam whereas the carbon black can either be sold or reprocessed to a commercially viable nano-carbon product or stored safely above the ground. Though hydrogen and carbon black production from thermal decomposition of methane is a well-known technology [6], this technology has been adapted for natural gas decarbonization (NGD) [7-10] and recently proposed for integration with oil sands operations to achieve ultra-low emissions of *in situ* bitumen recovery and upgrading [11, 12]. The conventional method to generate steam for bitumen recovery is by using once-through steam generators (OTSGs) and heat recovery steam generators (HRSGs) fuelled by natural gas (NG). Steam generation by these devices generates high GHG emissions.

Natural gas decarbonization integrated with *in situ* bitumen recovery is designed to generate steam for bitumen recovery using steam-assisted gravity drainage (SAGD) or cyclic steam stimulation (CSS) processes [13]. The motivation of research in this area stems from the fact that NGD process could offer significant improvements over existing conventional approaches of steam generation. These benefits include: i) a reduction of net carbon emission by >90%, ii) an increase in energy recovery efficiency by >30%, and iii) a decrease of the water consumption footprint by 0.6 m^3 water per unit m^3 oil produced [12].

In this study we assessed on an exergy basis the energetics and performance of the NGD-based processes: i) auto-thermal NGD for steam generation process and ii) integrated NGD and oxycombustion for steam generation process, and compared those to the performance of current steam generation method using OTSGs. The process was modelled using UniSIM Design® software and the material and energy balance results of each unit operations extracted and used to compute the exergy values and the associated process CO_2 emissions. The business as usual case where OTSGs are used to generate steam for *in situ* bitumen recovery was modelled and used as a basis for comparing the process exergy analysis results. We used exergy analysis results to compare energy analysis results on the basis associated of carbon reduction benefits, energy penalties and process efficiency losses. Additionally, we quantified exergy losses within and outside the system – the knowledge of these could help direct focus on areas to improve process efficiency.

2. Process description and modelling

2.1. Business as usual (BAU) SAGD steam generation case

The conventional process of steam production using OTSGs, referred as the BAU case was modelled and its results served as a basis of exergetic performance comparison for the NGD process. A UniSIM Design® simulation model of steam generation using OTSGs is presented in Fig. 2. The BAU case was modelled using Peng-Robinson as the equation of state, and the mass and energy balances calculated on the production basis of 33,000 barrels per day (bpd) of bitumen using SAGD. We assumed a steam-to-oil ratio (SOR) of 3 m^3/m^3 (cold water equivalent).
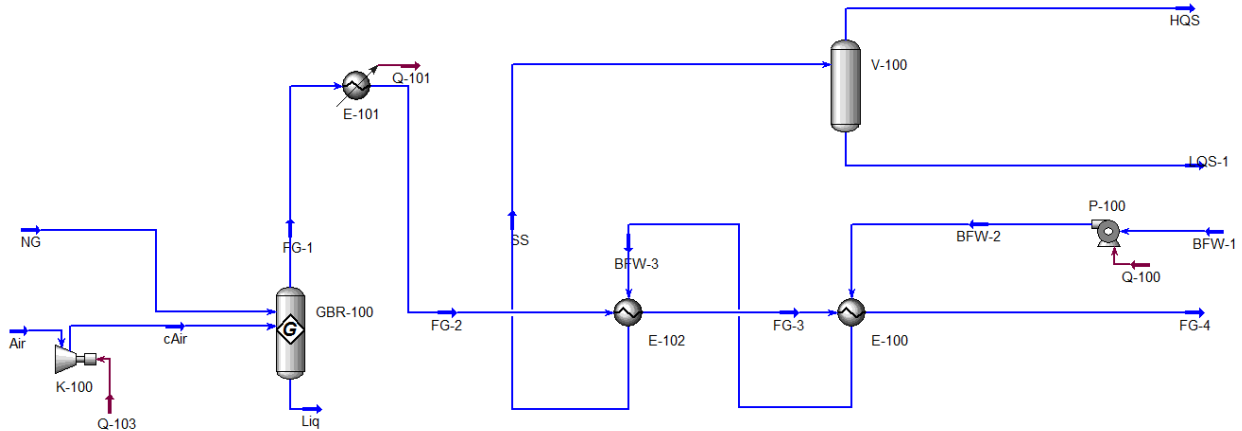

Fig. 2. PFD of the traditional SAGD steam generation method using OTSG boiler. Flow rates, temperatures and pressures of streams are listed in the Appendix.

Air is compressed from atmospheric conditions (15°C, 1 bar) to 2 bar using a compressor (Unit K-100). Natural gas (NG) and compressed air (cAir) were fed to a boiler where NG combustion was simulated with a Gibbs reactor (Unit GBR-100). Using heat exchanger E-100 (Stream FG-1), we accounted for energy loss from combustion due to radiation (~1.5% of combustion heat). Boiler feed water stream (Unit BFW-1) pressure was raised from 1 bar to 35 bar using a feed water pump (Pump P-100) and was preheated with a flue gas stream (Stream FG-3) in a heat exchanger (Unit E-100). The BFW stream (Unit BFW-3) was heated in Unit E-102 to generate saturated steam (SS) with properties: pressure, P=35.6 bar, temperature, T=244°C and quality, x=79%. The SS stream was flashed in Unit V-100 to produce high quality steam (HQS, quality = 1) which is used in SAGD. The saturated steam stream (LSQ-1) from Unit V-100 can further be flashed to produce steam for utilities (not included in this model).

