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Techno-Economic Comparison of Chemical Looping Combustion (CLC) in Canada and Sweden

Author:

Frida Söderström
frisod@chalmers.se
frida.soderstrom@ucalgary.ca

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Supervisors:

Professor Henrik Leion
henrik.leion@chalmers.se
Gothenburg, Sweden

Professor Nader Mahinpey
nmahinpe@ucalgary.ca
Calgary, Canada

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Abstract

In order to mitigate global warming by reducing industrial emissions, use of CO_2 capture technologies is critical. Chemical Looping Combustion (CLC) is a promising, yet uncommercialized, method with potentially improved environmental benefits compared to conventional carbon capture technologies. The aim of this work is to investigate the commercialization potential of a 100 MW CLC power plant in Sweden and Canada through simulations using Aspen Plus and a techno-economic analysis.

A reference simulation was developed, which was based on the Örtofta combined heat and power (CHP) plant in Sweden, that was used as a basis for building the CLC cases. For the Canadian CLC case, sub-bituminous coal was used as the fuel and district heating was excluded, while the Swedish CLC case used bark as fuel and co-generated electricity and district heating. Ilmenite was selected as the oxygen carrier (OC) in all CLC scenarios. Key assumptions included an investment return rate of 9% and a 30 year plant lifetime.

When comparing the CLC scenarios, significantly higher commercialization potential could be observed for the Swedish CLC plant, with a payback period (PBP) of 11.45 years and a net present value (NPV) of -€37.05 million, compared to 41.02 years and -€231.74 million for the Canadian case. The primary factor behind this difference was identified to be the choice of including district heating or not. Of all simulation scenarios, the Canadian CLC case had the by far highest electricity yield (2127.7 kWh/tonne fuel) but generated the least profit (€7.18 million/year). The Swedish CHP plant without CO_2 capture had the lowest levelized cost of electricity (LCOE) at €0.0899/kWh, as well as superior results compared to the CLC scenarios regarding the NPV and the PBP at €173.86 million and 5.74 years.

Sensitivity analysis showed that capital expenditure (CAPEX) had the most significant influence on NPV in all simulation cases, while electricity price came as the second most important. Comparatively, fuel and OC costs had minor impacts. To make the NPV of the Canadian CLC plant positive, the incentive for CO_2 capture in Canada would need to be increased by 3.6 times (to €220/tCO_2). In order to make the NPV of the Swedish CLC case positive, the incentive would need an increase of about 1.2 times (to €110/tCO_2). For the Swedish CLC scenario to have a higher NPV than that of the Swedish CHP plant without CO_2 capture, either the incentive would need to be increased by 2.4 times (to €220/tCO_2) or the carbon tax for biofuels would need to be increased by 4.6 times (to €160 /tCO_2).

Contents

Acronyms

Below is the list of acronyms that have been used in this work, listed in alphabetical order:

AR	Air Reactor
BCLG	Biomass Chemical Looping Gasification
CAPEX	Capital Expenditure
CCUS	Carbon Capture, Utilization and Storage
CEPCI	Chemical Engineering Plant Cost Index
CFB	Circulating Fluidized Bed
CHP	Combined Heat and Power
CLC	Chemical Looping Combustion
CLR	Chemical Looping Reforming
CO_2	Carbon Dioxide
CRF	Capital Recovery Factor
C_xH_y	Hydrocarbon (general formula)
DH	District Heating
ESI	Emergy Sustainability Index
FR	Fuel Reactor
H_2O	Water
iG	In-situ Gasification
IRR	Internal Rate of Return
kWh	Kilowatt Hours
LCOE	Levelized Cost of Electricity
LHV	Lower Heating Calorific Value
MCINCPSD	Stream class containing: Mixed, Conventional Inert & Nonconventional with Particle Size Distribution
MSW	Municipal Solid Waste
MW	Megawatts
MWe	Megawatt Electrical
MWh	Megawatt Hours
MWth	Megawatt Thermal
NPV	Net Present Value
O_2	Oxygen
OC	Oxygen Carrier
OPEX	Operational Expenditure
PBP	Payback Period
PCC	Post Combustion Carbon Capture
PCCS	Post Combustion Carbon Capture and Storage
PSD	Particle Size Distribution
PWG	Plastic Waste Gasification
tCO_2	Tonnes Carbon Dioxide
TEA	Techno-Economic Analysis
TRL	Technical Readiness Level

1 Introduction

Increased greenhouse gas emissions as a consequence of human activities, have increased the global mean surface temperature of our planet with more than 1.1 °C since the 1850s [1]. This has occurred due to radiative forcing, which is an energy imbalance in the atmosphere caused by greenhouse gases, to which the climate responds by increasing surface temperatures in order to restore the balance [2]. Increased temperatures have had consequences in terms of climate change such as: heat waves, drought, wildfires, floods, sea level rise, biodiversity loss and declined health of aquatic ecosystems [3]. After water vapour, carbon dioxide is the most abundant greenhouse gas in the atmosphere, making it the largest contributor to human-induced global warming [4]. Due to its abundance and relatively long atmospheric lifetime, even with the most effective measures it would take centuries to reach the pre-industrial CO_2 levels [5].

Without industries providing services, infrastructure and essential goods, societies would stop functioning. In order to continue forward while mitigating climate change, technological solutions such as carbon capture technologies need to be developed further. While conventional post-combustion carbon capture and storage (PCCS) technologies are on the market, there are other technologies that could have higher environmental benefit, that are currently being researched but are not yet commercialized. One of these is the process of Chemical Looping Combustion (CLC), which is investigated in this study. CLC is a promising technology, not yet used on large scale. This study seeks to answer what factors can be most important in commercialization of these systems, by comparing economic suitability in different geographic locations.

2 Background

2.1 CLC: cleaner combustion

Combustion can be explained as the exothermic chemical reactions between a fuel and an oxidant, which release energy while also converting chemical species, producing CO_2 and H_2O among other products, as explained by Equation 1 [6]. Combustion of solid fuels involves the processes of drying, pyrolysis and char oxidation [7]. Pyrolysis is defined as the heat-induced decomposition of organic materials in the absence of oxygen [8]. It is during the pyrolysis process that the materials are decomposed into volatile gases and char that can then undergo oxidation which can sustain the combustion process [7]. In the fuel conversion, char gasification is the limiting step and is what can lead to incomplete combustion [9].

$$C_xH_y + O_2 \rightarrow CO_2 + H_2O + heat + light \tag{1}$$

Chemical Looping Combustion (CLC), as seen in Figure 1, is a combustion technology with inherent CO_2 capture, in which a fuel can be combusted while avoiding reactions with nitrogen and in which nearly 100% of the CO_2 can be captured, which leads to considerably cleaner flue gases than most other combustion technologies [10]. In order to achieve a desirable exhaust gas that does not contain any nitrogen compounds, from which CO_2 can be extracted with high efficiency, it is appealing to use only oxygen for combusting the fuel and not air directly (which contains 78% N_2).

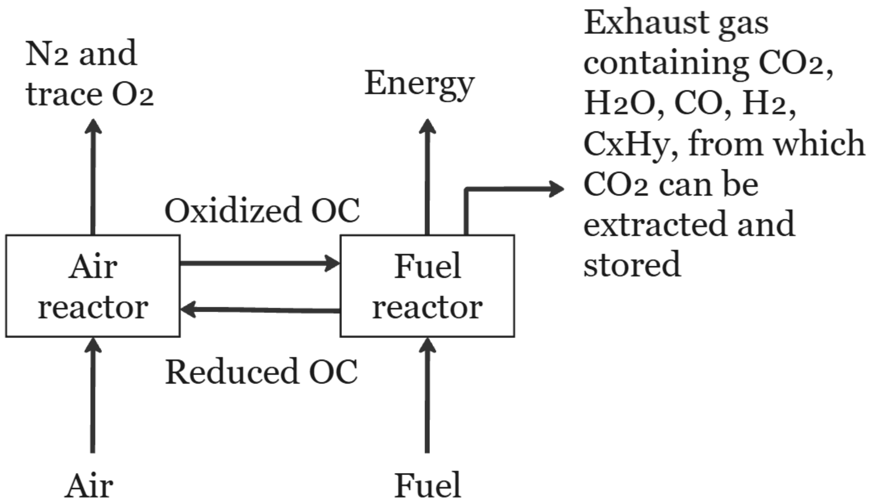

Figure 1: Simplified flowsheet of the CLC process, interpreted from Abuelgasim et al., 2021 [10].

In order to combust using only oxygen and not nitrogen, something called oxygen carriers (OCs) can be used. OCs are metal particles that are vital in CLC, as they are used for oxygen transfer between the fuel reactor and the air reactor, as explained by Equation 2 and Equation 3 below [11]. The oxidized OC is fed, along with the fuel, to the fuel reactor to be reduced (Equation 2), after which it moves to the air reactor to be regenerated (oxidized to its original state) due to an inflow of air (Equation 3).

$$C_xH_y + oxidized\ \ OC \rightarrow CO_2 + H_2O + reduced\ \ OC \tag{2}$$

$$reduced\ \ OC + O_2 \rightarrow oxidized\ \ OC \tag{3}$$

CLC requires OCs that are not too expensive but that have properties like high oxygen capacity and fast kinetics [12]. Other desirable qualities are high stability and low harm on the environment [13]. It is difficult to find OCs that meet all these requirements. Challenges with OCs include carbon formation in Ni-based OCs, sintering resistance in Fe-based OCs and sintering in Cu-based OCs [12].

2.2 Previous research about CLC

The history and current status of CLC, as well as the most relevant findings of recent research have been summarized in the following sections:

A promising technology not yet used on large scale

An idea of a concept similar to CLC was presented already in the 1950s, however research to use CLC to enhance combustion efficiency was first done in the early 1980s [10]. The first mention of the *term* Chemical Looping Combustion (CLC) occurred in 1987 [14]. In the early 2000s, significant experimental work in Chemical Looping Combustion began, such as testing of reactivity and durability of oxygen carriers [15].

CLC is not yet used on industrial scale. Gaseous fuels were initially prioritized in CLC studies, while solid fuels have become more of interest in later years [16]. The use of solid fuels in CLCs has achieved a technical readiness level (TRL) of 6 [9]. There has been an increased use of biomass as a fuel, as well as development in related technology which can include syngas production other than just the carbon capture [17]. A foundation for scale-up to commercial levels have been established, with over 10,000 hours of pilot operations with various OCs and reactor designs [18]. In more than 40 units over the world, the testing in CLC has occurred from lab-scale (0.2 kWth) to semi-commercial (4 MWth) [10]. Large-scale designs could be feasible already and would be even more energy efficient using biomass, which enables the possibility of negative emissions [19]. CLC can achieve near 100% capture efficiency and low energy penalties [10]. However, CLC is still not commercially available, even though there have been significant research in materials and operational challenges. Challenges for scale-up include reducing unburnt compounds and recovering oxygen carriers from the ash stream [9]. The technology can offer cost advantages, however, incentives for CO_2 capture are still limited as there are investment risks, since full-scale performance cannot be guaranteed [18].

CLC is the leading chemical looping process

Other than CLC, there are also other chemical looping processes. An example is biomass chemical looping gasification (BCLG), where China is in the lead in terms of research and development, with pilot systems of 10 kWth [20]. CLC and BCLG have similarities in that they both use OCs and both include energy conversion while mitigating greenhouse emissions, but in BCLG, the goal is to convert biomass into high-quality syngas [21]. The process shows potential for commercialization, however it is still in the development phase with a technical readiness level (TRL) between 3 and 5 [22], which can be compared with CLC that has a TRL of 6 [9]. Another chemical looping process that also produces syngas is chemical looping reforming (CLR), where the most recent progress has been mainly in the field of OCs, where a focus has been on metal-based OCs like Ni, Ce, Fe as well as composite OCs (like perovskites) and catalytic OCs [23]. The TRL of CLR was not found in the literature. Although processes like BCLG and CLR are progressing, CLC is currently the most advanced chemical looping technology. More techno-economic studies are needed to validate the vitality of any of these processes to be pushed towards commercialization, where CLC is closest to achieving that.

Natural ores can lead to cost-effective OCs

The research about OCs has been extensive in previous works. Alkaline earth metal-modified OCs like iron ore variants have been shown to have benefits, like improved combustion efficiency and less harmful emissions, however they still have the drawbacks of pollution and ash interactions [24]. A mixture of ilmenite and calcium manganite as OC can be more effective than the use of ilmenite or manganese ore alone [25]. Using electronic waste components as OCs for CLC can enhance economic and energetic feasibility of CLC for combined cycle power plants [26]. Generally, there has been a shift from Ni-based materials towards more environmentally friendly metal oxides such as natural ores, of which ilmenite is an example [17]. Natural ores, i.e., naturally occurring mineral deposits that are not engineered for specific purposes, can be a cost-effective type of OCs due to their low cost, while mixed ores is another alternative [27]. However, low cost OCs have lower lifespan and thermodynamic limitations, so synthetic OCs is another relevant alternative [28].

Post-combustion CO_2 capture: less sustainable than CLC

Several studies compare post-combustion CO_2 capture (PCC)-systems with CLC. In a comparison of a large scale (550 MWe) CLC-power plant and a coal-fired power plant, where both used Victorian brown coal, the conventional plant had higher net efficiency and lower levelized cost of electricity (LCOE) [29]. Still, the CLC plant had much potential for reducing CO_2 levels at a relatively low additional cost compared to the conventional plant. In another study comparing CLC and PCC-systems, ESI (Emergy Sustainability Index) was looked at, where a high value indicates greater sustainability [30]. CLC systems could reach as high as 0.835, to be compared with 0.565 as the highest value among the PCC systems. A different study showed that for bark combustion, CLC was superior compared to PCC in achieving negative CO_2 emissions, partly due to the increased energy efficiency [11]. It has also been showed that CLC can give a lower levelized cost of electricity compared to post-combustion capture systems, when it comes to methane conversion [31].

Polygeneration and waste use can enhance feasibility

Polygeneration (i.e., production of both power/heating as well as valuable products) can be a way to make CLC more profitable, for example a CLC process which includes both power generation and ethylene production, has been shown to have higher efficiency *and* lower total investment cost, compared to conventional CCUS (carbon capture, utilization and storage) processes [32]. CLC can also be integrated with methane reforming, to convert CO_2 into industrially relevant chemicals like methanol [33]. A process like that can lead to significant energy savings due to the heat integration.

CLC of Municipal Solid Waste (MSW) can be a promising way to convert energy while reducing environmental pollutants [24]. CLC power plants can be enhanced by incorporating solar energy systems as well as waste heat utilization for cooling needs [34]. Waste heat utilization outperform plants without waste heat recovery in terms of heat efficiency. When comparing CLC in a combination with plastic waste gasification (PWG) or with the copper-chlorine (Cu-Cl) cycle, in both cases to produce power and hydrogen while capturing CO_2, higher thermal efficiency and hydrogen production was observed in the CLC-PWG system compared to the CLC-CuCl system [35].

Biomass enables negative emissions

In a plant with gasification within a chemical looping combustion process (iG-CLC), integrated with a supercritical CO_2 cycle and an Organic Rankine cycle, the net efficiency slightly increased when the biomass share grew and negative CO_2 emissions could be achieved, which can lead to a competitive LCOE compared to conventional CO_2 capture technologies, depending on the carbon tax [36].

Other findings regarding CLC and fuel inputs are that higher alkali content in biomass fuels affects the performance of a CLC plant negatively, since greater alkali release can cause agglomeration, fouling and OC deactivation [25].

2.3 Canada and Sweden: resources, size and politics

Geographic location may play a significant role in the viability of CLC commercialization. Available resources in the region where the project is developed, as well as politics and trends, are factors that impact the commercialization potential. In order to evaluate the obstacles for CLC commercialization potential, techno-economic analysis (TEA) is a useful tool. The previous research shows that a lot of effort in CLC research has been directed towards optimizing oxygen carriers or integrating CLC with other processes, such as polygeneration and utilization of waste. To the best of our knowledge, there is no previous research where techno-economic factors for CLC commercialization in different geographic locations are compared. This study is intended to contribute to bridge this research gap.

In this study, the commercialization potential of CLC in *Sweden* and *Canada* have been considered. These countries have similar climate, both belonging to the circumpolar boreal zone, while having other interesting differences such as: size, available resources and politics [37].

Canada is located in North America, with a land area of 9,093,510 km^2, which makes it the second largest country in the world by area after Russia [38], with a population density of 4 people / km^2 [39]. Sweden, located in northern Europe, has a land area of 410,340 km^2 (number 56 in the world [38]) and a population density of 26 people / km^2[40]. The differences in size and population density are expected to give higher transportation costs and lower district heating suitability in Canada.

In this work, electricity production is of particular interest. Since CLC is most advanced for solid fuels [16], only solid fuels are considered in this work. As of 2024, 58% of Canada's electricity was produced from hydropower but the most common *solid* fuel for electricity production in Canada was coal, producing approximately 4.3% of the total electricity in the country [41]. Coal is also the most common fuel in solid fuel-fired heat and power plants in the world [42]. In Sweden, hydropower was also the largest source of electricity in 2024, contributing with 40% [43]. Biomass was however the most common *solid* fuel for electricity production at around 5% of the total electricity production, with coal contributing to only 0.3% of the electricity. Sweden can be considered a pioneer in biomass utilization. With this in mind, coal was selected as the solid fuel for the Canadian case and bark was selected as the solid fuel for the Swedish case.

Coal is a combustible sedimentary type of rock that generally contains more than 50 wt% of carbon (and varying amounts of elements like sulphur, oxygen, hydrogen and nitrogen), that is formed by a process that takes millions of years, where plant materials decay and transform in deep burial [44]. In Canada, coal can be found in the forms of anthracite, bituminous coal, sub-bituminous coal and lignite, with the majority of the coal deposits located in western provinces [45]. Sub-bituminous coal is often used for electricity power generation and has lower sulphur content than lignite, bituminous and anthracite, which leads to cleaner emissions. Sub-bituminous coal is also the most commonly used type of coal in the world [44]. For these reasons, sub-bituminous coal was selected as the coal type in the Canadian CLC case.

Ilmenite is considered a natural ore and is a common choice of oxygen carrier in CLC studies [17]. Due to its attractive characteristics like good physical durability, while having low cost [46], ilmenite was selected to be the oxygen carrier in both the Swedish and Canadian CLC case. Canada is among the top three producers of ilmenite in the world, alongside China and Australia [47].

Table 1 shows the daily average temperatures per month in Canada and Sweden, based on statistics from the period 1991-2020 [48][49]. Based on this, a temperature of -0.6 $^\circ$ C can be considered an average temperature for the whole year in Canada and 5.95 $^\circ$ C can be considered the yearly average in Sweden.

Table 1: Average daily temperatures in Canada and Sweden per month [$^\circ$ C] [48][49].

	Jan	**Feb**	**Mar**	**Apr**	**May**	**Jun**	**Jul**	**Aug**	**Sep**	**Oct**	**Nov**	**Dec**
Canada	-16.4	-15.3	-10.5	-2.4	5.7	12.0	15.2	14.0	8.9	1.5	-6.8	-13.0
Sweden	-3.2	-3.2	-0.1	4.6	9.6	14.0	16.8	15.7	11.4	6.0	1.5	- 1.7

In both Canada and Sweden it is possible to receive compensation from the government for initiatives related to carbon capture technologies. How large these can be generally depends on how much it costs to capture the CO_2. An exact value for the Canadian case was not found since it can vary from case to case, so it was assumed to be the "price of carbon emissions", for which the 2025 value is €60.84 [50]. In Sweden, it is possible to get incentives of around €91.60 per tonne CO_2 [51].

Since April 2025, there is no carbon tax on consumers in Canada [52]. However, for industries the minimum national carbon pollution price of 2025 is €60.84 per tonne CO_2e. There are also other regulations for emissions in place, varying in the different states. The tax rate for both consumers and industries of fossil CO_2 in Sweden is, in 2025, €138.3 per tonne [53]. This regards fossil fuels such as coal and natural gas, with a varying rate for other fuels. The value can also vary depending on if the fuel is used for district heating or only for electricity. The exact value for biomass for both electricity and district heating was not found. However, in this work it was assumed to be 25% of the rate for fossil fuels, which was used in the base cases and Swedish CLC cases of this work.

District heating systems, i.e., systems that distribute heat from a central heat source to many users, are a way to effectivize heating to save energy and it is commonly used in Sweden and the rest of Northern Europe [54]. District heating, which requires a high enough density of heat demand to decrease distribution losses, is not suitable for vast lands with spread out population, such as in Canada. In Sweden, district heating supplies more than half of the building heating demand [54]. Figure 2 gives an idea of the difference in scale between the two countries, which can partly explain why district heating is not common in Canada [55].

Figure 2: Sweden if it was placed on top of Canada [55].

In this work, it was assumed that the captured CO_2 in the CLC cases was transported through pipelines to remote underground storage, where it was subject to geological sequestration. The transport and storage cost can vary a lot regionally but as of 2021 it was found to typically be within the range of €3.5 to €40 / tCO_2 [56]. A cost of €10 was assumed for the Swedish CLC case. Since the distances of transportation are expected to be longer in Canada, the cost of transportation and sequestration can be assumed to be higher. A cost of €15 was assumed for the Canadian CLC case.

Electricity prices are constantly fluctuating, as are the prices of fuels and materials. For the sake of cost estimations, fixed prices were assumed based on the market values on the day when they were collected, in May 2025. For the biomass price, the prices of piece peat and return wood in Sweden 2024 were looked at, which were €16.76 /MWh and €24.73 /MWh, respectively [57]. Since the bark biomass that would be used in the processes of this study would likely be a by-product of saw mills the price would be expected to be even lower than that of piece peat. Since no exact number was found, a price of €16.5 /MWh was assumed for the biomass. These prices, as well as the information previously provided for the two countries, are summarized in Table 2. This can be considered the background on which the reasoning in this work is based.

Table 2: Factors that can impact CLC potential, for Sweden and Canada.

	Canada	**Sweden**
Land area [km^2]	9,093,510 [39]	410,340 [40]
Pop. density [people/km^2]	4 [39]	26 [40]
Incentive for CO_2 capture [€/tCO_2e]	60.84 [50]	91.60 [51]
Carbon tax for industries, fossil fuels [€/tCO_2e]	60.84 [52]	138.3 [53]
District heating	Uncommon [54]	Very common [54]
Average temperature [°C]	-0.6 [48]	5.95 [49]
Most common solid fuel for electricity	Coal [41]	Biomass [43]
Electricity price [€/kWh]	0.1152 [58]	0.1949 [58]
District heat price [€/kWh]	Not relevant	0.1008 [59]
Biomass price [€/kWh]	Not relevant	0.0165 [57]
Ilmenite price, globally [€/tonne]	318.9 [60]	318.9 [60]
Sub-bituminous coal price, North America [€/tonne]	16.42 [61]	Not relevant
Cost of CO_2 sequestration [€/tCO_2]	15 [56]	10 [56]

3 Methodology

For the simulations of this work, the simulation software Aspen Plus (version 14) was used. Aspen Plus was selected because it is a leading process simulation software in the chemical industry, due to its capability in modelling complex chemical processes [62].

A total of five simulation cases were constructed in Aspen Plus v.14:

- A Swedish combined heat and power (CHP) plant of 115 MW, based on a real CHP plant in Sweden, created for validation purposes.
- A Swedish combined heat and power (CHP) plant of 100 MW, created to compare with the CLC cases.
- A Swedish CLC plant of 100 MW.
- A Swedish CLC plant of 30 MW, created for verification purposes.
- A Canadian CLC plant of 100 MW.

The models representing unit operations in Aspen Plus are called *blocks*. Something that requires only one unit operation in the real process may require several blocks in Aspen Plus. Before modelling the solid combustion in Aspen Plus, the guides *Getting Started Building and Running a Process Model* [63] and *Getting Started Modelling Processes with Solids* [64] were studied. Some of the methods explained there were used in this work, like the combustion and water calculators, which will not be explained in this methodology section.

In this work, all economic estimates have been converted to €(EUR) for clarity. This currency was chosen because it is a commonly used currency for international comparisons. It was also preferred above USD, another common currency for comparisons, due to political and economic volatility at the time of this study. To convert literature values to €, the rates given by Markets Insider Currency Converter [65], on May 2nd, 2025 were assumed, as shown in Table 3:

Table 3: The currency conversion rates that were used.

	$1 CAD	**1 SEK**	**$1 USD**
€(EUR)	0.6404	0.0916	0.8858

3.1 Overall methodology

The overall simulation method of this work can be described by Figure 3:

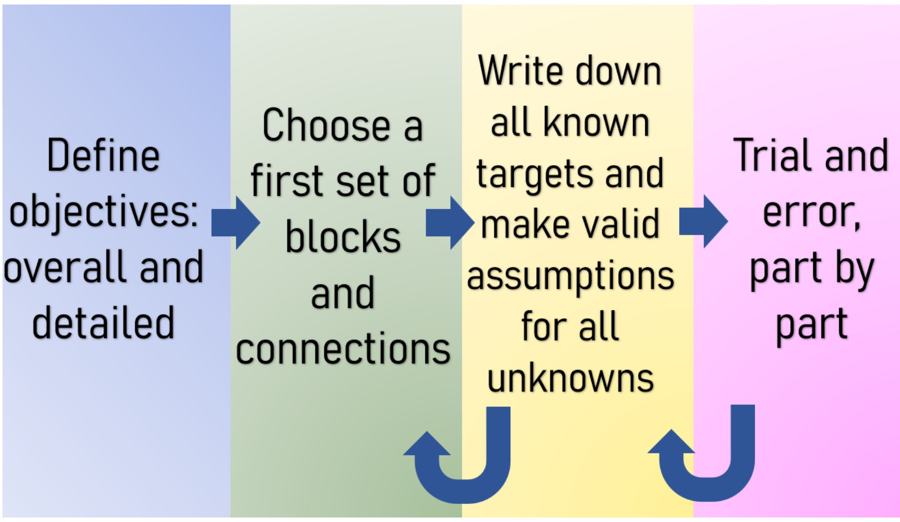

Figure 3: The four parts of the overall simulation approach.