2.2. Low emissions bitumen recovery – integrated NGD and steam generation processes

The purpose of the NGD reactor is to produce hydrogen for steam generation. The reaction taking place at the NGD reactor can be represented by:

$$CH_4 + Heat \rightarrow C + 2H_2 \qquad (\Delta H = 75.6 \text{ kJ/mol}) \qquad (1)$$

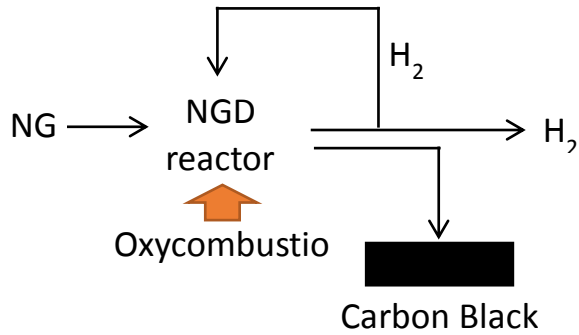which assumes that the NG consists of methane.

Fig. 2. A simple flow diagram of the NGD process illustrating the use of oxycombustion or a fraction of produced hydrogen used as the NGD heat source.

Hydrogen produced from NGD as depicted in Fig. 2 can be used in OTSGs or HRSGs to generate steam with the combustion product being water. As shown in Fig. 2, two possible methods to provide for heat requirements of the NGD reaction are via auto-thermal and the oxy-NG-fired processes. We carried out exergy analysis of both options and compared the results with those of the BAU process. The processes were modelled with UniSIM Design® software using the Peng-Robinson equation of state. The same basis of process calculation used for the BAU case was applied here.

2.2.1. The auto-thermal NGD process for steam generation

This method could be self-sustaining energetically by using ~14% of the produced hydrogen to produce the heat required for the NGD reaction. More so, by avoiding the use of external heat sources that generate CO_2 emissions, this method has considerably low emissions.

Figure 3 presents a UniSIM Design® PFD of the auto-thermal NGD process and steam generation for *in situ* bitumen recovery. Thermal decomposition of NG takes place in NGD reactor (CRV-100) to produce hydrogen and carbon black at ~890ºC. Natural gas (Stream NG-1) delivered to the plant at 15ºC and 5 bar is compressed to 35 bar (211ºC) by using compressor K-100. It was assumed that the NG feed is 100% methane. Carbon black (Carbon) is separated from the reactor bottom whereas a hot vapour stream (H2-1) containing hydrogen is used to heat the NG feed stream (NG-1+) to 800ºC (NG-1++) by using an exchanger (Unit E-100). After cooling, the hydrogen product (H2-2) from Unit E-100 is divided into two parts, Streams H2-2 and H2-4. Both hydrogen streams are combusted in air, a reaction modelled by using an equilibrium reactor (Units ERV-100 and ERV-101) as direct contact boilers. Air streams, Air-1 and Air-2 are compressed to 35 bar in Units K-103 and K-102, respectively, before being fed to the boilers. By using heat exchanger E-101, heat is extracted from the product stream (HT-heat) of Unit ERV-101 and used to supply the heat requirements of the NGD reaction.
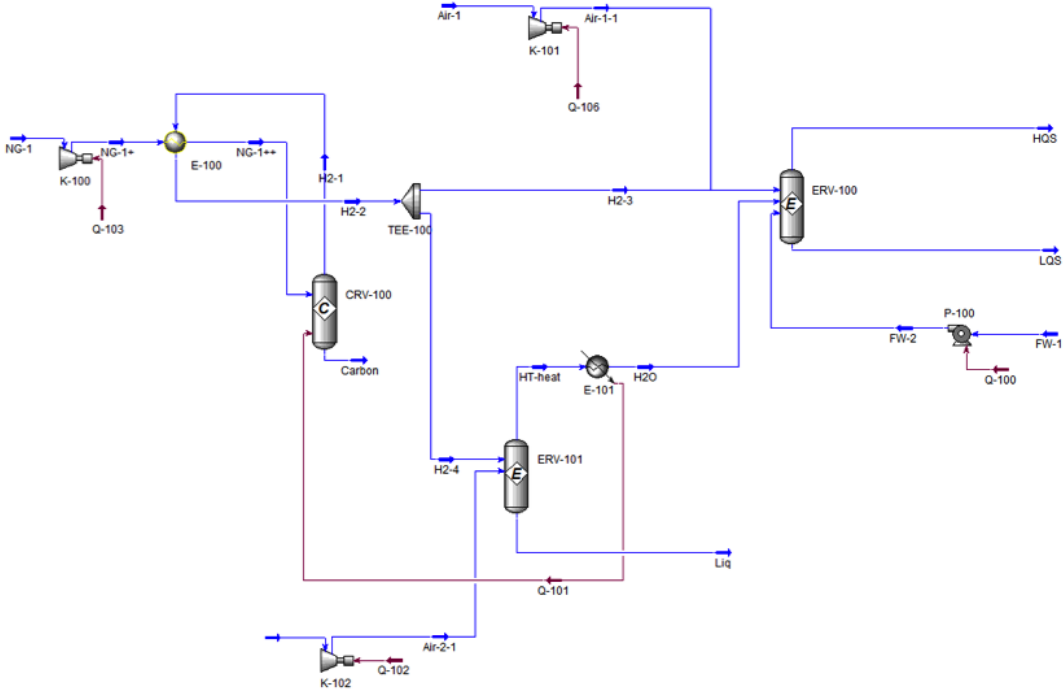
Fig. 3. Process flow diagram (PFD) for steam generation for in situ bitumen recovery process using auto-thermal NGD and hydrogen combustion. Flow rates, temperatures and pressures of streams are listed in the Appendix.