Initially, the objectives of each case were considered. The core objective was determined by the available inputs and the desired outputs, in relation to the system boundary that had been selected, as described by Figure 4:

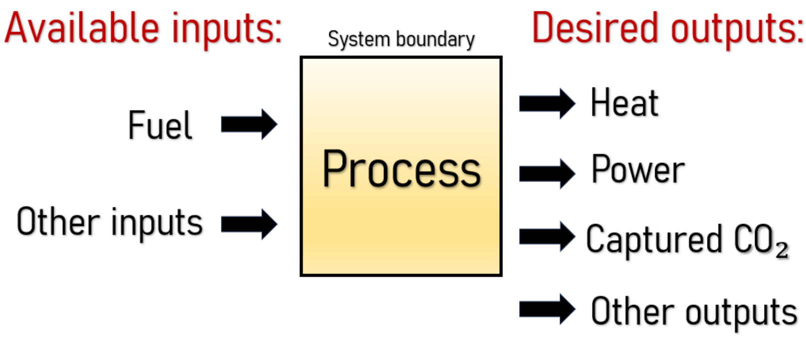

Figure 4: How available inputs and desired outputs form the core objective of a process.

In the base case, which was a combined heat and power plant, the main objective was to transform an input fuel to outputs in terms of heat and electric power. For the CLC plants, the main objectives again included transformation of fuels to create heat and power outputs, as well as CO_2 capture.

Next, unit operations were selected. For this, literature was consulted. In this work, various sources about CLC processes were considered, as presented in section 1. The main work that was used to build the simulations, both for the base and the CLC cases, was the work by Surywanshi et al. from 2023 [11]. After deciding what unit operations to include, the purpose of each unit was considered, for example if it was to separate components, to mix, to induce a chemical reaction, to compress the components or remove water.

After drawing a first flowsheet with preliminary unit operations and material streams (of the *real* process) and having defined the objective of each operation, step 1 of Figure 3 was considered complete. Then, it was time to choose the specific blocks in Aspen to represent each unit operation, which is the second step in Figure 3. For each unit operation of the process, the objective was considered related to what blocks are available in Aspen. For example, there are many available blocks in Aspen to separate components, where the choice of the block(s) depends on things like the phase of the components, the composition of the inflowing stream and how accurate and advanced the calculations need to be.

While the Aspen blocks were gradually being selected, a new flowsheet was drawn, of the *simulation*, with material streams connecting the blocks. All the blocks and streams were given names. After this, step 2 of Figure 3 was considered complete. As seen in Figure 3, this step is iterative. This means that after getting further with the simulation it might be noticed that other blocks are actually better suited to reach the objectives. In that case, the blocks were updated.

The third step of the overall methodology was to write down all known target properties for blocks and material streams, based on the literature, here the work by Surywanshi et al., 2023 [11]. Things that are *always* required for input streams in order to run the simulation in Aspen Plus are: temperature, pressure, mass flow and composition. For some streams or blocks these properties were not given in the literature. In those cases, reasonable assumptions had to be made. These assumptions are the reason why this step is also an iterative step, since the assumptions of the properties may have to be updated later, while some targets on the other hand are hard targets (i.e., more important). When finishing this step, this marked the completion of step 3 in Figure 3.

When all blocks and streams had been defined in terms of their properties, the only thing left was to enter it all into the simulation. When running a new Aspen Plus simulation the first time, it is likely that problems will arise, such as warnings or error messages, or undesired outcomes. Searching in literature was not the only answer to these issues. It was required to actually run the simulation repeatedly, using a trial and error approach, which is step 4 in Figure 3. For this, it helped to break it down into smaller parts. For example, the blocks were simulated one at a time until the desired outcomes had been reached, to understand what settings were important for each block before putting them together.

3.2 Simulation assumptions

Table 4 presents the global assumptions that were used in all the simulation cases, based on the settings in the work by Surywanshi et al., 2023 [11]:

Table 4: Global assumptions that were used for all simulation cases.

	Assumption
Ambient pressure [bar]	1.013
Air composition	21% O_2, 79% N_2
Type of flow, all components	Steady state
Overall method	Peng-Robinson, Boston-Mathias correction
Method for water and steam	STEAM-NBS
Pressure and heat losses	Negligible
RGIBBS calculation option	Chemical and phase equilibrium
Stream class	MCINCPSD
Operational year [hrs]	5300
Free water	Dirty water
Enthalpy method (NC components)	HCOALGEN
Density method (NC components)	DCOALIGT

NC components in Table 4 refer to nonconventional components, which is bark and coal in the simulations of this work. The stream class MCINCPSD means that conventional and nonconventional solids are both present, with a particle size distribution (PSD). This was chosen even though the particle size distributions for the solids were unknown, so that it would be possible to use the block RSTOIC. The PSD for the solids was in all cases assumed to be as is shown in Table 5, based on the assumption made in the AspenTech guide *Getting Started Modelling Processes with Solids* [64]:

Table 5: The assumption used to represent the PSD for the solids.

Lower limit [µm]	**Upper limit [µm]**	**Weight fraction**
120	140	0.1
140	160	0.2
160	180	0.3
180	200	0.4

Some of the global settings varied for the different cases, as is shown in Table 6, where the two base cases are the "validation" case and the "comparison" case:

Table 6: Global assumptions that varied for the different cases.

	Base case 115 MW	**Base case 100 MW**	**Swe. CLC 100 MW**	**Swe. CLC 30 MW**	**Can. CLC 100 MW**
Amb. temp. [°C]	8	8	5.95	5.95	-0.6
Fuel type	Bark	Bark	Bark	Bark	Coal
LHV at dry basis [MJ/kg]	19.6 [66]	19.6	19.6	19.6	26.18 [67]
Mass flow, dry fuel [kg/s]	5.87	5.10	5.10	1.53	3.82

The ambient temperature and the energy of the input fuel of the validation base case was set to equal the values in the work by Björnsson et al., 2021 [68]. For the comparison of this work, a scale of 100 MW was selected. The 30 MW case was created for validation purposes. The ambient temperatures for Sweden and Canada were set to be the average temperatures of the countries as explained in subsection 2.3.

The energies provided in MW and the lower heating calorific value (LHV) in Table 6 are not settings that were directly entered into Aspen Plus. Instead, they were used to calculate the mass flow of the dry fuel stream (in kg/s), as is shown by Equation 4 [11], which is a value that can be adjusted in the simulation:

$$\dot{m} = \frac{Energy\ \ of\ \ the\ \ fuel}{LHV} \tag{4}$$

Characteristics of bark

As explained in subsection 2.3, bark was selected to present the solid fuel for the Swedish CLC case. The characteristics of the dried bark are shown in Table 7:

Table 7: Characteristics of bark at dry basis [66], used in the simulation.

Proximate analysis	[%]	Ultimate analysis	[%]
Moisture	12.5	Total sulfur	0.04
Ash	3.0	Carbon	51.8
Volatile matter	77.3	Hydrogen	5.9
Fixed carbon	19.7	Nitrogen	0.4
		Oxygen	38.9

In the ultimate analysis of Table 7, sulfur makes up 0.04% of the mass at dry basis. In this simulation, it was assumed that all of this sulfur is organic sulfur. Another assumption that was made regarding the bark was that it was assumed to have 50% moisture before drying.

Characteristics of sub-bituminous coal

For the Canadian CLC case, sub-bituminous coal was selected as the solid fuel, for the reasons explained in subsection 2.3. Table 8 shows the proximate and ultimate analysis of the dried sub-bituminous coal [69]. Since the sum of ash, volatile matter and fixed carbon in the proximate analysis must equal 100% in Aspen Plus (as they are entered at dry basis in the simulation), and since the ash fraction is the same in the ultimate analysis, the volatile matter and fixed carbon were multiplied with a factor of 1.168 before entering into the simulation.

Table 8: Characteristics of sub-bituminous coal at dry basis [69].

Proximate analysis	[%]	Ultimate analysis	[%]
Moisture	13.54	Total sulfur	0.99
Ash	5.86	Carbon	54.41
Volatile matter	42.62	Hydrogen	5.19
Fixed carbon	37.98	Nitrogen	1.15
		Oxygen	32.40

As can be seen in Table 8, sulfur makes up nearly 1% in the ultimate analysis. In coal, there are three significant forms of sulfur: pyritic, organic and sulfate sulfur, where the pyritic and organic forms make up most of the bulk sulfur [70]. Therefore, in this simulation, it was assumed that the pyritic and organic sulfur each make up 0.45% of the ultimate analysis and that the sulfate sulfur makes up 0.09%, which adds up to the total of 0.99% as seen in the table above. It was also assumed that the coal had 30% moisture before drying.

Characteristics of ilmenite

Due to the advantages explained in subsection 2.2, ilmenite was selected as the oxygen carrier for all cases. The components of the reduced form of ilmenite in weight-% are presented in Table 9:

Table 9: Characteristics of the reduced form of ilmenite [71].

	TiO_2	FeO	Other
Wt%	50.77	42.14	7.09

Since TiO_2 and FeO make up to a total of 92.91 wt%, they are assumed to be the only reactive components of ilmenite. To make up for the missing 7.09 wt%, which affects the physical properties, it was for simplicity assumed that the rest of the material is composed of MgO, assumed to be inert. MgO is a component of ilmenite, even though there are also other components that make up the small fractions that are not TiO_2 and FeO.

3.3 Building the simulation cases

Before the CLC cases were constructed, a simulation of a combined heat and power (CHP) plant without CO_2 capture was created, based on the work by Surywanshi et al., 2023 [11] who based their work on the data provided by Björnsson et al., 2021 [68], regarding Örtofta combined heat and power plant; a real heat and power plant located in southern Sweden. The Örtofta simulation was then used as a base for the settings when creating the CLC cases. Detailed Aspen flow sheets of the three types of simulation cases can be found in Appendix.

I. Validation base case: Örtofta CHP plant in Sweden without CO_2 capture

As was explained in the introductory part of section 3, there is a distinction in the flowsheets of a *real* process and its Aspen Plus simulation. The flowsheets displayed here are the interpretations of the *real* processes, which means that not all blocks that were used in Aspen can be understood by just looking at the flowsheets. Instead, the explanation of how the flowsheets were translated into Aspen settings can be found in the text.

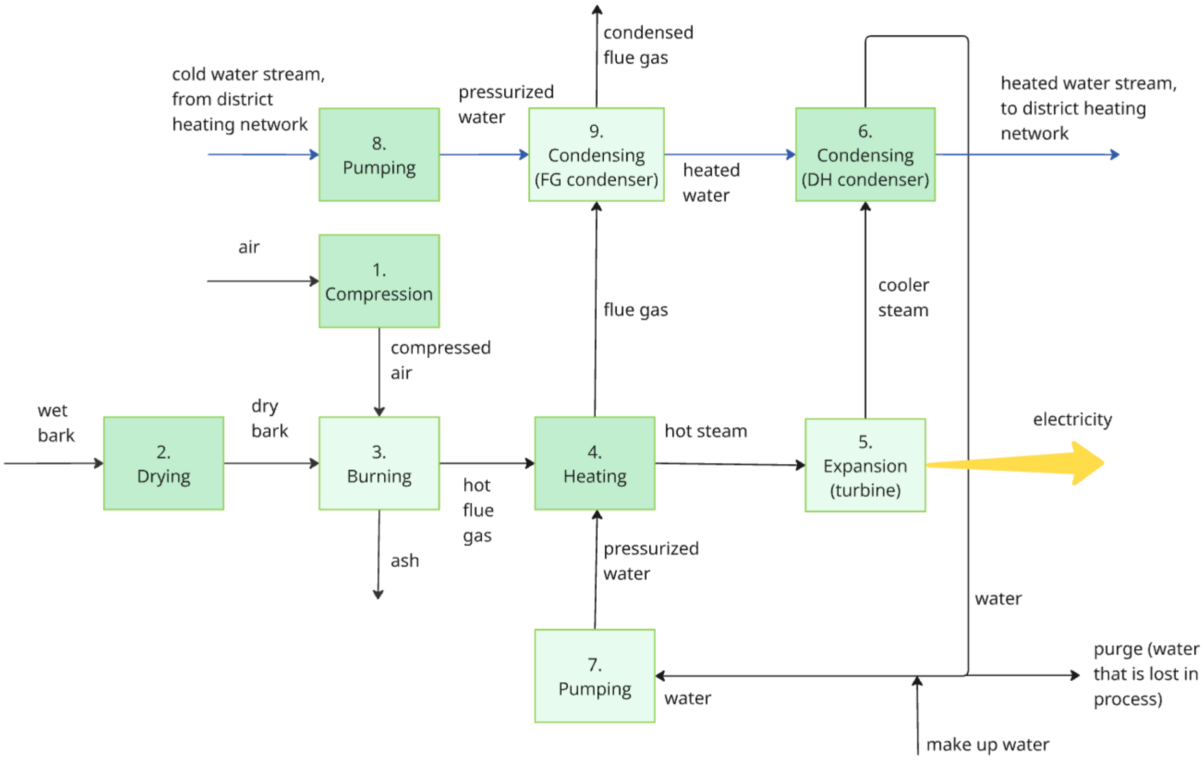

Figure 5: Flowsheet interpretation of the base case, based on the work by Surywanshi et al. [11] and Björnsson et al. [68].

For the validation base case, based on Örtofta CHP plant in Sweden, as seen in Figure 5, the wet fuel (bark), first goes through a drying step (unit operation 2). In this study, this was simulated by two consecutive blocks: RSTOIC and a FLASH2, with an inert nitrogen stream also entering at the same time as the fuel to facilitate the drying and combustion processes, exiting the system as an exhaust stream in the FLASH2 block. The dried bark has a mass flow of **5.87 kg/s**, which corresponds to **115 MW**. The burning step consists of a circulating fluidized bed (CFB) boiler (unit operation 3), which here was simulated using two blocks: RYIELD and RGIBBS. Air of ambient temperature and pressure enters a compressor (unit operation 1), after which it enters the CFB boiler. The products of the RGIBBS block (ash and flue gas) were separated by an SSPLIT block. The hot flue gas entered a heater (unit operation 4 in Figure 5), with the purpose to heat up an incoming water stream (up to a temperature of **540 °C**). This steam is expanded in the turbine (unit operation 5), which is where the plant generates electricity. The steam then continues to the district heating condenser (unit operation 6) where it is condensed, after which it is joined by an incoming water stream (entering at ambient and temperature and pressure) to make up for the lost water in the process. In the simulation, the split of the purge stream was done with an SSPLIT block. The mixed water stream is pumped (unit operation 7) to **1.11 bar**, before entering the heater again.

Simultaneously, the waste heat of the hot flue gas is utilized by heating up a water stream from the district heating network. This water stream enters the system at **44 ° C**, is pressurized in a pump (unit operation 8) to **107 bar**, then heated to **53.7 ° C** in the flue gas condenser (unit operation 9) and heated further to **93.3 ° C** in the district heating condenser (unit operation 6), after which it leaves the system and returns to the district heating network.

II. Comparison base case: 100 MW

The comparison base case is essentially the same simulation as the validation base case, with the main difference that the dry fuel has a mass flow of 5.10 kg/s instead of 5.87 kg/s, which corresponds to 100 MW, instead of 115 MW. This is in order to make a better comparison with the CLC cases, of which a size of 100 MW had been selected. Other streams were also adjusted to better suit this scale.

III. Swedish CLC case

The flowsheet describing the Swedish CLC case can be seen in Figure 6:

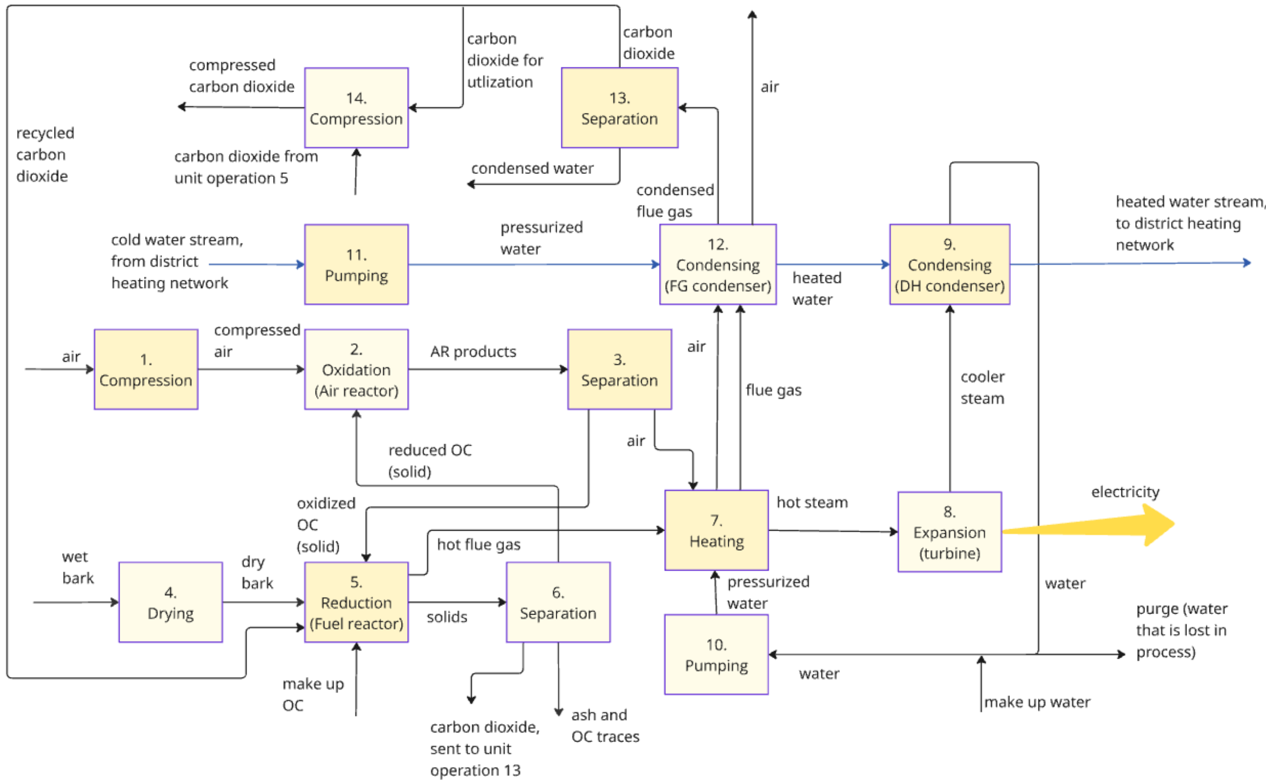

Figure 6: Flowsheet interpretation of the Swedish CLC case.

Again, the "as received" (wet) bark, first goes through a drying step (unit operation 4), simulated the same way as in the base case. The dried bark has a mass flow of **5.10 kg/s**, which corresponds to **100 MW**.The dried bark goes to the fuel reactor (unit operation 5), simulated with RYIELD and RGIBBS. The products of the fuel reactor are separated (unit operation 6), simulated using CYCLONE and SSPLIT, into four streams: one ash stream, one with the reduced OC, one hot flue gas stream and one stream containing mostly CO_2. The ash stream is separated from the system. The CO_2 is circulated to the compressor (unit operation 14). As in the base case, air of ambient temperature and pressure enters a compressor (unit operation 1), after which it enters the air reactor (unit operation 2). The separation of the components of the air reactor was done using SSPLIT twice (unit operation 3), to obtain two separate streams: one with the oxidized OC and one air stream.

The reduced OC is returned to the air reactor, and the oxidized OC is returned to the fuel reactor, in a loop. The air stream from the air reactor and the hot flue gas stream from the fuel reactor go into a heater (unit operation 7). These hot streams heat up a cold incoming water stream, just like in the base case, to **540 °C** and **107 bar**. This steam is again expanded in the turbine (unit operation 8), for electricity generation. The steam continues to the district heating condenser (unit operation 9) and make up water and lost water is dealt with the same way as in the base case. Like in the base case, the mixed water stream is pressurized through a pump (unit operation 11), before entering the heater again and going through the same loop. It is worth noting that the flue gas, air heating and heating of the district heating water stream are different loops that are not intended to be mixed. In reality, more consideration would be required for the heat integration.

In the Swedish CLC case, the waste heat of the hot flue gas is utilized to heat up a water stream from the district heating network. It enters and leaves the system at the same conditions as in the base case, being heated in unit operation 12 and 9. Water is separated from the cooled flue gas (unit operation 13), which gives a separate CO_2 stream. A part of this stream is recycled to the fuel reactor, while most of it is compressed and cooled for CO_2 sequestration (unit operation 14).

IV. Validation case, Swedish CLC: 30 MW

This case was essentially the same simulation as the Swedish CLC simulation, with the main difference that the mass flow of the dry fuel was 1.53 kg/s. This was created to compare the CLC model to a previous simulation of that scale, in the work by Surywanshi et al., 2023 [11]. The other input streams were adjusted to fit the scale better, such as the flow of air and make up water.

V. Canadian CLC case

Figure 7 shows the flowsheet describing the Canadian CLC case:

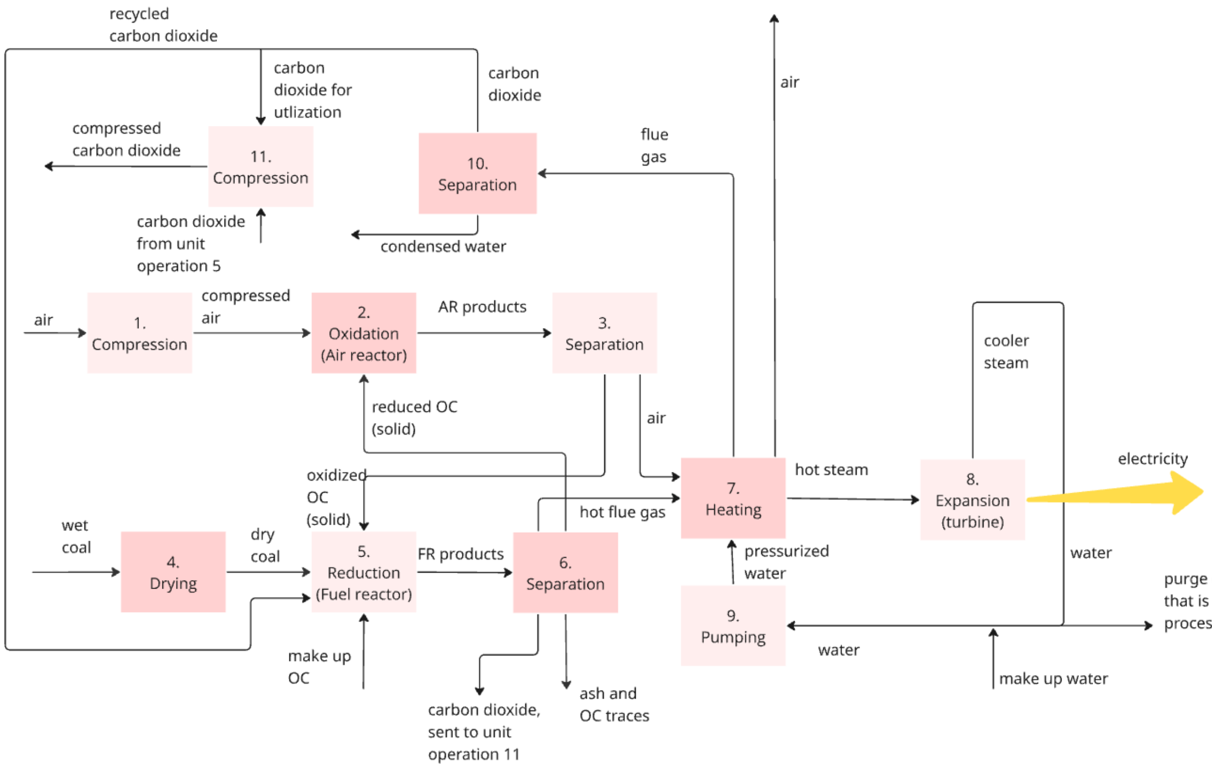

Figure 7: Flowsheet interpretation of the Canadian CLC case.

In this case, "as received" (wet) sub-bituminous coal, goes through a drying step (unit operation 4), simulated the same way as in the base case. The dried coal has a mass flow of **3.82 kg/s**, which corresponds to **100 MW**. All steps are simulated the same way as in the Swedish CLC case, expect that there is no district heating stream and therefore no FG condenser or DH condenser. Instead, the flue gas goes directly from the heater to a flash unit, where it is cooled and condensed water is separated from the CO_2. The CO_2 is handled the same way as in the Swedish CLC case.