The cold outlet stream of Unit E-101 is fed to Unit ERV-100. This stream can also be mixed separately with the high quality steam product of Unit ERV-100. The pressure of boiler feed water (FW-1) is raised from 1 to 51 bar by using a pump (P-100) and fed to the direct contact boiler Unit ERV-100. Stream FW-1 is used to generate additional steam (besides the steam from hydrogen combustion) by using the heat released from hydrogen combustion.

The boiler produces HQS and LQS at ~35 bar. The HQS (100% quality) is used for *in situ* bitumen recovery by injecting it into oil sands reservoir whereas the saturated, high pressure LQS can be flashed downstream to generate heat to meet process utility needs.

2.2.2. The integrated oxycombustion and NGD process

In this method, oxygen produced in a cryogenic air separation unit (ASU) is used to combust NG fuel to provide heat for the NGD reactor. Since the NGD process requires 75.6 kJ/mol CH_4 from a high temperature source, oxycombustion of NG was integrated with the NGD process to provide for its energy requirements. Figure 4 shows a UniSIM Design® PFD of the NGD process and its integration with steam generation for *in situ* bitumen recovery. Thermal decomposition of NG takes place in NGD reactor (Unit CRV-100) to produce hydrogen and carbon black at a reaction temperature of 920 °C. Natural gas (NG-1) delivered to the plant at 15°C and 5 bar is compressed to 35 bar (211°C) by using a compressor (Unit K-100). Carbon is separated from the reactor bottom whereas the hot vapour stream (H2-1) containing mainly hydrogen is used to heat the natural gas feed stream (NG-1$_+$) to 800 °C (NG-1$_{++}$) by using an exchanger (E-100). After cooling, the hydrogen from Unit E-100 is fed to a direct contact hydrogen combustor (Unit ERV-100). Oxygen, via Stream O2-2, with composition, 95 vol% O_2 and 5 vol% N_2, was delivered at 15°C and 5 bar and is compressed to 35 bar (O2-2$_+$) in a compressor (K-103) before it is fed to ERV-100.

Direct contact boilers avoid heat transfer losses to tube surfaces and achieve high thermal efficiencies (close to 100%) because they combine the combustion process and feed water heating in a single process unit. There is no flue gas stream and thus, no heat is lost to the stack. The pressure of boiler feed water (Stream FW-1) is increased from 1 to 50 bar by using a pump (P-100) and afterwards the stream is preheated (from 18 to 265°C) in Unit E-102 before it is fed to the direct contact boiler. The boiler is designed to produce high quality steam (HQS) and low quality steam (LQS) at 35 bar. The HQS is ~100% quality (for bitumen recovery) whereas the saturated, high pressure LQS can be flashed downstream to generate heat to meet plant utility needs.
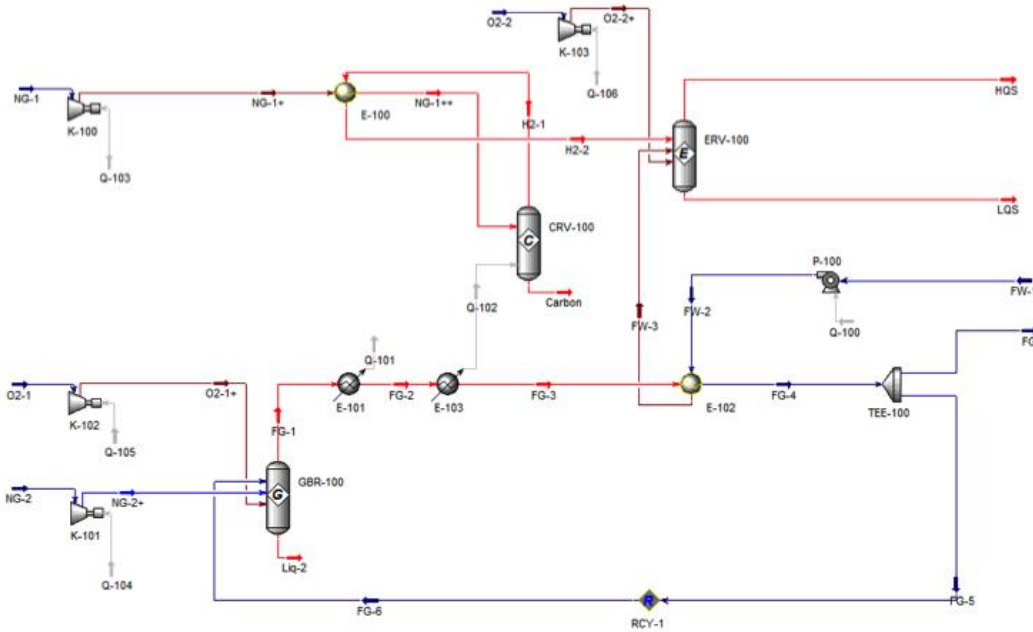

Fig. 4. Process flow diagram (PFD) for steam generation for in situ bitumen recovery process using integrated NGD, oxy-natural gas combustion and hydrogen combustion. Flow rates, temperatures and pressures of streams are listed in the Appendix.