3.4 Analysis methods

The generator efficiency was assumed to be 98.1% [72], which was used in the calculations of the net electricity output, as well as subtracting the generator electricity used in the pumps and compressors. The electric efficiency was calculated with Equation 5 [11]:

$$\eta_{el} = \frac{Net\ \ electric\ \ output}{Energy\ \ of\ \ the\ \ input\ \ fuel} \tag{5}$$

Similarly, the efficiency of the heating, relevant for the base case and the Swedish CLC case, can be found by Equation 6 [11]:

$$\eta_{DH} = \frac{Heat\ \ output}{Energy\ \ of\ \ the\ \ input\ \ fuel} \tag{6}$$

Another metric of comparison that was used was the payback period (PBP), defined as is shown in Equation 7 [11]:

$$PBP = \frac{CAPEX}{Annual\ \ plant\ \ profit} \tag{7}$$

The total capital expenditure (CAPEX) and the annual plant profit can be calculated as presented by Equation 8 and Equation 9 [11](where Equation 8 has been derived from the equations in the work by Surywanshi et al., 2023):

$$CAPEX = 3.4024536 \cdot Equipment\ \ Cost \tag{8}$$

$$Annual\ \ plant\ \ profit = Annual\ \ revenue - OPEX \tag{9}$$

The annual plant revenue and the total operating expenditure (OPEX) are calculated as is shown by Equation 10 and Equation 11 below [11], where the fixed OPEX includes labour, maintenance and administration, and materials includes fuels, sand and oxygen carriers, and where *Seq.* stands for sequestration:

$$Annual\ \ revenue = DH_{price} \cdot Heat_{Output/yr} + El_{price} \cdot El_{Output/yr} + CO_{2,captured} \cdot Incentive \tag{10}$$

$$OPEX = Fixed\ \ OPEX + Materials + Utilities + Tax + Seq. + Oxypolishing \tag{11}$$

Another metric that was used was the levelized cost of electricity (LCOE). It can be explained as the average cost of electricity over a plant's lifetime, when accounting for all costs [73]. It is the required price for electricity to break even on the investment. This was calculated as is shown by Equation 12, based on the literature [74], but modified slightly to not include the revenue and CAPEX costs related to district heating in the base case and the Swedish CLC case, since district heating is also produced in those cases.

$$LCOE = \frac{(CAPEX \cdot CRF + OPEX) \cdot Fraction\ \ for\ \ electricity}{Annual\ \ electricity\ \ generation} \tag{12}$$

The fraction for electricity was calculated as the net electricity output divided by the net total energy outputs (electricity and heat). In Equation 12, CRF is the capital recovery factor, given by Equation 13 below [11]:

$$CRF = \frac{i \cdot (i+1)^t}{(i+1)^t - 1} \tag{13}$$

where i is the interest rate and t is the plant lifetime, assumed to be 9% and 30 years, respectively [11], which leads to a CRF of 0.0973.

The net present value (NPV) is the value of all future cash flow over the life of an investment, discounted to the present, as is shown in Equation 14 [75]:

$$NPV = \sum_{n=1}^{n=t} \frac{CFn}{(1+i)^n} \tag{14}$$

where CFn is the net cash flow (the net money that goes in or out of the project, i.e., income minus costs) of the year n, i is the interest rate and t is the lifetime of the plant (assumed to be 9% and 30 years [11]). This metric takes the time value of money into account, to estimate the value of the project. The internal rate of return (IRR) is the interest rate that would give NPV = 0 [75]. It was assumed that year 0 there is no revenue and the costs equal CAPEX, while during year 1-29 the annual revenue is included as the income, and the costs equal OPEX. For year 30, the income was assumed to be the same annual revenue but the costs were assumed to be OPEX plus half of CAPEX as the scrap cost of the plant.

Assumptions for fixed OPEX and utilities

It is assumed that labour costs are similar in Sweden and Canada, as both countries are Western economies with markets of high-income and skilled labour. Maintenance, labour and administration are all part of the fixed operating cost, which can be assumed to be 3% of the CAPEX [76]. The other parts of OPEX, as shown in Equation 11, make up the *variable* operating cost. Utilities means things like water and electricity, which is here assumed to have a cost of 1% of CAPEX for the coal fired case 1.5% of CAPEX for the biomass fired case, because processes with biomass generally require more water and electricity. The rest of the variable operating cost are made up by the fuel and oxygen carrier costs (but should likely be around 2% of CAPEX).

Equipment cost estimation

For the cost estimations, the major parts of the process plants were included. The costs of the drying equipment and solid separators were not included since these were assumed to have smaller impact on the result, as was done by Surywanshi et al., 2023 [11].

The cost of each piece of major equipment was estimated with Equation 15 [11], where n is a scaling exponent:

$$Cost = \frac{CEPCI_{current\ \ year}}{CEPCI_{reference\ \ year}} \cdot Ref.\ \ cost(\frac{Current\ \ capacity}{Ref.\ \ capacity})^n \tag{15}$$

CEPCI is the Chemical Engineering Plant Cost Index. At the time of writing the most recent available CEPCI was from June 2024, at 798.8 [77], so this value was used as the CEPCI representing "the current year". The scaling exponents, reference costs and reference capacities used can be seen in Table 10:

	Scaling exp. n	**Ref. cost [M€]**	**Ref. year**	**Ref. capacity**	**Source**
Pump	0.14	0.01772	2019	250 m^3	[78]
Compressor	0.67	6.466	2019	10 MW	[78]
Heater	0.9	1.453	2014	57.2 MW	[78]
Steam turbine	1	27.11	2008	67 MW	[78]
CFB boiler	0.74	238.04	2011	300 MW	[79]
Air reactor	0.66	238.04	2007	1618 MW	[80]
Fuel reactor	0.66	238.04	2007	1618 MW	[80]

Table 10: Reference capacities and costs for the equipment.

The boiler of the base case is a circulating fluidized bed (CFB) [68]. For this, sand is a requirement, which was assumed to have a cost of 6.2 €/tonne and a flow of 53.2 tonnes/hr for a plant of 100 MW, based on a previous work [81].

A step that is usually required in CLC in order to achieve complete combustion of the fuels, is oxygen polishing, which was assumed to have a cost of €6.5/tCO_2 [82]. In this work, the oxy-polishing step was not included in the simulations. However, the costs for this were added after the simulations.

Model validation and verification methods

There are various simplifications in the simulation model assumptions, such as the composition of the components in the fuels, the oxygen carrier and air. For the combustion, RGibbs was used instead of a block where kinetics and other details are entered manually. Many other assumptions were made, most often based on the literature but sometimes settings were assumed when no other option was available. This can impact the results considerably.

Verification and validation are important steps to ensure accuracy when creating models. Verification involves checking for errors in the calculations, while validation means making sure the correct calculations were chosen by looking at if they can describe real world phenomena, which can be done by comparing with experimental data [83].

The accuracy of the base case simulation could be validated, since the real plant data are available in the work by Björnsson et al., 2021 [68]. The other cases could not be validated since there are no experimental data to compare with. The Swedish CLC simulation, however, was verified by adjusting the scale to 30 MW and comparing with a previous CLC simulation of 30 MW in the work by Surywanshi et al., 2023 [11]. A sensitivity analysis was conducted on key variables to assess the robustness and reliability of the results, which involved testing how high and low values of different parameters impacted the result. This way, it was possible to determine which parameters impacted the results the most, i.e., which were the most critical.

In this work, sensitivity analysis was done by changing the value of a single parameter and noting down the difference in input (as a fraction) and the difference in output (as a fraction) and dividing them by each other to calculate an impact factor, which describes how much that parameter impacts the output that is being looked at. A high value indicates that the parameter has a lot of impact on the output. Interacting effects (i.e., changing multiple parameters at once) were not tested in the sensitivity analysis.

4 Results and discussion

4.1 Validation and verification

Validation of the base case simulation

For the base plant, CHEMCAD values based on real experimental data from the 115 MW Örtofta CHP plant in Sweden are available in the work by Björnsson et al., 2021 [68]. In the work by Surywanshi et al., 2023 [11] and in the present work, simulations of the same plant were created in Aspen Plus. Table 11 and Table 12 below show the validation of the base case simulation of 115 MW compared to the values of the previous studies of the same plant:

Table 11: Stream property validation for the Swedish base case without CO_2 capture.

	Björnsson et al.[68]	**Surywanshi et al.[11]**	**Present work**
Compr. air to boiler [m^3/hr]	143 028	142 524	143 027
Hot steam to turbine [bar]	107	107	107
Hot steam to turbine [$^\circ$ C]	540	540	539.94
Hot steam to turbine [kg/hr]	141 012	141 480	141 012
Cold DH water, in [$^\circ$ C]	44	44	44
DH water, partly heated [$^\circ$ C]	53.7	53.7	53.71
Heated DH water, out [$^\circ$ C]	93.3	93.5	93.3
FG out of system [m^3/hr]	182 808	177 984	189 185

As is seen in Table 11, it was possible to achieve very similar stream conditions as those in Björnsson et al. The largest discrepancy is the volumetric flow rate of the flue gas leaving the system. As can also be observed, the value for this in the work by Surywanshi et al. is also deviant from the value of Björnsson et al. This can mean that a small change of some other setting in Aspen Plus might lead to large differences for the flow rate of that stream. In Table 12, the electricity and heat data, of the base case simulation of this work and the previous works, are gathered:

Table 12: Heat and power validation for the Swedish base case without CO_2 capture.

	Björnsson et al. [68]	**Surywanshi et al. [11]**	**Present work**
Energy of input fuel [MW]	115	115	115
Generator output [MW]	33.03	33.06	32.26
Heat duty, FG cond. [MW]	18.02	17.54	16.72
Heat duty, DH cond. [MW]	74.26	74.45	74.49
Heat gained in DH stream [MW]	92.28	91.99	91.21

The generator output of the present work, shown in Table 12, was calculated by multiplying the turbine output of the validation base case simulation, which was 32.89 MW, with a generator efficiency of 0.981.

As seen in Table 12, a bit less electricity and heat were produced in the simulation of the present work than in Björnsson et al. Since both are less this could mean that some global setting in the present work has been misinterpreted, leading to lower energy outputs. In general however, the values of the base case simulation are close to the values of Björnsson et al., indicating a trustworthy simulation. Since the CLC cases were built on this simulation, this means that all simulation parts that are not the air and fuel reactor can be expected to have realistic physical conditions.

Verification of Swedish CLC case compared to previous CLC simulation

Experimental data of large scale CLC plants are not available. This means that for the CLC cases, the simulations could not be validated against real data. However, a previous simulation of a CLC case of 30 MW in Sweden was available [11]. To compare, a version of the Swedish CLC case of the present work was adjusted to also be of 30 MW. The comparison is shown in Table 13:

Table 13: Validation of the Swedish CLC case of 30 MW, compared to a previous simulation of a CLC plant with ilmenite at 30 MW [11].

	Surywanshi et al. [11]	Present work
Turbine output [MW]	7.59	7.22
Generator output (98.1% efficiency) [MW]	7.44	7.09
Air compressor consumption [MW]	0.13	0.12
Pumps consumption [MW]	0.11	1.33
CO_2 compressor consumption [MW]	0.87	2.45
Total power consumption [MW]	1.11	3.89
Net electricity output [MW]	6.33	3.19
DH energy [MW]	22.71	20.76
Net energy output [MW]	29.04	22.95
CO_2 generated in FR [kg/s]	2.30	2.49
CO_2 captured [kg/s]	2.30	2.49

As can be observed in Table 13, most values are in a similar range in the present work compared to the previous CLC simulation. The major differences are the power consumption of the pumps and CO_2 compression, which leads to a lower net electricity output for the present work.

4.2 Techno-economic analysis of the cases

In Table 14 the collected data from the three main simulations, that were later used in the techno-economic analysis, are presented:

Table 14: The collected data from the simulations, for the cases at 100 MW.

	Swedish CHP plant without CO_2 capture	**Swedish CLC plant**	**Canadian CLC plant**
Net electric output [MW]	23.72	23.32	37.62
Heat output [MW]	79.15	27.31	0
Generated CO_2 [tCO_2 / year]	163 649.16	163 649.16	122 573.19
Captured CO_2 [tCO_2 / year]	0	163 649.159	122 573.162
Emitted CO_2 [tCO_2 / year]	163 649.16	0.001	0.028
OC usage [tonnes / year]	0	22 896	22 896
Dry fuel usage [tonnes / year]	97 308	97 308	72 887

Table 14 highlights that the CLC cases had lower net energy outputs than the CHP plant but that more than 99 % of all CO_2 emissions generated were captured, in the CLC scenarios. The combustion was complete in the CHP plant without CO_2 capture. Since it was assumed that an oxygen polishing step was included in the CLC scenarios, the combustion was assumed to be complete there too, which has been adjusted in the table shown above (although this was not included in the simulation). However, as can be seen, a lower level of CO_2 was generated in the coal based case. This was because less fuel was used in the Canadian CLC case, since the amount was selected based on the higher LHV (i.e., higher energy content per mass) of coal, in order for it to represent an amount of fuel that is equal to 100 MW. This had the consequence that less CO_2 was generated in that scenario. In Table 15, the results of the techno-economic analysis are seen:

Table 15: The estimated cost metrics, for the cases at 100 MW.

	Swedish CHP plant without CO_2 capture	**Swedish CLC plant**	**Canadian CLC plant**
Equipment cost [M€]	67.88	77.57	86.52
CAPEX [M€]	230.95	263.93	294.37
Fixed OPEX [M€/yr]	6.93	7.92	8.83
Cost of utilities [M€/yr]	3.46	3.96	2.94
Fuel cost [M€/yr]	8.74	8.74	1.54
OC cost [M€/yr]	0	7.30	7.30
Sand cost [M€/yr]	1.75	0	0
Oxygen polishing cost [M€/yr]	0	1.06	0.80
Tot. OPEX [M€/yr]	26.54	30.62	23.25
LCOE [€/kWh]	0.0899	0.2098	0.2603
Annual revenue [M€/yr]	66.79	53.67	30.43
Annual profit [M€/yr]	40.25	23.05	7.18
CO_2 seq. cost[M€/yr]	0	1.64	1.84
Carbon tax [M€/yr]	5.66	2.11E-8	1.72E-6
Incentive for CO_2 capture [M€/yr]	0	14.99	7.46
NPV [M€]	173.86	-37.05	-231.74
IRR [%]	17.21	7.20	-
PBP [yrs]	5.74	11.45	41.02
Electr./fuel [kWh/tonne]	717.9	705.7	2127.7

Table 15 shows the key comparison of this work. As can be seen, the equipment costs and therefore the CAPEX of the CLC scenarios were higher than that of the CHP plant, which is due to the assumption of the AR and FR reactors. Both CAPEX and the fixed OPEX were observed to be highest for the coal-based plant, which can be explained by the use of a larger turbine. This led to a higher CAPEX, which in turn induced a higher fixed OPEX since this was based on CAPEX in the economic assumptions. The total OPEX for the coal based case was however lower, which makes sense as combustion of biomass is a more complex and expensive process than coal combustion. However, the revenue for the Canadian case was so low that the profit was considerably smaller than for the other cases.

A low LCOE is typically good since this indicates that an investment is likely to become profitable even without a very high electricity selling price. As can be observed, it was the lowest for the Swedish CHP plant and the highest for the Canadian CLC plant. LCOE of this work should however be viewed with caution, since district heating was also involved in the two Swedish cases. This was accounted for by calculating the electricity fraction of the total output energy, but this may not be a completely representative way to consider only the electricity.

The CO_2 sequestration cost was higher in Canada than in Sweden due to the longer transportation costs. The carbon tax was significant in the CHP plant but was so small it can be considered negligible in the CLC cases, which is because almost 100% of the generated CO_2 was captured in those cases. The gains from incentives for CO_2 capture were higher in the Swedish CLC case for two reasons: because the incentives were higher than in Canada and because more emissions were generated and thus more was captured.

The NPV of the Swedish CHP plant was the highest, which was expected. The critical assumptions that lead to this will be discussed in the sensitivity analysis. As can be observed, the NPV of both the CLC scenarios were negative, while the case without carbon capture had a positive NPV. The interpretation of this is that in the current conditions, CLC is not a good investment in either country. However, the prospect of CLC looks considerably brighter in Sweden than Canada.

A high IRR and low PBP can indicate that there are strong chances of profitability and good performance, in which the Swedish CHP case is again superior. As can be observed though, the electricity per mass unit of fuel is by far much higher for the coal than for the biomass. This means that there is potential for optimizing the electricity production in the Canadian CLC plant in order to gain more revenue. It makes sense that the yield is higher for the case with coal, as biomass is less easy to deal with due to its complexity. The electricity yield for the two Swedish cases are similar to each other, which was expected. The discrepancy also makes sense as the addition of CO_2 capture requires extra electricity which lowers the yield.

The electric and heat efficiencies compared to the literature

Table 16 shows the electric and heat efficiencies of the simulations at 100 MW, calculated by using the net electricity outputs and heat outputs shown in Table 14 and dividing them by the energy of the input fuel, which is 100 MW:

Table 16: The electric and heat efficiencies, for the cases at 100 MW.

	Swedish CHP plant without CO_2 capture	**Swedish CLC plant**	**Canadian CLC plant**
η_{el}	0.24	0.23	0.376
η_{DH}	0.79	0.27	0

For the base case, which is a CHP plant without CO_2 capture, the electric efficiency η_{el} was expected to be around 0.2872 and the district heating efficiency η_{DH} was expected to be 0.8024 [11]. For the Swedish CLC case, where bark was also used, the electric efficiency should likely be lower, since the addition of a carbon capture technology requires energy. A reasonable value may be around 0.21 [11]. For the Canadian CLC case, coal is used as the input fuel. Since coal has more suitable properties for combustion than biomass, the electric efficiency for this plant is expected to be higher. In a previous study that looked at a coal fired chemical looping combustion system the electrical efficiency was 0.42 [84].

As is observed in Table 16, both efficiencies are close to the literature values for the CHP plant. For the Swedish CLC and Canadian CLC cases, the electric efficiencies are also close to the literature values. η_{DH} for the Swedish CLC plant is lower than for the Swedish CHP plant, which may be because less heat is generated from the fuel reactor and from the CFB boiler in the CHP plant. In the Canadian CLC case, η_{DH} is 0 since district heating was not included.

Consideration of the equipment costs

The total equipment costs of the CLC cases were both higher than that of the Swedish CHP plant. Table 17 shows the costs of the individual pieces of equipment for the cases at 100 MW:

Table 17: Equipment costs of the major pieces of equipment, for the cases at 100 MW.

	Swedish CHP plant without CO_2 capture	**Swedish CLC plant**	**Canadian CLC plant**
Pump 1 (water) [M€]	0.040	0.053	0.044
Pump 2 (DH water) [M€]	0.030	0.026	0
Compr. 1 (air) [M€]	0.61	0.98	0.97
Compr. 2 (CO_2) [M€]	0	2.97	7.36
CFB boiler [M€]	44.57	0	0
AR + FR [M€]	0	54.18	48.97
HX 1 [M€]	3.47	2.35	2.53
HX 2 (FG cond) [M€]	0.72	0.15	0
HX 3 (DH cond) [M€]	2.35	0.92	0
Steam turbine [M€]	16.09	15.94	26.65
Total [M€]	66.88	77.57	86.52

Table 17 shows that the CFB boiler of the CHP plant had a cost of €44.57 million, which is 65.7% of the total cost, while the air and fuel reactors of the CLC cases had costs around €50 million, which is 55-70% of the total cost. This was considered reasonable, based on the complexity of the CLC equipment that is required [85]. It can also be observed that the steam turbine of the Canadian CLC case was considerably more expensive than the steam turbine of the Swedish cases. This was because it produced much more electricity, and the equipment cost is estimated based on capacity. So high electricity generation, which is good for economic feasibility, also means increased CAPEX, which is bad for economic feasibility.

Sensitivity analysis of key parameters in the simulations

A sensitivity analysis was done to see how changes in key input variables in the simulations affected the results, to identify critical factors. In the simulations, key parameters were considered to be the conditions of the input streams. For these, the mass flow, temperature and pressure - and for the fuel, also the amount of moisture - were varied.

- Largest impacts on: electricity generation

  For the turbine output, increased mass flow and decreased moisture of the fuel as well as increased air flow generally increased the turbine output. Another observation was that if the mass flow of the cold incoming district heating water stream was increased, the turbine output was decreased.

- Largest impacts on: heat generation

  Regarding impacts on the district heating energy in the Swedish cases, increased flow of cold district heating water resulted in more heat transfer from the hot flue gas, which lead to a higher heat output.

- Largest impacts on: generated CO_2

  In the CHP cases, the factors that increased the CO_2 emissions the most were: increased air flow and pressure. In the CLC cases however, the air did not have the same impact. In those cases, the factors that increased the emissions the most were: increased fuel and OC usage, which is reasonable since these actions lead to that more can be burned.

- Largest impacts on: captured CO_2

  The amount of captured CO_2 in the CLC cases was most dependent on mass flow and moisture content of the fuel, which was because those factors determined how much CO_2 was generated. If more was generated, more was also captured.

The sensitivity analysis of the simulation parameters can give an idea of what parameters to adjust to increase performance. However, even though it is possible to increase the temperature or mass flow by 100 times in the simulation, it may not be physically realistic. Therefore, all streams need to be carefully considered when making such a change.

It was tested to change the ambient temperature to very high and low temperatures but this proved to have negligible impact on the performance of any of the simulations.

Sensitivity analysis of key economic parameters

There may be a discrepancy between what is commonly believed to be the most important factor in achieving CLC commercialization and what is actually most important. For example, the cost of OCs have had a large focus in previous CLC research - but is a cheap OC really the factor that would make commercialization possible?

Figure 8-Figure 13 show the impact factor (calculated as explained in subsection 3.4) that different economic parameters had on the following economic metrics: OPEX, annual profit, annual revenue, LCOE, NPV and PBP, for the cases of 100 MW. A high impact factor means that the parameter had a strong influence on the economic metric.

Figure 8, shows the parameter impacts on the total annual OPEX:

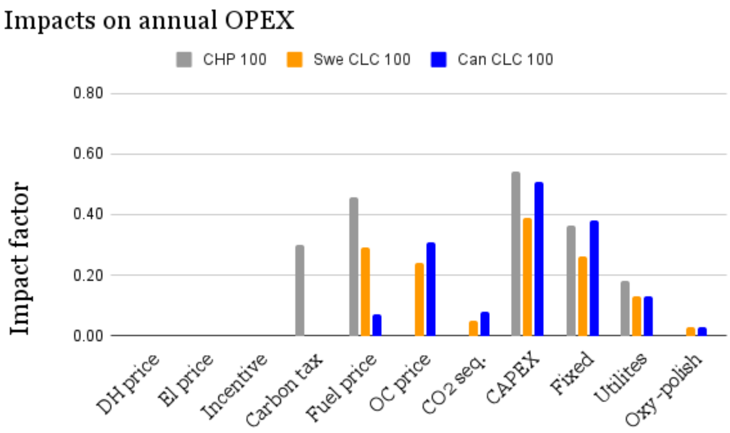

Figure 8: Parameter impacts on annual OPEX, for the cases of 100 MW.

As can be discovered in Figure 8, CAPEX had a big influence, especially in the CHP case. For the biomass-based cases, the fuel price had a lot of influence, while the CLC cases had in common that the OC price was important for the OPEX. In Figure 9, the parameter impacts on the annual revenue are displayed:

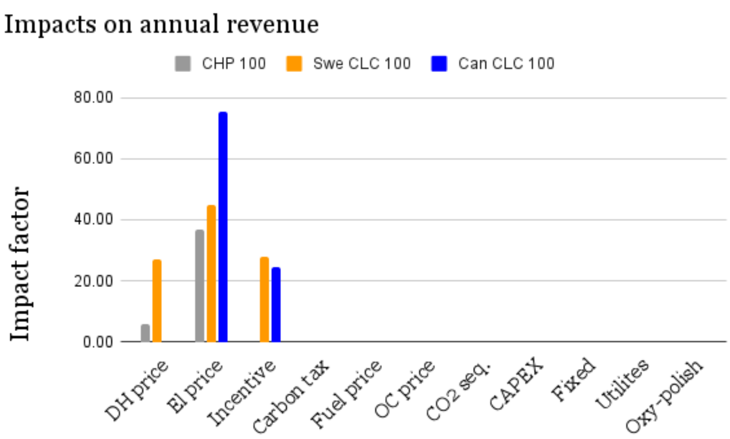

Figure 9: Parameter impacts on annual revenue, for the cases of 100 MW.

It is evident from Figure 9 that the electricity price was the most important factor impacting the revenue. When the OPEX is subtracted from the revenue, the profit is obtained. The results for the annual profit can be seen in Figure 10:

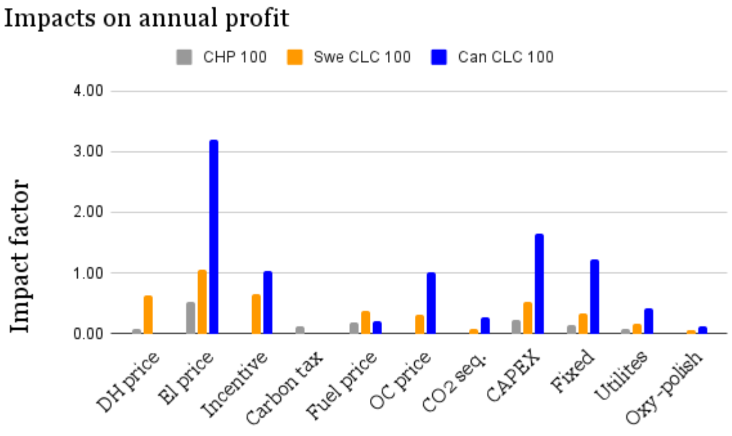

Figure 10: Parameter impacts on annual profit, for the cases of 100 MW.