Oxycombustion supplies the heat requirements of NGD via heat exchanger E-103. Oxycombustion is modelled by feeding compressed NG (NG-2→NG-2$_+$) and oxygen (O2-1→O2-1$_+$) streams into a Gibbs free reactor (GBR-100). In this reactor the combustion reactions of NG components (95 vol% CH_4, 2.5 vol% C_2H_6, 0.2 vol% C_3H_8, 1.6 vol% N_2 and 0.7 vol% CO_2) occurs with the oxygen stream containing 95 vol% O_2 and 5 vol% N_2. Heat losses to radiation in the combustor, assumed to be 1.5% of the heat of combustion, is accounted for by using a cooler (E-101). After supplying the heat requirements of NGD, the hot flue gases stream (FG-3) are used to heat the boiler feed water stream (FW-2→FW-3) to 265°C (50 bar, vapour fraction=0). At Unit ERV-100, Stream FW-3 is vaporised. To regulate the combustion flame temperature, 80% of the flue gas stream (FG-4→FG-5) is recycled to the oxycombustor whereas the rest (FG-4→FG-6) can be flared or co-injected with steam into oil sands reservoirs for EOR and CO_2 storage. In this study, the flue gas stream is flared.

3. Exergy analysis

Unlike energy analysis which is based on the First Law of Thermodynamics (FLT), exergy is a construct based on the Second Law of Thermodynamics (SLT). Exergy analysis is useful to identify the causes, locations and magnitudes of process inefficiencies [14]. Exergy of a closed system can be expressed as the summation of the physical (Ex_{ph}), chemical (Ex_{ch}), kinetic (Ex_{kin}) and potential (Ex_{pot}) components of the flow system.

$$Ex = Ex_{ph} + Ex_{ch} + Ex_{kin} + Ex_{pot} \tag{2}$$

For a closed system, the non-flow physical exergy can be expressed as:

$$Ex_{non-flow} = (U - U_0) + P_0(V - V_0) - T_0(S - S_0) \tag{3}$$

whereas for a flowing stream of matter the physical exergy is expressed as:

$$Ex_{flow} = (H - H_0) - T_0(S - S_0) \tag{4}$$

and for an ideal gas, the physical exergy is expressed as:

$$Ex_{ideal\ gas} = (H - H_0) + RT_0 \cdot \ln(\frac{P}{P_0}) \tag{5}$$

where $-(S - S_0) = R \cdot \ln(\frac{P}{P_0})$ $\tag{6}$

The system whose exergy is expressed in (3)-(6) has at temperature T and pressure P, a specific entropy S, and a specific enthalpy H, whereas at these properties at environmental state are T_0, P_0, S_0, and H_0, respectively [14]; R = 8.314 J/mol-K. For Q amount of heat extracted from an external heat source at a temperature T (K) and environmental conditions, T_0 = 298.15 K, P_0 = 1.01325 bar, the exergy the heat transferred is given by

$$Ex = Q \cdot \frac{T - T_0}{T} \tag{7}$$

This represents the conversion of heat to work using the Carnot efficiency factor ($1 - T_0/T$). For power, P, the exergy is equal to the energy. The chemical exergy E_{ch} of compounds is the exergy content of the chemical substance at environmental conditions. The standard chemical exergy of a chemical compound or element can be calculated by means of the exergy balance of a reversible standard reference reaction [15]. Given that the reactants taking part in a standard chemical reaction appear separately in standard state, chemical exergy can be expressed as:

$$Ex_{ch} = \Delta_r G^o + \sum_{i=1}^{n} Ex_{products} - \sum_{i=1}^{n} Ex_{reactants} \tag{8}$$

where $\Delta_r G^o$ is the normal standard free energy of the reference reaction, $\sum_{i=1}^{n} Ex_{products}$ and $\sum_{i=1}^{n} Ex_{reactants}$ are the sums of the normal standard chemical exergies of the products and reactant species, respectively [15]. The chemical exergy values of compounds were extracted from refs. [15] and [16]. The effects of pressure on the Ex_{ch} of liquids and solids are assumed to be negligible since the enthalpies of these states are independent of pressure. Exergy destruction (or exergy losses, Ex_{loss}) of a system is the measure of the amount by which a resource is irreversibly consumed or degraded. Ex_{loss} is essentially the sum of the entropy generation S_{gen}, for process units multiplied by T_0:

$$Ex_{losses} = T_0 \sum(S - S_0) = T_0 \sum S_{gen} \tag{9}$$

3.1. SAGD steam generation – The BAU bitumen recovery case

The exergy inputs and outputs of the BAU case are shown in Fig. 4.