What is observed in Figure 10, is that the profit of the Canadian CLC case was more sensitive to the parameters than the Swedish cases. This means that in the Canadian coal-based scenario, an increased electricity selling cost, OC price, or incentive for CO_2 capture could have a big influence on the feasibility of the technology. In the Swedish biomass-based cases, a big change in any single parameter would not be as critical to the total annual profit. The electricity price was the most significant parameter for all the cases, which aligns with that it was the most important for the revenue. In Figure 11, the parameter impacts on LCOE are shown:

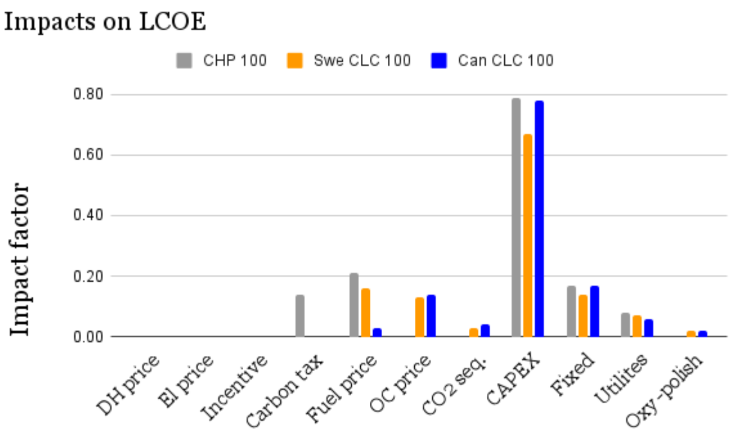

Figure 11: Parameter impacts on LCOE, for the cases of 100 MW.

The levelized cost of electricity can be interpreted as the selling price that electricity needs to have in order to break even on an investment, where a low LCOE is good since it means that the investment is less dependent on the electricity price. In the LCOE definition (Equation 12), both CAPEX and OPEX are included in the nominator. As can be seen, CAPEX has the by far most significant impact on LCOE. In Figure 11, the fuel price, OC price, cost of sequestration, the fixed OPEX and utilities are all part of the total annual OPEX. Even though a large change in OPEX would likely impact LCOE significantly, the impact of each singular parameter such as the OC price is less important by itself. Also, since OPEX is generally smaller than CAPEX it makes sense that it has less impact. Figure 12 displays the parameter impacts on the NPV:

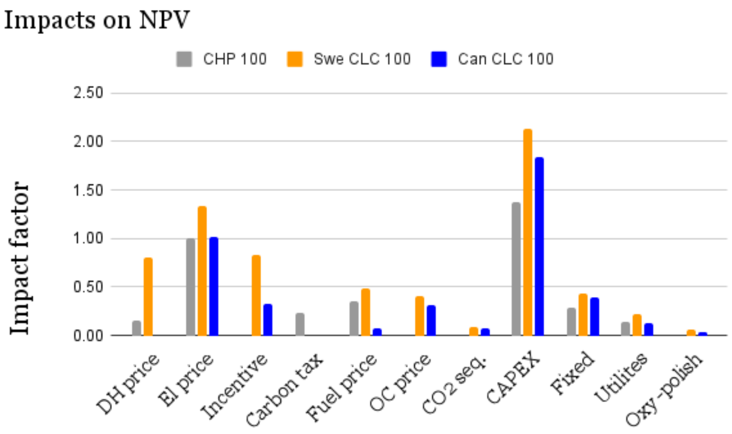

Figure 12: Parameter impacts on NPV, for the cases of 100 MW.

As can be seen in Figure 12, CAPEX and the electricity cost were of most importance for the NPV, not the OC. This graph displays that there was larger sensitivity in the Swedish CLC case for the NPV. Finally, Figure 13 shows the parameter impacts on the PBP:

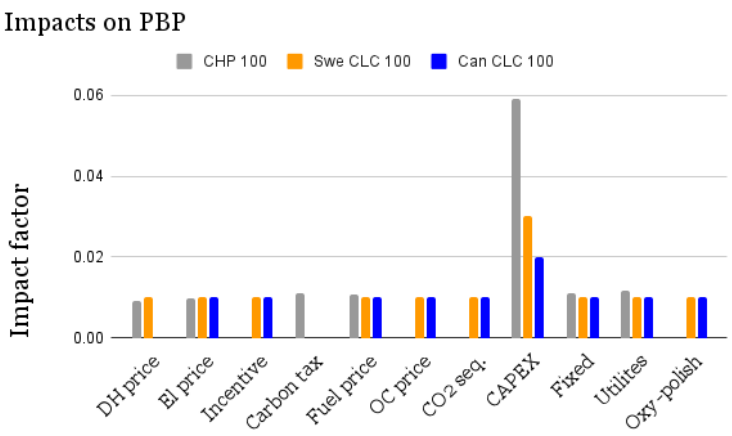

Figure 13: Parameter impacts on PBP, for the cases of 100 MW.

It was noticed that CAPEX had the largest impact on PBP for all cases, but especially for the Swedish cases. In summary, what the sensitivity analysis of the economic parameter impacts shows, is that even if the CLC scenarios had used an OC that was a 100 times cheaper this would not be sufficient to make it economically feasible. Instead, the CLC cases likely require improvement in several parameters to help push it to a positive NPV, where low price OCs are included, but where there are also lots of other examples such as: increased electricity capacity, increased electricity selling price, increased incentives for CO_2 capture and decreased equipment costs, so that CAPEX is decreased.

How politics plays a role in CLC commercialization

As was seen in Table 15, the NPV of the Canadian CLC case was estimated to be €-231.74 million, while the Swedish CLC case had a value of €-37.05 million. The sensitivity analysis of the economic parameters showed that there are many different measures that could impact the NPV of the CLC scenarios (as seen in Figure 12). However, while there are lots of things to do to improve the technology, like getting the fuels or the oxygen carriers to perform better at lower costs, political decisions can also impact the commercialization potential considerably. Figure 14 shows how an increased CO_2 capture incentive in Canada could increase the NPV of CLC to become positive:

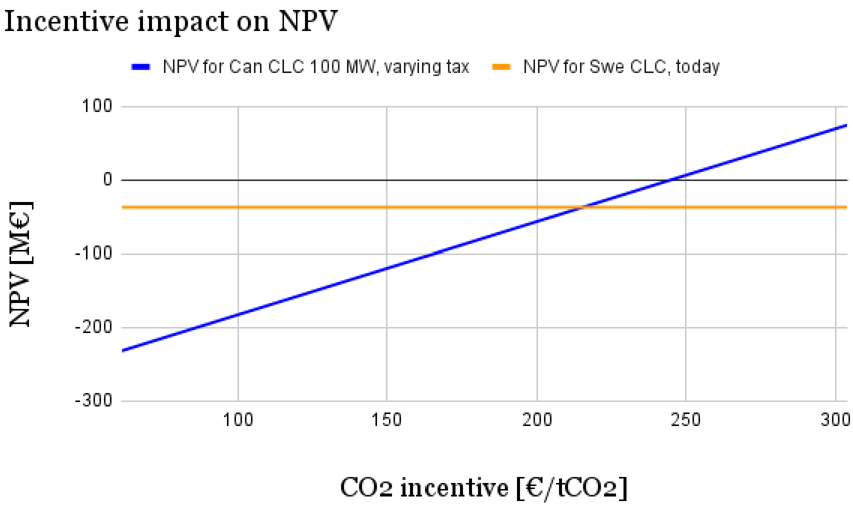

Figure 14: Incentive impact on NPV in Canada.

If the incentive for carbon capture would be increased by 3.6 times (to €220/tCO_2, which is a lot higher than the incentive available in Sweden), the NPV of the Canadian CLC scenario would become equal to the Swedish NPV. For the NPV to be positive however, the incentive would need to be increased more than 4 times, to around €243/tCO_2. That this would happen is not realistic. This shows that the political decisions regarding the incentives for CO_2 capture is not enough to make CLC possible in Canada. The incentives could also play a role in making CLC more viable in Sweden, as is shown in Figure 15:

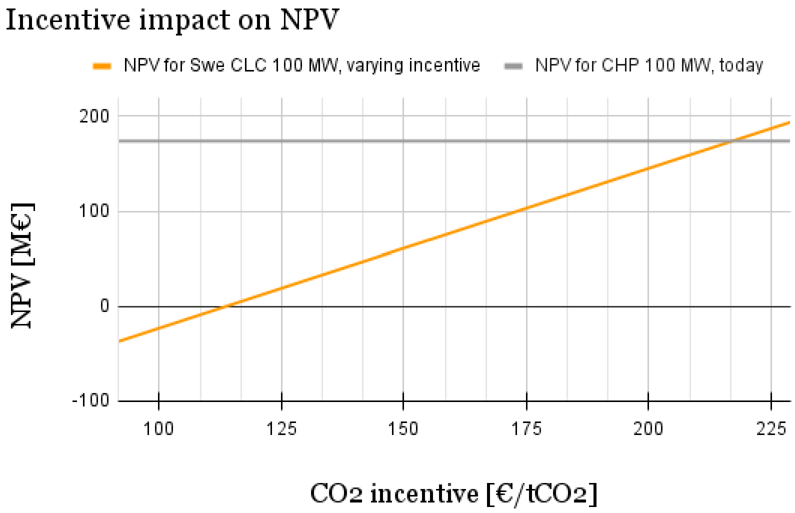

Figure 15: Incentive impact on NPV in Sweden.

Figure 15 shows that in order for the Swedish CLC case to have a positive NPV, an increase of 1.2 times would be required in the incentive, to €110/tCO_2. In order to reach a higher NPV than the Swedish CHP case without CO_2 capture, the incentive would need to be increased 2.4 times, to around €220/tCO_2. Another important political tool is taxes for carbon emissions. How carbon tax can decrease the NPV of the CHP case without CO_2 capture in Sweden was investigated, as is shown in Figure 16:

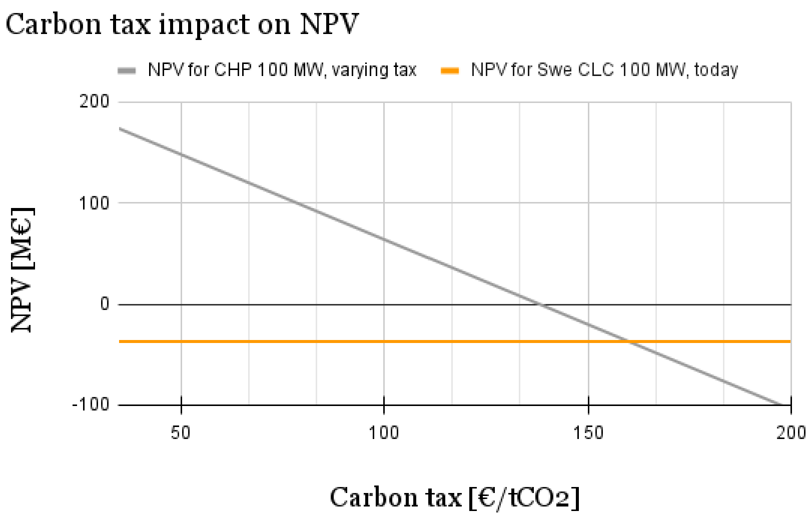

Figure 16: Carbon tax impact on NPV in Sweden.

From Figure 16, it is evident that the carbon tax would need to be increased 4.6 times (to approximately €160/tCO_2) for the NPV of the case without CO_2 capture to be lower than for the Swedish CLC scenario.

The taxes in Canada may also play an important role for CLC commercialization. However, within the scope of this work, it was difficult to test what impact higher taxes would have. There was no simulation of a Canadian CHP plant without CO_2 capture and the carbon tax was negligible in the CLC cases since nearly all was captured. Otherwise, it would have been reasonable to also test increasing the taxes to the point where it would no longer be economically justifiable to have a plant without CO_2 capture, in Canada.

Other ways the NPV of the Canadian CLC case could be increased

In all the scenarios, the amount of input fuel was decided based on the energy content per mass. For this reason, in the Canadian CLC scenario where coal was the fuel, a lower mass flow of coal was used than the mass flow of biomass used in the Swedish cases. Both coal and waste biomass can be considered cheap fuels and both cases used ilmenite as the OC. Still, since so much less coal was used, OPEX was significantly lower for the coal based scenario. As can be understood, the amount of fuel used in the Canadian CLC case could be increased a lot before the annual fuel costs would be nearly as expensive as in the biomass based cases. The electricity yield is about 3 times higher for coal than biomass, so if the Canadian scenario used more fuel, more electricity could likely be produced, which would provide more profits. Therefore, the Canadian CLC case could easily become more economically justifiable.

However, the more electricity that the Canadian CLC plant produces, the more expensive the turbine will become (at least with the current method for equipment costs), which would mean a major increase in CAPEX. So there is a fine balance between high electricity generation and increased CAPEX.

Should coal really be phased out?

In Canada, Sweden and many other countries around the world, fossil fuels are being phased out [41]. This likely has two major reasons: that combustion of fossil fuels leads to very high emissions, and that it is considered unsafe to rely on fossil fuels since they can run out, making the use of them unsustainable. Renewable fuels and a circular economy is the solution. However, the amount of renewable fuels, such as trees, are not unlimited on the planet even though they are renewable. It will be a massive challenge to to rely on biomass for *both* food and fuels.

It is commonly known that combustion of coal *does* lead to large amounts of CO_2 emissions compared to combustion of other fuels. However, the number of years until the coal deposits have run out (if the amount of coal usage would remain the same as it is now) was estimated to be 139 years in 2020 [86]. The coal deposits in the world are considerably more abundant than oil and natural gas deposits. The reason coal is being phased out faster than the other fuels in the Western economies is not because the stock is nearly run out. Rather, it is being phased out first due to its major contribution to global warming. However, with a technology like CLC, the capture efficiency is nearly 100%. Thus, it is a fuel that is very abundant compared to other fossil fuels, that can be combusted with very low emissions, if investments were made in this field. However, mining of coal likely also leads high to emissions, which cannot be captured directly by CLC and which was not considered in the calculations of this work. Also, 139 years is not infinite, so to rely on coal is not wise in the long run but could only be a temporary solution to slow down global warming while long term solutions are being figured out.

5 Conclusion

The simulations showed that the Swedish and Canadian CLC cases of 100 MW produced only 35-50% of the net output energy compared to what the Swedish CHP case of 100 MW produced. However, the CLC cases emitted nearly no emissions to the atmosphere, since the capture efficiency was near 100%.

When comparing the cases at 100 MW (Swedish CHP, Swedish CLC and Canadian CLC), the results showed that the Swedish CHP plant had the highest NPV at €173.86 million, compared to -€37.05 million for the Swedish CLC case and -€231.74 million for the Canadian CLC. The PBP was also the shortest for the Swedish CHP scenario at 5.74 years (compared to 11.45 and 41.02 for the Swedish CLC and Canadian CLC). The sensitivity analysis showed that CAPEX and the electricity selling price generally had the largest impacts on NPV while CAPEX alone had the highest impact on PBP. OC and fuel costs were minor in comparison.

The results of this work indicate that the potential for CLC commercialization is significantly better in Sweden than Canada for multiple reasons. A major reason was that there were more ways to gain revenue. In the Canadian case, revenue from selling electricity was the main income. In Sweden, which is a country with shorter distances and higher population density, district heating is common - which can massively increase the commercialization potential. The incentives for CO_2 capture and the taxes for CO_2 emissions in Sweden are also among the highest in the world. The ambient temperature was insignificant for the CLC potential.

If the incentives for carbon capture in Canada would be increased by approximately 4 times, the NPV of the Canadian CLC case would become positive. However, it is evident that increased incentives alone are not sufficient to make CLC possible in Canada. Increased fuel use to generate more electricity is another option (since the fuel is cheap and more could be used), but that comes with the trade-off against a more expensive turbine, which has a massive impact on CAPEX.

In Sweden, either the incentives would need to be increased 1.2 times to reach a positive NPV, or the carbon tax would need to be increased 4.6 times, in order for the NPV for the Swedish CLC case to be higher than the Swedish CHP plant without CO_2 capture.

The Aspen Plus simulations were judged to be satisfactory as they aligned well with the literature. However, there were larger uncertainties regarding the economic assumptions. As could be observed from the sensitivity analysis, important metrics like NPV and PBP were strongly influenced by the CAPEX. Since real equipment cost data for air and fuel reactors on large scale don't exist, assumptions based on the literature, that were difficult to verify, were used.

6 Suggested future outlook

For future works it would be interesting to compare a Canadian coal based CLC plant with a Canadian coal based plant without CO_2 capture, to be able to investigate how the political tool of taxes for carbon emissions could influence the CLC potential in Canada. The CLC simulation of the Canadian case in this work could also have been optimized. It would have been interesting to investigate how an increased amount of fuel could be balanced against an increased CAPEX (as a larger turbine would be required), to find the optimum capacity for the Canadian CLC plant.

Since the economic data regarding air and fuel reactors are so limited, a thorough review to investigate different cost estimations for air and fuel reactors in past works would be an interesting future outlook.

In the literature review of this work, it was observed that a lot of previous research has been directed towards polygeneration, which is the production of both heat/power *and* valuable products. Since CLC technology is so dependent on having multiple sources of revenue, the research in polygeneration is very justifiable and should be investigated even further.

Conflicts of interest

There were no conflicts of interest related to this work.

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Appendix

In figures A1-A3, the flowsheets for the three different types of simulations that were created in Aspen Plus can be seen:

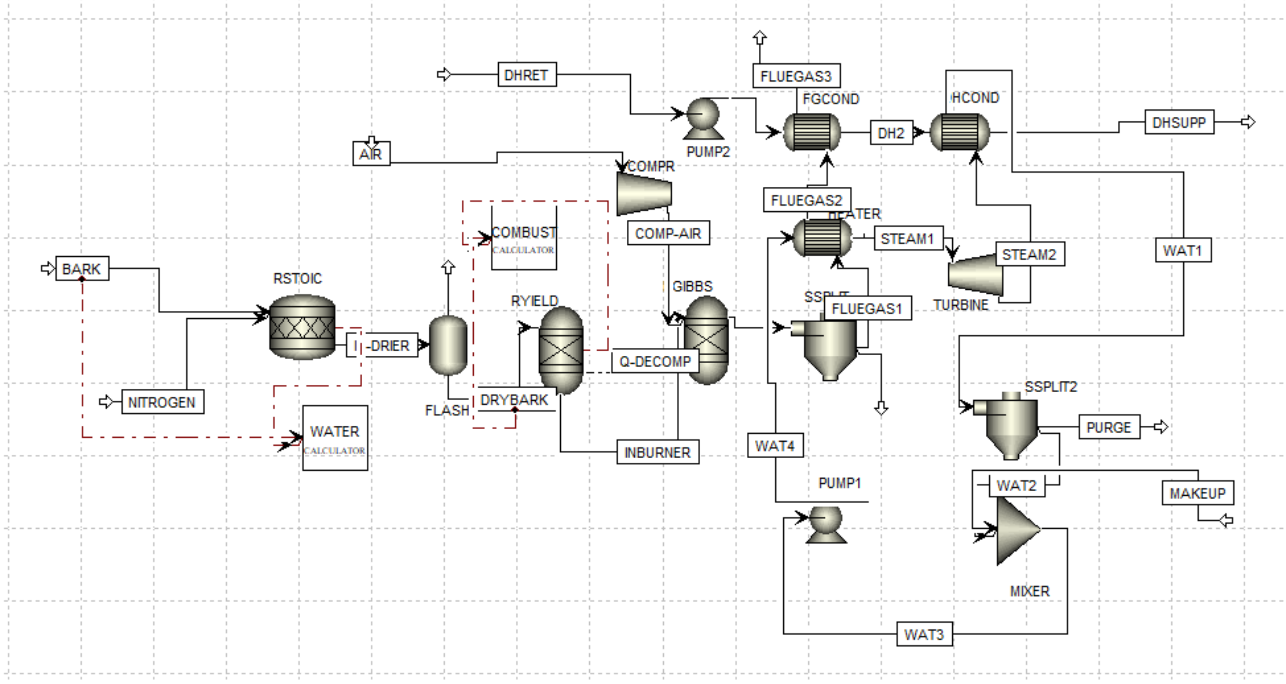

Figure A1. The Aspen flowsheet of the Swedish CHP simulation; the "base case".

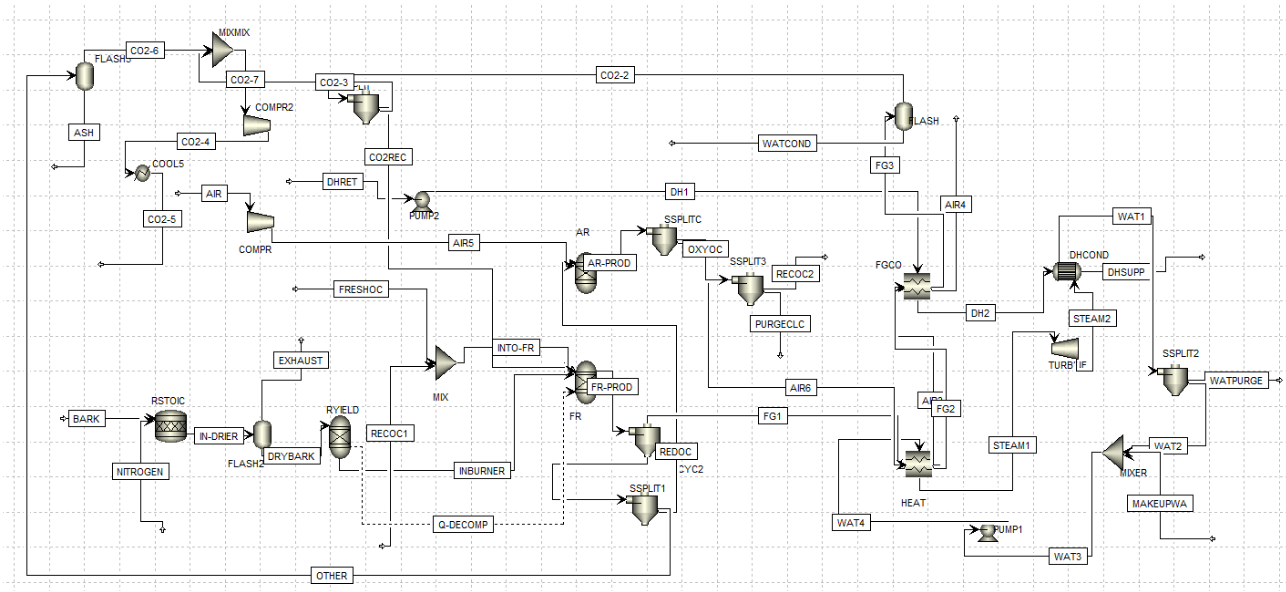

Figure A2. The Aspen flowsheet of the Swedish CLC simulation.

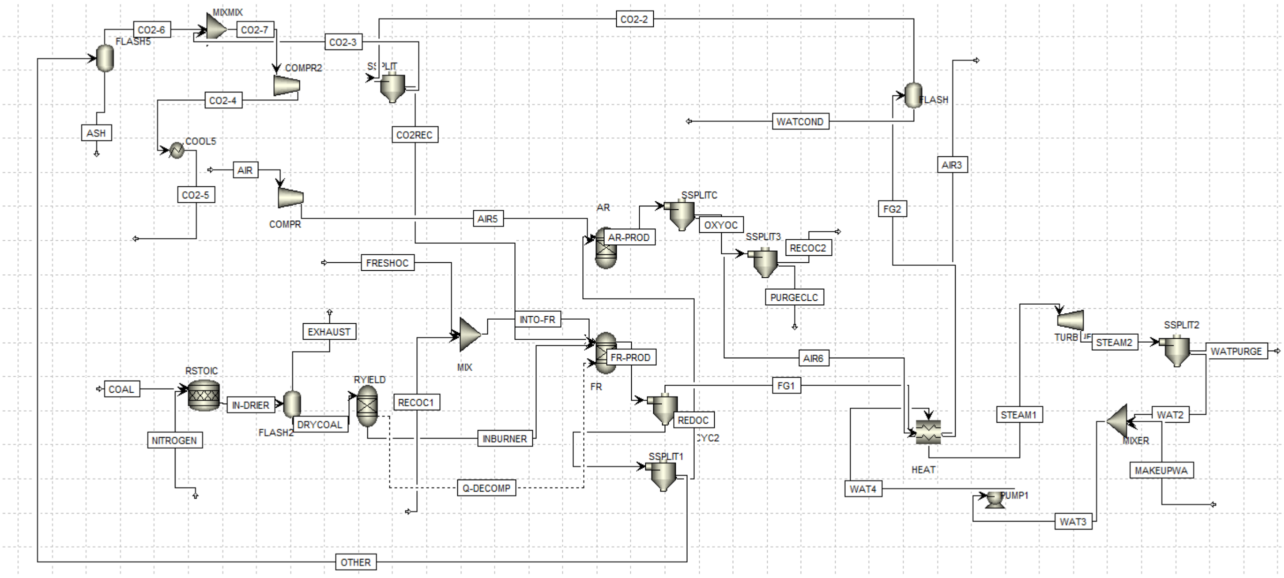

Figure A3. The Aspen flowsheet of the Canadian CLC simulation.