The exergy balance around the OTSG can be presented as:

$$Ex_{NG} + Ex_{air} + Ex_{power} + Ex_{water} = Ex_{FG} + Ex_{HQS} + Ex_{LQS} + Ex_{loss} \tag{10}$$

The main inputs to the OTSG (Fig. 4) are NG, air, feed water and electricity, and their exergy contents are represented by Ex_{NG}, Ex_{Air}, Ex_{Power} and Ex_{Water}, respectively.
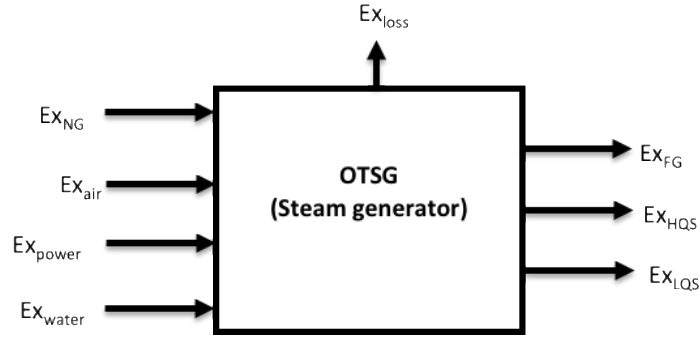
Fig. 4. Block diagram of the exergy balance around a traditional steam generation plant. HQS, LQS and FG signify high quality steam, low quality steam and flue gases, respectively.

On the other hand, the main outputs are HQS, LQS, flue gases and losses (both external and internal system losses) represented by Ex_{HQS}, Ex_{LQS}, Ex_{FG} and Ex_{loss}, respectively.

3.2. Low emissions bitumen recovery – integrated NGD and steam generation processes

The exergy balance around the direct contact boilers, shown in Fig. 5, can be presented as:

$$Ex_{NG} + Ex_{O_2/Air} + Ex_{power} + Ex_{water} = Ex_{FG} + Ex_{HQS} + Ex_{LQS} + Ex_{loss} \qquad (11)$$
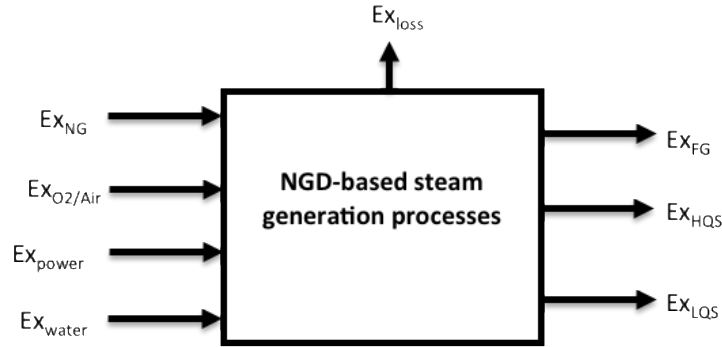
Fig. 5. Block diagram of the exergy balance around the NGD-based steam generation processes. HQS and LQS signify high quality and low quality steam, respectively.

The main inputs to the low-emissions NGD-based steam generation processes are NG, oxygen or air, feed water and electricity, and their exergy contents are represented by Ex_{NG}, $Ex_{O2/Air}$, Ex_{Water} and Ex_{Power}, respectively (Fig.5). The main outputs are flue gas, HQS, LQS and losses (both external and internal) are represented by Ex_{FG}, Ex_{HQS}, Ex_{LQS} and Ex_{loss}, respectively.

3.3. Process exergy efficiency and CO_2 emissions intensity

We compare the results of the FLT energy efficiency to SLT exergy efficiency of the process. The performance of SAGD process can be well predicted by the thermal efficiency of the steam

generation system [12]. Based on an energy balance, the energy efficiency of a steam generation process is given by:

$$\eta_S = \frac{\dot{m}_{steam} \cdot H_{steam}}{\dot{m}_{NG} \cdot LHV_{NG} + E_{other}} \tag{12}$$

where $\dot{m}_{steam} \cdot H_{steam} = \dot{m}_{LQS} \cdot H_{LQS} + \dot{m}_{HQS} \cdot H_{HQS}$ (13)

The first and second numerator terms represent the mass flow (kg/s) and mass enthalpy (kJ/kg) of steam, respectively whereas the first and second denominator terms represent mass flow (kg/s) and low heating value of NG, respectively.

$$E_{other} = \dot{W}_{Compressor} + \dot{W}_{Pump} + E_{ASU} \tag{14}$$

The term E_{other} is the sum of energy requirements related to auxiliary equipment such as compressor work ($\dot{W}_{Compressor}$), pump work ($\dot{W}_{Pump}$), and oxygen production energy requirements (E_{ASU}). On the other hand, exergy efficiency of a steam generation system based on using exergy analysis is given by:

$$\eta_{Ex} = \frac{Ex_{LQS} + Ex_{HQS}}{Ex_{NG} + Ex_{O_2/Air} + Ex_{power} + Ex_{water}} \tag{15}$$

Carbon dioxide intensity was calculated as the mass (kg) of CO_2 emitted per unit volume (m^3) of bitumen produced from oil sands. This is the sum of the CO_2 emitted by the process and that from electricity used by process water treatment plants and other auxiliaries. The electricity used by water treatment units and other auxiliaries (downhole pumps, pad auxiliaries, glycol system, evaporators, etc.) are estimated to be 6.4 kWh/bbl bitumen processed. We assumed that NG cogeneration was used to generate electricity, with CO_2 intensity of 87 kgCO_2/GJ electricity.

4. Results and discussion

We present in this section comparative results of energy, exergy and CO_2 emissions intensity of the BAU case, integrated oxycombustion and NGD process, and the auto-thermal NGD process.

It can be observed from Fig. 6 that the exergy efficiencies of steam generation are lower than efficiencies calculated on an energy basis. The exergy efficiencies values are a better reflection of the thermodynamic minimum energy requirements of the system. Exergy losses are calculated as the sum of the internal and external system losses divided by the net exergy inputs expressed as a percentage. Results show that the exergy losses are a reflection of the exergy efficiency performances. The sum of the exergy efficiency (%) and the exergy losses (%) should be equal 100. On the other hand, exergy penalty and CO_2 reduction potential account for the reduction (%) in exergy efficiency and process CO_2 intensity relative to the BAU case resulting from the application of the novel NGD processes, respectively. As a result of using BAU case as a basis, its energy penalty and CO_2 reduction potential values are zero in Fig. 6.

4.1. SAGD steam generation – The BAU bitumen recovery case

The production basis of a 33,000 bpd bitumen (with SOR=3 m^3/m^3) from oil sands using the BAU case requires about 1,037 tons/d NG. About 19879 ton/d of feed water were transformed into 79% quality steam, which were separated into 15,755 tons/d HQS and 4,124 tons/d LQS. The exergy analysis results of the BAU case are presented in Table 1. Both the physical and chemical exergy components of the inputs and outputs of the process are presented. Ex_{inputs} is ~2104.2 GJ/h whereas $Ex_{outputs}$ is ~1243.5 GJ/h, resulting in ~547 GJ/h exergy losses within the system.
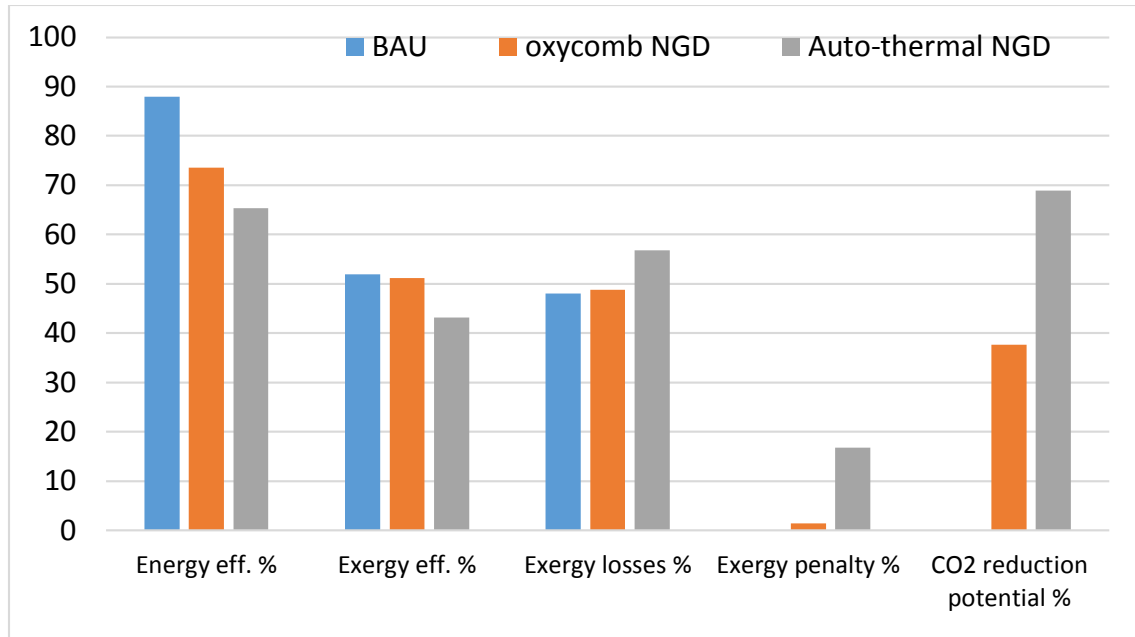
Fig. 6. Energy, exergy and CO_2 emissions performance of steam generation using the integrated oxycombustion and NGD process and the auto-thermal NGD process compared with the BAU case.

The flue gas stream is considered an out of the system albeit it is a waste heat stream. If the exergy in the flue gas is not included in the total output exergy of the process, then the exergy losses are ~1004.2 GJ/h. This reduces the total output exergy to ~1100.1 GJ/h, which is the exergy of HQS and LQS. The results show that internal and external exergy losses constitute about ~47.7% of the energy of the process. Figure 6 shows that the energy efficiency of this steam generation process is 52% whereas the exergy efficiency was calculated to be 87.9%. It was not included in the energy efficiency calculation. The process CO_2 intensity of the BAU case is 567 kg-CO_2/m³ bitumen with no potential for CO_2 emissions reduction.

4.2. The auto-thermal NGD process

In the auto-thermal NGD process, ~1,576 tons/d NG is fed to the NGD reactor. 20% of the hydrogen products was combusted separately in ERV-101 and used to generate 400 GJ/h of high temperature heat supplied to the NGD reactor. The remaining 80% of the hydrogen product was combusted in ERV-100. About 30,557 tons/d of feed water were fed directly into ERV-100 together with the combustion products from ERV-101.

The main outputs from the process are HQS and LQS streams. The LQS streams contains saturated steam whereas the HQS contains superheated vapor comprising of 61.6% steam, 38.8% N_2 and 1.5% oxygen. The mixed composition of the HQS stream comes as a result of the air used as oxidant streams for the hydrogen combustion boilers ERV-100 and ERV-101. We accounted for the mixed composition of HQS by considering the exergy contents of each of the components. Hydrogen produced from the auto-thermal NDG process generates 3,548 tons/d steam via combustion in addition to the steam generated from the feed water stream. The quantity of steam in the HQS stream is 12,942 tons/d whereas in the LQS stream it is 21,163 tons/d. The results of

the exergy analysis of the auto-thermal NGD process are listed in Table 2. In Table 2 we list both physical and chemical exergy components of the inputs and outputs of the process. The net exergy inputs, Ex_{inputs} is ~3,784.9 GJ/h whereas the net exergy outputs equal ~1,607.4 GJ/h. This implies that ~1,2177.4 GJ/h were exergy losses within system. Here, there is no flue gas generated. The products of the process were usefully deployed either for in situ bitumen recovery or for bitumen processing utilities. The energy efficiency of this steam generation process is 65.3% whereas the exergy efficiency is calculated as 42.5%. If we consider only steam generated, that is if we exclude the exergy content of other gases (N_2 and O_2) in the HQS stream, the exergy efficiency reduces by 3%-points. Our results show that the auto-thermal process results in an exergy efficiency reduction of ~9.8%-points relative to the BAU case.

Table 1. Exergy inputs, outputs and losses in the OTSG

	Ex_{ph}	Ex_{ch}	Ex
	GJ/h	GJ/h	GJ/h
Inputs			
NG	28.2	2181	2209
Air	3.1	43.0	46.1
Water	-668	436.7	-231.7
Pump	3.8		3.8
Compressor	76.9		76.9
Ex_{inputs}			**2104.2**
Outputs			
HQS	644.8	328.0	1860
LQS	41.5	85.9	299.8
Flue gases	59.9	83.6	143.5
$Ex_{outputs}$			**1243.5**
Ex_{losses} (system)			**860.7**
Ex_{losses} (system+external)			**1004.2**

The efficiency reduction comes mostly as a result of the NGD reaction where significant energy stored in carbon was not released through combustion as in the BAU case, rather the energy is still contained in the carbon black product. Another major area of exergy losses is the use of some fraction of the produced hydrogen to generate heat to meet the energy requirements of the endothermic NGD reaction. Also significant in this case is the compression power. Unlike the BAU case where compression power accounted for only 397.8 GJ/h, in the auto-thermal NGD process requires ~5 times more compression power. This process has the capacity to dramatically reduce CO_2 emissions; the CO_2 intensity of this process is 176 kg CO_2/m^3 bitumen. This is a ~70% reduction relative to that of the BAU case.

Table 2. Exergy inputs, outputs and losses for NGD-derived processes

Integrated NGD and oxycombustion process			Auto-thermal NGD process		
Ex_{ph}	Ex_{ch}	Ex	Ex_{ph}	Ex_{ch}	Ex

	GJ/h	GJ/h	GJ/h	GJ/h	GJ/h	GJ/h
Inputs						
NG	6.6	2627.3	2633.9	39.2	3381.2	3420.4
O_2	-42.6	103.1	60.3	-	-	-
Air	-	-	-	283.3	31.1	314.5
Water	-663.8	433.7	-230.1	356.2	671.2	-356.2
Pump	3.7		3.7	8.4		8.4
Compressor	72.3		72.3	397.8		397.8
ASU			121.2			
Ex_{inputs}			**2662.2**			**3784.9**
Outputs						
HQS	708.1	322.9	1031	753.0	197.4	950.4
LQS	207.5	123.8	331.3	216.5	440.6	657.0
Flue/other gases	6.0	29.3	35.3			
$Ex_{outputs}$			**1397.7**			**1607.4**
Ex_{losses} (system)			**1263.5**			**2177.4**
Ex_{losses} (system+external)			**1298.8**			**2177.4**

4.3. The integrated oxycombustion and NGD process

When using the integrated NGD and oxycombustion process, about 518 tons/d and 730 tons/d NG are fed to the oxycombustor and NGD reactor, respectively. 19,742 tons/d of feed water are preheated, fed into hydrogen combustion, and then converted to 73% quality steam. Hydrogen combustion produced ~1,945 tons/d steam in addition to the steam generated from the feed water stream. The quantity of HQS produced is 15,741 tons/d whereas that of LQS is 5,946 tons/d.

Table 2 also shows the exergy analysis results of the integrated NGD and oxycombustion process, indicating both physical and chemical exergy components of the inputs and outputs of the process. The net exergy inputs, Ex_{inputs} is ~2,662 GJ/h whereas net exergy outputs is ~1,398 GJ/h, resulting in ~1,264 GJ/h net exergy losses within the system. If the exergy in the flue gas is considered as an external loss, i.e., excluded from the net output exergy of the process, then the exergy losses (external and internal) become ~1,299 GJ/h. This reduces the total output exergy to ~1,362 GJ/h, which equals the summation of exergy of the useful products, HQS and LQS. Commercial vendor data suggest that oxygen production supplied at 95% purity has energy requirement below 220 kW h/ton O_2 – thus, specific electricity requirement of 200 kWh/ton-O_2 (=0.72 GJ/ton-O_2) was used.

The energy efficiency of this process is 74% whereas the exergy efficiency is 51%. Relative to the BAU case, results show that an integrated NGD and oxycombustion process results in an energy efficiency reduction of ~14%-points and an exergy efficiency value similar to that of the BAU case. The efficiency reduction is a result of the NGD reaction where significant energy stored in carbon is not released but rather stored in the carbon product. Another contributor to efficiency loss is the energy required by the endothermic NGD reaction.

The CO_2 intensity of this process is 354 kg CO_2/m^3 bitumen. This implies that the integrated NGD and oxycombustion process achieves a CO_2 intensity reduction of 213 kg CO_2/m^3 relative to the

BAU case. The CO_2 intensity of this process can be further reduced by co-injecting steam and the CO_2-rich stream from oxycombustion into oil sands reservoirs; this has been modelled and results showed that up to 60% of the CO_2 emissions of the process can be stored while at the same time improving bitumen recovery performance [11]. This implies that with marginal energy inputs the CO_2 intensity of the integrated NGD and oxycombustion process can be reduced to 212 CO_2/m^3 bitumen, which brings it close to the CO_2 intensity of the auto-thermal NGD process.

The integrated NGD and oxycombustion process is less energy intensive than the auto-thermal NGD process and has potential to reduce emissions to levels close to that of the auto-thermal NGD process if CO_2-rich flue gas stream is co-injected with steam into the reservoir. There are two main reasons why oxycombustion can be considered. First, oxy-combustion produces a high CO_2-concentration flue gas stream (~90 wt.% CO_2 on a dry basis) from natural gas combustion. A recent study demonstrated that injection of CO_2-rich flue gas into oil sands reservoirs can improve bitumen production performance and sequester 30-60% of the injected CO_2 [11]. Second, oxy-combustion can provide the heat required by the NGD process. If the goal is to achieve dramatic reductions in process emissions, the auto-thermal NGD process is highly competitive.

5. Conclusions

We have used exergy analysis in this paper to determine the energetic performance of natural gas decarbonisation applied to steam generation for low ultra-low CO_2 emissions in situ bitumen recovery. Two major methods were analysed and compared with business as usual (BAU) method of generating steam via the use of once-through steam generators. The NGD-based methods are i) auto-thermal NGD for steam generation process and ii) integrated NGD and oxycombustion for steam generation process. From the results, it can be concluded that the NGD-based methods analysed in the paper have the capacity to significantly reduce carbon emissions of bitumen recovery from oil sands (by 38-69%-points relative to the BAU case) while incurring some exergy penalty (9-20%-points relative to the BAU case), relative to the current operations. On a process CO_2 intensity (or emissions reduction) basis the auto-thermal NGD-based process is most superior among the cases analysed whereas on an energy intensity/efficiency basis the BAU case is most competitive followed by the integrated NGD and oxycombustion process. Depending on the prevailing policy or regulatory environments, the decision on which process to adopt will be influenced by process energetics, economics and net emissions reduction targets. This study and its findings are relevant to determine the thermodynamic minimum energy and operating cost requirements of using NGD-based technology to decarbonize the oil sands industry.

Acknowledgments

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Appendix

Table 1A.

Integrated oxycombustion and NGD process				Auto-thermal NGD process				BAU steam generation case			
Name	T ºC	P kPa	Molar Flow kgmol/h	Name	T ºC	P kPa	Molar Flow kgmol/h	Name	T ºC	P kPa	Molar Flow kgmol/h
NG-1	15	500	1897	NG-1	15	500	2585	NG	15	500	864
O2-2	15	500	2294	Air-1	15	100	517	Air	15	100	173
H2-1	920	3480	3794	H2-1	892	3480	17988	Liq	1743	200	346
Carbon	920	3480	1897	Carbon	892	3480	17988	FG-1	1743	200	346
NG-1++	800	3480	1897	NG-1++	800	3480	17988	FG-2	1732	190	328
H2-2	314	3460	3794	H2-2	285	3460	17885	FG-3	303	170	294
HQS	242	3460	36100	HQS	213	3440	17782	BFW-1	18	100	173
LQS	242	3460	13752	LQS	213	3440	17782	SS	244	3560	6154
NG-2	25	100	1282	FW-1	18	100	517	BFW-3	80	3580	6188
O2-1	15	500	2985	FW-2	18	5100	26362	HQS	243	3490	6033
Liq-2	897	550	0	NG-1+	211	3500	18092	LQS-1	243	3500	6050
FG-1	897	550	21428	Air-1-1	642	3500	18092	BFW-2	18	3600	6223
FG-2	881	540	21428	H2-3	285	3460	17885	FG-4	100	150	259
FG-4	100	500	21428	H2-4	285	3460	17885	cAir	98	200	346
FW-1	18	100	45661	HT-heat	1722	3460	17885				
FW-3	261	5080	45661	Liq	1722	3460	17885				
FG-3	684	520	21428	H2O	274	3440	17782				
FG-5	100	500	17142	Air-2	15	500	2585				
FG	100	500	4286	Air-2-1	292	3500	18092				
FG-6	100	550	17142								
FW-2	18	5100	45661								
NG-1+	211	3500	1897								
NG-2+	361	2500	1282								
O2-1+	231	2500	2985								
O2-2+	287	3500	2294								